

## VI. MICROWAVE SPECTROSCOPY

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### A. NOISE SPECTRUM OF PHASE-LOCKED OSCILLATORS

Sometime ago, a program for studying the sources of noise in phased-locked oscillators was established. The following remarks are a summary of the results of this project at its termination.

Since the first demonstration (1) of the simple utility of phased-locked klystron techniques in the microwave region, there has been considerable concern about the purity and validity of the resulting spectrum. Sources of noise, either amplitude-modulation noise or phase-modulation noise, can arise at many points in the servo-control loop. The importance of the various sources of noise is most convincingly demonstrated through experimental measurement. For this reason, a test facility consisting of two Felch oscillators (2), at 1 mc and 5 mc, was built, active tube multipliers, multiplied to 300 mc and 700 mc, were constructed, and electronic phase-locking equipment for K-band and X-band klystrons was assembled or constructed. The final system is shown in the block diagram of Fig. VI-1. It consists essentially of a silicon-diode harmonic generator, balanced mixer and 455-kc i-f amplifier, phase demodulator, dc amplifier, and phase-correcting network to the klystron repeller. Previous phase-stabilization circuits measured in this laboratory have indicated a carrier noise level of approximately 60 db per cycle per second (3). It was our intention to use the measurements for comparing the noise amplitude as a function of the multiplication ratio and as a function of the low-frequency crystal-reference oscillator frequency.

Two klystrons were stabilized in this fashion and were beaten against each other in a superheterodyne receiver; the resulting beat note (arising from a small difference in the fundamental crystal-oscillator frequencies) could be displayed on an oscilloscope and the carrier-to-noise ratio could be analyzed with a General Radio harmonic distortion analyzer.

Unfortunately, when the system reached satisfactory operating conditions, it was found that even with a frequency multiplication of 10,000 from a low-level Felch crystal oscillator, the noise power per frequency interval was much too low to be measured on the distortion analyzer. The clarity of the beat note under these conditions can be seen from Fig. VI-2. Although no definite measurements are available,

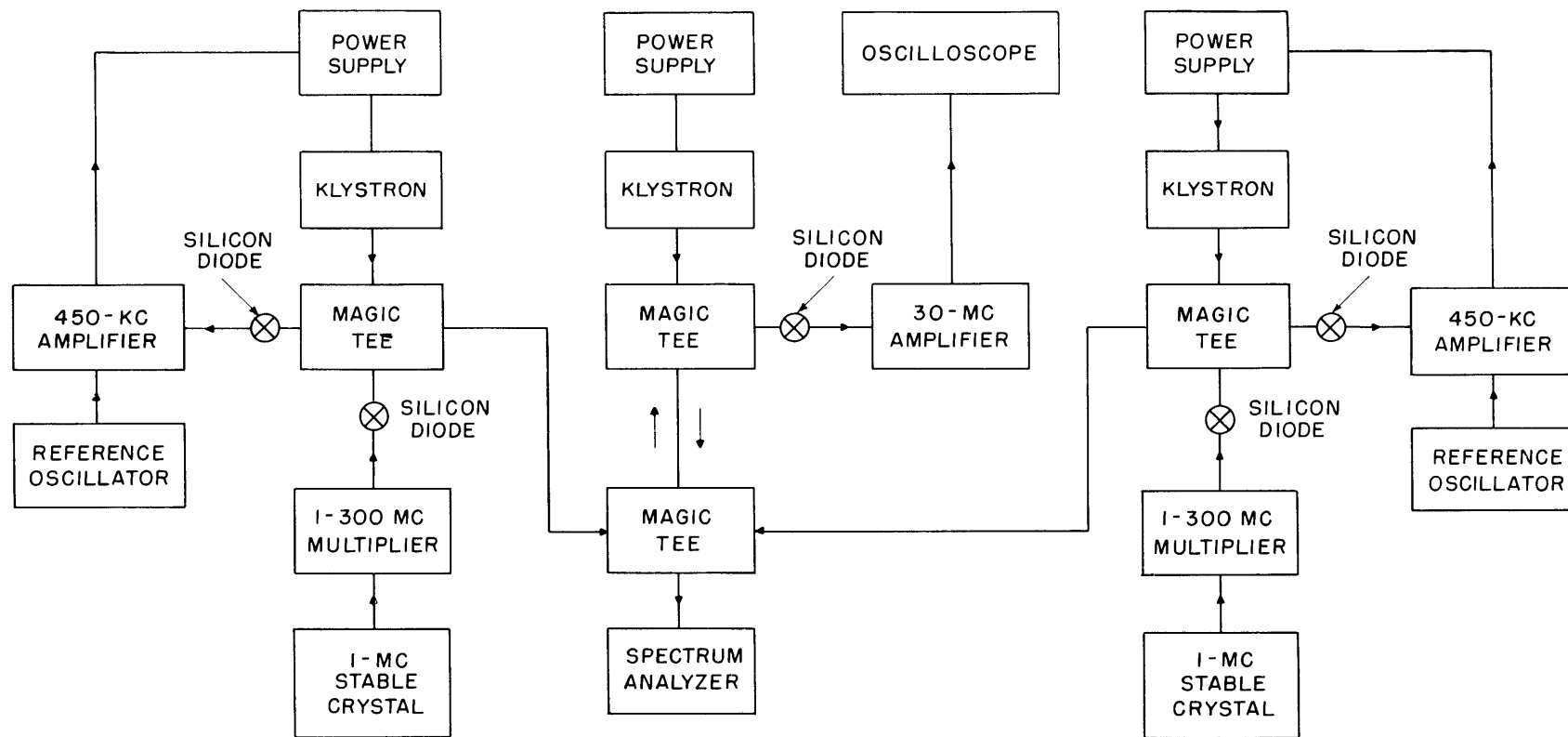
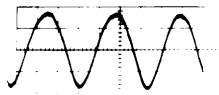


