

negative temperature dependence is observed at low temperatures with a marked transition at 210°K; both effects gradually disappear at higher oxidation states. Above 210°K the line width remains independent of temperature.

The integrated signal strength which is presumably proportional to the ratio of the spin-center concentration to the absolute temperature rises slowly with temperature below 210°K, increases rather abruptly near 210°K, more or less inversely as the line width (Fig. 2), and also remains constant between 210°K and 300°K.

The resonances are not characteristic for all lamellar compounds, since they were not found in saturated graphite bromide lamellar compounds whose oxidation state as estimated by Hall measurements was 0.018, nor in a series of bromide residue compounds. They

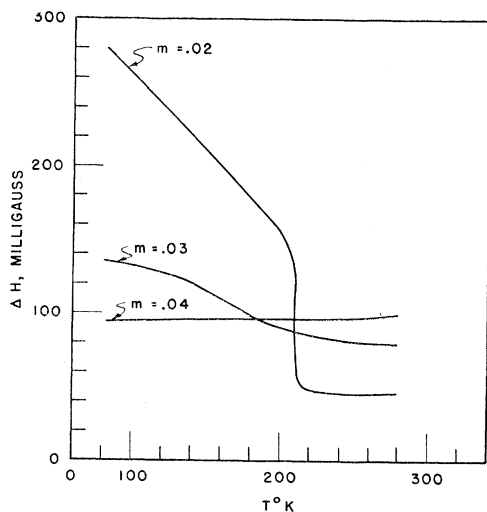


FIG. 2. Temperature dependence of the resonance line width.

were also absent in a lamellar compound of graphite with UCl_4 of oxidation state 0.007. The resonance is thus apparently not caused by positive holes in the valence band of graphite (the compounds discussed here are *P*-type conductors) because otherwise all compounds with equal oxidation state should manifest the same resonance. The spin centers are probably positive holes trapped at the acceptor states, i.e., bisulfate radicals. In graphite compounds with bromine or uranium halides, trapping either does not occur or results in molecule formation, i.e., $2\text{Br} \rightarrow \text{Br}_2$. Narrowing of the line in the bisulfate compounds with increasing concentration is probably due to exchange narrowing, possibly by coupling with the holes in the valence band,⁴ and/or by a progressive increase in the dielectric constant of the compound due to the large polarizability of H_2SO_4 . This increased dielectric constant would increase the overlap between wave functions of holes on adjacent traps and might split the energy levels of the traps into a band of states thus also ac-

counting for the nearly metallic temperature dependence of the resonance intensity.

The abrupt change in line width at 210°K may well be associated with the onset of rotation⁵ of sulfate molecules and/or ions and consequent disappearance of dipole broadening from proton nuclear moments.

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Measurement of the Spin Paramagnetism of Conduction Electrons*

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THE static magnetic susceptibility χ_0 of a metal is usually slightly paramagnetic. The principal contributions to χ_0 in many cases are a paramagnetic part due to polarization of conduction electrons spins, and a diamagnetic contribution due to orbital motion of the conduction electrons and due to ion cores. The diamagnetism is of the same order of magnitude as the paramagnetic contribution. The high degree of degeneracy of the electron gas makes χ_0 small and makes its measurement difficult and sensitive to impurity effects. Unless one can measure one or the other of the two contributions separately, one must depend on a theoretical relationship to determine the contributions of each part. Different theories give different answers for the relative size. Stimulated by the recent major theoretical advances of Bohm and Pines¹ in treating electrons in metals, we have performed a measurement of that part of χ_0 arising from the spin polarization *alone* by studying the conduction-electron spin resonance. We report initial results for lithium in this Letter.

The spin resonance arises from transitions of the electron spins among the Zeeman levels produced by a static field H . In practice the resonance is plotted at fixed frequency as H is varied through the resonance condition. Our experiment involves essentially the measurement of the total area under the curve of absorption *vs* magnetic field H . This area is simply related to the static spin susceptibility χ_p in many cases. Limitations of space do not permit a discussion of the exact relationship here, but, for example, a resonance line obeying the Bloch equations² satisfies the relation

$$\chi_p = \frac{2\gamma}{\pi\omega} \int_0^\infty \chi'' dH, \quad (1)$$

where ω is the frequency of the linearly polarized

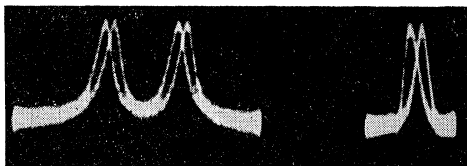


FIG. 1. Oscilloscope pictures of resonance absorption in lithium vs magnetic field. *Left*: Electron spin resonance. Zero field at the center of the sweep. The four peaks arise from a small phase shift between the horizontal and vertical axes. Sweep amplitude about 40 gauss. *Right*: Li^7 nuclear resonance at about 10 000 gauss. Sweep amplitude approximately 3 gauss.

