NUCLEAR POLARIZATION VIA "HOT" CONDUCTION ELECTRONS

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Overhauser[1] has shown that under certain conditions the saturation of the electron spin resonance absorption of conduction electrons in metals produces a sizable nuclear polarization. It was subsequently pointed out by several authors[2] that this phenomenon is not limited to metals but should occur in other paramagnetic substances in which the nuclei relax via the electron spin system that has been saturated by a radio-frequency field

The purpose of the present note is to point out a polarization scheme similar in principle to the Overhauser effect but which does not require an external radio-frequency field in order to saturate the electron spin. It is based on the difference between the electron spin temperature and the temperature corresponding to the mean kinetic energy of the electrons accelerated in a dc electric field. A preliminary experiment on electrons in silicon demonstrates the existence of a large temperature difference.

We consider for simplicity a system of electrons with $S=\frac{1}{2}$ coupled to nuclei[2,3] with $I=\frac{1}{2}$ via the hyperfine interaction $A\vec{I}\cdot\vec{S}$. Let $W(+-\rightleftharpoons -+)$ be the probability that this interaction produces a mutual nuclear-electron spin flip (the first sign refers to m_S and the second to m_I), $N_\pm$ the number of electron spins in the $m_S=\pm\frac{1}{2}$ states, and $n_\pm$ the number of nuclei in the $m_I=\pm\frac{1}{2}$ states. Assuming that each nuclear flip can proceed only via a simultaneous electron spin flip, one obtains for the steady-state condition

$$N_+ n_- W(+-\rightleftharpoons -+) = N_- n_+ W(-+\rightleftharpoons +-). \tag{1}$$

If the nuclei are in thermal equilibrium with the electrons, the ratio of the transition probabilities is

$$\frac{W(+-\rightleftharpoons -+)}{W(-+\rightleftharpoons +-)} = \exp[2(\mu_e-\mu_N)H/kT_R]$$

$$\simeq 1+(2\mu_e H/kT_R)-(2\mu_N H/kT_R),$$

where H is the applied magnetic field and T_R is the temperature of the reservoir that supplies the energy for the simultaneous electron nuclear flip. In the case of conduction electrons it is supplied by their kinetic energy. The electron and nuclear spin populations may be characterized by spin temperatures T_S and T_N defined by the relations

$$N_+/N_- = \exp[(2\mu_e H)/(kT_S)] \simeq 1+2\mu_e/(kT_S)$$

and

$$n_+/n_- = \exp[(2\mu_N H)/(kT_N)] \simeq 1+2\mu_N H/(kT_N).$$

From these definitions and Eq. (1) it follows that

$$n_+/n_- = 1-2\mu_e H/(kT_S)+2\mu_e H/(kT_R) + 2\mu_N H/(kT_R).$$

Therefore

$$\frac{1}{T_N}=\frac{\mu_e}{\mu_N}\left(\frac{1}{T_R}-\frac{1}{T_S}\right)+\frac{1}{T_R}. \tag{2}$$

The above relation shows that the nuclei may be polarized along or opposite to the external magnetic field (positive or negative nuclear temperature) depending on the difference between the electron spin temperature and the temperature corresponding to their mean kinetic energy. In the regular Overhauser effect it is assumed that $T_R=T_L$, where T_L is the lattice temperature and $T_S>T_L$. The last condition is obtained by saturating the electron spin transitions. In the case of hot electrons $T_R\neq T_L$ and nuclear polarizations may be attained without the application of a saturating microwave field.

Figure 1 illustrates the nuclear polarization

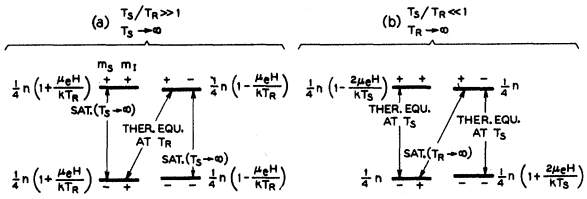

FIG. 1. Illustration of the nuclear polarization for the two limiting cases: (a) $T_S\to\infty$, nuclei are preferentially in the $m_I=+\frac{1}{2}$ state; (b) $T_R\to\infty$, nuclei are in the $m_I=-\frac{1}{2}$ state (this corresponds to a negative nuclear temperature). n in the diagram represents the total number of nuclei interacting with the electrons ($n=n_+ + n_-$). The quantity n_+/n_- for any value of T_R and T_S is given by Eq. (2).

for the two limiting cases: (a) $T_S \to \infty$ and (b) $T_R \to \infty$. It is seen that the magnitude and sign of the polarization correspond to the values given by Eq. (2). The energy levels in Fig. 1 correspond to an electron-nuclear system when the electron is in the vicinity of the nucleus. (For simplicity the nuclear Zeeman term has been neglected in the diagram.)

