

IV. MICROWAVE SPECTROSCOPY

Prof. M. W. P. Strandberg
Prof. R. L. Kyhl
Dr. C. F. Davis, Jr.
S. A. Collins, Jr.
D. H. Douglas, Jr.

J. G. Ingersoll
B. Josephson, Jr.
J. D. Kierstead
S. Krongelb
D. M. Lazarus

R. D. Mattuck
J. J. McNicholl
R. E. Norris, Jr.
H. G. R. Venkatesh
G. J. Wolga

A. AMMONIA MASER*

The maser has now been installed in the laboratory. After testing the vacuum chamber with liquid-helium detectors and fixing the major leaks, the pressure was less than 10^{-5} mm Hg. This should be low enough for our purpose.

Discriminators (1) for stabilizing the operating frequency of the klystron have been constructed.

The voltages that will be applied to the focuser rods were determined from electrostatic fields for multipole focusers (2). We shall operate the maser with ± 14 kv applied to alternate rods.

H. G. R. Venkatesh

References

1. G. Wolga, Discriminators and AFC circuit, private communication.
2. F. O. Vonbun, A multipole separator for masers, Engineering Report NR. E-1203, U.S. Army Signal Corps Engineering Laboratories, Fort Monmouth, N.J.

B. NUCLEAR RELAXATION

Observation of the F^{19} resonance in lithium fluoride (LiF) was noted in the Quarterly Progress Report of July 15, 1957, page 34. The Pound oscillator, used as a resonance detector in our experiments, was the subject of an exhaustive analysis by Watkins (1). His analysis indicated that the poor sensitivity of our oscillator was the result of operation under unfavorable conditions. We reexamined the F^{19} resonance in the colored LiF crystal referred to in the last report, and now find the signal-to-noise ratio to be greater than 100. We also were able to detect the resonance in uncolored lithium fluoride. The spin-lattice relaxation time, measured at room temperature by direct observation of the recovery from saturation, is approximately 10 sec for the uncolored crystal and 100 sec for the irradiated crystal.

Attempts to measure F-center concentrations with an ultraviolet spectrometer proved unsuccessful because of the unsuitability of the available crystals. New crystals with more suitable geometry for the nuclear resonance experiments, as well as for the

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optical determinations, have been ordered.

For the time being, measurements will be restricted to lithium flouride.

B. Josephson, Jr.

References

1. G. D. Watkins, An rf spectrometer with applications to studies of nuclear magnetic resonance absorption in solids, Ph.D. Thesis, Department of Physics, Harvard University, 1952.

C. MEASUREMENT OF PARAMAGNETIC SPIN-LATTICE RELAXATION TIMES WITH PULSE TECHNIQUES*

Paramagnetic relaxation measurements have been made by a new technique in an attempt to answer some of the fundamental questions of relaxation phenomena. The relaxation of the spin system is undoubtedly a multistep process (Fig. IV-1). This is believed to be a spin to Debye lattice vibration relaxation, followed by lattice energy transport to the crystal boundary and thermal conduction into the surrounding medium. Presumably, the lattice energy conduction is limited only by phonon-phonon and phonon-boundary collisions. In the over-all relaxation process is there a single "bottleneck," or more than one? And which "bottleneck" is measured in a given experiment?

There are well-known techniques for measuring spin relaxation time. The mutual inductance method of Gorter and Casimir was the earliest. In their experiments a time phenomenon in the frequency domain is measured without continual saturation power in

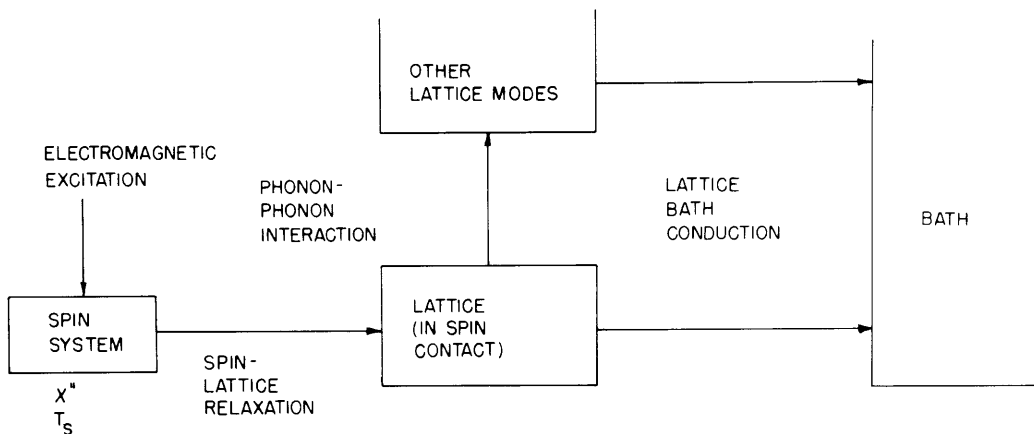


Fig. IV-1. Postulated mechanism of spin relaxation.

