VI. MICROWAVE SPECTROSCOPY

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A. RUBY LINEWIDTH[*]

The theory of the linewidth of paramagnetic ions in crystals is sufficiently well understood to permit quantitative description of the observed resonance spectrum. Factors contributing to the paramagnetic-resonance linewidth are the spin-spin interaction with identical paramagnetic spins in the lattice, the dipole interaction with other nonsimilar magnetic moments in the lattice, and the statistical variation of the internal crystalline field splitting that results from crystalline imperfections.

It has been our observation that the linewidth of the chromium ion in corundum is unduly large for simple description by way of these effects. The linewidth in "pink" ruby is between 15 and 17 gauss. From measurements of the magnetic Q of this material we conclude that the lattice includes approximately 1 part in 10^4 chromium which is substituted for the diamagnetic aluminum ion. Such a density would lead to a spin-spin linewidth from identical particle interactions of approximately 2 gauss. The width caused by the magnetic field from the neighboring aluminum ion of mass 27 is of the same order, as has been observed in the nuclear magnetic resonance spectrum of that aluminum isotope. The remaining factor, crystalline imperfections giving rise to variations in the zero-field splitting of the chromium paramagnetic levels, is thus the only mechanism by which one can explain the observed ruby linewidth. Unfortunately, as Davis and Strandberg (1) pointed out, one would expect to find a great variation in linewidth among the observed ruby lines because of the variation in line frequency dependence on the zero-field splitting parameter. This strong variation has not been observed; in fact, Frank Huppe, of this laboratory, observed the spectrum with a paramagnetic resonance apparatus operating at 200 mc, and found that the resonant line at low magnetic field, which should be almost totally independent of the zero-field splitting, has the same typical pink ruby linewidth.

These facts are very interesting and exciting. They suggest that ruby is indeed very poor for maser material because it has a low ratio for the number of ions per cubic centimeter to the intrinsic linewidth. If chromium had the theoretically expected ratio for the quantity, it should be possible to increase present gain-bandwidth products for ruby

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amplifiers by a factor that is in excess of 10. For example, this would mean that an X-band amplifier operating at 1.2° K could have a gain-bandwidth product in excess of 1200 mc in a simple regenerative cavity mode of operation. Indeed, the simple, or unloaded, type of waveguide traveling-wave amplifier would be quite feasible with a theoretically understandable paramagnetic crystal. Unfortunately, ruby falls short of this mark by a factor greater than 10.

We have also observed the ruby transition in the 55° orientation at a particular operating point of approximately 2.2 kilogauss and at 5740 mc, where the line frequency is essentially independent of the magnetic field. In this case the only line broadening arising from other factors than the chromium spin-spin relaxation time results from the variation from point to point in the crystal of the angle θ that measures the inclination of the static magnetic field with respect to the crystalline axis. The actual frequency width of the line is given as

$$\Delta\nu = \frac{\partial\nu}{\partial\theta}\,\Delta\theta + \frac{\partial\nu}{\partial H}\,\Delta H \tag{1}$$

where θ is the angle between the applied magnetic field H and the crystalline c-axis, and ΔH is the random (non-spin-spin) magnetic field, caused, for example, by the nuclear magnetic moments of the neighboring aluminum Al^{27} nuclei. The operating point is so chosen that the partial derivative of the frequency with respect to H is zero, and therefore the magnetic field inhomogeneity resulting from the surrounding nuclei can only change the resonant frequency by changing the direction of the applied magnetic field at the position of the paramagnetic ion. This amounts to an effective $\Delta\theta$ term given by

$$\Delta\theta_H = \frac{\Delta H_{Al}}{H_o} \quad \text{(radians)} \tag{2}$$

where ΔH_{Al} is the random field attributed to the surrounding neighboring nuclei, and H_o is the operating field. The partial derivative of the frequency with respect to the angle can be computed as approximately 8400 mc/radian. The expected value of $\Delta\theta$ from the magnetic field inhomogeneity would yield a linewidth of approximately 8 mc for this transition. We observed the transition at room temperature in a cavity that has a Q of approximately 1000 at C-band. The resonant absorption was observed as considerably broader than the cavity width.

The next step was to use a ruby crystal at low temperatures in a coaxial transmission structure of greater bandwidth. The absorption could be observed directly and, again, was broad compared with the width of the klystron mode which, in this case, was 10 mc. Indeed, the resonance was so broad, compared with 10 mc, that the only observations that could be made were of the angular sensitivity of the absorption. One-sixtieth

of a radian, measured crudely, was necessary to pass through the resonance, which would give a width greater than 100 mc.

From these considerations, we conclude that the actual spin-spin width for the resonance is the limiting factor; and, contrary to expectation from the average concentration of the chromium ions, the spin-spin relaxation time is much shorter, by a factor of 10 or more, than we anticipated. Since the spin-spin relaxation time is a function of the distance between ions, we are forced to conclude that the ions have a specific concentration that is at least 10 times greater than the mean concentration determined from the magnetic susceptibility measurements. This can only be true if clustering of the ions occurs.

We are now at the point at which we are trying to understand the causes of this clustering and how it can be modified. Although it must be said that the ruby crystal that is conventionally used in solid-state amplifiers is rather poor material, as compared with a perfect, nonclustered ruby, the observation is still heartening because there are possibilities for improvement. Probably, other ions that fit better into the corundum structure will not be clustered, or, by annealing, it may be possible to distribute the ions more uniformly over the lattice. A ruby crystal with a linewidth of 17 gauss has been annealed at a temperature of 1500° C by Dr. Perry Miles, of the Laboratory for Insulation Research, M.I.T. We observed a reduction of the linewidth to 14 gauss after this annealing process. A higher temperature oven is now being sought in order to extend this annealing process to its ultimate limit.

