Section 3  Optics and Devices

Chapter 1  Optics and Quantum Electronics

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Chapter 3  High-Frequency (> 100 GHz) and High-Speed (< 10 ps) Electronic Devices
Chapter 1. Optics and Quantum Electronics

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1.1 Modelocking of Fiber Lasers

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In our laboratory, we have pursued successfully the generation of modelocked pulse trains from additively pulse modelocked (APM) fiber lasers. The polarization in the fiber is made elliptical, and rotation of the polarization ellipse via the Kerr nonlinearity, followed by a polarizer, can produce intensity dependent loss. Effective saturable absorber action is produced resulting in the generation of mode-locked trains of pulses without the use of any other modulation mechanism. This principle has been demonstrated both in fiber rings supporting solitons, where the fiber in the ring has negative dispersion; and also in fiber rings with almost balanced positive and negative dispersive fiber segments. The latter led to so-called stretched pulse operation. Stretched pulse lasers can deliver up to two orders of magnitude larger output powers than the soliton ring lasers, because the fiber nonlinearity is self-limiting: as the pulses get shorter they experience

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greater stretching, and their average peak intensity is reduced.

The stretched pulse operation has been perfected to a degree that a prototype of a commercial model was built at MIT and delivered to Clark-MXR, Inc., which has commercialized the MIT design.

Active, synchronous modelocking of a fiber ring laser can produce pulses much shorter than those predicted by the Siegman-Kuizenga theory. This is possible when the pulses are soliton-like, shaped by group velocity dispersion and the Kerr nonlinearity of the fiber. We have checked experimentally our theoretical predictions of a stability theory that compares the gain required by the standard modelocked pulses of the Siegman Kuizenga theory, extended to include group velocity dispersion, with the gain required by the filtered solitons. As long as the latter is smaller than the former, the soliton pulses are stable. The experimental results confirmed this simple theory. Pulses four times shorter than those predicted by the Siegman-Kuizenga theory were observed. This should be compared with the theory of Haus and Silberberg, which, neglecting dispersion, predicted pulses only about two times shorter.

In 1994, asynchronous harmonic modelocking of a fiber ring laser supporting solitons was discovered. The system required no stabilization of the fiber ring, contrary to schemes that use a modelocking drive that is synchronous with a multiple of the roundtrip frequency of the fiber ring. A theory has been developed for the range of detuning over which a stable pulse stream can be obtained. There is a lower limit on the detuning frequency set by capture of the pulse repetition frequency by the modelocking frequency. In addition, there is a maximum allowed detuning set by the inability of the modelocking drive to affect the pulse stream. The larger the dispersion in the ring, the larger the allowed detuning, since the pulse-stream can change its roundtrip frequency by changing the carrier frequency.

In the study of nonlinear fiber loops used as intensity sensitive reflectors or transmitters, William S. Wong has developed a suppressor of narrowband back-ground radiation in a nonlinear fiber loop with positive and negative group velocity dispersion in the loop. Narrow-band radiation is unaffected by the dispersion, whereas short pulses of broad-bandwidth are affected. In this way, the loop can transmit pulses while suppressing the narrow-band background even if the latter is of comparable intensity.

1.2 Long-Distance Nonlinear RZ Communications

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The study of short pulse generation in fiber ring lasers is closely related to the problem of long distance pulse propagation, since the recirculation in a ring bears a close resemblance to unidirectional propagation along a fiber.

We have studied a possible interaction mechanism between solitons that has not received attention in the past: the effect of the radiation shed by one soliton as it gets amplified periodically, on the carrier frequency and the position of the solitons in the pulse stream. It was found that the interaction is of second order in the amplitude of the

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In joint work with a Bell Laboratories group, we have developed a supervisory control for soliton WDM communications. The use of solitons in long distance fiber communications is handicapped by three of its aspects: (1) the supervisory control sacrifices one of the WDM channels; (2) the lumped amplification of solitons leads to shedding of radiation that can cause instabilities, unless the amplifier spacing is of the order of 25 km (a smaller distance than for NRZ); and (3) the sliding guiding filters call for different amplifier-pods along the cable. The NRZ format has none of these three disadvantages. However, it has also been shown that the RZ format of pulse communications gives a 2.8 dB detection advantage as compared with the NRZ format.

We have initiated the study of a new nonlinear pulse propagation for long distance RZ communications. The idea evolved from the stretched pulse ring laser principle. The fiber consists of alternating positive and negative dispersion segments. The pulse propagating along this fiber stretch and compress alternately. It was found both analytically and numerically that the stretched pulses are less prone to shedding of radiation than the solitons. The amplifier spacing can thus be made larger than for solitons. Simulations have shown it can be made as large as 100 km for a bit-rate of 10 Gbit/s. Further, the average dispersion can be made smaller than fabrication tolerances allow for solitons by proper balancing the dispersion of the two segments. This reduces the Gordon-Haus effect. One may forego the sliding guiding filters, a measure that has two further advantages: (1) the amplifier pods can be all identical; and (2) the supervisory control need not sacrifice a WDM channel. Further, by maintaining the RZ format one can take advantage of the 2.8 dB detection advantage mentioned earlier.

It is also likely that the stretched pulse propagation scheme will find application in passive, end-pumped cables whose length is limited by nonlinear effects at the input. A soliton format has been proposed for alleviation of the nonlinearity at the cable input. The stretched pulse format may perform better.

1.3 Squeezing in Optical Fibers

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Squeezed quantum states of the electromagnetic field are states that have reduced fluctuations in one phase of the field at the expense of increased fluctuations in the quadrature phase. If employed in the measurement of either phase, or amplitude of an electromagnetic field, they lead to detection at sub-shot noise level. We have pursued the generation of squeezed states in fibers for one important reason: since squeezed states are very sensitive to loss, their use in a measurement has to be accomplished with minimum loss. Single mode fibers and their interconnections can be made with very small insertion losses. The squeezed radiation can be applied conveniently to fiber interferometers (e.g., fiber gyros) for reduced-noise phase measurements.

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Thus far, we have accomplished squeezing in a fiber Sagnac loop of our invention. The shot noise reduction achieved was 5.1 dB using gaussian pulses at the zero dispersion point of the fiber at 1.3 mm wavelength. For this purpose, a diode pumped modelocked Nd:YLF laser was constructed operating at 1 GHz repetition rate. The high repetition rate is required to prevent the guided acoustic wave Brillouin scattering from entering the spectral window of the detector. More recently, we have concentrated on squeezing in the negative dispersion regime to utilize the higher level squeezing predicted for solitons. The squeezing experiments using solitons at IBM Almaden and at MIT have not been particularly successful. We believe that the "soliton" lasers used as sources in these experiments had excessive noise. For this purpose, we have developed low noise fiber ring lasers. We have measured amplitude noise and pulse-to-pulse jitter. We found that the timing jitter in both laser systems was quantum limited; i.e., the amplified spontaneous emission was solely responsible for the jitter. The stretched pulse had smaller timing jitter, because its net group velocity dispersion could be adjusted to be smaller than that of the soliton laser. The pulse timing follows a random walk, the mean square displacement grows linearly with time. For the soliton laser we found a walkoff of 650 fs for a pulse-width of 250 fs in a time interval of 0.1 seconds. The amplitude noise was the lowest ever reported (0.2 percent), but of classical (environmental) origin. For the stretched pulse laser, less than 80 fs jitter, 0.05 percent amplitude noise, and a minimum pulse width of 85 fs. In order to understand the experimental results, a perturbation theory we developed.

1.4 Pulse-Excited Fiber Gyro

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Project Staff

Professor Hermann A. Haus, Professor Erich P. Ippen, Patrick C. Chou, Ravindra V. Dalal

We study the behavior of pulse excited fiber gyros in anticipation of their use with squeezed radiation. Limits are imposed on the maximum power that can be used in a fiber gyro by Brillouin scattering and Raman scattering. In such cases, the sensitivity can be improved by the use of squeezed radiation. Since the only practical way of producing squeezed radiation is through the use of pulses, the application calls for pulsed excitation of fiber gyros. Hence, it is necessary to explore the operation of fiber gyros under pulsed excitation.

Conventional excitation of Sagnac loop fiber gyros is via broad-band, incoherent cw radiation from an LED or the amplified spontaneous emission from a fiber amplifier. A short coherence time is required to reduce the effects of back scattering. Pulsed radiation can also provide a short coherence time. Scrambling of the phase among successive pulses via a phase modulator avoids the backscatter from all but the pulse pair at the symmetry point of the Sagnac loop. However, the question arises whether new nonlinear effects due to the high peak power of the pulses would introduce their own errors and noise. We have run experiments on a fiber gyro made available by Draper Laboratory designed to operate at 1.3 mm, and an updated version for 1.54 mm operation. We have shown that up to conventional average power levels, the signal to noise ratio of cw operation and pulsed

operation is comparable. One advantage of pulsed operation is that one may raise the power to levels that are not achievable with cw operation when the coherence time or length is to be maintained short (of the order of 100 μm coherence length). At higher power levels, we have a deterioration of the signal to noise ratio the cause of which is not yet determined.

In a related experiment, trying to eliminate these novel nonlinear effects, we spread the pulses by propagating them over 20 km of dispersive fiber. In this way, quasi-cw radiation was produced and the system behaved as expected when excited by cw radiation. Whereas this technique of pulse spreading may not be applicable to the use of squeezed radiation because of the unavoidable loss, it is an option for conventional fiber gyro operation. The use of coherent radiation allows center frequency control not possible with ASE. The spreading could be accomplished more conveniently by grating reflectors.

1.5 Optical Resonant Structures

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Project Staff

Professor Hermann A. Haus, Dr. Jay N. Damask, Mohammed J. Khan, Christina Manolatou

For several years, our research has focused on the design and realization of a resonator-based optical filter commonly called the channel-dropping filter. While we have focused much attention on the resonance structure of the quarter-wave shifted distributed Bragg reflector (QWS-DBR) gratings, only recently has attention been paid to the crosstalk characteristics of the filters outside the Bragg stopband. This year we developed this additional avenue of design to realize large improvements in overall filter performance and found a fundamental link between resonance filter linewidth and out-of-band crosstalk.

Figure 1 illustrates the channel-dropping filter as seen in previous RLE Progress Reports. A center bus waveguide lies between two side-coupled optical resonators. The resonators are constructed by etching QWS-DBR gratings on the surface of channel waveguides. The farthest right resonator is within its resonance bandwidth designed to reflect light back along the bus. The left-most resonator is designed to partially reflect, transmit, and pick-up light as it travels along the length of the resonator. The two resonators are separated along the bus by a net quarter-wave shift so that a resonant cavity is built up along the bus. This cavity acts to couple all of the light which excites the resonators from the bus to the upper tap-off waveguide.

The associated calculated spectral responses show the filter impulse response at the pick-up arm. On resonance there is full pick up. Off resonance but within the bandwidth of the Bragg gratings, there is little optical power transfer between the bus and left-most resonator waveguide. However, outside of the Bragg stopband, significant optical power is exchanged between the two waveguides, resulting in a high excess loss in transmission and a high crosstalk level at the tap-off waveguide.

There is a way of reducing the crosstalk. One degree of freedom is the shape of the bus waveguide as it couples to the resonators. The field envelope in the resonators is an exponential decay in either direction from the quarter-wave shift. If the coupling strength to the bus approximates this exponential decay, then it is reasonable to believe that the total coupling on resonance can remain the same while the coupling outside of the stopband, where normal evanescent waveguide-waveguide coupling exists, can be reduced.

Figure 2 illustrates an arc-shaped bus that tapers the coupling strength between the bus and
resonator. The sweep of the arc is about 15 degrees to either side of the quarter-wave shift. The optical loss associated with the bending of the bus waveguide is essentially zero by design. Care is taken to ensure a net quarter-wave phase shift is accrued along the bus between the two resonators. The calculated impulse response of the tapered-bus channel-dropping filter is shown below. There is significant improvement in the excess loss in transmission and crosstalk level at the pick-up port. Finally, it is important to recognize that the resonant linewidth is maintained between figures 1 and 2; in this way, a fair comparison of crosstalk reduction is made.

![Resonant Channel-Dropping Filter](image)

**Figure 2.** A resonant channel-dropping filter with an arc-shaped bus which threads through the two resonators. The shape of the bus creates a varying coupling strength which approximates the field envelope shape in the side-coupled resonators. The linewidth of the filter is maintained by moving the two resonators closer to the bus. The out-of-band crosstalk is reduced by the decrease in power exchange between the bus and resonator waveguides. However, there is a fundamental limit to the crosstalk reduction.

There is a limit to the reduction of crosstalk. One may consider reducing the radius of curvature of the arc-shape bus to gain in some proportion in crosstalk reduction. However, a fundamental limit exists. While the radius of curvature is reduced, in turn reducing the overlap integral between the bus and resonator outside of the Bragg stopband, the separation at closest proximity between the two guides must be decreased so as to maintain the linewidth of the resonance; put another way, the decrease in minimum separation as the radius is reduced is necessary to maintain the absolute level of power escape per cycle from the resonator to the bus. The mechanism of overlap-integral reduction via radius reduction is in competition with the requisite increase in peak coupling necessary to maintain the resonant linewidth.

