

trostatic potential has a saddle point at the midpoint. With the electron present, it is reasonable to assume that the jumping still takes place but that the electron, in trying to adjust adiabatically to the configuration of the center, associates itself as closely as possible with the positive ion. The effect of the electrostatic saddle point is thus at least partially washed out and the ion may spend more of its time in the central position, as sketched in Fig. 1 (b). Once this configuration is established the electron will be drawn toward the positively charged corners of the "pillbox" in which it now finds itself. Thus the electronic, as well as configurational, symmetry of the center is changed from C_{2v} to D_{2h} . One may consider the central ion-electron pair as simply an alkali atom, distorted toward the nearest neighboring positive ions.

Overhauser and Růchhardt⁵ tentatively conclude that centers responsible for the M absorption band have inversion symmetry on the basis of the absence of a large differential Stark effect as measured by line broadening at high external electric fields. Centers whose configurations are that of Fig. 1 (a) have permanent dipole moments and such a Stark effect is likely, according to these authors' computations. Jacobs⁶ was unable to find dielectric loss which could be associated with jumping of the M -center positive ion across a sizeable potential barrier. The model of Fig. 1 (b) is suggested in an attempt to reconcile these negative results with the original model, which is a plausible result of coagulation of F centers and neutral vacancy pairs.¹ Since the basic composition and the predominantly (110) symmetry of the center have been left unchanged, no reappraisals of experiments on formation and optical bleaching of the center¹⁻³ are necessary, while the lack of a sizable potential barrier and the presence of inversion symmetry seem to be complementary to each other on the revised model.

Detailed computations are necessary to establish the quantitative plausibility of the inversion symmetry model, but spin resonance experiments will probably be of more immediate assistance in its evaluation. A resonance associated with M centers has already been identified,⁴ but this resonance is hardly resolved from that of F centers and double resonance measurements⁷ (presently in progress at the University of Illinois⁸) are necessary to obtain more definite information on the center's structure.

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teresting discussions of his experiments and to Professor F. Seitz for his kind interest in this work.

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SPIN-LATTICE RELAXATION FROM STATE OF NEGATIVE SUSCEPTIBILITY*

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A state of negative magnetic susceptibility has been demonstrated in potassium chromicyanide by using a 180° pulse technique. The spin-lattice relaxation from this state has been observed in the time domain, and the complex susceptibility shows no change in slope as it passes through zero.¹

Negative paramagnetic susceptibility has been demonstrated in nuclear magnetic resonance with the use of both rapid passage² and 180° pulse techniques.³ Negative electron paramagnetic susceptibility has been produced in the three-level maser⁴ and in the two-level maser by means of rapid passage.^{5,6} The feasibility of generating a negative susceptibility in semiconductors by using spin-echo techniques has also been demonstrated.⁷ We have produced negative electron magnetic resonance susceptibility by means of a 180° pulse. The working material was $K_3Co(CN)_6$ containing 0.1% Cr^{3+} . The $-\frac{1}{2} \rightarrow +\frac{1}{2}$ transition at 9000 Mc/sec in a field of 3150 gauss, oriented parallel to the crystalline c -axis, was used.

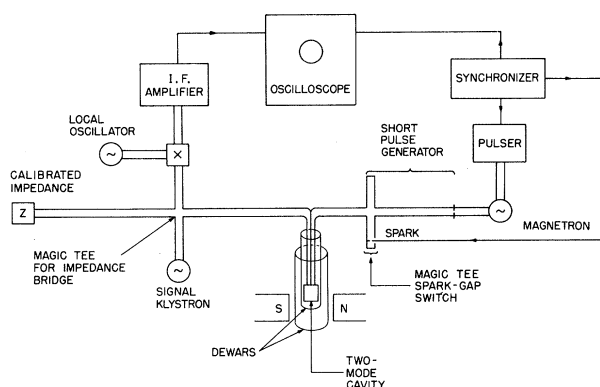


FIG. 1. Block diagram of equipment.

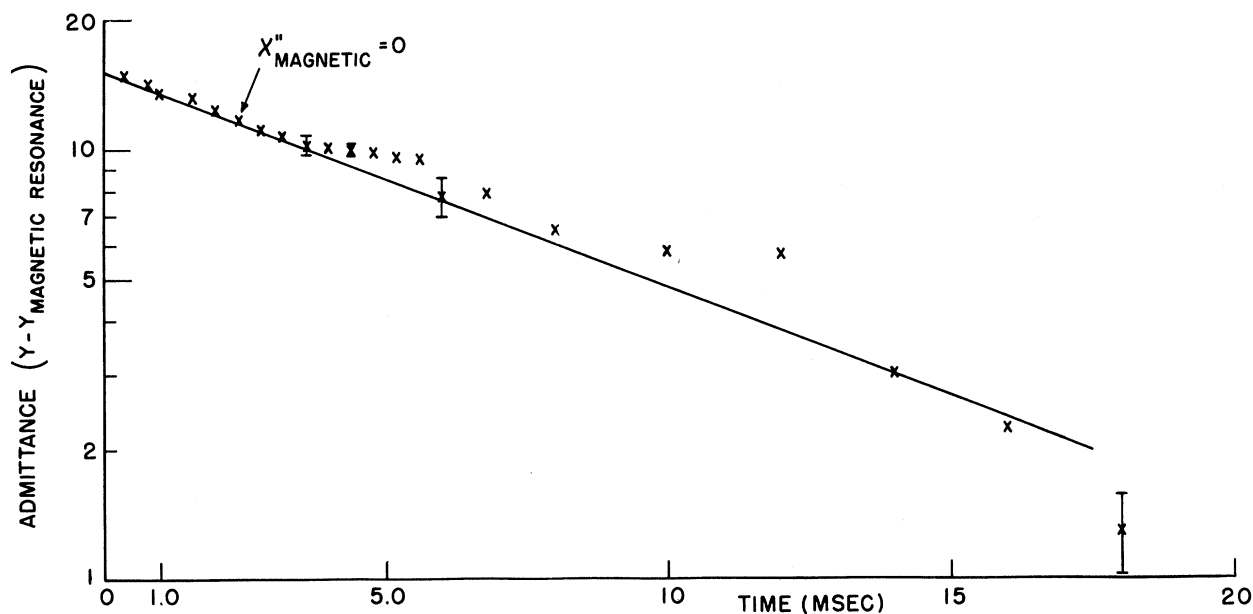
The equipment has been described elsewhere.⁸ The general arrangement is shown schematically in Fig. 1. A microwave pulse, roughly $10 \mu\text{sec}$ in duration, is generated from energy stored in a cavity formed from a length of waveguide excited by an $0.5\text{-}\mu\text{sec}$ magnetron pulse. One end of the waveguide is opened by firing a spark which unbalances a magic tee "bridge" by changing a plane of reflection. The sample is placed in a cavity that possesses two degenerate, or orthogonal modes. The mode to which the short pulse is applied has a very low Q (roughly 30). The other mode, which has a high Q , is used to observe the magnetic resonance of the sample. In order to use a small measuring signal, a superheterodyne detection system (30-Mc/sec i.f.)