Fig. VI-1. Phase-locked stabilization of microwave frequencies.

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(a)

Fig. VI-2. Beat note: (a) higher sweep rate; (b) lower sweep rate.



(b)

the noise level must be less than 100 db below carrier per cycle per second.

We conclude that with reasonably decent feedback electronics, the noise arising from the fundamental crystal oscillator or the multiplier chain is negligibly small, even with a frequency multiplication ratio of 10,000. Stated another way, this means that there should be no difficulty in operating a phase stabilizer from a 10-mc crystal in the 100-kmc frequency region, and so on.

There is no doubt that the system as we have operated it is far from optimum. The i-f frequency is low. This was governed by a desire to have it less than the fundamental crystal-oscillator frequency. Even with this relatively poor design, these experiments have removed any doubt about the feasibility of phase-locking techniques – in the face of the argument that in the process a catastrophic amount of noise will also be generated. There is no doubt that this noise limitation will be met as the multiplication ratio becomes greater. As far as we can foresee, a multiplication ratio of 10,000 should be quite sufficient for most purposes, and should still be adequately remote from the multiplication ratio that would yield a noisy spectrum in the stabilized klystron output.

M. W. P. Strandberg, J. G. Ingersoll

### References

1. M. Peter and M. W. P. Strandberg, Phase stabilization of microwave oscillators, Proc. IRE 43, 869 (1955).
2. F. P. Felch and J. O. Israel, A simple circuit for frequency standards employing overtone crystals, Proc. IRE 43, 596 (1955).
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## B. THE NUMBER OF F-CENTERS IN QUARTZ

In connection with the phonon absorption experiments in F-center quartz (1), it was considered important to measure the number of F-centers. This was done by comparing

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the integrated line intensity of F-center quartz with that of a calibrated POV sample at X-band. The F-centers were produced in a 15-mc X-cut quartz transducer by gamma irradiation from a cobalt 60 source. The total dose of  $1.5 \times 10^7$  r, from the results of other experiments (2), seemed sufficient to cause saturation in the production of F-centers.

Measurements were made at room temperature and at liquid-nitrogen temperature with the x-axis of the transducer perpendicular to the dc magnetic field. At room temperature only two quartz lines were found. They had linewidths of approximately 2 gauss, and g-values of 1.999 and 2.002. The integrated intensity indicated that the number of F-centers was  $3 \times 10^{15}$  per cubic centimeter. At liquid-nitrogen temperature, there were 20 lines with g-values ranging from 1.990 to 2.044 over a field of from 3400 to 3300 gauss.

The three strongest lines had g-values of 2.008, 2.006, and 1.995, and linewidths of approximately 2 gauss (as did the rest of the lines in the spectrum). A line with g equal to 1.999 at liquid-nitrogen temperature appeared to be the same as the line at room temperature with the same g-value. The integrated line intensity of the entire spectrum indicated that the number of F-centers was  $2 \times 10^{17}$  per cubic centimeter. We feel that the liquid-nitrogen value of  $2 \times 10^{17}$  F-centers per cubic centimeter should be accepted, since the lower value obtained at room temperature probably resulted from the fact that many of the weaker lines were hidden in the noise.