We have shown, following the above argument, that the ratio of crosstalk levels between the straight- and arc-shaped bus designs cannot be reduced below \((kL)^2\), where the \(k\) is the field-envelope decay in the resonators and \(L\) is their length. This product, in turn, is inextricably associated with the resonant linewidth.

The importance of this limitation is illustrated with a few numbers. Consider a design where the crosstalk level is limited to -20 dB, and the best crosstalk reduction is available through proper bus design. The grating strength required to have a 20 GHz wide linewidth is about 1600 inverse cm. Such a grating strength is both unavailable today and poses many fabrication problems as well as poor optical performance. A more reasonable grating strength of 80 inverse cm yields only a 1 GHz filter linewidth. This filter is so narrow the number of relevant applications is potentially limited. However, on the other hand, given an application where ultranarrow band filters in compact form are essential, the resonant channel-dropping filter may perform well.

### 1.6 Efficient Frequency-doubling of a Stretched-pulse Fiber Laser

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**Project Staff**

Professor Erich P. Ippen, Lynn E. Nelson, Siegfried B. Fleischer, Gadi Lenz

As reported previously, stretched-pulse additive-pulse mode-locked (SP-APM) fiber lasers can produce ultrashort pulses (100 fsec) with pulse energies of over 2 nJ. One application of these high-power pulses is as a seed for Ti:Sapphire

regenerative amplifiers. The output pulses from the fiber laser at 1.55 μm are compressed and passed through a nonlinear crystal to generate pulses at 775 nm which can then seed the amplifier. This frequency-doubled SP-APM fiber laser is an inexpensive, compact seeding source for regenerative amplifiers and could replace the argon-pumped Ti:Sapphire laser. Transfer of the technology for this laser to Clark MXR, Inc., Dexter, Michigan, took place in 1996; and a commercial product is now being manufactured and sold.

We have optimized the SP-APM laser for doubling efficiency and frequency-doubled pulse width, rather than for increasing the fundamental pulse width and quality. With the laser configuration shown in Nelson et al.\(^\text{24}\) the net dispersion of the SP-APM laser was + 0.13 ps\(^2\) and the 1.7 m-long piece of erbium-doped fiber was backward pumped. We obtained average output powers of > 95 mW at 31.8 MHz and compressed the highly stretched ~ 1 psec pulses to 105 fs with 90 percent transmission through a sequence of four silicon Brewster prisms. The pulse spectrum was centered at 1552 nm and had an approximate width of 67 nm. Grating compression produced fundamental pulse widths as short as 70 fs, but due to the lower efficiency of gratings (~ 30 percent) frequency doubling was undertaken with prism compression.

By focusing the compressed fundamental pulses onto a 1 cm-long AR-coated BaBO\(_4\) (BBO) crystal, frequency-doubled powers as high as 8.7 mW were achieved, corresponding to 10 percent conversion efficiency and pulse energies of up to 270 pJ. The frequency-doubled pulse width was 86 fs and the spectrum was 7.3 nm wide centered at 771 nm, resulting in a time-bandwidth product of 0.32, which is near transform-limited, assuming secant hyperbolic pulse shapes. These frequency-doubled pulse pulse energies are sufficient to seed high-repetition-rate regenerative amplifiers and avoid amplified spontaneous emission background.\(^\text{25}\)

Frequency doubling was also performed with a 1.5 mm KNBO\(_3\) (potassium niobate) crystal and with a 7 mm LiB\(_3\)O\(_5\) (LBO) crystal. Potassium niobate has a nonlinear coefficient which is two orders of magnitude larger than BBO, and we obtained conversion efficiency of 1.5 percent with this short crystal. We obtained 6 percent conversion efficiency with LBO, which is advantageous because it can be temperature tuned for 90-degree phase matching with no spatial walk-off. In both crystals, though, the frequency-doubled pulse widths were > 170 fs, indicating that although high efficiencies were obtained with short crystals, the smaller phase-matching bandwidth of these crystals would not allow efficient doubling of 100 fs pulses.

Frequency-resolved optical gating (FROG),\(^\text{26}\) which allows the direct determination of the intensity and phase of an ultrashort pulse, was used to better characterize both the fundamental and frequency-doubled pulses and to explore why the doubled pulse spectrum was offset from the center of the fundamental spectrum. FROG data on the 1.55 μm pulse was taken by measuring the spectrum of the second harmonic generation (SHG) as the pulse delay was changed in a background-free autocorrelation configuration. Figure 3 shows the measured SHG FROG trace of the 1.55 μm pulse along with the calculated fit. The FROG trace indicates that the lower-frequency components are contained in the pulse wings; thus efficient frequency doubling occurs at shorter wavelengths. This explains why the SHG spectrum is centered at 771 rather than 776 nm, which would be predicted based on the first moment of the fundamental spectrum. We confirmed that our FROG data accurately represented the pulse by comparing the experimental 1.55 μm spectrum with the Fourier transform of the calculated pulse intensity and phase from the calculated FROG fit. As shown in figure 3, there was excellent agreement between the two spectra, indicating that no filtering occurred in the pulse compression or nonlinear crystal.

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The frequency-doubled pulses were also characterized with an SHG FROG measurement. The FROG trace was round and smooth, indicating that the pulse was near transform-limited with flat phase across the entire pulse. Amplitude noise measurements were performed on the frequency-doubled pulses and compared with those of a commercial Ti:Sapphire laser pumped by an Argon-ion laser. The amplitude fluctuations on the frequency-doubled pulses rolled off more quickly and were at \(-138\) dBc/Hz at 100 kHz. The Ti:Sapphire noise is typically at least 40 dBc/Hz higher over the range 3 - 100 kHz.27

\[1.7\] **Wavelength Shifting in Passive InGaAsP/InP Quantum-well Waveguides**

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**Project Staff**

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We have developed an analytical theory for nonlinear conversion by four-wave mixing (FWM) in the presence of two-photon absorption, and demonstrated its predictions experimentally.28 Theoretically, we show that while nonlinear loss enhances small-signal FWM, it places a fundamental upper limit on the conversion efficiency. Expressions for the optimum input pump intensity and associated maximum conversion are obtained, and optimum waveguide lengths determined. Experimentally, we used picosecond pulses from two synchronized color-center laser for pump and input signals, tunable around 1560 nm. The waveguides were 7.5 mm-long, AR-coated, InGaAsP/InP ridge guides incorporating single quantum wells with a bandedge of 1490 nm. The two-photon absorption coefficient was measured by nonlinear transmission to be \(B = 23\) cm/GW, and a small-signal conversion efficiency of \(10^{-4}\) cm^4/MW^2 was obtained. A maximum conversion of about 10 percent was achieved with peak pump powers of about 300 W, or picosecond pulse energies of about 1 nJ. Figure 4 shows a plot of experimental conversion efficiency along with the theoretically predicted trace indicating limiting by two-photon absorption. New structures with multiple quantum wells for enhanced conversion, and p-n junctions for residual carrier sweep-out, are being designed and fabricated for future experiments.


1.8 Investigation of Air-Bridge-Waveguide Photonic Bandgap Structures

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National Science Foundation/MRSEC

Project Staff
Professor Erich P. Ippen, Professor Leslie A. Kolodziejski, Dr. Günter Steinmeyer, Constantine N. Tziligakis, Kuo-Yi Lim, Dr. Gale S. Petrich

Properly designed sub-micron dielectric structures offer the possibility of confining and controlling light in novel ways. Such structures, with photonic bandgap characteristics, are being designed by the groups of Professors John D. Joannopoulos and Hermann A. Haus and fabricated by the groups of Professors Lionel C. Kimerling, Leslie A. Kolodziejski, and L. Rafael Reif with dimensions appropriate to near-infrared wavelengths. Our group has been developing wavelength-tunable-femtosecond (WTF) techniques for the infrared and are employing them to investigate actual device characteristics. The first experiments are to characterize one-dimensional air-bridge photonic bandgap devices at wavelengths in the 3-5 micron regime. In these structures, an appropriate defect (absence of a hole) in the middle of the periodic structure creates a microcavity characterized by a high-Q resonance within the photonic bandgap.

The experimental setup used to study these structures takes advantage of the recent advances in femtosecond Ti:Sapphire laser and optical parametric oscillator (OPO) technologies. By synchronously-pumping an OPO with the femtosecond output of a Ti:Sapphire laser, we obtain two well-synchronized pulse trains with on the order of 100 mW of average power. These two trains, the signal and idler, are tunable in wavelength with respect to each other in the 1-2 micron range. Femtosecond pulses at longer wavelengths are then obtained by difference frequency generation in a nonlinear crystal. For our immediate purposes we have demonstrated wavelength conversion to the 3.5-5 micron regime with pulse durations of 200 fs and average powers of about 100 microwatts. Figure 5 shows spectra of pulses produced at three different wavelengths. The broad bandwidth inherent to the ultrashort pulses, about 200 nm, provides enough spectrum to obtain essentially single-shot measurement of the microcavity filtering characteristics. Wavelength tuning makes it possible to map out the bandgap.

Figure 4. Wavelength conversion efficiency versus normalized pump intensity. (Theory-solid line).

Figure 5. Femtosecond pulse spectra generated by difference frequency mixing.
1.9 Femtosecond Optical Nonlinearities in ZnSe and Characterization of ZnSe/GaAs Heterostructures

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Project Staff

Professor Erich P. Ippen, Professor Leslie A. Kolodziejski, David J. Dougherty, Siegfried B. Fleischer, Emily L. Warlick, Jody L. House, Eason Ho, Dr. Gale S. Petrich

ZnSe is an important material for fabricating laser diodes in the blue spectral region. Understanding its bulk optical and interface transport properties can lead to improved device design. Because ZnSe is a II-VI semiconductor, its properties provide useful comparison to the body of knowledge about III-V semiconductor nonlinear optics.

Pump-probe experiments were performed to investigate the nonlinear optical mechanisms in ZnSe. Frequency doubling of pulses from a modelocked Ti:Sapphire laser provided 60 fs probe pulses tunable across the ZnSe bandgap. For above-band pumping and large carrier injections, screening of the Coulomb attraction between electron-hole pairs was seen to be the dominant probe absorption saturation mechanism. The thermalization and cooling dynamics of the injected electron and hole distributions were completely masked by the screening reduction. This situation represents a qualitative difference in the bandedge nonlinear response compared to III-V semiconductors such as GaAs where the exciton binding energy is much smaller than kT. By using a below-band pump and doped ZnSe films, we were able to heat the carrier distributions by free carrier absorption without population density changes. The carrier cooling dynamics were thus isolated. Figure 6 illustrates observed dynamics for intraband excitation in a doped sample where distributional dynamics are revealed and the carrier heating signal is identified by its change of sign at the electron Fermi level. Relaxation times of 500 fs for electrons and 900 fs for holes were measured independently, in N and P-type samples respectively. A delayed heating response was also seen in the N-type samples. This was found to be due to electrons returning from the satellite L-valley in the conduction band, and a L-Γ scattering time of 1.8 ps was determined. By tuning the pump photon energy it was also possible to measure the energy of the L-valley minimum to lie 1.31 eV above the Γ-valley. This is arguably the most accurate measurement of this quantity so far. An instantaneous bleaching was also seen and attributed to the optical Stark effect. The optical Stark effect in semiconductors is well known; however, two unique features of this work are (1) a clear demonstration of the state repulsion caused by the pump field, and (2) a determination of the contribution of Coulomb enhancement to the polarizability of the continuum states by the measurement of samples doped above and below the exciton Mott density.

Characterization of ZnSe/GaAs heterojunctions were carried out with contactless electro-and photo-reflectance modulation spectroscopy techniques. Fields much larger than can result from modulation doping effects were measured. This points to large interface charges in the \(10^{12}\) cm\(^{-2}\) range. The electro-reflectance technique unambiguously determined the sign of the charges to be negative for:

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both n-type GaAs and intrinsic GaAs substrates. This leads to large conduction band bending on the GaAs side of the junction and formation of a large effective barrier for electrons to enter the ZnSe. This barrier’s existence was confirmed by photoreflectance saturation intensity measurements and a new technique using a tunable pump to directly measure the conduction band offset at the interface. These results demonstrate that band bending due to interface charges must be taken into account for any study of the effect of growth and nucleation conditions on this interface and for understanding carrier transport in ZnSe based devices using GaAs substrates.

Additional pump-probe experiments were performed to extend the conventional photoreflectance measurements. The mechanism of photoreflectance techniques is screening of built-in surface fields by surface trapping of minority carriers. This reduction in the electric field changes the Franz-Keldysh contributions to the refractive index and, therefore, the sample reflectivity. Pump-probe experiments were performed to time-resolve these subpicosecond trapping dynamics for the first time. Both the intensity and spectral dependence of the reflectivity signals support this interpretation. A simple model is proposed for explaining these results which shows that this technique may be used as a non-contact method for measuring surface trap densities and recombination velocities. Extension of this technique to the technologically important Si/SiO₂ interface should be straightforward.

