VI. MICROWAVE SPECTROSCOPY

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A. INTERACTION OF PARAMAGNETIC SPINS WITH LATTICE PHONONS[*]

Ruby with 0.01 per cent Cr^{+++} impurity has been used as a paramagnetic crystal in an attempt to excite and detect lattice phonons. The experiment was first tried with one magnetic resonance, and with the two microwave cavities resonant at the same frequency. The power incident on the monitoring cavity was approximately 20 microwatts. Then it was found that with 5 milliwatts power incident on one cavity (the pumping cavity), the resonance in the other cavity (the monitoring cavity) could be reduced by a factor of 5. This reduction falls off rapidly as the frequency of one cavity is varied with respect to the other. When the frequencies are 20 mc apart the reduction is only 1 per cent. This would be expected as a result of phonon transmission through the crystal from the pumping cavity to the monitoring cavity. However, the possibility that it arises from direct microwave coupling between the cavities must not be ruled out, since some microwave power does, in fact, leak from one cavity to the other. To eliminate the effect of microwave coupling, the crystal is being cut to 1/4 of its cross-section area. Then the isolation between the two cavities should be better than 100 db.

Calculations were made for estimating the magnitude of the expected phonon effect. This work is based on the spin-lattice equations derived by Strandberg (1). His equations contain several unknown quantities, for example,

τ_L, the phonon-phonon relaxation time;

τ°_s, the spin-lattice relaxation time when the spins are far from saturated; and

$\Delta\nu$, the frequency interval over which spins and phonons interact appreciably.

If we make reasonable estimates for these unknown quantities at 4.2° K (liquid-helium temperature); for example, $\tau_L = 10^{-6}$ sec, $\tau^{\circ}_s = 10^{-3}$ sec, $\Delta\nu = 100$ mc, then we can make certain predictions. For instance, it should be possible to raise the average phonon number $<n>$, at f = 9400 mc, by a factor of 3. Roughly speaking, if the temperature of the spins in the pumping cavity is made very high by saturation, say, raised from 4.2° K to 50° K, then the phonons in contact with those spins can be raised to 10° K. Now if phonon scattering from one end of the crystal to the other (a distance of approximately

1 cm) is not too great, these phonons will have a very great effect on the spins in the monitoring cavity. Furthermore, use of these values in the spin-lattice equations is not inconsistent with observing the spin-lattice relaxation as an exponential function of time — at least within the accuracy of this kind of measurement.

B. W. Faughnan

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B. PHONON-SPIN ABSORPTION COEFFICIENTS IN PARAMAGNETIC CRYSTALS[*]

Experiments were performed in an attempt to detect phonon-spin absorption in F-center quartz and in potassium chromicyanide.

1. F-center Quartz

An 8-mc X-cut quartz crystal was irradiated for 48 hours by a cobalt 60 source to produce interstitial F-center electrons. The quartz was substituted for the ordinary tank circuit of a Pound-Watkins rf oscillator and placed in a dc magnetic field. We hoped to detect the slight decrease in crystal Q, caused by the absorption of the phonons by the electron spins, by using a conventional 50-cps modulation method. The experiment was tried at liquid-nitrogen temperature for various orientations of crystal relative to the field, with negative results. From paramagnetic resonance experiments, we estimated that the smallest value of the sound absorption coefficient, σ, that could be measured by this method is approximately 10^{-6} cm^{-1}. Thus, $\sigma_{quartz} < 10^{-6}$ cm^{-1} at T = 70°. Since $\sigma \sim 1/T$, an experiment was tried at T = 4°, but the oscillator would not drive the crystal at this temperature.

2. Potassium Chromicyanide

If a spin system in a magnetic field is irradiated with phonons of the Larmor frequency from an external generator, partial saturation of the spin populations will take place. If we assume a linewidth of approximately 10^7 cps (chromicyanide) at T = 4°, and that at these low temperatures the effect of Raman processes is small, it can be shown that the equilibrium population difference decreases by a factor of 2 when the acoustic phonon-energy density in the crystal is approximately 10^{-8} ergs/cm^3. Such

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a condition is easily achieved by cementing a quartz crystal to the chromicyanide and driving it with a signal generator. The saturation can be detected by monitoring the cyanide resonance with a Pound-Watkins spectrometer. This experiment has been tried at 4.2° K with negative results; it will next be tried at 1.4° K.

