Letters to the Editor

Observation of Exchange Interaction Effects in Ferromagnetics by Spin Wave Resonance*

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NO previous experiments at any temperature have revealed exchange interaction effects in ferromagnetic resonance, and semitheoretical estimates[1] have indicated that at room temperature the fractional "exchange shift," $\Delta H/H$, of the static resonance field is too small to be observable. The "spin wave resonance" experiments reported herein show, however, that an easily measurable $\Delta H/H$ ($\sim$20 or 30 percent) can be obtained at room temperature if appropriate experimental conditions are established. Our experiments also include an actual measurement of the important exchange factor (or "exchange stiffness constant") A, and show that in the present case both the width and shape of the absorption and dispersion curves can be described by using *not* the usual phenomenological damping terms (of the Landau and Lifschitz, or Bloch, type) but rather the physical mechanism of exchange stiffness in the skin depth of a ferromagnetic metal. Thus the energy losses near resonance are, in this case, almost exclusively due to eddy current dissipation.

The ideas underlying our experiments are the following. We increase $\Delta H/H$ by making the resonance field H rather small ($\sim$10² oersteds) and ΔH reasonably large, the latter being accomplished by producing a fairly small skin depth δ. Because of the relatively low frequency necessitated by the small H, a small δ requires a very large μ_2 ($\sim$10³) at resonance. We obtain a resonance despite the small H, and achieve a very large μ_2, by bringing about in our present polycrystals a magnetocrystalline anisot-

ropy, K, of about zero; in monocrystals, however, a small $K (\neq 0)$ should be usable.

We used a long, electropolished rod of a 66 percent nickel-34 percent iron alloy (by weight) which had been slowly cooled (at $\approx$60°C/hr) to obtain[2] $K \approx 0$, then covered with gold foil except near the center, and mounted in a strain-free manner as the inner conductor in a (half wave or full wave) coaxial cavity. Using an improved version of a pulse method,[3] supplemented by the axial static H which is normal to the microwave field, we measured the complex "equivalent isotropic permeability," $\mu_{equ} = \mu_1 - i\mu_2$, as a function of H.

In Fig. 1 the measured results for 3000 Mc/sec, corrected for static demagnetization,[4] are shown by the points on the solid lines, and the calculated results, based on the reasonable constants $g = 2.42$ and $A = 3.17 \times 10^{-6}$ erg/cm, are shown by dashed lines. These calculations, by Ament and Rado,[5] required a combined solution of the precession equation [including the spin-wave, or exchange, term $(2A/M_s^2)\mathbf{M} \times \nabla^2\mathbf{M}$] and Maxwell's equations, because the perturbation approach[1] is not valid in our case and does not yield the line shape. The agreement between the data and the exchange effect calculations is seen to be good, particularly when one considers that neither the characteristic asymmetries of the μ_1 and μ_2 curves, nor their width for a given height, could have been represented by means of the phenomenological Landau-Lifshitz damping. Furthermore, even though the samples are not quite saturated for small H ($<$15 oersteds), the unprecedented negative values of μ_2 obtained both experimentally (dotted lines) and theoretically (not shown) over part of that range do not violate thermodynamics but constitute, in fact, evidence of exchange effects. Since in the presence of exchange effects μ_{equ} is not an induction/field ratio but a measure of the surface impedance, μ^2 can be negative and only the attenuations must be (and are) positive.

We obtained similar line width and line shape agreement with the exchange stiffness picture at 6000 Mc/sec. Here we found $\mu_{equ} \approx 1$ for $H = 0$, and a g value only slightly above 2. However, within the present experimental error of A ($\approx$30 percent), A is the same at 6000 as at 3000 Mc/sec. These and other matters (e.g., the determination of A from the Bloch $T^{\frac{3}{2}}$ law) are discussed by us elsewhere.[6]