1.10 Femtosecond Continuum Studies of PbTe Quantum Dots at 1.5 μm

Sponsors

Defense Advanced Research Projects Agency/ National Center for Integrated Photonics Technology

U.S. Air Force - Office of Scientific Research

Project Staff

Professor Erich P. Ippen, Dr. Günter Steinmeyer

With their strong quantum confinement, semiconductor quantum dots in glass matrices offer the potential of application as efficient nonlinear optical elements. Previous work has shown that Cd-compound nanocrystallites exhibit strong optical nonlinearities in the visible. The lower bandgap of PbTe provides opportunity to produce quantum dots absorbing in the important telecommunication band at 1.5 μm.

We have used our recently demonstrated source femtosecond continuum around 1.5 μm to investigate optical nonlinearities of PbTe nanocrystallites in a glass. The quantum dots (obtained through collaboration with C.L. Cesar at UNICAMP, Campinas, Brazil) are produced in a glass matrix by a heat treatment close to the softening temperature of the glass. The mean radii of the dots were controlled by annealing time and temperature, and the quantum confined energy level was varied from 1300 to 1800 nm, respectively.

For our pump wavelength of 1.52 μm, the different samples make possible pump-probe experiments over a range of above and below band excitations. Induced transmission is observed both above and below the excitation wavelength within the response time of our system, consistent with ultrafast screening and carrier-carrier scattering. Partial recovery, particularly of the lowest electronic states, occurs on a timescale of 10 ps. Similar partial recovery in CdS quantum dots has been explained previously as due to trapping by surface states. Complete electron-hole recombination follows on a longer timescale. Experiments to further elucidate these dynamics are underway.

1.11 Ultrafast Dynamics of K₃C₆₀ and Rb₃C₆₀ in the Normal and Superconducting Phase

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Project Staff

Professor Mildred S. Dresselhaus, Professor Erich P. Ippen, Siegfried B. Fleischer, Boris Pevzner

The transient optical response of high-Tₜ and metallic superconductors to impulsive optical excitation by sub-picosecond light pulses gives important information on the dynamics of Cooper pair breaking and recombination. For this reason, we have performed pump-probe studies on supercon-

Figure 7. Pump-induced change in the reflectivity $\Delta R/R$ of $K_3C_{60}$ at a series of nominal temperatures above and below the superconducting phase transition of $T_c = 18$ K.

The samples used for our studies were found to exhibit a superconducting transition of $T_c = 18$ K for $K_3C_{60}$ and $T_c = 28$ K for $Rb_3C_{60}$, in good agreement with literature values. Both materials show a three- to five-fold increase in the signal size $\Delta R/R$ when cooled down from room temperature to about 50 K, which is consistent with the decreasing electronic specific heat. At high temperatures the signals are characterized by a positive transient which decays within 1 ps for $K_3C_{60}$ and 0.7 ps for $Rb_3C_{60}$. This normal state decay slows by about a factor of two for both samples as the temperature is lowered from room temperature to about 50 K. Such behavior is typical for metallic systems in which non-equilibrium carriers cool via inelastic collisions with phonons. In figure 7, we plot the change of the reflectivity for $K_3C_{60}$ at a series of nominal temperatures above and below the superconducting phase transition. A similar set of measurements was also taken for $Rb_3C_{60}$. The most striking feature in the low temperature response of the potassium doped film is the dip of about 50 percent of the fast initial response around the transition temperature. This reduction in signal size (most pronounced at 16 K) might be due to the change in the electronic heat capacity below $T_c$ or a change in the optical penetration depth due to the superconducting electrons. Below $T_c$, an additional slower component with opposite sign, not present in the normal state, is observed. This slow exponential decay characteristic of the quasi-particle dynamics in the superconducting state shows a considerably faster decay rate as the temperature approaches the phase transition from below. At the same time, the amplitude of this response becomes diminishingly small and the quasi-particle response disappears in the normal state ($T>T_c$).

Femtosecond-pulse-induced coherent phonon oscillations were also studied in doped fullerenes with 20 fs time resolution (see figure 8). We observed a remarkable number of oscillations (200 periods of the $A_{1g}$ mode) with a minute signal modulation of only $10^{-7}$ on the probe beam. This is to our knowledge the largest number of phonon oscillations reported in a time-resolved measurement on any system. A slight dependence of the frequency on dopant is observed (a $2.5 \text{ cm}^{-1}$ difference of the coherent phonon frequency between $K_3C_60$ and $Rb_3C_60$). Our measurements also revealed, for the first time, a vibrational mode at about $150 \text{ cm}^{-1}$ which we attribute to a beating between the $C_{60}$ anion and the alkali-metal cation. The dephasing time for this oscillation is very fast (1-2 periods) so that it would be impossible to observe such a highly damped mode in Raman spectroscopy. Thus, time-resolved techniques such as pump-probe are much superior for such studies.

1.12 Ultrashort Pulse Generation and Ultrafast Phenomena

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1.12.1 Compact Ultrashort Pulse Sources

Advances in signal processing, high-speed communications, and the investigation of ultrastart nonlinear processes in semiconductor materials and devices require the development of compact ultrashort pulse laser sources. These sources must be technologically simple, robust, and cost effective, in order to enable applications in a non-laboratory environment. During the last few years, significant advances have been made in the development of ultrastart laser sources utilizing the Kerr effect (nonlinear refractive index). Kerr lens modelocking (KLM),35 has achieved the generation of the shortest pulses ever produced directly from a laser oscillator.36 A theoretical model for Kerr lens modelocking has been developed by Professors Erich P. Ippen and Hermann A. Haus in conjunction with our group.37

We have applied our theoretical understanding of KLM to the optimization of modelocking performance. This has resulted in the extension of this simple pulse-forming mechanism to novel resonator
geometries. Two femtosecond Ti:Sapphire KLM lasers and a femtosecond Cr:Forsterite laser in our laboratory are the direct result of these studies.

KLM lasers utilize self-focusing in the presence of soliton-like pulse shaping arising from self-phase modulation and the net negative intracavity group velocity dispersion (GVD) to generate chirp-free ultrashort pulses. Negative GVD is most commonly achieved in KLM lasers by use of an intracavity prism pair. This, however, places constraints on laser geometry and size. X- and Z-cavity geometries commonly used in KLM lasers result in repetition rates not exceeding the 100 MHz range. In the past few years we have advanced our theoretical understanding of KLM to include compact resonator geometries. We have demonstrated new techniques for dispersion compensation in KLM lasers that use a novel compact resonator design in conjunction with a prismatic output coupler. This approach achieves a pulse repetition rate of 1 GHz, to our knowledge the highest repetition rate achieved in a KLM laser to date, and 100 fs pulses. We have demonstrated novel compact cavity designs that use an intracavity lens and have investigated the effect of various compact resonator geometries on Kerr-lens action and self-starting of modelocking.

In the past year, we have worked on extending our previous results from Ti:Sapphire operating at wavelengths around 800 nm to a wavelength region of 1.3 microns which is of high importance for high speed communications applications as well as for investigation of ultrafast nonlinear processes and for optical coherence tomography. The system is based on the Cr:Forsterite laser crystal which has a broad gain bandwidth and is tunable from below 1.2 microns to above 1.35 microns. By combining numerical simulations with experimental work, we have designed a compact laser system that would lead to pulse repetition rates up to the GHz range and output powers in excess of 100 mW. An additional advantage of a Cr:Forsterite laser is a potential for diode pumping. This system could eventually be a portable, tunable, low-cost, 1.3 micron-femtosecond laser source which could be used outside a laboratory environment.

### 1.12.2 Solid-State Laser Development for the Near-Infrared

Chromium-doped forsterite (Cr:Mg 2 SiO 4 ) laser crystals are of interest because this material permits the development of tunable solid-state sources in the 1.3 µm wavelength range. Since the first demonstration of laser operations several years ago, the spectroscopic and laser properties of forsterite have been extensively investigated, and modelocked femtosecond operation has been achieved in this material. Optimal diode pumped laser performance necessitates the use of laser crystals with short absorption lengths. This allows the highly divergent output from the diodes to be efficiently coupled to the intracavity mode. Short absorption lengths result from a high density of the dopant laser ion and the use of diode lasers with emission wavelengths matched to the peak of the absorption cross-section. Typically, achieving high doping densities is frustrated by the presence of excited state absorption or parasitic loss due to low crystal quality.

During the past year, we have further explored the performance and design options of lasers based on Cr:Forsterite crystals. Our efforts were directed toward the development of systems where thin crystals can be used. Typically Cr:Forsterite is pumped at 1.06 µm using well established Nd:YAG lasers. Due to typical absorption coefficients of (2 cm⁻¹), these systems require long crystals and can be subject to considerable parasitic absorption effects. Spectroscopic studies performed in our laboratory indicated that the excited states that give rise to the broad emission band centered at 1.28 µm can be populated through absorption at 740 nm. At this wavelength the absorption cross-section is nearly an order of magnitude stronger than at 1.06 µm. It is possible, then, to use crystals with a doping density identical to those used with 1.06 µm excitation while reducing the length of the crystal nearly ten-fold. Since the figure of merit of a laser crystal is the ratio of the desired net absorption to the parasitic net absorption, this approach leads to a dramatic increase in the effective quality of currently available material.

To test the efficacy of these ideas we have demonstrated laser operation with pump wavelengths.

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ranging from 700 to 820 nm.\(^{41}\) The reduction of parasitic loss resulting from the use of a short, 3 mm Cr:Forsterite crystal provided a significant enhancement of the range over which this laser can be tuned. The measured cw tuning range from 1175 to 1375 nm is, to our knowledge, the largest cw tuning range reported for a Cr:Forsterite laser. Initial research on obtaining femtosecond Kerr-lens modelocked operation was conducted using an asymmetric z-cavity and dispersion compensating prisms. Pulses with durations of 52 fs were generated using 2.0 W of pump power.\(^{42}\) The spectral bandwidth was 34 nm FWHM, corresponding to a time-bandwidth product of 0.325. While this pulse duration is longer than the best result achieved with 1.06 \(\mu m\) excitation,\(^{43}\) we believe that we are currently limited by the relatively weak self-phase modulation induced at the low, 2.0 W pump power. In general, for femtosecond pulse generation, the use of short crystals provides better performance by reducing the deleterious effects of high-order dispersion. Thus, the choice of near-IR pump wavelengths has significant advantages over the standard pump wavelength of 1.06 \(\mu m\) and will be further investigated for improvement in the design of Cr:Forsterite laser systems.

In order to investigate the possibility of developing a compact forsterite laser for future diode pumped applications, a compact version of the cw laser was implemented where the z-cavity had arm lengths of only 10 cm. For this laser the pump beam diameter in the crystal was not modified from that optimized for the longer cavity used above. This configuration produced a cw output power identical to the larger cavity designs. We also investigated the operation of this compact laser without water cooling (room temperature operation with heat sinking and air cooling). Stable operation with only a modest, \(< 10\) percent, decrease in output power was achieved. We are currently exploring the use of high brightness 670 nm diodes as excitation sources. We believe that this work will result in a compact diode pumped source of high power, tunable laser emission in the important wavelength range of 1.3 \(\mu m\).

### 1.12.3 Carrier Dynamics in InGaAs Strained Layer Diodes

Femtosecond nonlinear gain and transient carrier dynamics in laser diodes play a central role in understanding laser linewidth, modulation bandwidth, amplification, and short pulse generation. Previous studies by our group have used a new heterodyne nondegenerate pump-probe measurement technique for InGaAs/AlGaAs graded-index single-quantum well diode lasers to investigate nonlinear gain dynamics.\(^{44}\) In our earlier work, the pump and probe wavelengths were fixed and differential transmission curves were measured for several different values of the injected carrier concentration.\(^{45}\) The experimental technique has since been modified to allow for the measurement of time-dependent differential transmission spectra over multiple probe wavelengths. In addition, we extended our theoretical formalism to include carrier-carrier scattering with a relaxation-time approximation model.

The transient photogeneration of electron-hole pairs by the pump pulse and subsequent relaxation of carriers by polar optical phonon and carrier-carrier scattering were investigated. Transient measurements showed a pump-induced transmission decrease in both the gain and loss regions. The carrier-carrier scattering rates are determined using the quasi-equilibrium distribution functions for a given background carrier density. As the background carrier concentration is raised, the relaxation rate for carrier-carrier scattering is dominant in comparison with polar optical phonon scattering. Estimates indicate that for carrier concentrations above \(10^{12} \text{cm}^{-2}\), coefficients of \((2 \text{ cm}^{-2})\), carrier-carrier scattering is important and must be included. When carrier-carrier scattering is turned on, the phonon replicas disappear in the time dependent distrib-


ution and the carrier-carrier scattering acts to thermalize the distribution. The inclusion of carrier-carrier scattering suppresses the dip observed in the differential transmission around the pump frequency and smoothes out sharp structures in the phonon replicas.