P. H. Carr

### References

1. Research on Paramagnetic Resonances, Fifth Quarterly Progress Report on Contract No. DA36-039-sc-74895, Research Laboratory of Electronics, M.I.T., July 15, 1958 - Nov. 15, 1958.
2. R. W. Ditchburn, E. W. J. Mitchell, E. G. S. Paige, J. F. Custers, H. B. Dyer, and C. D. Clark, The optical effects of radiation damage in diamond and quartz, Report of the Conference on Defects in Crystalline Solids, University of Bristol, July 1954, p. 92.

### C. ANTIFERROMAGNETIC RESONANCE IN MANGANOUS CHLORIDE

The resonance spectrum of single-crystal manganous chloride has been determined as a function of temperature, frequency, and orientation. From these data the molecular field quantity  $(2H_E H_A)^{1/2}$  was determined. (See Fig. VI-3.) The curve extrapolates to a value of 13 kilogauss at  $0^\circ$  K, from which it is determined that  $H_A$  equals 5 kilogauss. Notice that this curve does not go to zero above the Néel temperature ( $1.96^\circ$  K). At temperatures below  $30^\circ$  K, demagnetization effects were significant and had to be taken into consideration.

The linewidth was also determined as a function of these parameters. There was no

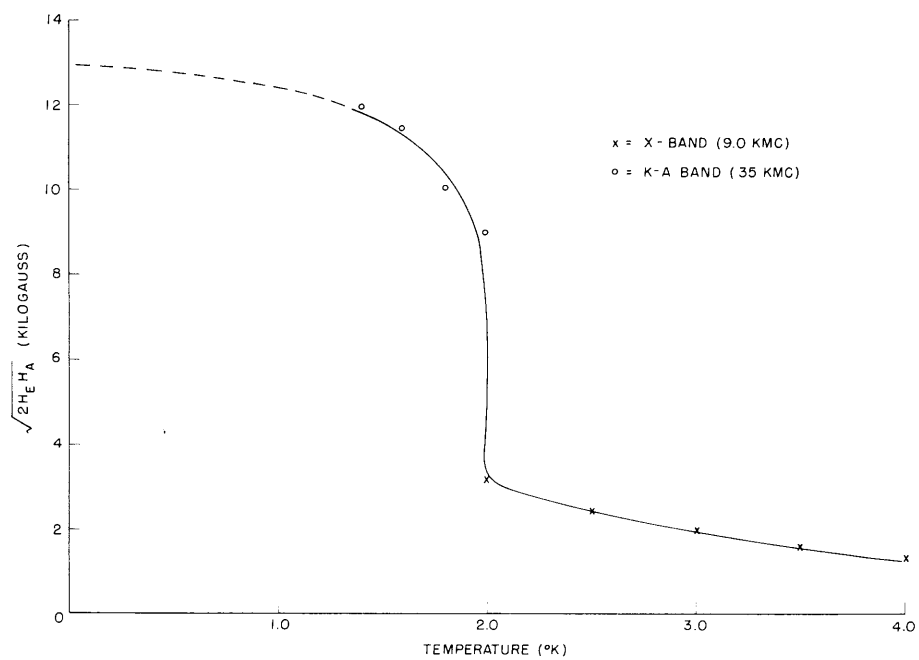


Fig. VI-3. Determination of  $(2H_E H_A)^{1/2}$  from low-field data on manganous chloride.

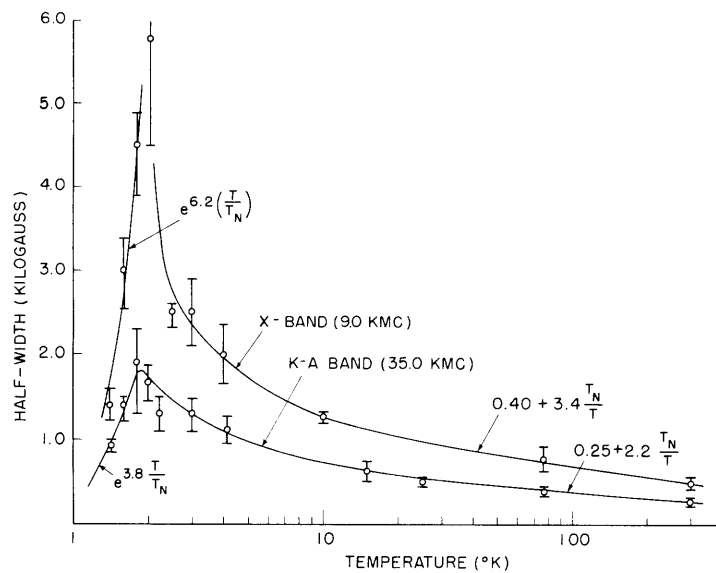


Fig. VI-4. Half-width versus temperature.