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(IV. MICROWAVE SPECTROSCOPY)

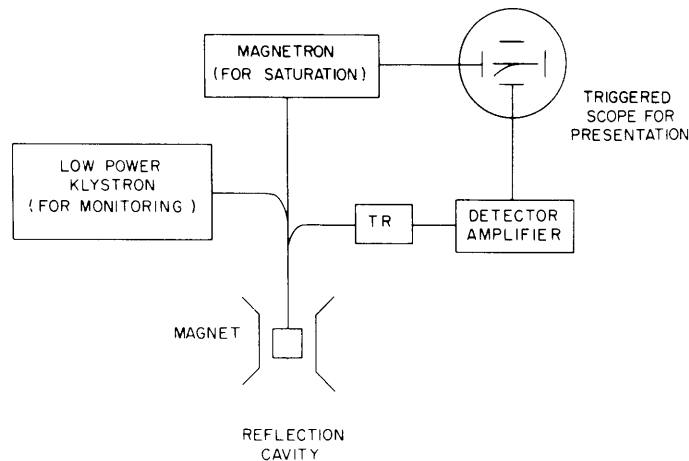


Fig. IV-2. Block diagram of relaxation measuring equipment.

the spin system. But only a weighted mean time for the available transitions at the experimental temperature and field is measured. In fact, it is uncertain that the "bottleneck" is between spins and lattice (1); it might be attributed to the small number of lattice oscillations that are on "speaking terms" with the spins and hence provide insufficient thermal conduction.

Resonance power-saturation measurements have been made by Eschenfelder and Weidner, and by others. Interpretation of these data involves the Bloch spin-lattice relaxation model. Time measurements are made in a restricted frequency domain, generally at a single frequency, and interpreted by making a change in power. This has the same effect as varying the spin temperature. We must assume that the lattice is a large thermal reservoir or a very good thermal conductor, or else the process will heat the lattice and invalidate the assumption of the equality of lattice and bath temperatures. Conventional measurements contradict the former assumption.

We shall describe the technique used in our experiments (Fig. IV-2). We make a low-power paramagnetic-resonance experiment. A klystron is stabilized to a wave-meter adjusted to the resonant frequency of the experimental cavity, and the magnet is then adjusted to the specimen resonance. Pulsed magnetron power, at the same frequency as that of the klystron, is fed directly into the experimental cavity. The magnetron duty cycle is low so that a relatively low average power is fed to the specimen. The reflectivity of the cavity is measured after the magnetron pulse by means of the low-power klystron monitoring signal, and it is presented on the oscilloscope as a function of time. Susceptibility is measured as a decrease in cavity Q ; it is simply related to cavity reflectivity. Figure IV-3 shows an oscilloscope photograph used for data interpretation. Other required data, which are determined directly from an oscilloscope trace, are the reflection coefficients immediately after, and a long time after,

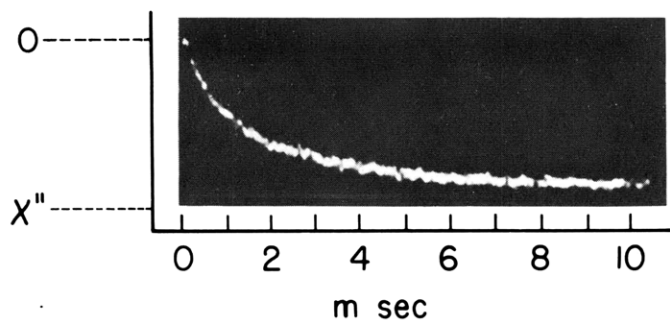


Fig. IV-3. Oscilloscope photograph of susceptibility recovery curve: Gadolinium ethyl sulfate diluted (lanthanum) 1:200. $T = 4.2^\circ \text{ K}$, $f_0 = 8745 \text{ mc}$, $H = 2240 \text{ g}$, $\mathcal{E} \parallel \mathcal{H}$, $+5/2$, $+3/2$ transition.