In collaboration with theoretical physicists at the University of Florida, Gainesville, detailed theoretical models for the gain dynamics for InGaAs strained-layer diodes were developed. In our model, transient gain and differential transmission are computed in a multiband effective-mass model including biaxial gain, valence subband mixing, and scattering both within and between bands. The carrier-carrier scattering rates are determined using quasi-equilibrium distribution functions for a given background carrier density and the Fermi-Dirac distribution function where the chemical potential and temperature are self-consistently chosen so that both particle number and energy are conserved. Polar optical phonon scattering rates by Boltzmann equation and carrier-carrier scattering rates by an effective relaxation operator are solved using an adaptive Runge-Kutta routine. Our results show that (1) the inclusion of carrier-carrier scattering improves the agreement between experiment and theory where only carrier-phonon scattering was included and (2) carrier-carrier scattering is necessary to produce the heating of carriers in the high-energy tails. This formalism aids the full understanding of the obtained experimental results and might aid future device design.

1.12.4 Publications


1.13 Laser Medicine and Medical Imaging

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1.13.1 Optical Coherence Tomography Technology

Optical biopsy, or micron-scale, cross-sectional, optical imaging of tissue microstructure in situ, would aid the diagnosis and clinical management of many diseases. Optical coherence tomography (OCT) is a new optical imaging technique that uses low-coherence interferometry to perform high-resolution, cross-sectional imaging in biological systems. OCT is analogous to ultrasound B mode imaging except that this technique uses an infrared light source and low-coherence interferometry to perform micron resolution ranging and imaging. For an interferometric signal to be detected, the optical path lengths of the object and reference beam must be matched to within the coherence length of the source. Since multiply scattered photons from the object have traveled different optical path lengths than the reference beam, multiple scattering effects are minimized in the OCT image. Tissue reflectance is obtained axially by varying the reference arm delay and digitizing the magnitude of the interference. A cross-sectional image is produced by recording axial reflectance profiles while the beam on the tissue specimen is scanned.

OCT has been extensively applied in ophthalmology to provide tomographic images of the transparent structures in the eye. Clinical studies have shown that OCT provides high resolution, cross-sectional images of the retina and can be used to diagnose a wide range of retinal macular diseases. OCT imaging in other human tissues is made difficult due to optical scattering. However, recent in vitro studies have shown that OCT can image architectural morphology in highly optically scattering tissues. An important application of OCT is imaging the vascular system to identify atherosclerotic lesions that are prone to rupture. OCT has ten times greater resolution than clinically available, catheter based ultrasonography, intravascular ultrasound (IVUS). Additional research has shown that OCT can perform high resolution imaging of other organ systems such as the gastrointestinal and urinary tracts.

Research in this laboratory, performed to advance optical coherence tomography technology, has yielded significant improvements in image resolution, acquisition speed, and the development of endoscopic imaging techniques. One of the key technological advances which greatly enhances the performance of OCT is the development of high power, ultrashort coherence length light sources. Recently, OCT imaging with axial resolutions of 2 to 4 μm has been demonstrated using broad bandwidth, short pulse femtosecond laser sources which have a short coherence length.

Improvement in image acquisition times have also been recently demonstrated. The image acquisition times of OCT systems used in previous studies (60 seconds) is adequate for in vitro imaging but is insufficient for in vivo imaging because of motion artifacts. The development of higher power sources is critical to increasing image acquisition speed since the image acquisition rate directly trades off against detection sensitivity. To achieve image acquisition times below one second, alternatives to mechanical reference arm scanning technologies must also be developed. Recent work using these new technologies has achieved an acquisition rate of up to four images per second.

Another technology which is necessary in order to apply OCT for imaging of internal organ systems is a catheter/endoscope which can deliver, focus, scan, and collect a single spatial mode optical beam. The catheter must be flexible and have an

small diameter to facilitate its entry into internal channels such as coronary arteries, pancreatic or biliary ducts. A single mode fiber-optic scanning OCT catheter/endoscope prototype has been developed during the past year. This device is an enabling technology for developing a wide range of endoscopes and will permit the OCT imaging of the in vivo gastrointestinal tissues.

This work is an ongoing collaboration with researchers at MIT, MIT Lincoln Laboratories, Massachusetts General Hospital Cardiology Department, Massachusetts General Hospital Pathology Department, and the Wellman Laboratories of Photomedicine.

1.13.2 High-Resolution Optical Coherence Tomography

Chromium doped Forsterite (Cr\(^{4+}:\text{Mg}_2\text{SiO}_4\)) is a new tunable solid state laser material with a broad emission band near 1.2 \(\mu\)m. Cr\(^{4+}:\text{Mg}_2\text{SiO}_4\) has been modelocked using KLM, and pulse durations as short as 25 fs have been achieved. We have designed and constructed a short pulse Cr:Forsterite laser that utilizes a diode pumped Nd:YAG laser for excitation. The resulting all-solid-state source has superior stability and reproducibility compared to either Cr:Forsterite sources that rely on lamp excited Nd:YAG pump sources or argon ion pumped TiAlO\(_3\) oscillators. In addition, the diode pumped laser is significantly more efficient and economical compared with argon ion lasers.

The peak output power from the Cr\(^{4+}:\text{Mg}_2\text{SiO}_4\) laser is sufficient to generate significant spectral broadening within optical fiber. However, the overlap of the spectral emission from this laser with the zero dispersion point of standard single mode fiber results in a strongly modulated spectral output inappropriate for biomedical imaging. We have avoided this problem by using dispersion shifted fiber and can obtain high resolution, high contrast coherence ranging. The high average output power provided by this source enables high image acquisition speeds and high sensitivities, qualities essential to biomedical imaging.

Coherence ranging is performed with the self-phase modulated spectral source using a fiber optic Michelson interferometer. Wavelength flattened splitters were used to preserve the spectral extent of the source as it was routed to the scanning reference arm mirror, the biological sample, and back to the detectors. Dual balanced heterodyne detection enabled near shot-noise limited detection.

Ultrasound resolution OCT imaging using modelocked solid state lasers is a powerful technology for performing optical biopsy which can be applied for diagnosis, interoperative guidance, and clinical management in a wide range of medical applications.

1.13.3 High-Speed Optical Coherence Tomography

Currently, typical OCT systems used to image human tissue are implemented using superluminescent diodes (SLD) with center wavelengths of 850 or 1300 nm. These OCT systems have free space axial resolutions of 15-20 \(\mu\)m, a sample arm power of 50-150 (\(\mu\)W,) and signal to noise ratios (SNR) ranging from 90 to 110 dB. The image acquisition times of these systems (5-60 seconds) make their application to in vivo imaging difficult because of motion artifacts. To achieve image acquisition times below one second, alternative technologies to mirror translation scanning must be developed. Because a high SNR is necessary for imaging to significant depths within turbid media any increase of image acquisition rate must be accompanied by a commensurate increase in source optical power. We have constructed a novel OCT system which addresses both of these concerns and achieves an acquisition rate of four images per second. Each image consists of 250 X 250 pixels covering a sample area of 3 mm X 2.5 mm.

Previous OCT systems have utilized a mechanically translated reference arm mirror to enable axial scanning. The mirror velocity is typically in the range of 30 mm/s corresponding to a Doppler frequency of 50 kHz. To enable image acquisition rates of two frames per second the Doppler frequency must be increased ~100 fold to 5 MHz. This would necessitate a reference mirror velocity of 3 m/s. Additionally, this velocity must be held constant over distances of 3-5 mm and repetition rates of ~0.5 kHz. Because no cost effective trans-

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lator meeting these specifications is currently available; we have developed an alternative technology that uses piezo-electric transducers to induce stretch in an optical fiber. 40 m of single mode optical fiber is wrapped under constant tension about a piezo-electric modulator (PZM). As the PZM expands, the fiber is stretched inducing temporal delay on the light propagating within it. The long length of fiber allows the small expansion of the PZM to be magnified to a length of approximately 3 mm. The deleterious effects of static and dynamic birefringence incurred in the fiber are compensated using Faraday rotators placed in the free space region of the interferometer arms.

To maintain the high SNR necessary for deep imaging into turbid media such as human tissue, the decrease in image acquisition time must be matched by an increase in optical power. To enable high-speed, high-SNR imaging, we have employed the Cr:Forsterite laser source discussed above. At the high modulation frequency induced by the fast scanning piezo delay line the noise due to amplitude fluctuation in the modelocked laser is low, but prevents shot-noise limited detection. Heterodyne dual balanced detection is used to reduce the laser noise below the shot-noise level and allows for OCT imaging with a SNR of 112 dB. This dynamic range is sufficient to allow OCT imaging to depths of greater than 2 mm within human tissue.

1.13.4 Scanning Catheter/Endoscope for Optical Coherence Tomography

In order to apply OCT for imaging of internal organ systems, a flexible, small diameter, catheter/endoscope which is capable of delivering, focusing, scanning, and collecting a single spatial mode optical beam was constructed. In the past year, we have advanced our capability to perform catheter based imaging by constructing a second generation catheter that allows significantly higher optical throughput with low internal backreflection. The refined catheter design also produces a probing beam with superior focusing properties for imaging within small channels such as are present in the cardiovascular system. The OCT catheter consists of a drive motor at its proximal end, a single mode fiber running the length of the catheter, and optical focusing and beam directing elements at the distal end. During image acquisition, the catheter is inserted into an internal tissue channel, and the focused beam is scanned perpendicular to the axis of the catheter. An OCT image is acquired as the beam angle of rotation is varied over some range (usually 360 degrees). The speed of imaging depends on the speed of the rotation and the OCT unit acquisition speed.

In order to demonstrate imaging with the catheter, we have performed OCT imaging of an in vitro human saphenous vein. This study was designed as a direct comparison of catheter based OCT with clinically available intravascular ultrasonography. The results of this study indicate that OCT can provide significantly higher resolution while penetrating to depths that are relevant to the diagnosis of cardiovascular disease.

While many engineering issues still must be addressed in order to perform OCT imaging in vivo, the key enabling technologies required to achieve this objective have been developed and demonstrated by this laboratory. Improvement in image resolution, acquisition time, and the development of a fiber optic endoscope have been shown to be technically feasible. We believe that the results of this research will accelerate progress towards performing in vivo optical biopsies, and should have a significant impact on medical diagnosis and the management of disease.

1.13.5 Ophthalmic Imaging and Diagnosis With Optical Coherence Tomography

In collaboration with MIT Lincoln Laboratory, a clinically effective, ophthalmic imaging OCT system has been developed. A compact superluminescent diode light source operating at 840 μW is coupled via fiber optics through a standard slit-lamp biomicroscope. The operator is able to simultaneously view a video image of the scanning OCT beam on the retina and an OCT image on a computer monitor which is updated in real-time every 2.5 seconds. The system is fully computer controlled to allow positioning of a variety of scanning patterns on the retina. Image processing algorithms have been developed to minimize the effects of eye motion during scanning, to automatically extract the thickness of various retinal layers directly from the OCT images, and to provide topographic displays of quantitative parameters.

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such as total retinal and retinal nerve fiber layer thickness.61

Over 5000 patients with a variety of macular diseases and diseases of the optic nerve head have been examined at the New England Eye Center, Tufts University School of Medicine. In patients with macular pathology, OCT images have been correlated with the conventional clinical techniques of slit-lamp biomicroscopy and fluorescein angiography.62 The cross-sectional view of OCT is effective in the diagnosis and monitoring of macular holes, macular edema, and retinal detachments. In patients with glaucoma, the ability of OCT to directly measure retinal nerve fiber layer thickness with micron scale resolution may lead to the first truly objective diagnostic for the presence or progression of this degenerative disease.63

Macular hole is a frequently encountered retinal disease that progresses to involve a complete loss of the retina directly in the fovea leading to a devastating reduction in visual acuity. However, early surgical intervention can often prevent or correct vision loss. OCT is useful in identifying early macular holes, in staging macular hole progression, and in evaluating the risk of hole formation in the fellow eye of patients who already have a unilateral macular hole.64 Pre-operative assessments of macular hole structure with OCT correlate with the probability of hole closure after surgery. This information is useful in considering candidates for surgical treatment. OCT has been able to provide information on the pathogenesis of hole formation and the conditions which lead to the development of macular holes which may eventually lead to better treatment of this disease.

Diabetic retinopathy is the second leading cause of blindness in the United States. The development of macular edema is a major treatable cause of vision loss in patients with diabetes. Macular edema is also common manifestation of a variety of other retinal diseases including epiretinal membranes, uveitis, retinal vascular occlusions, and retinal inflammatory diseases. Intraretinal fluid accumulation may also be a complication of cataract surgery. OCT is a more sensitive and objective indicator of retinal thickening due to macular edema than either slit-lamp biomicroscopy or fluorescein angiography, is useful in objectively tracking the resolution of edema following therapy, and may be effective as a screening tool for the development of retinal edema in these patients.65 Macular edema is traditionally evaluated by assessing retinal thickening with slit-lamp biomicroscopy, and leakage of fluorescein dye from blood vessels into the retina, which is a relatively poor indicator of vision loss. OCT provides a sensitive, objective test for both the early detection of macular edema, and longitudinally monitoring retinal thickness over time in patients who have received laser treatment for this disease. A standardized, topographic method of displaying macular thickness has been developed which provides both a quantitative and intuitive means of assessing retinal thickening geographically. The use of a standardized protocol means that OCT may have significant public health implications as a screening technique for the development of retinopathy in all patients with diabetes.