consistent orientation dependence. The data are shown in Fig. VI-4. These curves show a frequency dependence which is explained by the "10/3 effect" theory of exchange narrowed lines (1). In the antiferromagnetic state ( $T < 1.96^\circ \text{K}$ ) the data can be fitted with an exponential, although different constants are needed for the two frequencies. Gerritsen (2) has also observed an exponential dependence in antiferromagnetic  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ . In the paramagnetic state the curves follow a  $T^{-1}$  dependence, again with different constants for the two frequencies. There is still no rigorous theory for the temperature dependence of exchange narrowed lines.

D. H. Douglass, Jr.

#### References

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#### D. PARAMAGNETIC-RESONANCE SPECTRUM OF RUBY AT LOW MAGNETIC FIELDS\*

The paramagnetic resonance spectrum of the triply ionized chromium ion ( $\text{Cr}^{3+}$ ) that appears as an impurity in  $\alpha$ -corundum (ruby) was obtained for a transition frequency of 188 mc at magnetic fields below 100 gauss. A graph of the resonant magnetic field as a function of the crystal c-axis orientation with respect to the external magnetic field is given. These values for the resonant magnetic field as a function of c-axis orientation are substituted in the secular equation for ruby and yield values of the spectroscopic splitting factor of  $g = 1.99$ , and for the zero-field-splitting parameter  $D$ , of  $0.191 \text{ cm}^{-1}$ , which values are in excellent agreement with results obtained at microwave frequencies. This agreement implies the validity of the usual spin Hamiltonian for low-field ruby calculations. The ruby linewidth is found to be approximately 14 gauss and is essentially independent of c-axis orientation. A discussion of our failure to observe predicted transitions at higher fields is included in the author's Master's thesis presented to the Department of Physics, M.I.T., June 1959.

F. F. Huppe

#### E. PHONON-SPIN ABSORPTION IN PARAMAGNETIC CRYSTALS\*

A theoretical and experimental investigation was undertaken to determine the feasibility of using ultrasonic techniques for examining spin-phonon interaction in paramagnetic crystals. A general theory of the interaction has been developed with the use of

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higher-order perturbation procedures. The results are presented (in the author's Ph.D. thesis, Department of Physics, M.I.T., 1959) in the form of an equivalent interaction Hamiltonian, involving sums over products of spin operators and phonon creation-annihilation operators, which is applicable to any iron-group spin. Because of the symmetry of certain tensors in this Hamiltonian, the spin operators that are important for  $S > \frac{1}{2}$  systems occur in the anticommutator combination,  $S_i S_j + S_j S_i$ . This gives rise to a general selection rule that is independent of the detailed structure of the spin system. A formula is derived for order-of-magnitude calculations of the interaction strength. It is applied to obtain numerical estimates of direct and Raman interactions of a spin with a monochromatic phonon wave, and it is shown that, ideally, the acoustic experiments should be conducted in the microwave region, but can still be performed, under restricted conditions, at lower frequencies.

Experimental attempts to saturate spin populations with 13-mc and 5-mc phonons, are also described in the thesis. Data are presented that give strong indication of a saturation of the "hole" spin populations in  $\gamma$ -irradiated natural crystalline quartz. The bandwidth of this saturation is much larger than the paramagnetic linewidth, and a tentative theory describing the observations in terms of an acoustic "maser" action that takes place in the crystal is offered. A critical experiment is proposed to test this hypothesis. Data are also given that indicate the presence of a small "antisaturation" effect in  $\gamma$ -irradiated fused quartz.

R. D. Mattuck

### F. INTERACTION OF PARAMAGNETIC SPINS WITH LATTICE PHONONS\*

#### 1. Experiment

Work has continued on the excitation and detection of phonons by their interaction with paramagnetic spins. The effect reported in the Quarterly Progress Report of October 15, 1958, page 42, was found to be caused by direct microwave coupling. To decrease this coupling the size of the crystal hole was reduced to  $2.2 \times 2.2$  mm. Two identical rectangular cavities were built and used, instead of the original circular and coaxial cavities. The microwave attenuation between the two cavities was then 50 db. To increase detection sensitivity the pumping power was amplitude-modulated at 5 cps. Then the reflected monitor power was beat with a local oscillator derived from the monitor klystron, and the resulting low-frequency signal was amplified by a narrow-band 5-cps amplifier and detected by a phase-sensitive detector with a bandwidth of 0.01 cps. Microwave leakage power was not directly detectable at the output of the

