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Memorandum of Project MICHIGAN

ELECTRON-NUCLEAR INTERACTION IN RUBY AND ITS EFFECT ON THE RUBY MASER

George Makhov
Robert Terhune
John Lambe
Lloyd Cross

July 1960

SOLID-STATE PHYSICS LABORATORY
Willow Run Laboratories
THE UNIVERSITY OF MICHIGAN
Ann Arbor, Michigan

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PREFACE

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Robert L. Hess
Technical Director
Project MICHIGAN

ELECTRON-NUCLEAR INTERACTION IN RUBY AND ITS EFFECT ON THE RUBY MASER

ABSTRACT

It has been found that changes in the polarization of the Al^{27} and Cr^{53} nuclei in ruby affect markedly the absorption or emission of microwave power associated with the electron-spin resonance of the Cr^{+++} ion. This effect has been used to observe weak nuclear resonances and to change markedly the operating characteristics of the maser.

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1 INTRODUCTION

One need hardly emphasize the advantages of a maser device which is capable of amplifying microwave energy and which, simultaneously, possesses low-frequency output. Such a device combines the practically noise-free amplification characteristics of the maser with the convenience of a radio-frequency output stage. In the case of solid-state masers, this device can be conceivably realized by making use of electron-nuclear interactions. These interactions couple the paramagnetic resonance, which occurs at microwave frequencies, with the nuclear resonance which occurs at frequencies of the order of megacycles or of tens of megacycles.

While electron-nuclear interactions have been observed in a number of paramagnetic materials, little is yet known of their exact nature. This is partly because of the complexity of the processes involved, but also partly because of the scarcity of pertinent experimental data. Accordingly, an experimental study is being conducted at Willow Run Laboratories which is directed at gathering data which may provide insight into the mechanism of electron-nuclear interaction. The initial results of this study are reported here.

2
ELECTRON-NUCLEAR INTERACTION WITH ELECTRON-SPIN SYSTEM AT POSITIVE TEMPERATURE

The polarization of nuclear spins throughout a crystal by saturation of an electron-spin resonance of paramagnetic ions present as impurities has recently been reported (References 1 and 2). An apparent inverse of this effect in ruby (Al_2O_3:0.05% Cr^{+++}) has been observed at Willow Run Laboratories; i.e., when the induced nuclear polarization is partially removed by saturating an aluminum nuclear spin-resonance transition, a large decrease in the power absorbed by the electron-spin resonance of the Cr^{+++} ions is observed. Effects associated with induced transitions between the hyperfine levels of the Cr^{53} ions have also been observed.

Magnetic-field modulation at 5 kc was used to observe the edge of the ($+1/2 \rightleftarrows -1/2$) electron-spin-resonance absorption line of ruby with a microwave power level about 20 db above saturation. The spectrum shown in Figure 1 was then obtained by scanning the frequency of a low-power r-f oscillator connected to a single turn of wire around the ruby sample. The five lines in the 3-mc region correspond very well to the nuclear-magnetic-resonance spectrum of the host aluminum nuclei in the crystal. The same dependence of the splitting between lines upon the angle of d-c magnetic field and line width were observed both here and in the nuclear-magnetic-resonance spectrums.