OCT is also effective in evaluating retinal detachments, such as those occurring with central serous chorioretinopathy.66 The ability to objectively monitor these detachments over time is useful in evaluating the necessity or effectiveness of treatment in this disease. Furthermore, OCT shows the defects in the retinal pigment epithelium which are associated with the pathogenesis of this illness.

Age-related macular degeneration is the leading cause of blindness in the United States. Vision loss in this disease is often due to the formation of

choroidal neovascularization (CNV). OCT may represent a new technique for visualizing the boundaries of occult CNV which is necessary for effective treatment. We have also found that OCT is effective in the identification and quantification of subretinal and intraretinal fluid, and is particularly useful in evaluating possible foveal involvement of fluid accumulation or CNV. In patients with untreated exudative AMD, OCT was compared with fluorescein angiography in the classification of CNV. A subset of eyes with poorly defined CNV on fluorescein angiography had well-defined boundaries on OCT suggesting that OCT could potentially be used to guide laser photocoagulation treatment in these eyes. These studies suggest that OCT may lead to the possibility of more effective treatment delivery in eyes.

Current diagnostic techniques for epiretinal macular membranes include slit-lamp biomicroscopy and fluorescein angiography. OCT images are useful in confirming the diagnosis of faint, diaphanous membranes, and in providing a cross-sectional assessment of factors contributing to vision loss, such as membrane opacity, retinal distortion, or tractional detachment, and macular edema. Many studies have attempted to define prognostic indicators, such as membrane thickness or the presence of pre-operative cystoid macular edema, to predict eventual visual outcome after epiretinal membrane surgery. OCT provides a means to evaluate the cross-sectional characteristics of an epiretinal membrane, allowing a quantitative measurement of retinal thickness, membrane thickness, and the separation between the membrane and inner retina. Thus, OCT may be useful in objectively evaluating the surgical prognosis in eyes with an epiretinal membrane.

Both serous macular detachments and schisis-like macular separations are associated with congenital optic nerve pits. The pathogenesis of these lesions, however, has been controversial. OCT is useful in structurally characterizing the development of these lesions. Neurosensory detachments appear as a complete elevation of the sensory retina overlying optically clear serous fluid. In contrast, schisis-like separations are associated with a splitting of the outer retina with vertical bridging elements between the outer and inner retina.

Glaucoma is the third leading cause of blindness in the United States. Current diagnostic techniques such as direct and indirect ophthalmoscopy, nerve fiber layer photography, and evaluation of the cup-to-disc ratio are subjective. Quantitative visual field testing may provide early identification of peripheral visual field defects; however, up to 50 percent of the retinal nerve fiber layer may be lost before detection by standard clinical techniques. OCT provides a quantitative method of directly measuring the thickness of the retina and retinal nerve fiber layer (NFL) with high resolution and may lead to an objective, early diagnostic for the onset of glaucoma and the progression of glaucomatous nerve damage. NFL thickness as measured by OCT correlates well with the functional status of the optic nerve as measured by traditional visual field examination, and correlates better with visual field loss than either cupping or neuroretinal rim area. Normal and glaucomatous eyes show a significant difference in NFL thickness as measured by OCT. Furthermore, measurements of NFL thickness using OCT are reproducible to within 10 to 20 μm. These studies indicate that OCT is potentially able to detect the onset of glaucoma or glaucomatous progression before significant and irreversible damage to the retina occurs.

1.13.6 Optical Biopsy using Optical Coherence Tomography

Biopsies are routinely used in medical practice to assess the state of human tissue and accurately determine the cause and extent of disease, or confirm its absence. The results of these biopsies usually decide the course of future medical action, therapeutic modalities used and the prognosis for each particular case. Inaccessibility to certain

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organs, risk of injury, patient discomfort and cost, however, prohibit the extensive use of biopsies and reduce the diagnostic capabilities of the procedure by limiting the area and extent of coverage. OCT could be used to acquire cross-sectional tissue images, depicting tissue microstructure, with micrometer scale resolution thus providing an additional diagnostic tool that could supplement and in some cases replace conventional biopsies. 71 Since OCT works in real, or near-real, time a large number of optical biopsies could be taken in a short time thus increasing the predictive value of the procedure. 72 The system's fiber optic design also allows easy integration into existing medical imaging devices, such as catheters and laparoscopes, while maintaining a small system profile and a low cost. 73 Several studies have been undertaken by our laboratory and our collaborators to demonstrate the feasibility of the application of OCT in such areas as vascular, urinary and gastrointestinal pathology and surgery. The resulting images were compared with histologic cross-sections from the associated sites to confirm consistently high correlation.

The general consensus in the medical community today is that most myocardial infarctions, known to the public as "heart attacks," are the result of the rupture of mild to moderate size atherosclerotic plaques, which initiates a cascade of events that leads to thrombosis and vessel occlusion. Plaques with large lipid cores and weak fibrous caps are characterized by the highest risk of rupture. Current imaging modalities, such as angioscopy, ultrasound and MRI, are unable to definitively identify these high-risk plaques. Angioscopy can only image the surface of the artery and only with a clear field of view in the absence of blood. Ultrasound and MRI suffer from low resolution and dynamic range and, in the case of the latter, expensive apparatus add to the overall cost and complexity of the procedure. A study comparing ultrasound and OCT resolution dramatically illustrated the marked improvement in image quality OCT provides. A series of in vitro experiments has shown that OCT can be employed to visualize structural features such as intimal wall thickness and lipid content and even image through highly calcified plaques with a resolution and dynamic range unavailable with any other imaging modality. This technique provides the capability to image small atherosclerotic plaques well in advance of possible rupture as well as the extent of existing fissures. 75 These properties, in conjunction with the fiber optic design of the system which can be adapted for delivery through the channels of existing catheters, would make this tool very attractive to clinical practice. It can also be used to guide surgical intervention to reduce patient risk and improve prognosis of patients undergoing atherosclerotic plaque manipulation or other intervention close to sensitive tissue such as nerves. A number of technological issues regarding the in vivo application of OCT will be addressed during the animal study which uses a catheter-delivered OCT fiber and is currently under way. 76

We have investigated the capability of OCT to differentiate the architectural morphology of gastrointestinal tissue with the long term objective of extending OCT to endoscopic based diagnostics. Normal and diseased gastrointestinal tissues were taken postmortem and imaged using OCT. The images were compared to corresponding histology to confirm tissue identity and qualitatively suggest the mechanisms which produce tissue contrast. Microstructure was delineated in different tissues,

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including the esophagus, colon, liver, gallbladder, common bile duct, and pancreas, at $16 \pm 1 \mu m$ resolution, higher than any clinically available cross-sectional imaging technology. Differentiation of tissue layers, such as the submucosa, muscularis, and serosa was achieved due to their different optical properties. It was also possible to illustrate the limitations of diagnostic assessments based on visual inspection and gross pathology. One of the samples was diagnosed as a colonic adenoma but the OCT based diagnosis, which was confirmed by histologic results obtained later, indicated the correct nature of the problem, i.e., inflammation. The ability of OCT to provide high-resolution imaging of gastrointestinal micro-structure, without the need for excisional biopsy, suggests the feasibility of using OCT as a powerful diagnostic imaging technology which can be integrated with conventional endoscopy.

During the past year we have investigated the capability of OCT to image in vitro microstructure of the urinary tract, with the focus on a critical region consisting of the prostatic capsule, surrounding adipose tissue, and neurovascular bundles. Urologic tissues were taken postmortem, dissected, and imaged using OCT. Microstructure was delineated in different urologic tissues, including the prostatic urethra, prostate, bladder, and ureter, with an axial resolution of $16 \pm 1 \mu m$ higher than any clinically available endoscopic intraluminal imaging technology. The ability of OCT to provide non-contact high-resolution imaging of urologic tissue architectural morphology, without the need for excisional biopsy, suggests the potential of using OCT to obtain information on tissue microstructure that could only previously be obtained with conventional biopsy. A role for OCT in guiding the resection of hyperplastic prostatic parenchyma has also been suggested. OCT images, with an axial resolution of $16 \mu m$, sharply delineate the prostate capsule border. Furthermore, neurovascular bundles were identified in close approximation to the prostatic capsule. Since postoperative impotence and incontinence have been linked to traumatic transsection of these neurovascular bundles, OCT guidance may substantially reduce the morbidity associated with mechanical interventions.

Improved methods for visualizing microanatomy, in addition to technical refinements in tissue manipulation, have been the cornerstone of the advance in microsurgical interventions. The OCT technology could provide the surgeon with superior, high resolution, real-time information about tissue microstructure in conjunction with a low profile within the operative field. In addition, OCT is attractive for surgical diagnostics due to its ability to perform imaging through air and capacity to perform imaging through small optical fibers. We examined the feasibility of optical coherence tomography for surgical guidance and diagnostics by demonstrating that high resolution imaging can be obtained in tissue with important surgical implications. Tissue was selected from the nervous and vascular systems since microsurgical interventions have become increasingly important in these organ systems. Imaging of structures within both the central and peripheral nervous system was possible. Since injury of small nerves during surgical procedures can result in iatrogenic injury, intraoperative real-time high resolution imaging may substantially reduce the morbidity associated with tissue dissection of vulnerable regions. The ability of OCT to perform high resolution transmural imaging of blood vessels also demonstrated that it could substantially reduce the morbidity associated with vascular repair.

1.13.7 Optical Coherence Tomography in Developmental Biology

Imaging technologies offer numerous possibilities to investigate the processes involved in neural development. In the past year, we have examined the potential of using OCT for the investigation of developing neural morphology. To demonstrate the capabilities of this technique in accessing neural development, we have chosen to image early normal and abnormal neural morphology in a common developmental biology model, *Xenopus laevis* (African frog). *In vivo* images clearly identify gross and subtle differences in neural structure and may offer an alternative to the costly and time-consuming process of repeated histological preparation for neural developmental studies. Because

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imaging can be performed rapidly and repeatedly, the morphological changes of single specimens can be followed throughout development.

To demonstrate the potential of OCT to image in vivo developing neural morphology in a specimen which is highly scattering, a sagittal section through a Xenopus brain is shown in figure 9. This image shows high-resolution detail of internal brain morphology. Structure corresponding to the cerebellum, choroid plexus, and medulla oblongata are identified as well as a longitudinal section of the nasal tube and olfactory nerve as it enters the nasal placode. The dark internal regions correspond to the low-backscattering cerebral spinal fluid within the lateral, third, and fourth ventricles. Posteriorly, a longitudinal section of the spinal cord is observed.

Optical coherence tomography fills a niche between confocal microscopy and imaging modalities such as US, MRI, and CT. Specimens or morphology too large for light and confocal microscopy can still be imaged with high resolution using OCT. The cost, complexity, or size of other imaging technologies is sometimes prohibitive for routine, benchtop imaging. We have demonstrated a diode-based benchtop OCT instrument which provides 12 μm resolution with 3 mm of imaging penetration in Xenopus laevis tadpoles. The imaging capabilities of OCT will greatly extend the ability to investigate neural development. OCT complements the en face view of light microscopy by allowing imaging to be performed from the transverse, cross-sectional perspective. OCT also enables the visualization of neural development in more highly scattering, optically opaque specimens, and in some adult animal models. The ability of OCT to image with high penetration depths in highly-scattering specimens beyond the ranges of confocal and light microscopy enhances the identification of in vivo and in vitro morphological structure. The ability to recognize both normal and abnormal developing neural morphology has the potential for identifying mutations and neurological defects resulting from genetic disease, environmental toxins, or predetermined genetic manipulations.

High-quality histology is often difficult to obtain, costly, and time consuming for these small, fragile specimens. It is also impractical to histologically prepare the large numbers of specimens typically needed for genetic and developmental studies. OCT offers a significant alternative for rapidly assessing morphological changes. Used as a research tool for near real-time optical histology, OCT may reduce the need to sacrifice specimens. The use of OCT may also reduce the uncertainty associated with artifacts often attributed to histological preparation by identifying the in vivo orientation of the specimen and the biological structure prior to processing.

Significant technological advances will only serve to improve the ability of OCT to image and identify subtle structural features. New laser sources at other wavelengths in the near-infrared will poten-

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Figure 9. In vivo sagittal OCT image of Xenopus brain, nasal placode, and surrounding structure. Internal neural morphology in this highly-scattering tissue is clearly identified. Grey-scale bar indicates intensity of optical backscatter. Neural tissue appears more highly backscattering (grey to white) compared to the low backscattering cerebral spinal fluid (black). Abbreviations: cb=cerebellum, cp=choroid plexus, mo=medulla oblongata, ot=olfactory tract, sc=spinal cord.