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monitor cavity because it mixed with the local oscillator and no coherent signal resulted. However, it could give rise to an observable signal by partially saturating the monitor cavity resonance. This would be detected as a change in the amount of monitor power absorbed. But this leakage-power saturation behaves in a known way as a function of pump power, and the pump power can always be reduced sufficiently so that the leakage power causes negligible saturation in the monitor cavity, while the pump power is strong enough to saturate the pump cavity resonance. This is possible because of the large microwave attenuation between the two cavities. If the sensitivity of the apparatus is described by the minimum rise in phonon temperature detectable in the monitor cavity, then we have, in the most favorable case,

$$\Delta T/T|_{\min} = 0.001$$

## 2. Results

The phonon excitation and detection experiment was tried, using  $\text{Cr}^{3+}$  ions in  $\text{K}_3\text{Cr}(\text{CN})_6$ ,  $\text{Al}_2\text{O}_3$  (ruby), and magnesium oxide. An unidentified ion was also observed in  $\text{MgO}$ . For one resonance at  $H = 3345$  gauss, with c-axis parallel to  $H_{\text{dc}}$ , and with a spin-lattice relaxation time of approximately  $10^{-3}$  second, the buildup of phonon occupation number with pumping power was observed (see Fig. VI-5). The maximum value

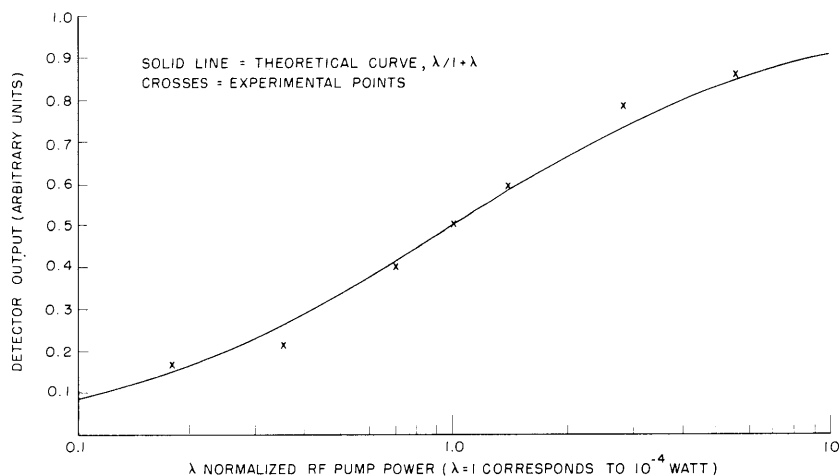


Fig. VI-5. Phonon buildup as a function of pump power for  $\text{MgO}$ .

of the phonon occupation number indicates a deviation from thermal equilibrium of  $\Delta T/T \approx 0.05$ . By using a second unidentified (titanium?) transition at  $H = 3357$  gauss, with c-axis parallel to  $H_{\text{dc}}$ , phonons were again observed. This line has a relaxation



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time of approximately one second and a  $\Delta T/T$  of 0.01. The results with ruby and cobalticyanide were negative. For ruby, an upper limit of  $\Delta T/T < 0.005$  was obtained.

### 3. Discussion

The experimental points of Fig. VI-5 follow quite well the theoretical curve for the buildup of phonons in contact with spins when the phonon temperature rise is small. The bandwidth of the spin-phonon interaction has not yet been determined but this will soon be done. This will tell whether the excited phonons are confined to the spin-resonance linewidth or whether the excess energy from the spins is spread over the whole phonon spectrum. The latter case would lead to an observable effect only if the thermal contact between the crystal and its surroundings is very poor. The experiments described above were performed at  $T = 4.2^\circ \text{K}$  with the crystal in contact with the liquid helium. The main thermal contact of the crystal was probably with the metal between the two cavities. By repeating the experiment at a temperature below  $2.2^\circ \text{K}$  it should be possible to determine whether or not thermal contact of the crystal with its surroundings is an important consideration. This is because the very great thermal conductivity of helium II will greatly increase the thermal contact of the crystal.

A detailed theoretical analysis of spin-phonon interaction, as well as an analysis of the experiments described here, is contained in a Ph.D. thesis submitted to the Department of Physics, M.I.T., May 18, 1959.

B. W. Faughnan