Since the above polarization scheme depends on the relative magnitudes of T_S and T_R, a preliminary experiment was performed to measure these quantities. The measurements were performed at 1.3°K on a phosphorus-doped silicon sample ($N_D = 2\times10^{16}/\text{cm}^3$, $N_A = 8\times10^{14}/\text{cm}^3$). Carriers were introduced by illuminating the sample through a germanium filter, thereby insuring that only electrons were produced.[4] A dc electric field was applied between large-area n^+ electrodes[5] and the resulting current measured. The electron spin resonance signal from the bound donor electrons was observed simultaneously at ~9000 Mc/sec. The electrical characteristic of the sample is shown in Fig. 2. At a field strength of ~150 v/cm the sample was found to break down.[6] This breakdown is believed to be the result of impact ionization of the donors by the accelerated electrons. This means that an appreciable number of electrons had acquired a kinetic energy in excess of the donor ionization energy (0.044 ev), i.e., T_R at the breakdown field will be of the order of 500°K. The deviation from Ohm's law at lower fields is an indication that the electrons are "hot," i.e., that they are not in thermal equilibrium with the lattice.[7]

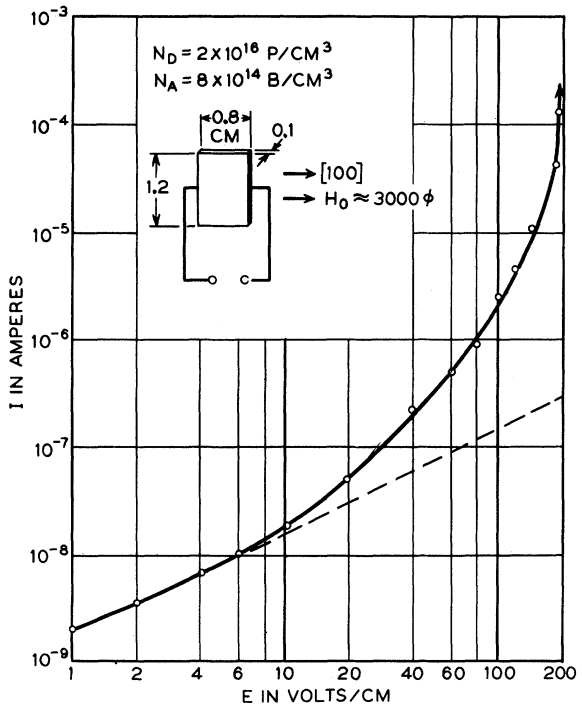

FIG. 2. Current-voltage characteristic of phosphorus-doped silicon at a lattice temperature of 1.3°K. The deviation from Ohm's law (see dashed line) indicates that the electrons are "hot." The electron spin resonance signal from the bound donors was measured simultaneously. The results are illustrated in Fig. 3.

The spin temperature of the free carriers T_S was obtained by measuring the spin resonance signal from the bound donors in the presence of hot electrons. There is good evidence that the spin relaxation time of the bound donor electron proceeds by a spin exchange with the free carriers,[4] which in turn relax by some other process (e.g., interactions with paramagnetic impurities). The two spin systems acquire therefore the same Boltzmann factor. The amplitude of the electron spin resonance signal from the bound donor, which is proportional to the Boltzmann factor $\mu_e H/(kT_S)$, can then be used as a convenient thermometer to measure T_S. Figure 3 shows the experimental results. The amplitude of the resonance signal at a field of 150 v/cm has decreased by a factor of two corresponding to $T_S \simeq 2.6$°K which is about two orders of magnitudes lower than T_R. However, the number of electrons in these experiments (10^8 - $10^{10}/\text{cm}^3$) was too small to have produced a sizable nuclear polarization. In germanium hot electrons may be produced at lower electric fields,[7] and therefore higher current densities may be tolerated without

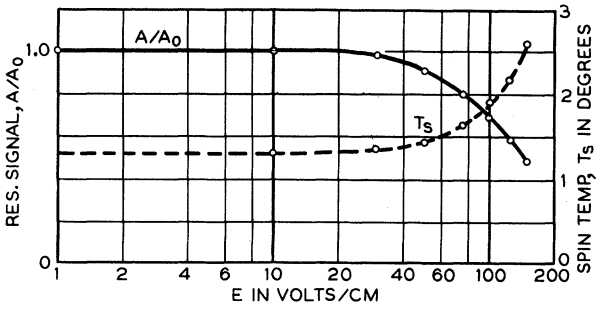

FIG. 3. Amplitude of the electron spin resonance signal from bound electrons in phosphorus-doped silicon at a lattice temperature of 1.3°K. The spins of the bound electrons are assumed to be in equilibrium with the spins of the hot electrons. The amplitude of the signal may therefore be used to measure T_S. The electrical characteristic of the same sample is illustrated in Fig. 2.

appreciably heating the lattice. Experiments on germanium are in progress.

