negative mu mesons in this momentum interval is 4700. Since none of these particles was absorbed in 147 g/cm² of lead the removal path length must be greater than 6.9×10^5 g/cm². The upper limit for the corresponding removal cross section is 2.4×10^{-30} cm²/nucleon. The statistical significance placed on these limits is the same as that placed on the previously determined upper limit for the intensity of pi mesons.

This result is more direct than the results obtained by the earlier experiments in the sense that no corrections have to be made for instrumental errors or for nuclear events produced by particles other than mu mesons, e.g., pi mesons and protons in equilibrium with the mu meson flux. However, it should be pointed out that the cross section corresponding to a removal path length refers only to those collisions in which a large fraction of the incident energy is distributed among several heavily ionizing particles.

Polarization of Nuclei in Metals

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(Received June 25, 1953)

A new method for polarizing nuclei, applicable only to metals, is proposed. It is shown that if the electron spin resonance of the conduction electrons is saturated, the nuclei will be polarized to the same degree they would be if their gyromagnetic ratio were that of the electron spin. This action results from the paramagnetic relaxation processes that occur by means of the hyperfine structure interaction between electron and nuclear spins. A shift of the electron spin resonance due to the same interaction will occur for large amounts of polarization and should provide a direct indication of the degree of polarization.

I. INTRODUCTION

A NUMBER of methods[1] have been proposed for producing polarization or alignment of nuclear spins. A new method, applicable to the nuclei of atoms of a metallic solid, is made possible by the existence[2] of the electronic spin resonance from electrons in the conduction band. The interaction between the electron spin magnetic moment β_e and the nuclear spin magnetic moment β_n that is responsible for the polarizing action is that of the hyperfine coupling of an S state,[3]

$$H = -(8\pi/3)\beta_e \cdot \beta_n \delta(\mathbf{r}). \tag{1}$$

Here, $\delta(\mathbf{r})$ is a delta function of the relative coordinate of the electron and nucleus under consideration. The polarization is produced when the metallic sample, in a constant magnetic field $\mathfrak{K}_0$, is irradiated with a perpendicular microwave magnetic field, $2\mathfrak{K}_1 \cos\omega t$, of frequency such as to satisfy the electron spin resonance criterion,

$$\hbar\omega = 2\beta_e \mathfrak{K}_0. \tag{2}$$

The metal is then in a nonequilibrium condition, and we shall show that the dynamical processes which tend to restore the system to its equilibrium state induce nuclear transitions via Eq. (1) in predominantly one

direction, with a resulting steady state nuclear polarization. This action cannot be attributed to a large Boltzmann factor for the energy separations of adjacent nuclear magnetic states, and the spin-temperature concepts, familiar in the literature of magnetic resonance, do not apply.

The magnitude of the polarizing effect depends upon the degree to which the electron resonance can be saturated, and hence upon the microwave power available, the paramagnetic relaxation time, and the resonance line width. A number of spin relaxation processes have been studied by the writer in a paper,[4] hereafter referred to as A, and the methods used for those calculations will be followed here. We shall consider first the problem of resonance absorption in the electron spin system.

II. ELECTRON RESONANCE ABSORPTION

If a metal is placed in a magnetic field $\mathfrak{K}_0$, say in the z direction, the spin magnetic moments of the conduction electrons tend to line up parallel to the field. If N_+ and N_- are, respectively, the number of electrons per cc with spin up and down, the degree of bulk magnetization is proportional to the difference, $D = N_- - N_+$, and the equilibrium value of D is

$$D_0 = 3N\beta_e \mathfrak{K}_0/2\epsilon_f. \tag{3}$$

If, in addition, the metal is subjected to a perpendicular, alternating magnetic field, $2\mathfrak{K}_1 \cos\omega t$, of frequency such

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[1] C. J. Gorter, Physica 14, 504 (1948); M. E. Rose, Phys. Rev. 75, 213 (1948); B. Bleaney, Proc. Phys. Soc. (London) A64, 315 (1951); R. V. Pound, Phys. Rev. 76, 1410 (1949); A. Kastler, J. phys. et radium 11, 255 (1950), Compt. rend. 233, 1444 (1951).

