

21. Atomic Resonance and Scattering

Academic and Research Staff

*Prof. D. Kleppner, Prof. D.E. Pritchard, Dr. R. Ahmad-Bitar²⁵, Dr. D. Derouard²⁵,
Dr. T. Ducas²⁵, Dr. R.G. Hulet, Dr. G. Lafyatis, Dr. P. Moskowitz, Dr. J-L. Picque²⁵*

Graduate Students

V. Bagnato, P.P. Chang, R. Flanagan, T. Gentile, P.L. Gould, E. Hilfer, B.J. Hughey, M.M. Kash, P.D. Magill, P. Martin, A.L. Migdall, W.P. Moskowitz, S. Paine, E. Raab, T.P. Scott, B.A. Stewart, R. Stoner, R. Weisskoff, G.R. Welch

Undergraduate Students

S-K. Chan, M. Lee, S. LePage, I. Nir, S. Osofsky

21.1 Resonant Multiphoton Ionization of Hydrogen

National Science Foundation (Grant PHY83-06273)

National Bureau of Standards (Grant NB83-NAHA-4058)

Lawrence R. Brewer, Martin Ligare, Daniel Kelleher, Daniel Kleppner

Multiphoton processes are playing an increasing role in atomic, molecular, and optical physics. Hydrogen, because of its simplicity, lends itself to rigorous theoretical analysis and provides a natural benchmark system. We have completed a study of 4-photon ionization of hydrogen in the vicinity of 365 nm. Three photons are resonant with the 1s–2p transition: the fourth ionizes the 2p state at threshold.

The major experimental challenge was to generate 365 nm light and to understand its spectral and temporal properties in detail. The generation system was based on a pulsed Quantal 581 Nd:YAG laser at 1.06 μ and a pulsed dye laser and oscillator at 554 nm. The 1.06 μ and 554 nm light were combined in a KDP crystal to generate light pulses at 565 nm with the following properties: energy — 10 mJ; pulse length, approx 8 ns; spectral width, 0.25 cm^{-1} .

The special distribution of the laser light was monitored with a Reticon diode array. The temporal behavior was studied using a high speed oscilloscope. The properties of the laser were analyzed in terms of a model for multimode operation of the Nd:YAG light source and found to be in good agreement with the model. The power was measured on a shot-by-shot basis using a carefully calibrated Scientech 362 power meter.

²⁵Visiting Scientist

Measurements were made in a simple atomic beam apparatus in which hydrogen could be produced at temperatures as low as 5 K. The experiments were carried out at 78 K. The photoions were swept out by a small field and detected with an EMI D233 electron multiplier.

The ion yield was measured as a function of frequency for a variety of pulse energies and spectral profiles. The results were compared to the prediction of the theory of Holt, Raymer, and Reinhardt.¹ This theory is nonperturbative, and include processes such as stimulated recombination, which are often negligible. Fig. 21-1 compares theory and experiment for one value of pulse energy.

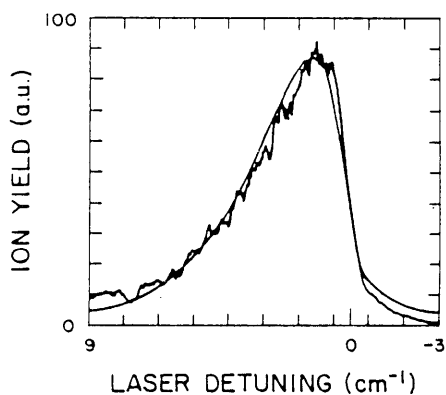


Figure 21-1: Comparison of theory¹ and experiment on the ionization yield for 4-photon resonant ionization in hydrogen. The pulse energy is 7.1 mJ. Zero on the laser detuning scale is the ionization threshold.

This work confirms the validity of the theory of Holt, Raymer, and Reinhardt, and demonstrates that multiphoton processes in hydrogen can be understood in detail for powers up to 3×10^{10} watts cm^{-2} .