R. D. Mattuck

C. A TUNABLE RUBY MASER WITH LARGE BANDWIDTH[*]

A ruby maser was operated under conditions that begin to show the expected performance figures for solid-state masers. In some respects, performance has even exceeded expectations. The maser has a 20-mc bandwidth at 10-db gain with a bath temperature of 4.2° K and is tunable from 8400 mc to 9700 mc.

Four-level cw pumping was used for excitation — a modification of the common three-level pumping method. It is also known as "push-pull" pumping. The circuit uses a reflection cavity with an external circulator to provide the desired unilateral gain.

The maser crystal is ruby ($Al_2O_3 \cdot Cr_2O_3$), chromium being the paramagnetic ion. The chromium concentration is approximately 0.01 per cent. Maser action in ruby has been reported by Makhov and others (1). The Cr^{+++} ion spin 3/2 system produces four energy levels in an applied magnetic field. For the 54° 44' polar orientation of the c-axis, the 1-3 and 2-4 transition frequencies are degenerate for all fields. The maser is operated in this orientation at fields of approximately 4000 gauss. The 1-3 and 2-4 transitions are saturated by pumping power in the frequency range of 23 kmc; and population inversion is achieved between levels 2 and 3 with subsequent amplification at approximately 9100 mc.

A large ruby crystal (1 cm × 1 cm × 1/2 cm) is placed in the short-circuited end of a 1-cm square waveguide. A moving plunger provides the other wall. See Fig. VI-1. Since the crystal dielectric constant is high (2) at X-band, the perturbed TE_{10} mode propagates in the crystal and is cut off in the rest of the waveguide. The X-band fields are almost entirely confined to the crystal. Since the fields near the plunger are small, a Q_0 of 2000 is realized without plunger-wall contact. Coupling to the feeding K-band and X-band waveguides is furnished by the coupling slits shown in Fig. VI-2. Plunger displacement tunes the TE_{10} resonance over a large frequency range. To provide variable coupling and resulting gain and bandwidth control, the X-band waveguide is fitted with a sliding quarter-wavelength impedance transformer. Maser operation is accomplished throughout this range by appropriately tuning the magnetic field and pumping frequencies as the cavity is being tuned.