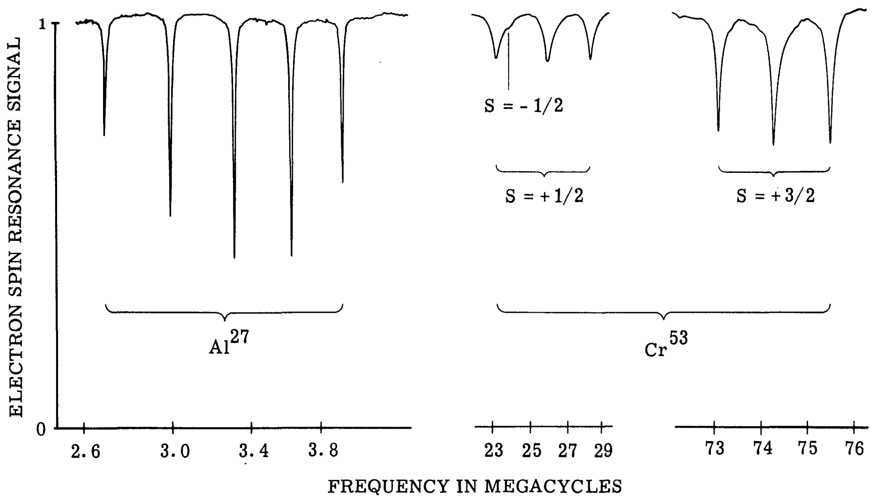

FIGURE 1. Al^{27} AND Cr^{53} NUCLEAR RESONANCES IN RUBY. $\theta = 0^{o}$.

The lines near 25 and 75 mc are associated with the I·S splitting of the Cr^{53} ions (Reference 3) which have a nuclear spin of 3/2 and a natural abundance of 9.5%. The other isotopes of chromium all have zero nuclear spin. The +1/2 and +3/2 electron-spin states are very close together at our operating point. As a result they interact strongly as the angle of the d-c magnetic field is varied, causing large changes in the hyperfine splitting (Figure 2). Also, the second-order correction to their hyperfine splitting is appreciable and leads to the observed triplets. The strength of the lines reduced very rapidly with departure from zero degrees. A weak triplet associated with S = -3/2 was also observed with the lines falling at 71.93, 72.15, and 72.37 mc at zero degrees. It is interesting to note that though all the Cr^{53} data were taken by observing the S = -1/2 to +1/2 transition, saturation of the hyperfine levels in the S = +3/2 state caused the largest changes in the electron-spin-resonance signal.

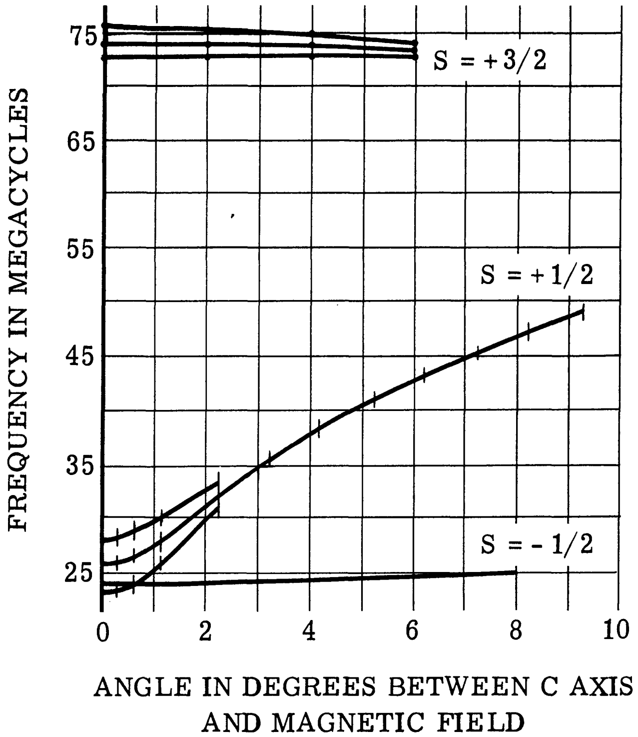

FIGURE 2. ANGULAR DEPENDENCE OF THE Cr^{53} SPECTRUM

The strong effect of nuclear polarization on the electron-spin resonance can easily be observed in ruby without applying r-f power. At microwave power level well above saturation, if one rapidly moves from one part of the resonance line to another a large transient with a decay time of about 5 seconds is observed. The same decay time is observed for the transient following removal of r-f power sufficient to saturate one of the aluminum nuclear transitions. Further, the observed decay time of the aluminum nuclear polarization is also 5 seconds and T_1 (spin-lattice relaxation time) for the electron-spin-resonance transition is approximately

0.2 second. The decay time of the effects associated with the Cr^{53} nuclei was also near 5 seconds.