M. W. P. Strandberg

References

1. C. F. Davis, Jr. and M. W. P. Strandberg, Paramagnetic-resonance spectrum of ammonium chromium alum, Phys. Rev. 105, 447-455 (1947); Technical Report 242, Research Laboratory of Electronics, M.I.T., Dec. 9, 1955.

B. PARAMAGNETIC AMPLIFIERS[*]

A nonreciprocal microwave structure with a circularly polarized cavity has been designed, constructed, and operated satisfactorily at X-band. We intend to continue this work, using the structure at low temperatures as a unilateral, nonreciprocal paramagnetic amplifier. The operation will be that of a three-level amplifier with the use of a pump at K_A-band. An analogous structure has been designed and constructed to operate at L-band (1420 mc), the frequency of the hydrogen line used in radioastronomic observations. This structure will have adequate gain-bandwidth product for observation

of the hydrogen-line spectrum, and being unilateral and nonreciprocal, it will require no circulator at L-band. Although circulators probably will be available at L-band, they are cumbersome devices, and we prefer the operating simplicity of our own device. A paper that discusses these amplifier constructions and their properties is being prepared.

The analysis shows, for example, that not only do these nonreciprocal, circularly polarized structures have the advantage that an additional circulator is not required, but they actually have uniquely favorable characteristics in that they are less dependent on input and output VSWR.

One interesting use that has been made of the X-band circularly polarized structure is that a casual examination of the ruby spectrum disclosed absorptions other than the one intended for the actual amplifier levels. Subsequent computations indicate that most of the transitions in the ruby spectrum preserve a high degree of circular polarization sensitivity for a wide range of the angle θ (which measures the angle between the ruby c-axis and the applied magnetic field) of $0°-80°$.

The work on circularly polarized structures has been confined to regenerative cavity design, although the cavity Q can be of any value; for example, a value approaching that of slow wave structures of current design. The general disadvantages of a single-port cavity structure usually arise from the regenerative nature of the amplifier itself. In work directed toward making circularly polarized nonreciprocal amplifiers, the regenerative character of the amplifier simplifies the problems of matching and construction of the device. This is probably the first time that the regenerative effect is an advantage rather than a disadvantage.

M. W. P. Strandberg

C. PHONON-SPIN ABSORPTION IN PARAMAGNETIC CRYSTALS[*]

An experiment was made in an attempt to detect ultrasonic saturation of spin populations in irradiated fused quartz. The result was negative.

Other experiments were performed on a crystal of irradiated Brazilian quartz in which the saturation effect has already been observed. The effect has the peculiar property that in the range considered (12 mc to 19 mc) the degree of saturation is independent of the difference between the spin resonance and phonon-generator frequencies. This will be investigated further by increasing the spin-resonance frequency to 200 mc.

R. D. Mattuck

D. SPECTROSCOPY OF FREE ATOMS[*]

An accurate g_J-value has been obtained for the ground state of free atomic Cl^{35} (see Quarterly Progress Report No. 52, p. 28). The value obtained is $g_J = 1.33379 \pm 0.00007$ in which the uncertainty given is the mean-square deviation. This is to be compared with the value $g_J = 1.3341$, obtained by assuming L-S coupling in the $^2P_{3/2}$ state, with $g_L = 1.0000$ and $g_s = 2.0023$. The result is an average over seven different $\Delta M_J = \pm 1$ and $\Delta M_I = 0$ hyperfine transitions at 8993 mc.

The measurements of resonant frequency and field for each transition were interpreted with numerical solutions of the secular equations. The calculations employ numerical values for a and b, the magnetic dipole and electric quadrupole interaction constants for Cl^{35}, obtained from atomic-beam experiments [1]. The mean-square deviation of g_J is within the uncertainty of the atomic-beam values.

The transition frequencies were measured by observing an audio beat less than 10 kc wide between the klystron signal and an X-band signal obtained by harmonic generation from a 6-mc signal monitored with a frequency counter.

The transition field was measured by measuring the resonant frequency for protons that are a short distance away in the magnet gap. A correction for the field difference between the position of the protons and the chlorine was made. The correction was obtained by repeating the experiment with atomic hydrogen. The free atoms and protons in this experiment occupy the same positions in the field as in the chlorine experiment. A field correction for the hydrogen run was obtained by comparing the calculated and observed transition frequencies and deducing the field correction required to reconcile the data. A connection at the chlorine field strength was made by a linear interpolation of the hydrogen field correction.

G. J. Wolga

References

1. J. H. Holloway, B. B. Aubrey, and J. G. King, Quarterly Progress Report, Research Laboratory of Electronics, M.I.T., April 15, 1956, pp. 35-36.

E. ANTIFERROMAGNETIC RESONANCE IN MANGANOUS CHLORIDE

Experimental data has been taken at K_A-band and is being analyzed. It has been found that the deviations from paramagnetic behavior above the Néel temperature can be quantitatively explained as arising from demagnetization effects that result from the shape of the sample. This is very interesting because demagnetization effects are usually unimportant in paramagnetic and antiferromagnetic materials.

Below the Néel temperature, the analysis of the data is complicated by these same demagnetization effects. A complete analysis will be presented in the next quarterly progress report.

D. H. Douglass, Jr.