* Presented at the Detroit meeting of the American Physical Society, March 18, 1954 [Bull. Am. Phys. Soc. 29, No. 3, 13 (1954)].
[1] C. Kittel and C. Herring, Phys. Rev. 77, 725 (1950).
[2] R. M. Bozorth and J. G. Walker, Phys. Rev. 89, 624 (1953).
[3] M. H. Johnson and G. T. Rado, Phys. Rev. 75, 841 (1949); the pulsing method described in this reference was used in the present work to establish a reference point ($\mu_{equ} = 1$) for the Q and frequency shift measurements by saturating the rod parallel to the microwave field.
[4] R. M. Bozorth and D. M. Chapin, J. Appl. Phys. 13, 320 (1942).
[5] W. S. Ament and G. T. Rado, Bull. Am. Phys. Soc. 29, No. 3, 13 (1954). Also, paper in preparation.
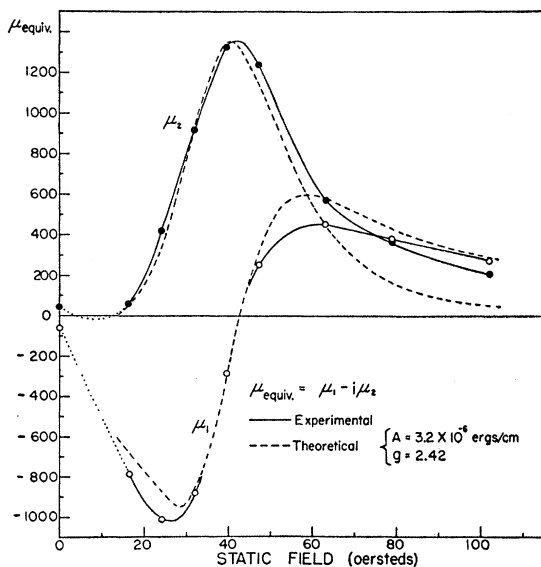[6] G. T. Rado and J. R. Weertman (paper in preparation).

FIG. 1. Measured and calculated values of $\mu_{equ} = \mu_1 - i\mu_2$ at 3000 Mc/sec.

Bound States in Dislocations

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SHOCKLEY[1] has pointed out that an edge dislocation in germanium may give rise to acceptor levels, and recent measurements[2] support this prediction. Dislocations may, however, have other effects in semiconductors, in addition to providing Shockley's "dangling" levels. Dislocations can affect the distribution of impurities.[3] Furthermore, edge dislocations are accompanied by lattice dilations and contractions, which in turn cause the top of the valence band and the bottom of the conduction band to be displaced from their normal positions. We have therefore attempted to estimate the trapping energy associated with the elastic distortions. In the case of a semiconductor in which a volume change causes the bottom of the conduction band and the top of the valence band to move in opposite directions, these

elastic distortions will provide traps for holes and electrons which coincide spatially, and therefore may influence recombination processes and exciton motion.

In the case of both metals[4] and semiconductors[5] the potentials involved are of the form $-A \sin\theta/r$, where θ is the angle with the slip plane and r is the distance from the dislocation axis. It can be shown that this two-dimensional dipole potential distribution contains an infinite number of bound states. This, however, is true only if the dislocation field is really infinite in extent. To get more realistic information we have applied the variational method to this problem. The minimum value of

$$\int \psi^* \left(-\frac{\hbar^2}{2m^*} \nabla^2 + V \right) \psi d\sigma$$

was found, for a trial function to be described, and for $V = -A \times \sin\theta/r$. The integration here is over the two-dimensional plane perpendicular to the dislocation. The trial function we have used is $(2/\pi)^{\frac{1}{2}}\alpha \exp(-\alpha^2 s^2)$, where s is the distance from a line parallel to the dislocation axis. This second line is taken to be at $\theta = \pi/2$ and at a distance d from the dislocation axis. The undetermined parameters are α and d. The integrals that have to be evaluated in this variational procedure can be made trivial by applications of Green's theorem. The minimum expectation value of the energy occurs when

$$d = 1.76\hbar^2/Am^*, \quad \alpha = 0.451Am^*/\hbar^2.$$

The energy at this minimum is

$$-0.204m^*A^2/\hbar^2.$$

These results are illustrated in Fig. 1.