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tially reveal new information since tissue scattering and absorbance properties in specimens depend highly on wavelength. Shorter coherence-length laser sources have already permitted resolutions on the order of 2-5 μm. These higher resolutions permit the imaging of individual cells. Advances in diode-pumped fiber laser sources may provide the larger optical bandwidths for cellular resolutions in a compact instrument no larger than the current benchtop diode-based OCT system. The fiber-based design of OCT allows for very small sample arm beam delivery systems such as a fiber-optic imaging catheter. Such a catheter can be used in utero to examine neural development in live-bearing species.

Optical coherence tomography represents a multifunctional, investigative, benchtop tool that could complement many of the existing imaging technologies and instruments available today. This technique has the potential to contribute significantly to the investigation of the developing nervous system.

1.13.8 Non-Invasive Assessment of the Dynamic Developing Cardiovascular System Using Optical Coherence Tomography

Abnormal cardiac function remains a leading cause of morbidity and mortality worldwide. Developmental animal models have been used to assess molecular and physiologic mechanisms of not only congenital cardiac abnormalities, but also genetic predisposition to acquired abnormalities. However, although imaging technologies exist which allow the changes of human adult cardiac function to be followed with time, comparable imaging does not exist for following cardiac function in the developing animal embryo. Previous technologies which have been applied to the visualization and quantitation of morphology and function in embryonic hearts include confocal microscopy, video light microscopy, magnetic resonance imaging, and high frequency ultrasound. The investigation of cardiac development in animal models, utilizing the powerful technologies of molecular and cellular biology, has recently been aggressively pursued to understand the mechanisms underlying cardiac dysfunction. However, severe limitations exist with current methods for assessing cardiac structure and dynamics in these animal models, frequently forcing investigators to analyze phenotypes with postmortem histopathology. The disadvantages of this approach include an inability to assess dynamic information and the sacrificing of mutants which typically have limited availability. The high-speed, high-resolution imaging which is possible with optical coherence tomography is well suited to overcome the limitations inherent in imaging the developing heart. OCT has also been applied to developmental biology where it can permit repeated in vivo imaging of a developing system in order to track developmental changes.

Optical coherence tomography have been used in the past year for the high-resolution assessment of structure and function in the developing *Xenopus laevis* cardiovascular system. Microstructural details were delineated with OCT, such as ventricular size and wall thickness, at 16 μm resolution and correlated with histology. Three-dimensional representation of the cardiovascular system was also achieved by repeated cross-sectional imaging. In addition to structural information, OCT, unlike technologies such as computed tomography and magnetic resonance imaging, provides high-speed in vivo imaging, allowing quantitative dynamic activity such as ventricular ejection fraction to be assessed. Optical coherence tomography is an attractive new technology for assessing cardiovascular development due to its high resolution, ability to image through nontransparent structures, and inexpensive portable design. In vivo and in vitro imaging is performed at a resolution approaching that of histopathology without the need of animal sacrifice.

We have demonstrated the imaging advantages of OCT for imaging cardiovascular development and assessing cardiovascular function by first acquiring in vitro images of the Xenopus cardiovascular system, generating three-dimensional datasets with projections, and comparing images to the corresponding histology. A high-speed OCT system with a laser source was then used to rapidly acquire in vivo images of a beating heart to illustrate the


future potential of this technology. In a manner similar to ultrasound M-mode echocardiograms, in vivo optical cardiograms were obtained from embryonic hearts and quantitative measurements of heart rate, end diastolic/systolic dimension, and ejection/filling times were made from cardiac chamber dimensions and wall displacements (figure 10). Finally, to illustrate the ability of OCT to detect variations in cardiac function, an inotropic agent was administered to alter the functional state of the heart. The high resolution, high-speed imaging provided by OCT should represent a powerful tool in assessing and understanding the molecular basis for abnormal cardiac development.

1.13.9 Publications


Brezinski, M.E., G.J. Tearney, S.A. Boppart, E.A. Swanson, J.F. Southern, and J.G. Fujimoto. "Optical Biopsy with Optical Coherence Tomog-
Chapter 1. Optics and Quantum Electronics

1.14 Transverse Dielectric Response from QED

Project Staff

Professor Peter L. Hagelstein

We are familiar with the notion that material effects can be included in electromagnetics problems through the introduction of a transverse dielectric constant. In Quantum-Effect Devices (QED), the coupling between radiation and matter is mediated through a $-\hat{j} \cdot \hat{A}$ coupling, which in turn must have something to do with the dielectric response. We have been curious about how the dielectric response can be developed directly from QED, and what the form of the resulting theory will be.

Surprisingly enough, that transverse dielectric response can be developed simply from QED through the use of infinite order Brillouin-Wigner theory. The basic idea is that if one has a Schrödinger equation of the form

$$E\Psi = (H_0 + V)\Psi$$

where the interaction $V$ creates or destroys a quantum, then the inclusion of single quantum creation or annihilation can be included to lowest order according to

$$E\Psi = H_0\Psi + V(E - H_0)^{-1}V\Psi$$

A Schrödinger version of linear response theory for the transverse dielectric can be generated directly using this approach, by taking the expectation value of the effective potential for photon exchange. The transverse radiation theory that results in this linear response theory is

$$\hat{H}_{\text{rad}} = \int \frac{1}{2} \varepsilon_0 |\hat{E}_{\text{T}}(r)|^2 + \frac{1}{2} \mu_0 |\hat{H}_{\text{T}}(r)|^2 \, d^3r + \int \int \hat{A}(r) \cdot (\hat{j}(r)[E - H_0]^{-1} \hat{j}(s)) \cdot \hat{A}(s) \, d^3r d^3s$$

Within QED, the interaction between the radiation field and matter is mediated through the vector potential. Consequently, the inclusion of the transverse dielectric response appears in the Hamiltonian in terms of vector potential operators instead of electric field operators.
A local transverse dielectric model can be obtained directly from the linear response Hamiltonian by assuming that the matter response is of short range; in this case the result can be written in the form

\[
\varepsilon(r, \omega) = \varepsilon_0 - \frac{1}{\omega^2} \int \left( \hat{j}(r)[E - \hat{H}_0]^{-1}\hat{j}(s) \right)_+ + \left( \hat{j}(s)[E - \hat{H}_0]^{-1}\hat{j}(r) \right)^T d^3s
\]

1.15 Maxwell's Equations and Configuration Space Photon Equations

Project Staff

Professor Peter L. Hagelstein

Quantum radiation theory is based on the second quantization of Maxwell's equations, analogous to the use of second quantization of the Schrödinger equation to develop many-body quantum theories. While we think of Maxwell's equations as being purely classical, we recognize that they must be closely related to single-photon configuration space equations. Various fundamental questions arise that relate to these issues; for example, are Maxwell's equations themselves fundamentally quantum mechanical? What evolution equations are satisfied by a single photon wavefunction? What is the connection between electromagnetic field solutions and the photon wavefunction? These questions are of current interest, as many researchers in quantum optics have developed models that are written in terms of photonic quantities rather than in terms of field quantities.

For free-space electromagnetic propagation, clear answers are available to these questions. Maxwell's equations are identical to single photon equations, and the single photon wavefunction can be derived directly from solutions to Maxwell's equations. Certain technical issues arise: the classical fields are real, the photon wavefunction is complex; and only solutions that correspond to positive energy states are allowed in the photon theory. If we use six-component photon wavefunctions, then the connection formula is

\[
\left( \frac{-i\sqrt{\varepsilon_0} E(r, t)}{\sqrt{\varepsilon_0 - \varepsilon_0 \omega^2}} \right) = \sqrt{\hbar c} G(r) \otimes \left[ \psi(r, t) - \psi^*(r, t) \right]
\]

where \( G(r) \) is an Amrein function

\[
G(r) = \lim_{\varepsilon \to 0} \frac{1}{(2\pi)^3} \int e^{i\mathbf{k} \cdot \mathbf{r}} \sqrt{\varepsilon - \varepsilon_0} |\mathbf{k}| d^3k
\]

\[
= -\frac{3}{4} \frac{1}{(2\pi)^{3/2}} \frac{1}{|\mathbf{r}|^{7/2}}
\]

The photon wavefunction is nonlocal in the electromagnetic fields, which is a nonintuitive result. It is possible to develop mathematical solutions for which the fields are strictly confined to a restricted volume, and where the photon wavefunction is finite outside of the region. Intuitively, we would expect to find the photon with a probability that corresponds to the local energy density; this intuition is not supported by the physical theory, and is in fact incorrect.

We have developed configuration space equations for a single photon in the presence of a local dielectric. There are a number of technical issues involved, but perhaps of interest here is an example of what the photon equation corresponding to Ampere's law looks like in a plane-wave basis; we may write

\[
i\hbar \frac{\partial}{\partial t} (\psi_E r) = \hbar c (\nabla \times \psi_E)
\]

\[
+ \mu_0 \hbar c \sum_{jk} \delta_{jj}(r) \otimes \frac{1}{|k|} \otimes K_{jk}(r) (\psi_E + \psi_{E*})
\]

where

\[
K(r) = \int \left( \hat{j}(r)[E - \hat{H}_0]^{-1}\hat{j}(r') \right)_+ + \left( \hat{j}(r)[E - \hat{H}_0]^{-1}\hat{j}(r') \right)^T d^3r
\]

and where

\[
\delta_{ij}(r) = \frac{1}{(2\pi)^3} \int \left[ \delta_{ij} - \frac{k_i k_j}{|k|^2} \right] e^{i\mathbf{k} \cdot \mathbf{r}} d^3k
\]

The interaction of the photon wavefunction with matter is explicitly nonlocal. This feature of the theory has generally not been taken into account by those working with photon models in the presence of linear or nonlinear dielectric response. This is one of a number of such technical issues associated with photon wavefunctions that need to be addressed when developing theoretical models based on photons.
1.16 Possible New Tetragonal Phase in PdH

Sponsor
Defense Advanced Research Projects Agency
Contract N66001-96-C-8634

Project Staff
Professor Peter L. Hagelstein, Steve Crouch-Baker, Michael McKubre, Mitchell Swartz

Advances in electrochemical techniques over the past 10 years have allowed palladium to be loaded with hydrogen by electrolysis to a loading ratio exceeding 0.95 with some reproducibility. As hydrogen primarily fills octahedral sites in fcc Pd at loadings below unity, there is the possibility that occupation of more highly excited tetrahedral sites begins to occur. At low loading, the excitation energy for the tetrahedral site occupation is on the order of 0.2 eV; at higher loadings this excitation energy is reduced somewhat. Significantly large tetrahedral occupation would be expected to put a strain on the lattice, and perhaps lead to a stretching of the lattice as we propose. Phase changes from the fcc structure to an elongated tetragonal version closely related to the fcc structure are known, an example being the fcc to tetragonal phase change in iron with interstitial carbon addition. Thermodynamic evidence for the existence of such a possible phase change is shown in figure 11. Clarification of this situation will require x-ray diffraction performed on highly loaded samples, which presents some technical problems as these materials outgas rapidly when electrolysis is stopped.

The fcc structure for PdH at unity loading is shown in figure 12, together with an illustration detailing the proposed tetragonal structure. The tetragonal sites lie in planes half way between the three planes of the fcc cube. These sites will be occupied with low probability at high hydrogen loading; correspondingly, a minor reduction in the octahedral loading will occur. Sufficient occupation of the T sites is proposed to stretch the entire structure along one axis, which would have the effect of bringing down the tetrahedral site energy. Equivalent thermodynamic data for PdD is not presently available; however, following trends between PdH and PdD at a slightly lower loading, we would anticipate that a tetragonal phase should occur at a slightly lower loading (assuming that it forms at all). We note that the hydrogen in tetrahedral sites is in closer proximity to metal atoms and to other hydrogen atoms in the lattice.

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84 SRI Inc., Palo Alto, California.
1.17 Sub-surface Interstitial Diffusion Barriers

Highly loaded PdH is very unstable against rapid outgassing. Wire samples of 1 mm diameter that have been loaded to a hydrogen atomic fraction of 0.95 are observed to outgas a few percent of their hydrogen loading within a minute of the termination of electrolysis. It is known that surface blocking layers made up of gold, copper, chromium or other materials are effective in preventing rapid desorption at lower loadings. Results obtained to date at SRI indicate that the effectiveness of deposited surface layers is degraded when the loading exceeds 0.90. While the mechanism for the failure of the surface blocking layer is not presently understood, it is believed to be connected to the very high activity of the interstitial hydrogen. Molecular hydrogen formation at the surface become energetically favorable by several tenths of an eV from Pd, which perhaps allows the hydrogen to exploit holes and weak links in the metal surface coating.

An alternate approach to this problem was prompted by a consideration of an experiment reported by Lipson, where the Pd surface was pre-treated with high temperature oxygen prior to electrochemical loading. After some deliberation, we conjectured that the principal effect of the oxygen in this case was to serve as an interstitial barrier to hydrogen diffusion, consistent with the observation of increased loading times for the resulting samples. A schematic of this is shown in figure 13.