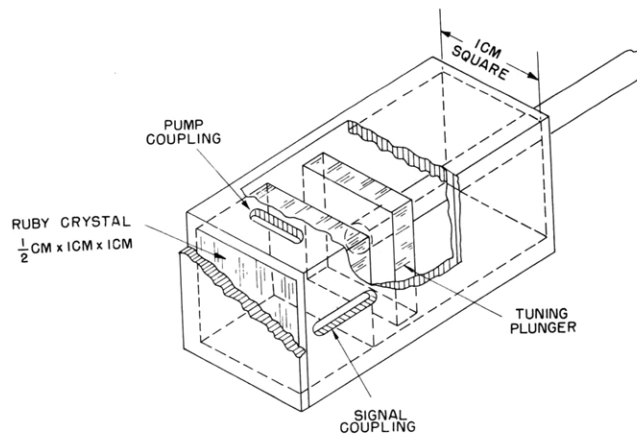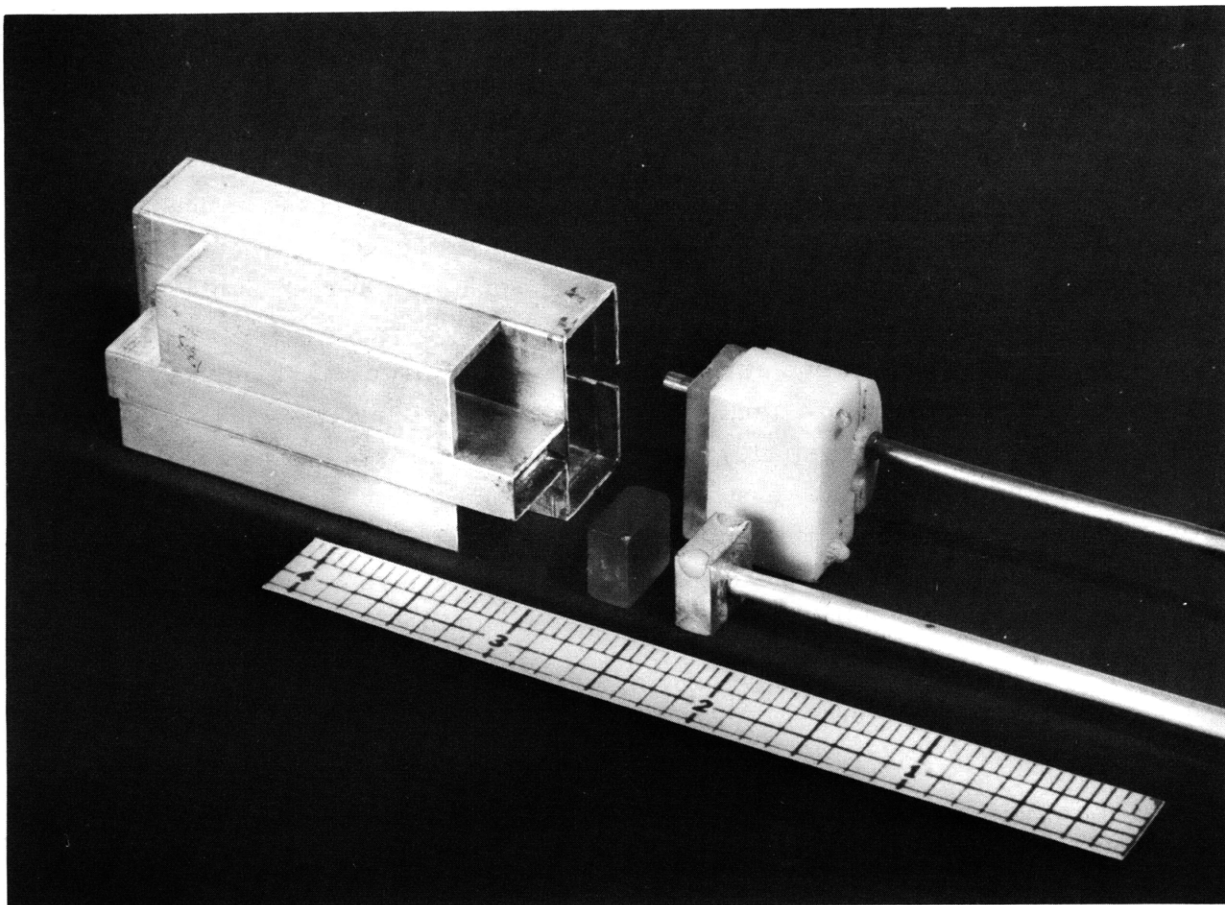
Fig. VI-1. Ruby maser cavity.

Fig. VI-2. Ruby maser cavity with feed waveguides and tuning plug for adjusting coupling coefficient.

We have observed that this device has the unusual and desirable feature of complete maser action in the absence of cavity resonance at the pumping frequency. It is this characteristic that allows the amplifier to be operated at any frequency in the signal-tuning range. We have observed that complete saturation can be achieved in the absence of a cavity resonance at the pumping frequency. Under these conditions, pumping power appears to be coupled to the crystal primarily by the fringing fields near the coupling hole.

The voltage gain-bandwidth product (3) was measured at several frequencies in the tuning range, and is essentially constant at 43 mc with a bath temperature of 4.2° K. At 1.5° K, the product increases to approximately 65 mc.

The use of push-pull pumping resulted in essentially complete inversion of the spin population. The negative resistance provided by the pumped ruby is numerically approximately equal to the positive resistance in the equivalent circuit representation of the paramagnetic absorption in the unpumped state. Cavity losses are relatively small. Under both of these conditions the effective noise temperature is expected to be comparable to the bath temperature. The other factor contributing to the large gain-bandwidth product is the large crystal size. The number of active spins is estimated to be 10^{18}.

The successful use of such a large crystal results from the fact that cavity resonance is not necessary at the pumping frequency. The required incident pumping power is 3 mw when the cavity is resonant, 15 mw when it is not. A mechanism must be present for obtaining good saturation with very nonuniform pumping fields. The phonon-saturation mechanism (4) offers a logical explanation. Spin-spin coupling mechanisms might also be considered.

The maser shows saturation effects at a signal input power of 23 microwatts. The dynamic range should be approximately 100 db above noise level.