For a given value of m^*, as the binding strength A is increased, the state becomes more strongly localized, and the binding energy increases more rapidly than A. In the case of a semiconductor, the equation for A has been given by Dexter and Seitz,[5] based on the theory of deformation potentials,[6] which neglects all the more recently suggested complications in the germanium band structure. To represent electrons in germanium we have used $\lambda = 4.0 \times 10^{-8}$ cm, $(1-2\nu)/(1-\nu) = \frac{1}{2}$ and an effective mass of $\frac{1}{4}$ the electronic mass. E_1 was taken to be 2 ev. These values give a bound state 3×10^{-3} ev below the bottom of the conduction band and centered at a distance $d = 8 \times 10^{-7}$ cm from the dislocation axis. In view of the uncertainty of the relevant constants these numerical results do not have much significance. For all reasonable combinations of the relevant constants we do find that the state is sufficiently localized (within 10^{-5} cm) so that it is within the field of a single dislocation, at the densities of dislocations which might be expected in semiconductors. Furthermore, the state is always sufficiently spread out so that it is not likely to be just a consequence of the fictitious singularity at the origin.

The method of deformation potentials as used by Dexter and Seitz takes into account how the band edges are displaced in the absence of holes and electrons. If holes or electrons are present, the carriers can redistribute themselves, and thereby screen the deformation potential. If we are well within a Debye length of the dislocation axis, then the method of deformation potentials, which predicts a band edge shift of $E_1\Delta$, is justified. Since the Debye length $(\epsilon kT/4\pi ne^2)^{\frac{1}{2}}$, for 1-ohm cm germanium is 10^{-5} cm and larger for purer material, our lowest state is well within this range for all reasonable purities.

Since the bound states can presumably have momentum along the dislocation axis, we cannot expect to find discrete states. If the edge dislocations are parts of sharply curved rings, the two-dimensional picture we have given breaks down completely, and there may be no bound states. In this discussion it has also been assumed that there are no extra "dangling" electrons to give a repulsive Coulomb field.

In the case of metals, bound states are implicitly assumed in the argument leading to the potential $-A \sin\theta/r$. Using the constants from earlier calculations,[4] we find that in Cu the lowest of these states is at an energy of 0.016 ev below the bottom of the
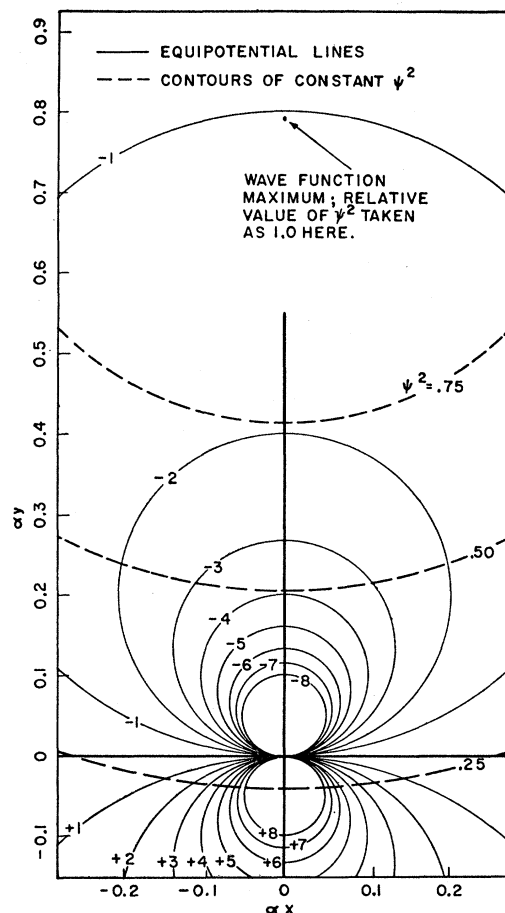

FIG. 1. Variation of the best trial wave function for the lowest state in a potential $-A \sin\theta/r$. The vertical axis y measures the distance perpendicular to slip plane; the horizontal axis x measures the distance in the direction of the slip vector. The dislocation axis is taken to be at (0,0) and perpendicular to the plane of the paper. The values given for ψ^2 are not normalized; they must be multiplied by $2\alpha^2/\pi$ to normalize them. The solid circles are equipotentials. The integral values with which they are labeled give relative potential values. These values must be multiplied by $1.25\alpha A$ to give actual potential values.