The effects of nuclear polarization upon the electron-spin-resonance signal were only observable when microwave power levels of the order of or greater than that needed to saturate the electron-spin transition was used. When the electron-spin-resonance signal was observed at low microwave powers, the application of r-f power had no effect upon the signal. Further, when the signal was observed during the nuclear-polarization relaxation time following a quick reduction in the microwave power level from above to well below saturation, only a 0.2-second time-constant transient was observed. The application of r-f power had no effect upon the transient, thereby indicating that T_1 is independent of the nuclear polarization.

A decrease in microwave power absorption with the application of r-f power was always observed. As a check, some observations were made using amplitude modulation of the microwaves rather than magnetic-field modulation. Also, the amplitude of the magnetic-field modulation was varied over a wide range with no significant effects being observed.

It does not seem that the effects reported here can be explained by any of the mechanisms proposed by Feher (References 4 and 5) to explain his observations in doped silicon. The slow relaxation after the removal of r-f power would seem to indicate that the host nuclei play an important role in either the induced transition probabilities or the relaxation mechanism between electron-spin states. If so, in order to get an appreciable percentage effect in the electron system by saturating the nuclei one would have to reduce the relative electronic polarization $\left(\frac{N_i}{N_j}\right)$ to the same order as the nuclear polarization $\left(\frac{n_k}{n_l}\right)$; i.e., $\frac{N_i}{N_j} = \frac{n_k}{n_l}$. This idea is consistent with our experimental results because the nuclear polarizations are increased by a factor of about 40 (Reference 1) and the electronic polarizations are decreased by a factor of about 100 under our experimental conditions; thus the two are made comparable.

3
ELECTRON-NUCLEAR INTERACTION WITH ELECTRON-SPIN SYSTEM AT NEGATIVE TEMPERATURE

The effect of electron-nuclear interaction on the behavior of the ruby maser is of considerable interest. In the basic experiment, a two-turn coil was wound about a ruby sample containing, nominally, 0.1% chromium. This assembly was placed in a doubly resonant microwave cavity and located in the d-c magnetic field in such a manner that the axis of the coil was perpendicular to the direction of the field, and the polar angle was approximately 60°. With the system cooled to 4.2°K, a K-band microwave pump was used to saturate the 1-3 transition, and stimulated emission at X-band frequency was obtained in the 3-2 transition. Subsequent application of r-f power at 4.2 mc produced an increase in the gain of the maser amplifier, and resulted in a change in the mode of operation of the maser oscillator. The effect was most pronounced at the resonant frequency of the free-aluminum nuclei; however, at higher levels of r-f power substantial interaction was obtained over a band extending from 500 kc to 20 mc. Typically, power levels required to produce a detectable effect were of the order of 10 mw on resonance, and about an order of magnitude higher off resonance.

When the performance of the maser amplifier with r-f power on and off is compared, as shown in Figures 3(a) and 3(b), respectively, the change in the magnetic Q due to the change in polarization of the aluminum nuclei may be calculated. The change in gain amounted to 20 db. The dependence of the maser amplifier's gain on the magnetic Q is given by

$$G = \frac{\left(\frac{1}{Q_c} - \frac{1}{Q_L} + \frac{1}{|Q_m|}\right)^2}{\left(\frac{1}{Q_c} + \frac{1}{Q_L} - \frac{1}{|Q_m|}\right)^2}$$

where Q_c is the coupling Q, Q_L is the loss Q, and Q_m is the negative magnetic Q. In our arrangement, Q_c and Q_L are typically 10^3. With this value, the decrease in Q_m due to the application of r-f power was computed to be approximately 20%. This change in magnetic Q was observed under conditions of partial saturation of the pumping transition, and on the low-field side of the EPR (electron paramagnetic resonance) line.