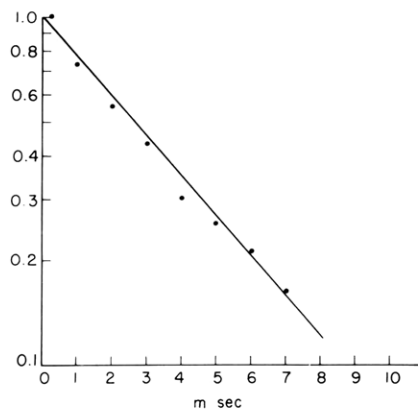


Fig. IV-4. Logarithm of relative susceptibility decrement as a function of time (from data of Fig. IV-3). Relaxation rate T_1 , 3.8 msec.

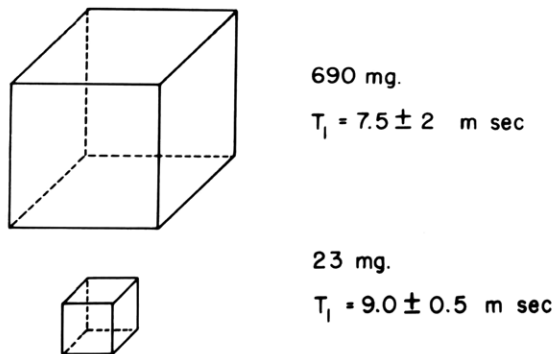


Fig. IV-5. Comparison of chrome alum specimen sizes.

(IV. MICROWAVE SPECTROSCOPY)

the saturating pulse. Photographic data are reduced to susceptibility decrement and plotted on semilogarithmic paper (Fig. IV-4). The slope of the line is the relaxation time.

The question of simple or multistep relaxation was studied by determining whether or not the decay of the susceptibility to thermal equilibrium deviates significantly from a simple exponential. In our measurements on chrome alum and gadolinium ethyl sulfate the logarithmic plots of the time-dependence of the susceptibility were concave upward – barely within experimental error. This indicates that a multistep relaxation may, indeed, exist. However, within the first time constant there was a well-defined straight line which was used for interpretation. All of the gadolinium lines were observed at one frequency, and the relaxation times were significantly different. These differences can only be accounted for in the spin-lattice step.

It seemed significant to ask whether or not lattice diffusion length could have an effect on relaxation times. To answer this question two chrome alum specimens were run (Fig. IV-5). They were roughly cubical and weighed 690 mg and 23 mg. The factor of 30 in volume (roughly 3 in linear dimension) was expected to show an effect of size on relaxation time, if it existed. Strangely enough, the smaller specimen exhibited the longer relaxation time, 9.0 ± 0.5 msec, as opposed to 7.5 ± 2 msec for the larger. This indicates that at low average-power levels, the relaxation time is not influenced by specimen size. The corollary is that no "bottleneck" appears to be created in thermal diffusion through the crystal. The chrome-alum relaxation times were of the order of those reported by Gorter.

Comparison of the relative relaxation times of the seven lines of diluted gadolinium ethyl sulfate shows the theoretically predicted longer relaxation times for the extreme lines of the spectrum. These experimental results give relaxation times 50 to 60 times as long as those reported by Feher and Scovil (2) from power-saturation experiments in which the susceptibility was reduced 10 db. In an attempt to resolve this inconsistency, relaxation times of two gadolinium lines were measured as a function of monitoring power. The relaxation time was relatively constant up to the point at which the monitor power began to saturate the resonance. Then, with approximately 6 db more power, the measured relaxation time dropped by a factor of ten.

In summary: By using a low average power, a relatively long relaxation time that exhibits, predominantly, the characteristics of the spin-lattice relaxation step can be measured.

M. W. P. Strandberg, C. F. Davis, Jr., R. L. Kyhl

References

1. L. C. Van den Marel, J. Van den Brock, and C. J. Gorter, *Physica* 23, 361 (1957).
2. G. Feher and H. E. D. Scovil, *Phys. Rev.* 105, 760 (1957).

(IV. MICROWAVE SPECTROSCOPY)

D. MICROMODULATOR

A compact tapered-core electromagnet with an air return path was constructed for the micromodulator. It produces 11,000 gauss at 1/4 inch pole separation (with 2.25 amp per coil) making it satisfactory for use up to 30,000 mc. The field is homogeneous within a fraction of a gauss over the sample volume (approximately $1/16$ inch³), and the linewidth is approximately 6 gauss at 22 kmc, which is the predicted value for powdered POV at this frequency. A Stark cell with a vacuum manifold has been set up and the micromodulator will be tested on ammonia lines at 18-30 kmc.

R. D. Mattuck