conduction band in the undeformed metal. The wave function is centered at a distance $d = 1.4 \times 10^{-7}$ cm from the dislocation axis. The corrugations in the bottom of the conduction band arising from dislocations may reduce the number of electrons which are free to move as conduction electrons. To estimate this total number of trapped electrons we shall try to find the number of bound levels in our potential $-A \sin\theta/r$. For lack of a better method we have used the Fermi-Thomas approximation (which has already been used implicitly in deriving the potential $-A \sin\theta/r$ in the metallic case). This tells us that there are $1.41\alpha R$ bound states in the two-dimensional potential, $-A \sin\theta/r$, between $r = 0$ and $r = R$. For a heavily cold-worked metal we can take $\pi R^2 = 10^{-12}$ cm^2 which gives about four bound states per dislocation in heavily cold-worked Cu. This does not take into account that each of these states in the two-dimensional potential can also have momentum along the dislocation axis. If we assume that the n electrons per cc in the occupied part of the conduction band have spatial wave functions which can be factored into one-dimensional wave functions, then there will be $(3n/\pi)^{\frac{1}{3}}$ different possible one-dimensional wave functions per unit length within the energy range occupied in the conduction band. For heavily cold-worked Cu this means that about 1 out of 500 electrons is confined to motion along the dislocation axis. The presence of jogs, solute atoms, and vacancies near the dislocation axis may

well prevent even this remaining degree of freedom from contributing to the conductivity. Wiener and Groetzinger[7] have found that heavily cold-worked Cu has a Hall coefficient about 1 percent higher than the annealed metal which implies that the number of carriers has decreased. While the exact contributions of the electrons trapped near the dislocation to the conductivity and the Hall effect involve complicated unsolved questions, the localization of $\frac{1}{5}$ of 1 percent of the electrons can hardly account for a 1 percent change in the Hall effect. It is possible, however, that the discrepancy is due to the crudeness of our remaining approximations.

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[3] A. H. Cottrell, Strength of Solids (The Physical Society, London, 1948), p. 30.
[4] D. L. Dexter, Phys. Rev. 85, 936 (1952). A more elaborate discussion of the metallic case, which takes into account explicitly not only changes in electron density but also the changes in band structure resulting from dilations, has been given by S. C. Hunter and F. R. Nabarro, Proc. Roy. Soc. (London) A220, 542 (1953). Dexter's values are used in our calculations.
[5] D. L. Dexter and F. Seitz, Phys. Rev. 86, 964 (1952). Undefined symbols that we have used in this letter are in accordance with the definitions of Dexter and Seitz.
[6] J. Bardeen and W. Shockley, Phys. Rev. 80, 72 (1950).
[7] B. Wiener and G. Groetzinger, J. Appl. Phys. 23, 441 (1952).

Orientation of Nuclei by Saturation of Paramagnetic Resonance

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THE orientation of metallic nuclei by saturation of the electron resonance in a magnetic field, predicted by Overhauser,[1] has its counterpart in paramagnetic substances. In the case of metals the essential points are: (1) one of the processes contributing to the electron relaxation is the coupling with the nucleus, which conserves the total spin vector; (2) this coupling is the predominant one in the nuclear relaxation; (3) the energy in such a process can be conserved because the states of the conduction electrons form a continuum; (4) in the saturated situation, the Pauli principle provides electron transitions with decrease of magnetic energy with a larger a priori probability than the inverse transitions.

We wish to point out here that the conditions prevailing in paramagnetic salts in which the nuclei of the paramagnetic ion have a spin, are such that requirements equivalent to those mentioned above are satisfied: (1) through the coupling $\alpha(\boldsymbol{\sigma}\cdot\mathbf{I})$ between the electronic spin $\boldsymbol{\sigma}$ and the nuclear spin $\mathbf{I}$, the nuclear and electronic systems can exchange energy under conservation of total angular momentum; (2) its contribution to the electronic relaxation process is a minor one, because, as is well known, the most probable electronic relaxation process comes from the interaction of the electron spin with the lattice vibrations; but it certainly is responsible for practically the entire heat-contact of the nuclear spins with the crystal. This is seen, for example, from the fact that the very long nuclear relaxation times found in pure diamagnetic crystals are greatly reduced[2] by small amounts of paramagnetic impurities; (3) in the processes of spin-exchange between nucleus and electron the energy can be conserved by a simultaneous absorption or emission of a lattice quantum $2\mu_0H$; (4) the difference in the a priori probabilities of absorption (increase of magnetic energy of the electron) and emission comes from from the fact that these probabilities are proportional to ν and $\nu+1$, respectively, where ν is the number of lattice quanta of magnitude $2\mu_0H$ present at the temperature T, which is given by the Bose-Einstein distribution law.