References

1. C.R. Holt, M.G. Raymer, and W.R. Reinhardt, "Phys. Rev. A 27, 2971 (1983).

Publications

- Kelleher, D.E., M. Ligare, and L.R. Brewer, "Resonant Four-Photon Ionization of Atomic Hydrogen," Phys. Rev. A, accepted for publication.
- Brewer, L.R., "Resonant Multiphoton Excitation of Atomic Hydrogen," Ph.D. Thesis, Physics Department, M.I.T., October 1984.

21.2 Atoms in Strong Magnetic Fields

National Science Foundation (PHY83-06273)

Michael M. Kash, George R. Welch, Ishai Nir, Daniel Kleppner

Understanding the diamagnetic spectrum of an atom with a single valence electron in a strong

magnetic field presents a formidable challenge to theory and experiment.¹ The classical and quantum mechanical equations of motion are easily constructed, but in spite of the fundamental simplicity of the system the solutions are elusive and our understanding is far from complete. Numerical techniques have been applied to both the classical² and quantum³ dynamics, but the results possess features which are difficult to interpret. Construction of an approximate constant of motion is a promising method for predicting the spectrum of a hydrogen atom in a uniform magnetic field.⁴

We have produced and detected Rydberg atoms in a lithium atomic beam using cw dye lasers. This represents substantial progress toward our research goal of studying highly excited atoms in a strong magnetic field by a cw laser spectroscopy. Our primary task is to measure the level anticrossing sizes and linewidths in the diamagnetic spectrum of lithium.

The excitation of lithium Rydberg atoms is performed with a two-step, three-photon process. The first step is a two-photon transition from the 2s to the 3s state. This process is detected by observing the cascade fluorescence ($3s \rightarrow 2p, 2p \rightarrow 2s$) through a fiber optic bundle with a sensitive photomultiplier tube. The transition is excited by 735 nm light from a Coherent ring dye laser. The $2p \rightarrow 2s$ fluorescence as a function of ring laser frequency is shown in Fig. 21-2.

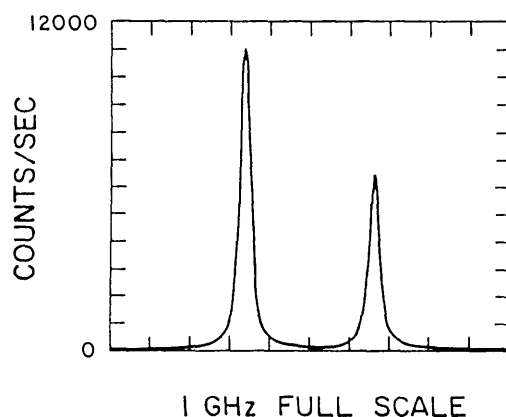


Figure 21-2: Cascade ($2p \rightarrow 2s$) Fluorescence as a function of a laser frequency. The two peaks reflect the hyperfine structure of the $3^2s_{1/2}$ and $2^2s_{1/2}$ states of lithium 7.

The ring laser is stabilized against slow drift by locking it to the signal. The second step is a one-photon transition from the 3s state to the np state. This has been observed by observing a decrease in the cascade fluorescence as a Coherent linear dye laser is scanned through the resonance. Digital lock-in techniques are used to enhance the signal-to-noise ratio. The decrease in the cascade fluorescence as a function of linear laser frequency for the 15 p state is shown in Fig. 21-3. To minimize the electric field in the atom's rest frame, the atomic beam is parallel to the axis of a solenoid superconducting magnet. The interaction region consists of an aluminum cylinder whose axis is parallel to the atomic beam. The cylinder also contains prisms to

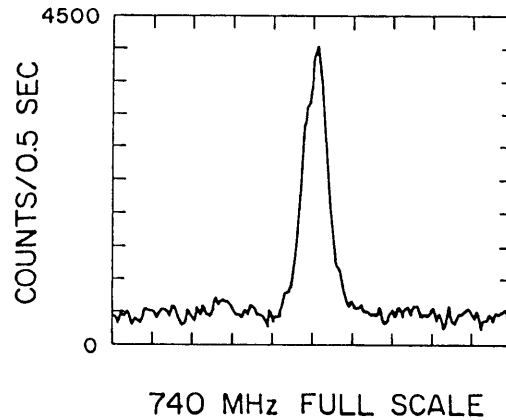


Figure 21-3: Decrease in cascade fluorescence as a function of laser frequency at the 15p state. The measurement time was 6 seconds and the chopping frequency was 665 Hz.