The cascading of several single-cavity elements of the type that has been described in a reflection array to provide even greater gain-bandwidth products presents no theoretical problems, but the experimental difficulties may be severe. This study was originally planned as part of a more complex program involving several cascaded cavities. Traveling-wave solid-state masers offer a similar approach which is being pursued in other laboratories.

R. L. Kyhl, R. J. Morris, M. W. P. Strandberg

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1. G. Makhov, C. Kikuchi, J. Lambe, and R. W. Terhune, Maser action in ruby, Phys. Rev. 109, 1399-1400 (1958).

2. ϵ/ϵ_o = 11.53, parallel to c-axis; and 9.53, perpendicular.

3. $(G^{1/2} - 1) \; Bw = 2f_o / |Q_{neg}|$ = "voltage gain-bandwidth product" is nearly constant for high gains. Q_{neg} is the net negative cavity Q, including losses Q_o; f_o is the operating frequency.

4. M. W. P. Strandberg, C. F. Davis, Jr., B. W. Faughnan, R. L. Kyhl, and G. J. Wolga, Operation of a solid-state quantum-mechanical amplifier, Phys. Rev. 109, 1988-89 (1958).

D. AMMONIA MASER

Work by the present investigator has been completed. A thesis (1) giving a unified treatment of the theory of the gas maser is summarized here.

The theory of the complex polarization of a molecular beam interacting with a microwave field, with the velocity distribution of the molecules in the beam taken into account, was presented. Explicit operational conditions were deduced. General formulas for the emitted power and the oscillation frequency were obtained. Although the integrals that are involved present no practical difficulties, the large-signal theory was systematically derived by means of a correspondence method by introducing an rms time of flight of molecules in the cavity. The expressions thus obtained for the minimal oscillation condition and for the saturation value of the energy stored in the cavity differ from those obtained by K. Shimoda (2). The device was examined analytically as a single concept, and, in practice, a maser can only be so designed.

The experimental part of this thesis was concerned with the design and operation of an ammonia maser. The maser was so designed that it can easily be adapted, with a few simple modifications, for the study of other types of molecular beams, such as an alkali-halide beam. A notable feature of the present design is the provision of a liquid-air jacket that enables the cooling of the whole vacuum chamber to a very low temperature. With this cooling the maser can be operated at a pressure of 10^{-7} mm for a few hours with a molecular beam of the requisite intensity.

H. G. R. Venkates

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E. ANTIFERROMAGNETIC RESONANCE

We have learned a great deal about antiferromagnetic manganese chloride. Experiments with neutron scattering carried out at Oak Ridge National Laboratory, have shown that in state I ($1.81°$ K $< T < 1.96°$ K) the crystal splits into 3 directions of antiferromagnetic domain growth. The domains that are thus formed make an angle of $120°$ with respect to each other around the hexagonal axis. If a magnetic field is applied normal to one of these domain directions, all the other domains are rotated or converted to this particular direction. Furthermore, each domain is made up of only two sublattices. State II ($T < 1.81°$ K) can be thought of as arising from state I by having each domain

direction split in two at an angle of 20° with respect to each other.

In the usual magnetic resonance experiment, in which the frequency is fixed and the field is swept, it follows that at some low value of the field the crystal is converted into a single domain and remains a single domain for higher fields. Thus the two-sublattice theory of a single domain is applicable. In the two-sublattice theory, the quantity in the form $2\lambda K$ is observed in the resonance experiments. The strength of the exchange field, λ, can be shown to be equal to the experimental quantity $\chi_{\perp}^{-1}$. Therefore, K, which is the anisotropy constant (i.e., anisotropy energy $= K \cos^2\theta$), can be determined as a function of temperature. It has been found that K follows a modified Brillouin function, except in a region close to the Néel temperature. The curve has a tail that extends to the region above this temperature. This is not surprising, since the susceptibility is anisotropic above the Néel temperature. The simple two-sublattice model assumes that there is no anisotropy. This tail is probably attributable to the short-range order that exists above the Néel temperature.

In the domain model there is six-fold symmetry about the hexagonal axis and two-fold symmetry about an axis perpendicular to the hexagonal axis, in both states I and II. This symmetry has been observed. The actual shape of the curve (resonant field versus angle) will be different for the two states but the differences that are expected at X-band are not large enough to resolve the two states. It is hoped that by going to K-A band the two states will be definitely resolved.