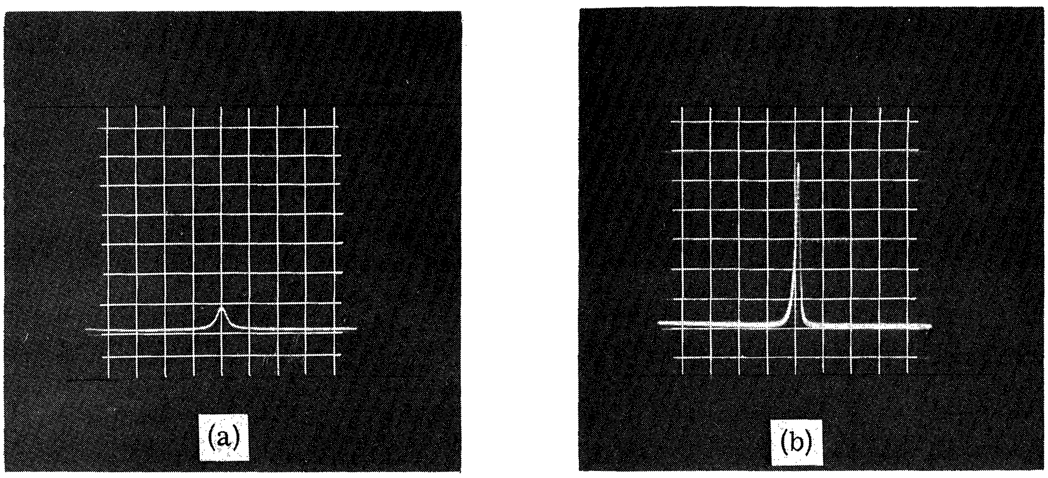

FIGURE 3. EFFECT OF CHANGE OF NUCLEAR POLARIZATION ON BEHAVIOR OF MASER AMPLIFIER. (a) r-f off; power gain = 15 db. (b) r-f on; power gain = 35 db.

The effect of decreased nuclear polarization on the behavior of the maser oscillator is shown in Figure 4. The application of r-f power tended to make the transient of the c-w mode of the oscillator (Figure 4a) less damped, and led eventually to the relaxation mode of operation (Figure 4b). The use of high r-f power levels tended to diminish or even reverse the effect.

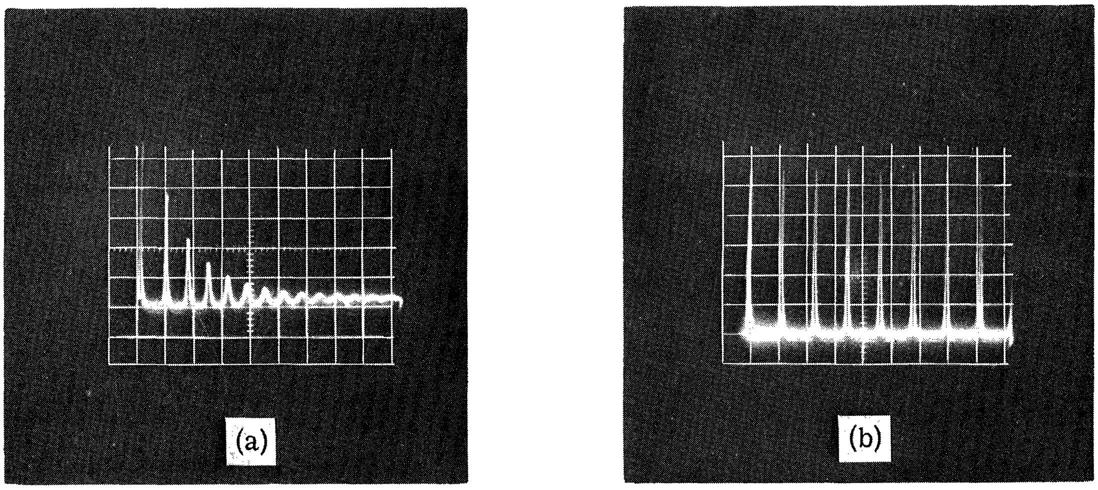

FIGURE 4. EFFECT OF CHANGE OF NUCLEAR POLARIZATION ON BEHAVIOR OF MASER OSCILLATOR. (a) r-f off; c-w mode of operation. (b) r-f on; relaxation mode of operation.