A temperature difference between T_R and T_S may also be obtained in systems in which the free carriers relax via spin-orbit coupling.[8] In this case the spin relaxation time may be several orders of magnitude longer than the electron-lattice collision time. As a result T_S will lag behind T_R. If the electrons are accelerated in an electric field gradient, a difference between T_R and T_S will be produced. The sign of the nuclear polarization will then depend on the sign of the electric field gradient.

Another possible application of the saturation of the electron spin resonance signal in the presence of hot electrons seems worth mentioning. If the hot electrons could preferentially saturate the outer levels of a three-level system, a maser would be realized which would not require the high pumping frequency that is presently used.[9]

It is a pleasure to acknowledge many helpful discussions with P. W. Anderson and H. Suhl which provided the stimulus for the above work, and to thank E. A. Gere for his assistance in the experiments and I. M. Mackintosh for the preparation of the n^+ contacts.

[1]A. W. Overhauser, Phys. Rev. 92, 411 (1953).

[2]A. W. Overhauser, Phys. Rev. 94, 768 (1954); J. Korringa, Phys. Rev. 94, 1388 (1954); F. Bloch, Phys. Rev. 93, 944 (1954); C. Kittel, Phys. Rev. 95, 589 (1954); A. Abragam, Phys. Rev. 98, 1729 (1955).

[3]This discussion, as well as the Overhauser effect, is of course not limited to electron-nuclear systems but is applicable to any two interacting spin systems (e.g., conduction electrons interacting with bound electrons).

[4]G. Feher and E. A. Gere, Phys. Rev. (to be published).

[5]The n^+ contacts were prepared by I. M. Mackintosh. 5×10^{20} P/cm^2 were diffused to a depth of ~ 0.0003 in. and a 500A gold layer evaporated on top of it.

[6]Such a breakdown in silicon was observed at 20°K by W. Kaiser and G. H. Wheatley (to be published). It is in contradiction with the conclusions of Lampert et al. [Lampert, Herman, and Steele, Phys. Rev. Letters 2, 394 (1959)].

[7]For a review article on this subject, see S. H. Koenig, J. Phys. Chem. Solids 8, 227 (1959).

[8]R. J. Elliott, Phys. Rev. 96, 266 (1954).

[9]N. Bloembergen, Phys. Rev. 104, 329 (1956).

CYCLOTRON RESONANCE IN ALUMINUM*

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We have observed cyclotron resonance in a single-crystal sample of aluminum at 24 kMc/sec and 4.2°K. The results indicate the presence of two distinct groups of carriers with properties consistent with the theoretical analysis of Heine.[1]

Changes in power absorption in the sample were observed using a standard microwave reflection spectrometer. Both absorption and absorption-derivative measurements were made, the latter using magnetic field modulation.

The sample was cut from a single crystal of zone-refined aluminum using a string saw wetted with $CuCl_2$ solution. The resulting plane surface was then smoothed and electropolished. This surface was a {110} plane and formed part of the end wall of a TE_{011} cylindrical cavity which was mounted with the magnetic field parallel to the sample surface. The cavity together with the sample could be rotated about their common axis.

Figure 1 shows experimental curves of absorption and absorption-derivative versus magnetic field taken with the magnetic field 10° from a ⟨110⟩ crystal axis. At fields above 2 kilo-oersteds the absorption curve shows a component periodic in $1/H$ superimposed on a monotonically decreasing component, a behavior similar to that previously reported in copper.[2] The variation is (not surprisingly) somewhat different from that predicted by Azbel' and Kaner[3] for a free-electron gas. The absorption-derivative curve shows the oscillations more clearly. These oscillations correspond to a cyclotron mass of $1.5m_0$ and an $\omega\tau$ of about 10.

For some sample orientations the hump which appears at 2 kilo-oersteds on the absorption curve in Fig. 1 is resolved into distinct peaks. Experimental curves for such an orientation are shown in Fig. 2. These curves were taken with the magnetic field parallel to a ⟨111⟩ crystal axis. The peaks correspond to a cyclotron mass