<div align="right">D. H. Douglass, Jr.</div>

F. PARAMAGNETIC RESONANCE

A radiofrequency transmission spectrometer has been constructed for the study of electron paramagnetic resonance in ruby at low magnetic fields.

Preliminary runs at room temperature have indicated that there is a resonance at approximately 235 mc and at 45 gauss for the crystal c-axis perpendicular to the applied dc magnetic field. At this frequency, this resonance occurs at approximately 75 gauss for the c-axis parallel to the applied magnetic field.

<div align="right">F. F. Huppe</div>

G. NUCLEAR RELAXATION

Measurements of the dependence of the spin-lattice relaxation time (T_1) of F^{19} nuclei in crystalline lithium fluoride upon the concentration of radiation-induced F-centers have been extended to higher F-center concentrations (1). The experimental results, including those previously reported (1), are presented in Fig. VI-3.

For concentrations below $(3 \times 10^{16}/f)$ centers/cm^3, the behavior of the nuclear-spin system agrees with the theoretical predictions of Bloembergen (2) and of Khutsishvili (3, 4). In this region the rate of energy exchange between the nuclear-spin system and
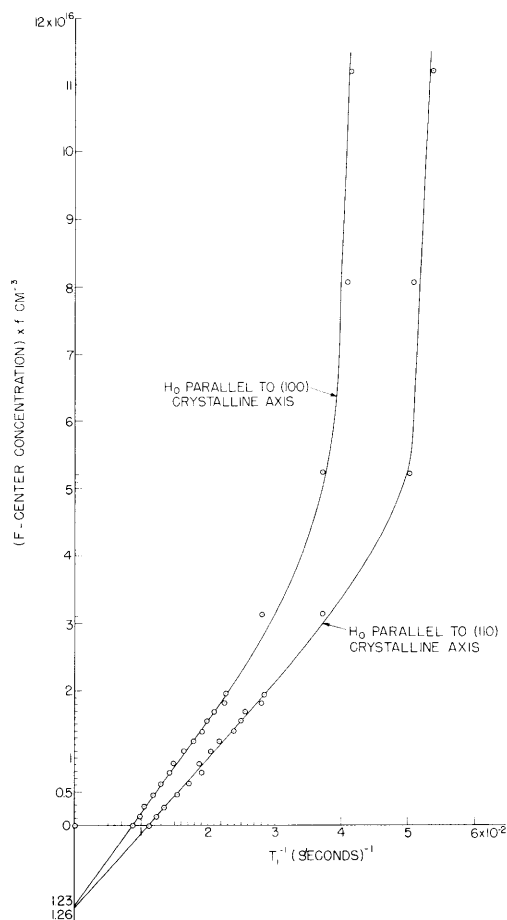
Fig. VI-3. F-center concentration versus the inverse of the spin-lattice relaxation time (T_1) of F^{19} nuclei in lithium fluoride for two orientations of the crystal in the dc magnetic field H_o.

the lattice is limited by the process of spin diffusion, and the slopes of the curves should be inversely proportional to the nuclear spin-diffusion constant. We have calculated the diffusion constant from an approximate expression derived by Bloembergen for the two orientations of the crystal in the dc magnetic field which are shown in Fig. VI-3. The calculated ratio D_{100}/D_{110} is 0.70, but the ratio of the slopes of the experimental curves is 0.81. Part of the discrepancy between the measured and calculated ratios can be attributed to misalignment of the crystal in the dc magnetic field. It is clear, however, that the directional character of the nuclear spin-spin interaction is responsible for the angular variation of T_1.

For F-center concentrations above $(5 \times 10^{16}/f)$ centers/cm^3, which corresponds to approximately one paramagnetic impurity center per 10^6 nuclei, T_1 becomes virtually independent of the F-center concentration. This behavior is expected for impurity

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concentrations that are large enough to allow the nuclei to relax by means of direct dipole-dipole interaction with the impurity spin, but previous estimates (2, 3) indicated that diffusion-controlled relaxation should predominate with paramagnetic impurity concentrations as high as one impurity center per 10^3 nuclei.

B. Josephson, Jr.

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