deflect the lasers so that the laser and atomic beams intersect at right angles, to minimize Doppler broadening. The interaction region combines efficient collection of cascade fluorescence with good geometric rejection of scattered laser light. Also, the entire cylinder may be floated at -20 kV to cause field ionization of Rydberg atoms just outside the interaction region. Using a magnetic field-insensitive surface barrier diode detector for the Rydberg electrons, field ionization is expected to provide a substantially higher signal-to-noise ratio detection technique than the decreased cascade fluorescence. Preliminary observation of field ionization confirms this.

We expect to start taking data in the near future.

References

1. J.C. Gay, "High-Magnetic-Field Atomic Physics," in H.J. Beyer and H. Kleinpoppen (Eds.), *Progress in Atomic Spectroscopy* (Plenum 1984).
2. J. Delos, S.K. Knudson, and D.W. Noid, "Trajectories of an Atomic Electron in a Magnetic Field," *Phys. Rev. A* 30, 1208 (1984).
3. J.C. Castro, M.L. Zimmerman, R.G. Hulet, D. Kleppner, and R.R. Freeman, "Origin and Structure of the Quasi-Landau Resonances," *Phys. Rev. Lett.* 45, 1780 (1980).
4. D.R. Herrick, "Symmetry of the Quadratic Zeeman Effect for Hydrogen," *Phys. Rev. A* 26, 323 (1982).
5. M.L. Zimmerman, M.M. Kash, and G.R. Welch, *J. de Phys. Col. C2*, 43, 113 (1982).

21.3 Cavity Quantum Electrodynamics

National Science Foundation (Grant PHY84-11483)

Joint Services Electronics Program (Contract DAAG29-83-K-0003)

U.S. Navy – Office of Naval Research (Contract N00014-79-C-0183)

Thomas R. Gentile, Eric Hilfer, Barbara J. Hughey, Daniel Kleppner, Randall G. Hulet

a) Spontaneous Emission in a Tuned Cavity

During the past year we have continued work on an apparatus to study spontaneous emission of a single Rydberg atom in superconducting cavity. The goal is to study a single excited atom in an effective two-level configuration interacting with the single mode of cavity in its photon ground state. At sufficiently high Q the atom-cavity system behaves like a pair of coupled oscillators, one man-made, the other atomic. The experiment will be carried out with calcium at a frequency of about 35 GHz.

b) Inhibited Spontaneous Emission

Inhibited spontaneous emission is being investigated using circular states of cesium and a time-of-flight apparatus to measure the spontaneous lifetime on the transition $n = 22 \rightarrow 21$. Spontaneous emission will be inhibited by two conducting plates separated by $\lambda / 2 = 0.25$ mm. Apparatus has been built and is being tested.

21.4 Trapping of Neutral Atoms

U.S. Navy - Office of Naval Research (Contract N00014-83-K-0695)

Joint Services Electronics Program (Contract DAAG29-83-K-0003)

Vanderlei Bagnato, Riyad Ahmad-Bitar, Gregory P. Lafyatis, Philip E. Moskowitz, David E. Pritchard, Eric Raab

An experimental apparatus has been set up to slow and trap neutral atoms. We have successfully slowed a Na-beam using the technique of Phillips and Metcalf.¹ Atoms in a specific magnetic sublevel are slowed by radiation pressure; a specially tailored magnetic field produces a Zeeman shift which compensates the changing doppler shift of the slowing atoms and keeps them resonant with the laser. The light scattered from the Na beam is observed by a photodiode array. Presently, we are modeling the dynamics of the deceleration to fit our observations of slowing as a function of laser frequency, laser intensity, polarization, and magnetic field strength. Our present apparatus has a 30 mK deep magnetic trap that should be able to confine some of the decelerated (paramagnetic) Na atoms.

In addition, we have designed and are constructing a second slowing and trapping apparatus which will use superconducting magnets. It will function in a fashion similar to the 1st apparatus, but will have a much deeper well — ~ 1 K for Na atoms — allowing for much greater densities of trapped atoms.