An upper limit for the resulting nuclear orientation can be obtained as follows: In the equilibrium state in an external magnetic field in the absence of electron resonance radiation, we have

$$|a|^2n_+N_m(\nu+1) = |a|^2n_-N_{m+1}\nu;\qquad(1)$$

here $n_\pm$ are the occupation probabilities for the electronic spin

states (we assume an effective spin 1/2 for simplicity) in thermal equilibrium, $n_\pm=\exp(\mp\mu_0H/kT)$; $N_m=\exp(-2m\mu_nH/kTI)\approx1$ are the corresponding quantities for the nucleus with spin I, and ν is the number of excited lattice vibrations of magnitude $2\mu_0H$; and $|a|^2$ is a combination of matrix elements for this transition. Equation (1) expresses the principle of detailed balancing. In the steady state formed under constant absorption of resonance radiation, we can write, when all other relaxation processes for the nucleus can be neglected,

$$|a|^2n_+'N_m'(\nu'+1) = |a|^2n_-'N_{m+1}'\nu',\qquad(2)$$

where the primed quantities are those reached in the steady state. Equation (2) is the condition for a steady state in this case. Under the assumption that (a) the electron resonance is completely saturated $(n_+'=n_-')$ and (b) the lattice-lattice relaxation time is so short that we can put $\nu'=\nu$, we find, from (1) and (2):

$$N_m'/N_{m+1}'=n_+/n_-,\qquad(3)$$

i.e., the nuclei are orientated at the temperature T to the extent as if they had a magnetic moment μ_0I. In view of the small mutual interaction of lattice vibrations, the validity of $\nu'=\nu$ is doubtful if the temperature is low; it would then perhaps only be approximated in dilute paramagnetic salts. At high temperatures the Raman-type process between the electron spin and lattice vibrations eliminates this difficulty, but then, of course, the nuclear orientation is always small. This limiting value[3] is the same as that obtained by Overhauser in the case of metals; a small energy leak for the nucleus, a shift in the distribution of lattice quanta, or an only partial saturation may decrease this orientation.

This nuclear orientation should be detectable by a narrowing and shift of the electron resonance line near saturation if the width of that line is predominantly produced by the interaction with the nucleus. If, in a dilute salt, the hfs is resolved, a change of the relative intensities of the components may be expected. Whether direct detection could be made by measuring the nuclear resonance absorption in the paramagnetic ion, depends on the rate of energy-exchange between the electron and the nucleus. The time average of the diagonal part of the interaction, i.e., the effective internal field for the nucleus, would be reduced to zero by saturation. This possibility of varying the internal field might be useful experimentally.

Note added in proof: After this manuscript was submitted, it was brought to my attention that a similar suggestion has been made independently by F. Bloch at the Stanford Meeting [Phys. Rev. 93, 944 (1954); see also Physics Today, April 1954, p. 30] and by A. W. Overhauser at the New York Meeting [Phys. Rev. 94, 768 (1954)].

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Electron Resonance in F Centers

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IN the preceding note attention has been drawn to the possibility of nuclear orientation by saturation of the electron resonance in paramagnetic salts. An application of particular interest is offered by the electrons of F centers in alkali halides. As a paramagnetic substance it is very dilute; therefore, the lattice oscillators will have the equilibrium distribution at low temperatures also, and the electron resonance line width is mainly due to the interaction with the 6 neighboring alkali nuclei. In this case the process of nuclear orientation is more complicated because it involves two different relaxation times. According to Kip and co-workers,[1] the interaction between the electron and the nuclei is given by $\alpha(\boldsymbol{\sigma}\cdot\Sigma\mathbf{I}_i)$, where $\boldsymbol{\sigma}$ is the electronic spin and $\mathbf{I}_i(i=1\cdots6)$ are the nuclear spins. Clearly, in any process due to