is used. An impedance bridge is used to measure the instantaneous complex impedance of the cavity.

The quantity plotted on the ordinate in Fig. 2 is proportional to the magnetic susceptibility. The cavity filling factor was not determined. The point at which the susceptibility passes from negative to positive values is indicated. It is interesting to note that there is no change in slope at this point. The observed T_1 is 8.65 msec.

As a check on the tuning of the cavity and magnetic field and on the correct interpretation of the experiment, it was also possible to display the complex reflection coefficient of the cavity on an oscilloscope by means of auxiliary equipment that is not shown in Fig. 1 (i.e., a high-speed Smith-chart plotter).

To our understanding, a simple relaxation process uncomplicated by anomalous phonon effects is to be expected in crystals that are as dilute as this at 4.2°K . This result indicates that spin spectral diffusion⁹ and the effect of the other levels do not strongly influence the simple decay of the susceptibility. That anomalies can occur in less dilute crystals can be seen, since others,¹⁰ using saturation techniques, obtain for this crystal with 0.5% Cr^{3+} a T_1 of 0.2 sec, independent of spin quantum numbers. Presumably, these measurements have determined a lattice-bath relaxation time rather than a true spin-lattice

FIG. 2. Semilogarithmic plot of a quantity proportional to magnetic susceptibility versus time.

relaxation time.¹¹

Much remains to be done experimentally to develop a sound understanding of the spin-lattice relaxation process, but we feel that this demonstration of negative susceptibility by 180° pulse techniques, and the direct observation of the associated relaxation time from a state of negative magnetic susceptibility shed new light on spin-lattice relaxation processes in paramagnetic crystals.¹²

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[†]Now with Sperry Gyroscope Company, Great Neck, New York.

¹Research on Paramagnetic Resonances, Third Quarterly Progress Report, Signal Corps Contract DA36-039-sc-74895, January - April 1958. (The complete research report, of which this work is only a part, has been unduly delayed, and will be presented later.)

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Bogoliubov, Tolmachov, and Shirkov² and by Anderson³ to include collective excitations of the electrons. Both make use of the random phase approximation. We have calculated the response of a superconductor at $T=0^\circ\text{K}$ to a static magnetic field described in a general gauge, by use of Anderson's method. As he points out, the theory satisfies the sum rules and therefore should be gauge invariant. Aside from a small correction from the transverse collective modes, results are the same as those of BCS, who used the gauge, $\text{div}A=0$, and included only quasi-particle excitations.

Longitudinal collective excitations (plasma oscillations if Coulomb interactions are included) do not play a direct role if $\text{div}A=0$ but must be considered if the calculation is performed in a general gauge. Their crucial part in the satisfaction of the longitudinal sum rule has been discussed by Anderson.⁴ Pines and Schrieffer⁵ have shown that the plasmons shield the electrons in such a way that quasi-particles do not contribute to the longitudinal current so that the latter is described entirely in terms of the plasmon variables.

In the present calculation it is assumed that the normal-state wave functions are plane waves of energy $\epsilon_k = (\hbar^2 k^2 / 2m) - \epsilon_F$ and that the superconducting transition is caused by a two-particle interaction

$$\sum_{k, k'} V(k, k') C_{k'\sigma'}^* C_{-k'+q, \sigma}^* C_{-k+q, \sigma} C_{k\sigma'}$$

To ensure the gauge invariance of the results, $V(k, k')$ is a function of $\vec{k} - \vec{k}'$, only.

One should really start with the original gauge-invariant Hamiltonian in which the electron-phonon interaction is still present. Wentzel⁶ has attempted to calculate the Meissner effect from Fröhlich's Hamiltonian⁷ in a completely gauge-invariant manner but his procedure does not introduce the collective excitations. His result disagrees with ours in the London limit. To get the correct result by his method it would presumably be necessary to sum, to all orders in the interaction V , appropriate terms in the perturbation expansion. This question is discussed by Pines and Schrieffer.⁸ The author⁹ has calculated the effect from the same Hamiltonian in the gauge $\text{div}A=0$ and has obtained the same result as BCS. This calculation shows that Fröhlich's Hamiltonian leads to essentially the same results as that involving the two-particle interaction.

COLLECTIVE EXCITATIONS AND THE MEISSNER EFFECT*

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The theory of superconductivity of Bardeen, Cooper, and Schrieffer¹ has been generalized by