Once we have trapped atoms, we will cool them. Proposed schemes² should produce cooling to 10^{-7} K. The trapped cooled atoms should be an excellent system for studying collective phenomena (superradiance, Bose condensation, and phase transitions), studying collisions such as Na-Na and Na-Na*, and performing ultra-high resolution spectroscopy.

References

1. W.D. Phillips and H. Metcalf, Phys. Rev. Lett. 48, 596 (1982).
2. D.E. Pritchard, Phys. Rev. Lett. 51, 1336 (1983).

21.5 High Precision Mass Measurement on Single Ions Using Cyclotron Resonance

National Science Foundation (Grant PHY83-07172-A01)

Joint Services Electronics Program (Contract DAAG29-83-K-0003)

Robert W. Flanagan, Jr., Phillip L. Gould, Gregory P. Lafyatis, Peter J. Martin, David E. Pritchard, Robert M. Weisskoff

In the past few years techniques for capturing, manipulating, and storing single electrons have been developed.¹ We intend to extend these techniques to permit the capture and storage of single ions in a Penning trap, which should permit measurement of (relative) masses to one part in 10^{11} . Such high precision will enable us to weigh chemical bonds, measure energy differences in isomer nuclei, provide more precise determination of physical constants,² and give a clearer understanding of many-body interactions.

The signal emitted by one single oscillating ion is expected to be quite low; consequently, we plan to conduct these measurements in a cryogenic environment using a SQUID detector. The SQUID is an extremely sensitive device with virtually no thermal noise limitation. Another advantage of low temperature, apart from noise considerations, is that it enables us to maintain an extremely high vacuum (less than one particle/cc),³ permitting long trapping times.

A custom, high-homogeneity NMR magnet has been designed and is nearing completion. A SQUID has been obtained and the development of associated superconducting circuitry is currently being undertaken. A Penning trap has been designed and is expected to be manufactured using computer-aided machining in the immediate future. The cryogenic envelope for the trap has been designed and plans for its construction finalized. Acoustic and RF isolation as well as the difficulties associated with operating a SQUID in proximity to a high field magnet are currently being considered.

References

1. R.S. Van Dyck, Jr., Paul B. Schwinberg, and Hans G. Dehmelt, in Daniel Kleppner and Francis Pipkin (Eds.), *Atomic Physics*, Vol. 17, (Plenum Press, 1981), p. 340; see also R.S. Van Dyck, Jr., and P.B. Schwinberg, Phys. Rev. Lett. 47, 395 (1981).
2. Johnson, private communication.
3. W. Thompson and S. Hanrahan, J. Vac. Sci. Technol. 14, 643 (1977).

21.6 Diffraction of an Atomic Beam by Transverse Standing Wave Laser Radiation

National Science Foundation (Grant PHY83-07172-A01)

Phillip L. Gould, So-Kuen Chan, David E. Pritchard

We are studying the momentum transfer to an atomic beam by a transverse standing wave laser radiation in an effort to gain a better understanding of the atom-field interaction. The force felt by an atom (and the resultant deflection) is a direct result of interactions of the induced electric dipole moment of the atom with gradients of the applied optical field. The effects of spontaneous emission depend on the length of time the atom spends in the field. For short times (relative to the natural lifetime), atomic "diffraction" is predicted¹ and has been observed by us.² For long times, momentum "diffusion" is expected.³ We are currently studying the transition regime between diffraction and diffusion. A preliminary result for an interaction time of 5 natural lifetimes is shown in Fig. 21-4. Diffraction is seen to persist, in contrast to existing theories.³