It should be noted that the transient form of the interaction, as in the case of resonant absorption, can be observed without the use of r-f power. When maser action is initiated by bringing the d-c magnetic field rapidly to the appropriate value, a transient results during which the amplifier gain rises quickly to a high value, then decays slowly to a lower steady-state value. The time constant of the transient is seconds in duration. The existence of the transient cannot be ascribed to spin-lattice relaxation since T_1 for cooled ruby is of the order of 0.1 second. Rather, it appears to be due to the relaxation of the polarization of aluminum nuclei.

Subsequent experiments have indicated that the application of r-f power does not affect thermal-relaxation processes to any perceptible degree. Rather, the application of r-f power can be thought of as added pumping, resulting in an increase in the magnetic Q. The degree of enhancement of the pumping process is of the order of the change in the degree of nuclear polarization, extimated to be about 1% under our experimental conditions. Under conditions of marginal saturation of the pumping transition, this may lead to a substantial decrease in the magnetic Q, as noted above.

These considerations suggest that the recovery of a solid-state maser device from saturation, or from fluctuations in pumping power, is characterized in general by two time constants. There is rapid recovery, of the order of 0.1 second, determined by spin-lattice relaxation; and slow recovery, seconds in duration, determined by nuclear-electronic relaxation. These effects have been observed experimentally. The latter effect, of course, becomes significant only when the on-period of the saturating signal, or variations in the degree of saturation of the pumping transition, are on a time scale comparable to that of nuclear relaxation.

In conclusion we would like to conjecture that the observed increase in gain of the maser amplifier due to application of r-f power may be used to advantage in detecting weak nuclear resonances as well as in studies of ENDOR-type (electron-nuclear double resonance) effects. In essence, the effect on the electron system produced by the resonant pumping of the nuclear system is amplified by the practically noiseless maser amplifier. When a high degree of regeneration is used, a small change in the magnetic Q results in a very considerable change in gain, or output (as attested by Figures 3a and 3b, and the corresponding calculation). A disadvantage of this method is that maser action cannot be obtained in ruby in the vicinity of $\theta = 0^{\circ}$, whereas nuclear resonance effects are conveniently observed at that orientation. Presumably, this can be remedied by using a paramagnetic material, such as iron-doped sapphire, which permits maser action at the above orientation.

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(Memo no. 2900-175-R)
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AD Div. 25/8

Willow Run Laboratories, U. of Michigan, Ann Arbor
ELECTRON-NUCLEAR INTERACTION IN RUBY AND ITS EFFECT ON THE RUBY MASER by George Makhov, Robert Terhune, John Lambe, Lloyd Cross. Memorandum of Project MICHIGAN. July 60, 7 p. incl. illus., 5 refs.
(Memo no. 2900-175-R)
(Contract DA-36-039 SC-78801) Unclassified memorandum

It has been found that changes in the polarization of the Al^{27} and Cr^{53} nuclei in ruby affect markedly the absorption or emission of microwave power associated with the electron-spin resonance of the Cr^{+++} ion. This effect has been used to observe weak nuclear resonances and to change markedly the operating characteristics of the maser.

(over)

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1. Maser — Operation
2. Synthetic ruby
I. Project MICHIGAN
II. Makhov, George, Terhune, Robert, Lambe, John, Cross, Lloyd
III. U. S. Army Signal Corps
IV. Contract DA-36-039 SC-78801

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Polarization
Al^{27}
Cr^{53}
Ruby
Electron-spin
Resonance
Maser

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