This conjecture has led to a more general consideration of the possible utility of sub-surface interstitial barriers for inhibiting transport of hydrogen to and

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from surfaces. For example, very high loading of such samples is expected from the experience of Lipson and others, but at a price of substantially increased loading time. Nevertheless, the outgassing rates of such samples are considerably reduced, which improves the ease with which further experiments can be done. Suppose a cathode undergoes a phase change to a tetragonal (or some other) phase at high loading, then it becomes practical to freeze the sample to liquid nitrogen temperature, transport it to another facility, and then take time for the analysis.

While oxygen appears to be suitable for this application, other light interstitial elements may also be suitable. SRI has identified a synthesis route for the development of interstitial carbon diffusion barriers which involves a chemical reaction involving ethylene at 200°C, which is considerably lower than the 1000°C oxygen flame deposition used by Lipson.86

1.18 Electromigration with Interstitial Diffusion Barriers

Sponsor
Defense Advanced Research Projects Agency
Contract N66001-96-C-8634

Project Staff
Professor Peter L. Hagelstein, Steve Crouch-Baker, Michael McKubre, Mitchell Swartz

Hydrogen in palladium behaves as if it has a fractional positive charge in the presence of an applied electric field within the metal hydride. This effect can be used to move hydrogen within a metal hydride to perhaps increase the peak loading in part of a sample. This approach is currently being pursued at a number of laboratories as a way to stimulate anomalies in metal hydrides and deuterides.
1.18.1 Possible Atmospheric Synthesis of Vacancy Phase PdH and NiH

Sponsor
Defense Advanced Research Projects Agency
Contract N66001-96-C-8634

Project Staff
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Vacancies are present in metals at very low concentrations, unless some procedure has been used specifically to generate vacancies. Metals that have been irradiated with MeV electrons at high dosages achieve a saturation density of defects that can be on the order of a few tenths of a percent. Consequently, there was much interest in a recent report\cite{Fukai1994} of the observation of the production of 25 percent metal vacancies in NiH and 18 percent metal vacancies in PdH in a high temperature cell at high pressure. These vacancies were found to be stable following outgassing of the hydrogen, resulting in the production of new low density pure metals.

The generation of high levels of metal atom vacancies is reasonably well understood theoretically. Essentially, hydrogen atoms in the vicinity stabilize the vacancy, by a few tenths of an eV per neighboring hydrogen. In NiH, a metal vacancy is energetically favored by roughly 0.5 eV; in PdH the vacancy is only a few tenths of an eV away from being energetically favored. These numbers are vastly different than the roughly 1.5 eV formation energy associated with vacancy creation in the pure metal. Consequently, thermodynamics predicts that host metal lattice vacancies should form spontaneously in highly loaded NiH and PdH, limited only by the vacancy diffusion rate from the surface or from internal sources of vacancies. The high temperature high pressure synthesis reported previously was observed to occur at a rate limited by this vacancy diffusion process.

Unfortunately, only small vacancy phase samples can be produced using a high temperature high pressure synthesis. These new materials will be of limited utility until an improved method for their syn-

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\cite{Cellani1996}

\cite{Fukai1994}
thesis is discovered. We have proposed several novel methods that may lead to the product ion of vacancy phase metal hydrides under atmospheric conditions. These proposed routes include:

1. Using electrolysis to produce highly loaded cathodes, and then freezing and transporting to an electron irradiation facility. Prolonged exposure of the loaded sample to electron irradiation will produce vacancies that will be stabilized by the hydrogen. The relaxation of interstitial metal atoms will perhaps produce other kinds of defects in the hydride, which may lead to further vacancy production.

2. Using irradiation to produce a high defect density sample, then using electrolysis to develop high loading. The hydrogen will stabilize the vacancies initially present, and the interstitial metal atoms will again produce other defects as above.

3. If Pd or Ni is deposited on a highly loaded substrate, the vacancy phase is thermodynamically favored, and can perhaps be grown directly. One speculative approach is to use electrolysis or electroless deposition on the metal hydride. Another related approach is to use electron beam deposition of metal atoms with codeposition of hydrogen using an ion beam, as suggested by G. Hubler of NRL.

As a first step toward working with irradiated metal samples, we have used the van de Graaf accelerator at the high voltage lab to produce irradiation damage in Pd. The presence of vacancies was determined through resistivity measurements performed before and after irradiation. A significant increase in the resistivity was caused by substantial defect production. This resistance increase was time-dependent due to room-temperature annealing as shown in figure 15.

1.19 Models for Anomalous Energy Transfer

Project Staff

Professor Peter L. Hagelstein

A variety of anomalies have been claimed to be observed in metal hydrides and metal deuterides. While not presently accepted by the scientific community, many of these experiments appear to be sufficiently solid to warrant some effort to understand them. To this end, we have endeavored over the past several years to explore new lattice-nuclear coupling mechanisms that might lead to a basic understanding of the anomalies. While many such proposed mechanisms have been examined by us, few of these have survived following detailed analysis.

For example, the elastic neutron hopping effect that we described last year has now been shown to require considerable lattice energy transfer in order to operate, and appears not to be observable. We have obtained some new results that pertain to the problem of anomalous energy transfer through phonon mode rearrangement, and we have shown, using moment theorems, that this mechanism as initially proposed for a harmonic lattice or weakly nonlinear lattice is not capable of transferring enough energy to account for the claimed anomalies.

When two nuclei approach each other closely, the Coulomb interaction and the nuclear interaction between them constitute an exceedingly nonlinear interaction in the phonon operators. For example, the relative position of the nuclei can be expressed in terms of the phonon mode amplitudes, and the close approach interactions are strongly nonlinear in the relative coordinate. We have considered the possibility that anomalous energy transfer might be mediated through this high order nonlinearity, and that such a mechanism might mediate energy transfer required for fusion reactions. A model for this process has been developed, and we have found that it is not possible to transfer a delocalized anomalus energy quantum to a linear lattice as would be required for the model to be relevant to the claimed anomalies. This result indicates that either lattice nonlinearities or other physics would have to be found to account for a $d + d \rightarrow 4\text{He}$ reaction mechanism as proposed by Fleischmann and Pons in 1989.

Over the course of our efforts, we have examined quite a large number of proposed mechanisms to account for the claimed anomalies. While the
anomalies have largely been dismissed as being impossible, very little in the way of analysis has been put forth to justify this seemingly obvious conclusion. Although we have endeavored to put the anomalies on some kind of solid theoretical basis, it has turned out that our results have confirmed this conclusion for essentially every model that we have considered seriously. Of all of the approaches that we are aware, only one approach presently remains under consideration as a possibly viable candidate explanation for the anomalies; this approach includes processes associated with anomalous energy transfer as outlined below.

1.19.1 Some General Conclusions

Some of the negative results we have obtained can perhaps be developed into more general conclusions or theorems that pertain to the anomalies, a few of which are listed below:

- Anomalies require energy exchange with the lattice. There is one exception to this: low-level neutron emission can perhaps result in principle from conventional lattice physics as discussed further below.
- Anomalous energy exchange with a linear lattice appears to be forbidden.
- The Coulomb barrier prohibits direct nuclear interactions between nuclei except for possible hydrogen isotope reactions at low levels.

While there may be other conclusions that could be stated, these are some of the more important ones relevant to our recent modeling.

If the anomalies are real, then within the framework of our approach they must occur as a result of anomalous energy transfer from the lattice to atoms or nuclei within the lattice. Whether anomalous energy transfer from the lattice can occur is in our view presently unknown. On the one hand, we have not succeeded in ruling out such an effect; on the other hand, if the experimental claims are right, then they appear to be consistent with and explainable through such a mechanism.

1.19.2 Nuclear Close Approach

If nuclei approach each other closely some small fraction of the time, how could it be proven? Certainly the nmr lineshape would contain information about nuclear close approach probabilities in the far wings, but it is not practical to do such measurements for such low probability configurations. From this perspective, using dd-fusion as a diagnostic on the short-range pair-correlation function appears to be an attractive option, in spite of the controversy surrounding such measurements.

Following the initial claims of the observation of neutron emission from metal deuterides in 1989, many experiments were done that placed low upper limits on the observed neutron emission rates. Supported by theoretical calculations for molecular D₂ that predicted very low reaction rates (on the order of 10⁻⁵⁰ sec⁻¹ per pair), theorists argued that the rates for metal deuterides would also be quite low, and the notion that dd-fusion could occur be observed in metal deuterides was rejected. Realistic computations specific to fcc metal deuterides were not completed until 1993, and in these computations the inclusion of screening effects on the Angstrom scale led to much higher predicted reaction rates (on the order of 10⁻³⁰ sec⁻¹ per pair).[^s2]

The experimental claims are for rates on the order of 10⁻²³ sec⁻¹ per pair.

While there may be other conclusions that could be stated, these are some of the more important ones relevant to our recent modeling.

If the anomalies are real, then within the framework of our approach they must occur as a result of anomalous energy transfer from the lattice to atoms or nuclei within the lattice. Whether anomalous energy transfer from the lattice can occur is in our view presently unknown. On the one hand, we have not succeeded in ruling out such an effect; on the other hand, if the experimental claims are right, then they appear to be consistent with and explainable through such a mechanism.

Observations of low level neutron emission from highly loaded PdD continue to be reported. Although the origin of such signals remains poorly understood, it would be of interest to clarify the situation for tetrahedral occupational at high loading, and the possible stimulation of a new tetragonal phase in PdH at high loading. The computation of the screened dd-fusion rates in PdD cited above were performed for octahedral occupation of the deuterium, but equivalent results are not yet available for other configurations; the screened fusion rates would be greatly increased for a tetrahedral-octahedral pair.

Observations of low level neutron emission from highly loaded PdD continue to be reported. Although the origin of such signals remains poorly understood, it would be of interest to clarify the situation for tetrahedral occupation, and to understand whether the pair-correlation function for screened O-T tunneling supports an observable fusion rate. If this effect is observable, then there is a possibility of investigating the short range pair correlation function using neutron signals as a diagnostic to study tetrahedral deuterium occupation. It is not obvious that there are other approaches that can access this quantity in condensed matter physics.

1.19.3 Effects Induced by Anomalous Energy Transfer

Anomalous energy transfer from the lattice would induce a number of effects, including atomic recoil events and nuclear reactions. A model for such processes would include a description of the lattice physics associated with the energy transfer, and microscopic physics that describes what effects occur as a result of the energy transfer. As the overall model includes two disparate physical phenomena, we can simplify the analysis considerably by separating the problem into two parts. The easier part focuses on what effects are induced if anomalous energy transfer occurs; more difficult is the part of the problem that derives an anomalous energy transfer effect.

Nevertheless, it is relatively straightforward to consider the easier part of the problem, and ask what effects are induced by anomalous energy transfer. We have examined this problem, and we find that the reactions that would be expected appear to be very relevant to the experimental claims. Anomalous energy transfer from the lattice would stimulate alpha and beta decay reactions, and we can begin to make tentative connections between claims and mechanisms; these are summarized below.

- Claimed excess heat effect: Perhaps due to lattice-induced alpha decay of heavy impurities with mass above 140 (see figure 16). For example, measured impurity levels of heavy isotopes vary between 10 ppm and 200 ppm; exothermic alpha decays of these impurities could perhaps lead to net energy production on the order of 100 eV per Pd nucleus (as claimed) with minor residual radioactivity (depending on which impurities are present). Sm and Ce are optimum "fuels" in this sense (see figure 17); cathodes doped with these elements have in some cases been claimed to be superior heat-producers. We note that a recent claim\(^\text{90}\) of quantitative \(^3\)He production, if correct, strongly supports a p+d fusion mechanism that also requires anomalous energy transfer.

- Claimed tritium effect: Perhaps due to alpha decay of \(^7\)Li. Tritium production is claimed to occur without observable 14 MeV DT fusion neutrons, which implies that it would be generated at low energy. Lattice-induced alpha decay of \(^7\)Li is computed to lead to a sufficiently slow tritium final state energy at modest generation rates to be a candidate reaction. We note that lattice-induced electron capture of \(^3\)He is an alternate route to tritium production.

- Claimed alpha emission: One of the basic reaction mechanisms within the model.

- Claimed \(^4\)He generation: Recent experiments have claimed the observation of quantitative \(^4\)He emission associated with energy production. This could be a signature of lattice-induced alpha decay, or perhaps fusion.

- Claimed induced radioactivity: Three Pd cathodes were observed become highly radioactive during electrolysis in LiOD. We conjecture that this may have occurred through lattice-induced beta decay of isotopes of Pd and of neighboring impurity elements.

\(^*\) Figure 16. Alpha decay energy (MeV) as a function of nuclear mass \(A\) for naturally occurring isotopes.

\(^*\) Figure 17. Lattice input energy required to induce a decay rate of 1 sec\(^{-1}\).

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\(^{90}\) Y. Arata and Y.-C. Zhang, "Helium (\(^1\)He, \(^1\)He) within Deuterated Pd-Black," Proc. Jpn. Acad. 73B: 1 (1997).
The experimental claims of anomalies are presently not accepted by the scientific community; the existence of an anomalous energy transfer effect is unproven. At present our view is that if the anomalies are right, then they would occur as a consequence of anomalous energy transfer from the lattice through the mechanisms outlined above.