alternating field inducing transitions, γ is the gyromagnetic ratio of an electron, and χ'' is the imaginary or "loss" part of the complex magnetic susceptibility. This same equation is satisfied by a line of any shape which is sufficiently narrow, and by a broad line of the type discussed by Garstens.³

The measurement of χ_p , which involves an absolute intensity measurement, appears difficult at first glance. Such experiments have been done by Hutchison and Pastor.⁴ The essential and new feature of our experiment is the elimination of most of the usual difficulties by comparing the absorption of the electron resonance in lithium with that of the nuclear resonance in Li^7 in the same sample and at the same frequency (17 Mc/sec). (See Fig. 1.) In order to observe the resonances of the electrons or nuclei we merely change H from 6 gauss to 10 000 gauss, leaving all circuits unchanged. We can measure the ratio of the areas under the absorption curve. With the use of Eq. (1) we can relate this ratio to the ratio of the electron and nuclear static susceptibilities. Knowledge of the nuclear susceptibility, the ratio of the absorption areas, and the gyromagnetic ratio of electron and nucleus is all that is needed to determine the electron susceptibility, since circuit parameters and the amount of sample cancel out in the ratio. The nuclear susceptibility can, of course, be calculated to a high degree of precision from the Langevin formula.

The value of spin susceptibility we obtain for lithium is $2.0 \pm 0.3 \times 10^{-6}$ cgs volume units. The theoretical spin value of Pines⁵ is 1.87×10^{-6} , while the free-electron (Pauli) value is 1.17×10^{-6} . The experimental value of Rao and Sarithri⁶ measured by conventional means is 1.35×10^{-6} , and includes, of course, a diamagnetic part associated with orbital electron motion and ion cores. Starr and Kaufmann⁷ find 1.95×10^{-6} , also by conventional means. Pines' theoretical value clearly agrees much more closely with experiment than does the free-electron value.

A fuller account of this research will be published at a later date, together with a discussion of the inherent limitations in accuracy. It is worth while pointing out here, however, that the reaction of the lithium to form nonmetallic compounds is not likely to effect accuracy to first order, since the nuclear resonance in the metals is displaced several gauss from that in a nonmetal⁸

(Knight shift), with the result that we measure only the ratio of electron absorption to that of metal nuclei. There is, of course, no electron resonance in the non-metal. In support of these statements, we have not found a sample dependence in our work.

We suggest that there are other systems in which a simultaneous nuclear and electron resonance can be found (e.g., the metal ammonia solutions and free radicals) where the comparison will make possible an improved determination of electron susceptibilities and may be used to determine the concentration of electron absorbing centers.

We are continuing the work on lithium and sodium. We hope that improvement of the signal-to-noise ratio will give adequate precision for reliable values of the diamagnetism.

We wish to express our appreciation to Dr. Pines for numerous interesting conversations. We have had many stimulating discussions with other members of our resonance group, particularly with Dr. Richard E. Norberg.

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Paramagnetic Susceptibility of Conduction Electrons*

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WE here report the results of a calculation of the effect of electron-electron interactions on the paramagnetic susceptibility of conduction electrons in metals using the collective description of electron interactions.¹ We have computed the paramagnetic susceptibility χ_p at 0°K, using the method of Sampson and Seitz,² which requires a knowledge of the way in which the energy of the electronic system changes as the population of electrons of spin up and spin down varies. Our calculation for χ_p differs from that of SS in that we use the expressions for the correlation energy developed in IV, rather than the earlier Wigner expression.

As shown in IV, the average energy per electron may be written as

$$E = E_0 + E_F + E_{\text{exch}} + E_{\text{corr}}^{1,r} + E_{\text{corr}}^{s,r}, \quad (1)$$



FIG. 1. Oscilloscope pictures of resonance absorption in lithium vs magnetic field. *Left*: Electron spin resonance. Zero field at the center of the sweep. The four peaks arise from a small phase shift between the horizontal and vertical axes. Sweep amplitude about 40 gauss. *Right*: Li^7 nuclear resonance at about 10 000 gauss. Sweep amplitude approximately 3 gauss.