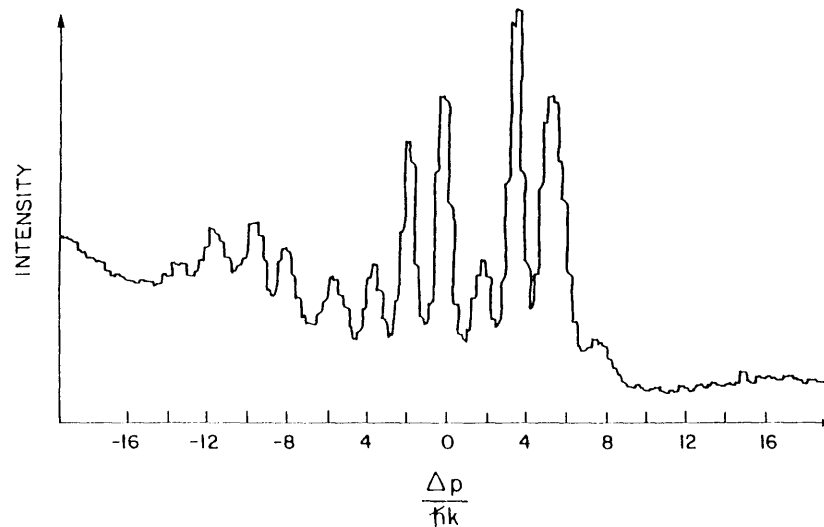


Figure 21-4:

The experiment was performed by crossing a sodium beam with a standing wave laser field and observing the momentum transfer downstream from the interaction region. The atomic beam is well collimated (2×10^{-5} rad) and has a narrow velocity distribution (10% FWHM). The atoms are optically pumped upstream from the interaction region with a second laser in order to prepare each atom as a two-level system. Our resolution is sufficient to detect the momentum transferred by a single photon (but note that in Fig. 21-4 we see momentum transferred in units of twice the photon momentum) — the Kapitza-Dirac effect.⁴

References

1. R.J. Cook and A.F. Bernhardt, Phys. Rev. A 18, 2533 (1978).

2. P.E. Moskowitz, Phillip L. Gould, Susan R. Atlas, and David E. Pritchard, Phys. Rev. Lett. 51, 370 (1983).
3. C. Tanguy, S. Reynaud, C. Cohen-Tannoudji, J. Phys. B 17, 4623 (1984).
4. P.L. Kapitza, P.A.M. Dirac, Proc. Camb. Phil. Soc. 29, 297 (1983).

21.7 Vibrational Dependence of Rotational Rainbow Structure in $\text{Na}_2\text{-Ar}$

National Science Foundation (Grant PHY83-07172-A01)

Warren E. Moskowitz, Brian A. Stewart, James L. Kinsey, David E. Pritchard

We have measured rotationally inelastic differential cross sections for collisions of atoms with highly vibrationally excited molecules. Specifically: $\text{Na}_2(j_i = 7&9, v_i = 31) + \text{Ar} \rightarrow \text{Na}_2(j_f, v_f = 31) + \text{Ar}$ at a center of mass energy of 0.3 eV. The Na_2 is in its ground electronic state but has vibrational energy equal to about 50% of dissociation. The measurements were made in crossed molecular beams. The initial Na_2 level was populated by optical pumping, and the final level was detected by laser induced fluorescence. The angular distribution was determined by our proven Doppler technique, called ADDS, in which a single mode, c.w. dye laser beam intersects the collision region along the relative velocity vector, and excites molecules which have been scattered into the final state with the correct Doppler-selected velocity component in the direction of the laser. The laser is tuned, and the resulting fluorescence signal yields the angle-differential cross section (Fig 21-5).

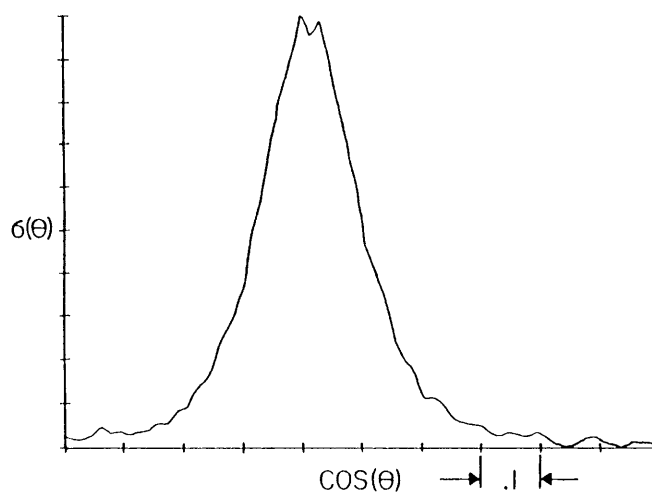


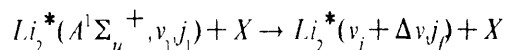
Figure 21-5: Scattering signal for $j_f = 15$.