1.19.4 A Model for Lattice-Induced Radioactivity

We have conjectured that there exists a route by which the lattice can transfer an anomalously large quantum of energy to the lattice, and we have outlined a theory above for a specific coupling mechanism that might mediate such anomalous energy transfer. Such a model, if successful, will ultimately provide predictions that might be characterized through an equivalent energy transfer probability. For example, we know more or less what reactions will follow if we can transfer a given amount of energy to a nucleus; the model above if successful will be most useful in providing estimates of how much energy what fraction of the time for a given lattice configuration. Some progress can be made toward making sense of the experimental data by simply parameterizing this probability of energy transfer in an empirical model.\(^91\)

For example, anomalous lattice energy transfer to nuclei would perhaps induce alpha and beta decay if the amount of energy transfer is sufficiently large. For a given amount of energy input, we can estimate the relevant nuclear decay rates from the Gamow tunneling model and from the Fermi beta decay model. If we denote one of these microscopic reaction rates (produced or enhanced with lattice energy transfer \(\varepsilon\)) through \(\gamma(\varepsilon)\), then the average reaction rate is

\[
\Gamma = \int P(\varepsilon)\gamma(\varepsilon) \, d\varepsilon
\]

where \(P(\varepsilon)\) is the probability that an energy transfer of \(\varepsilon\) occurs. We have made estimates of lattice-induced alpha and beta decay reactions for the stable nuclei.

We have applied this model to data collected from lattice-induced cathode activation that was claimed several years ago. In this experiment, three Pd cathodes were observed to have become radioactive during electrochemical loading in heavy water (the effect was discovered 45 days after the activation).\(^92\) Unstable isotopes near Pd were observed and identified from HPGe gamma measurements, in relative amounts that appear to be hard to account for. We postulated that the radioactivity is perhaps due to lattice-induced beta decay and electron capture of the Pd isotopes and low levels of Ru, Rh, Ag and Cd impurities. A Gaussian model for the energy transfer model was assumed

\[
P(\varepsilon) = f_0 e^{-\varepsilon^2/\delta^2}
\]

and the parameters \(f_0\) and \(\delta\) were least squares fit to the yield of unstable isotopes. A summary of the results is presented in table 1, where the optimized energy transfer is parameterized by \(\delta \varepsilon = 357\) keV. The model appears to be consistent with most of the observed radioactivity, but cannot account for the observation of \(^{99}\)Rh. A systematic analysis of possible lattice-induced reactions from all isotopes of these elements was performed, and the results are largely consistent with upper limits established on isotopes that were not observed. The result from the empirical model is for \(^{110}\)Ag; the fractional yield for \(^{110m}\)Ag is unknown but is expected to be small. The accuracy of the model numbers is much lower than the precision that the digits indicate; the accuracy is no better than factor of three.


### Table 1: Basic model results for unstable isotope production compared with experiment.

<table>
<thead>
<tr>
<th>Unstable Product</th>
<th>Stable Parent</th>
<th>Lattice-induced Mechanism</th>
<th>ΔE(MeV)</th>
<th>n_i (model)</th>
<th>n_i (exp’t)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{99}$Rh</td>
<td>$^{99}$Ru</td>
<td>$\beta^-$ decay</td>
<td>2103</td>
<td>0</td>
<td>$2.74 \times 10^9$</td>
</tr>
<tr>
<td>$^{101}$Rh + $^{103m}$Rh</td>
<td>$^{101}$Ru</td>
<td>$\beta^-$ decay</td>
<td>541</td>
<td>$4.7 \times 10^{10}$</td>
<td>$3.40 \times 10^{10}$</td>
</tr>
<tr>
<td>$^{102}$Rh + $^{102}$Rh</td>
<td>$^{102}$Pd</td>
<td>$e^-$ capture</td>
<td>1078</td>
<td>$2.9 \times 10^{10}$</td>
<td>$1.84 \times 10^{10}$</td>
</tr>
<tr>
<td>$^{103}$Ru</td>
<td>$^{103}$Rh</td>
<td>$e^-$ capture</td>
<td>767</td>
<td>$6.0 \times 10^9$</td>
<td>$3.67 \times 10^9$</td>
</tr>
<tr>
<td>$^{105}$Ag</td>
<td>$^{105}$Pd</td>
<td>$\beta^-$ decay</td>
<td>1348</td>
<td>$1.7 \times 10^{10}$</td>
<td>$2.62 \times 10^{10}$</td>
</tr>
<tr>
<td>$^{106m}$Ag</td>
<td>$^{106}$Cd</td>
<td>$e^-$ capture</td>
<td>290</td>
<td>$5.3 \times 10^9$</td>
<td>$4.51 \times 10^9$</td>
</tr>
<tr>
<td>$^{110m}$Ag</td>
<td>$^{110}$Pd</td>
<td>$\beta^-$ decay</td>
<td>997</td>
<td>$[1.2 \times 10^{13}]$</td>
<td>$6.56 \times 10^8$</td>
</tr>
</tbody>
</table>

### 1.19.5 Possible Anomalous Energy Transfer Mechanisms

There are experimental claims for observations of anomalies, and there appears to be a set of lattice-induced reactions that may be relevant as discussed above. However, the key to this problem lies in the question as to whether the lattice can transfer a large energy quantum. We discuss here some of the issues and approaches.

As an example, we consider the case of a lattice-induced alpha decay mechanism. There are, in principle, two general approaches to the problem: either the lattice (somehow) transfers energy directly into the nucleus, inducing a subsequent decay; or else the decay proceeds directly, with the final state lattice energy being significantly less than the initial state lattice energy. We currently have no experimental input on this point, but simply note that the difference between the approaches would show up directly in the associated gamma spectrum. Our theoretical efforts so far have led us in the direction of the second of these approaches, primarily as the impact of a nucleus disintegrating on the lattice is much greater than that of a nucleus being excited.

There appears to be two basic approaches to the problem of anomalous energy transfer and both appear to have associated advantages and disadvantages. In the first approach, we imagine that a light nucleus has tunneled some distances toward a heavy nucleus so that the effective interaction is strongly nonlinear. In this case, the anomalous lattice energy transfer is mediated by this strong nonlinearity directly as a phonon operator, with damping due to the electrons. The energy transfer in this case can perhaps be estimated from the construction

$$e^{-G(r_{12})} \psi_i(E_o) = \sum E C(E) \psi_f(E)$$

Here we have assumed that the principal effect of the disintegrating nucleus on the lattice wavefunction is to have induced a tunneling factor that prohibits the nuclei from approaching. After the nucleus has disintegrated, the lattice is projected into a superposition of final states with a spread in energy.

In the second approach, the disintegration of the nucleus causes a rearrangement of the lattice, and the energy is transferred as an energy fluctuation. This is similar to the energy transfer mechanism that we have described in previous reports. This mechanism is known not to be sufficient to account for anomalous energy transfer in a harmonic lattice, and we are assuming that substantial nonlinear effects will have to help.

The case can be made that neither of these mechanisms is capable of transferring enough energy in a harmonic lattice to induce either alpha or beta decays. The basic problem appears to be that the relevant interaction is simply not strong enough and does not involve enough lattice sites to do the job. Consequently, we are examining whether there are other effects that might produce a larger impact on the lattice. The most obvious candidate for this is a phase change. For example, if a local nuclear disintegration stimulates a lattice rearrangement over a large number of neighbors, then this will have a much greater impact on the electronic structure and the phonon mode structure than would the introduction of an isolated vacancy.

### 1.19.6 The Japanese NHE Effort

A major effort was initiated in Japan some years ago to investigate the claims of excess heat and to determine whether the excess heat effect could be useful for the Japanese energy economy. While this effort has produced numerous important and interesting results pertaining to the loading and met-
allurgy of palladium deuteride, to date no episodes of excess heat were observed in several hundred experiments using mass flow calorimeters. From these results, a conclusion is being reached in Japan that there is no excess heat effect; the program is scheduled to be terminated in early 1998.

The Japanese effort took as its premise that a successful program could be developed by reproducing the SRI and Pons-Fleischmann excess heat experiments. This was attempted in collaboration with the SRI staff and with Pons and Fleischmann. Very accurate calorimeters were developed, protocols were developed that result in reproducible loading of deuterium into Pd to high levels, and a serious effort was put into a careful attempt at replicating key experiments. That a positive confirmation was not obtained is the source of much concern and discussion for those in the field that believe that there is an excess heat effect; for the skeptics this result provides the strongest evidence to date that there is no effect.

Prior to the start of the program, SRI had reported more than 40 episodes of excess heat in mass flow calorimeters. Pons and Fleischmann had reported reproducible observations of very high levels of excess heat production in boil-off experiments. A strong effort dedicated to replicating these results seemed to be the right thing to do.

So what happened? Why did this effort fail to replicate an excess heat effect? The answer is obvious if in fact no excess heat effect exists; otherwise the answer is much less obvious. In any event, it is useful here to attempt to summarize some of the issues involved.

The basic Pons-Fleischmann experiment to be tested involved heat-flow calorimetry and a dynamic operating temperature over a wide range of temperatures. The current generation of mass flow calorimeters are isothermal, and cannot be used directly for these experiments. For the Japanese, heat-flow calorimetry is in general suspect as they have observed conditions under which a specific heat-flow calorimeter gave an excess heat effect inside of a mass-flow calorimeter that gave no excess. Whether this is the case or not for the Pons-Fleischmann heat-flow calorimetry, the Japanese have seen no episodes of excess heat in these experiments that are in excess of the accuracy that the Japanese associate with the measurements.

The initial SRI results appear to be free of these problems as they were done using mass flow calorimeters. In this case, a problem of a different nature has arisen. In the initial SRI work, highly loaded cathodes were run at high current density, and the largest excess heat results were obtained with cathodes that maximized both variables. The ability of a cathode to maintain high loading at high current density is a function of the cathode batch and associated processing, as well as the electrochemical protocol used. Most of the SRI positive results were obtained with an early batch from Engelhard, which appears to have been special; subsequent efforts over several years at SRI and in Japan have failed in efforts to maintain high loading at high current density using other cathodes. It could be said that the conditions of the initial positive SRI experiments have not been reproduced (although extensive efforts have been devoted to reproduce relevant conditions both at SRI and in Japan); whether this is important, or why it might be important, is presently unknown.

The Japanese results underscore the issue of reproducibility of the experiments. The SRI positive results appeared to be reproducible at SRI; it was not appreciated that serious difficulties would be encountered reproducing the loading and current characteristics. Reproducibility is also a serious issue in recent Pons-Fleischmann experiments at the IMRA laboratory in France, as discussed recently by Pons. Unfortunately, if there is a real excess heat effect and one does not know what it takes to make it work, then one also does not know why it doesn't work in any given experiment.

Ultimately, these issues will not be sorted out until whatever physical mechanisms are involved are clarified.

1.19.7 Where To Go From Here?

It was conjectured initially that fusion was somehow occurring via some mechanism that was not understood. This notion has been rejected by the scientific community. That fusion neutrons would be expected at very low levels by conventional processes is not in question; that the magnitude of the effect is calculated to be significantly larger than the molecular D$_2$ limit was perhaps unexpected. If substantial tetrahedral deuterium occupation in PdD can be produced at high loading, then the question of the magnitude of screened reaction rates must be revisited.

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The Japanese effort has in essence tested the conjecture that high deuterium loading produced electrochemically in Pd leads to an excess heat effect. Considerably more than this was tested by the Japanese, but for the purpose of this discussion, this is sufficient. There is no compelling physical mechanism that underlies such a conjecture. From the results of the Japanese work, the only conclusion possible is that this conjecture is incorrect.

SRI has proposed from their positive results that to produce excess heat that high loading is required, that it must be maintained for hundreds of hours, that a high current or current density is required, and that a flux of deuterium passing through the interface is needed. This proposal has not, in our view, been verified or disproven, largely because no one has been able to arrange for these conditions to be met simultaneously since the early SRI efforts. Why this protocol, as opposed to any other, should produce any anomalous effects is not understood.

So where do we go from here?

If the anomalies are real, it is essential to make progress on clarifying whatever underlying physical mechanism is responsible. Once the physical mechanism is understood and proven, we can decide what applications are feasible. For example, a lattice-induced alpha decay mechanism has the potential for energy production, radioactive waste reduction, and tritium generation.

Unfortunately, experiments that have the greatest potential for shedding light on mechanism have generally had the poorest record for reproducibility. Electrochemically-induced radioactivity clearly provides the most detailed results from which questions concerning mechanism can be addressed. While it appears to have been observed by accident, at present no one knows how to reproduce it.

Other anomalies are claimed to be observable with varying degrees of reproducibility. Strong evidence for quantitative helium generation associated with excess heat has been presented, and gamma emission near 90 keV is claimed with an associated yield of 0.5 percent has been claimed recently by an Italian group.\textsuperscript{94} Reproducible alpha and proton emission at MeV energies from PdD and from PdH has also been claimed recently.\textsuperscript{95} Further tests to confirm these effects are ongoing.