21.8 Vibrationally Inelastic Collisions

National Science Foundation (Grant PHY83-07172-A01)

Thomas P. Scott, Brian A. Stewart, Peter D. Magill, David E. Pritchard

We have thoroughly investigated the phenomenon of quasi-resonant vibration \rightarrow rotation energy transfer discovered in our laboratory¹ in the system



with target gases X = He, Ne, Ar, Xe. We have studied the dependence of this phenomenon on v_i , j_i , target gas, and collision velocity, measuring both thermally averaged rate constants and velocity dependent cross sections. Noteworthy features of the rate constant data include:

The quasi-resonant narrowing of the final rotational state distribution with increasing j_i is observed for each of the target gases; moreover, in the case of Ne, Ar, and Xe, the shape of this distribution is identical within experimental error (see Fig. 21-6). Increasing j_i not only induces the resonant peaking, but also increases the rate constants dramatically enough that the rotationally summed vibrationally inelastic rate constants increase with j_i despite the narrowing of the j_f distribution.

Increasing v_i also enhances both the peak rate constant and the rotationally summed rate constant which is about 50% larger for $v_i = 9$ than for $v_i = 4$.

Our velocity dependent rate cross section data indicate that:

The total vibrationally inelastic cross section is very large in this system, especially for low velocity, where for $\Delta v = -1$ alone it is 30\AA^2 . This implicates moderately long range collisions in the vibrationally inelastic collision process.

For $\Delta v < 0$ the position of the peak of the quasi-resonant distribution depends little on velocity or target gas, being given by $j_f = j_i - 4\Delta v$; this indicates that roughly 70% of the vibrational energy is exchanged with rotational energy. The vibrationally exoergic cross section decreases as $1/v_{\text{rel}}$ for all j_i .

The width of the quasi-resonant peak decreases as v_{rel} decreases; this increase in specificity is most pronounced in $Li_2^* - Ne$ collisions.

Because of this enhancement in specificity with decreasing collision velocity, we expect to see evidence of quasi-resonant transfer at low velocity for reduced j_i . Experiments are underway to test this hypothesis.

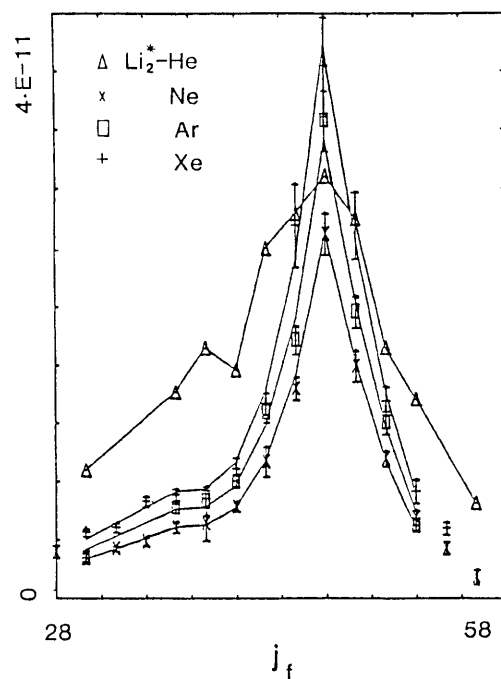


Figure 21-6: Rate constants $k_{v_i=9, j_i=42 \rightarrow v_i + \Delta v, j_f}$ versus j_f for Li_2^*-He, Ne, Ar and $Xe \Delta v = -1$ collisions. For Li_2^*-He , a dashed line connects the data points. For $Li_2^*-Ne, Ar, and Xe$ solid curves demonstrate the identical shape of the distributions for these systems. The solid curves are the average of the latter three systems, scaled for best fit to each system.

References

1. K.L. Saenger, N. Smith, S.L. Dexheimer, C. Engelke, and D.E. Pritchard, J. Chem. Phys. **79**, 4076 (1983).