[2] Griswold, Kip, and Kittel, Phys. Rev. 88, 951 (1952).

[3] E. Fermi, Z. Physik 60, 320 (1930).

[4] A. W. Overhauser, Phys. Rev. 89, 689 (1953).

as to satisfy Eq. (2), the time variation of D is given by

$$dD/dt = (D_0 - D)/T_1 - \alpha D. \tag{4}$$

The first term in Eq. (4) is due to the relaxation processes that are effective in establishing the equilibrium degree of spin magnetization. A number of such processes have been studied in A, and the total relaxation can be described by a term of the above type with a characteristic relaxation time T_1. The second term is due to the alternating field, which tends to decrease the amount of magnetization since more transitions are induced from the spin state of greater population. The value of α can be derived by a simple calculation using time dependent perturbation theory and is given by

$$\alpha = 4\pi\beta_e^2 \mathfrak{IC}_1^2/hw. \tag{5}$$

Here, w is the integrated width of the resonance line. That is, if the shape of the resonance as a function of energy is given by $P(E)$, having a maximum value $P(E_0)$, then w is defined by the equation

$$wP(E_0) = \int_0^\infty P(E)dE.$$

After the alternating field has been turned on, a steady state will be reached such that $dD/dt = 0$. A net rate of spin flips in one direction due to $\mathfrak{IC}_1$ will be balanced by a similar rate in the opposite direction due to relaxation interactions. Of these latter processes those due to Eq. (1) will bring about the nuclear polarization. From Eq. (4) the steady-state value of D is

$$D = D_0/(1 + \alpha T_1). \tag{6}$$

In the limit of complete saturation of the resonance, large $\mathfrak{IC}_1$, $D \rightarrow 0$. It is convenient to define a parameter s which is 0 when there is no alternating field and is 1 for complete saturation:

$$s = 1 - (D/D_0). \tag{7}$$

From (6) we obtain

$$s = \alpha T_1/(1 + \alpha T_1).$$

We shall see that the degree of nuclear polarization will be proportional to s, which is a function only of αT_1.

The electron spin resonance in lithium has been studied recently by Slichter,[5] using radio-frequencies and particles of the metal that were small compared to the skin depth. The resonance was found to be Lorentzian in shape. The width of the line between maximum slopes was 5 oersteds, corresponding to a transverse relaxation time T_2 of 10^{-8} sec. The relaxation time T_1 was measured by the method of saturation and found to be 10^{-8} sec. Thus, the expected agreement between T_1 and T_2 for conduction electrons was confirmed.

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[5] C. P. Slichter, private communication.

For a Lorentzian line the integrated width w is given by

$$w = \pi h/T_2.$$

The fractional saturation can then be written:

$$s = \gamma_e^2 \mathfrak{IC}_1^2 T_1 T_2/(1 + \gamma_e^2 \mathfrak{IC}_1^2 T_1 T_2). \tag{8}$$

Here, we have replaced $2\beta_e/\hbar$ by γ_e, the gyromagnetic ratio of the electron spin. From Eq. (8) we see that appreciable saturation results only if

$$\gamma_e^2 \mathfrak{IC}_1^2 T_1 T_2 \sim 1.$$

The amplitude of the microwave field that is necessary to saturate the resonance is inversely proportional to the relaxation times, and at room temperature $\mathfrak{IC}_1$ must be of the order of 5 oersteds. At liquid helium temperature $\mathfrak{IC}_1$ need be less, perhaps, if T_1 and T_2 are correspondingly longer. The shortest relaxation time that was found in A, due to the spin-current interaction, was inversely proportional to the absolute temperature. However, the observed relaxation time is smaller by a factor of 100 than the value given by theory. An important relaxation mechanism has probably been overlooked, so that the correct temperature dependence of T_1 is as yet unknown.

III. PRODUCTION OF NUCLEAR POLARIZATION

The nuclear polarization that results when $s > 0$ can be derived most easily from the results of Sec. IV of A, where the paramagnetic relaxation due to the hyperfine interaction (1) was studied. These calculations made use of the assumption that $\beta_e \mathfrak{IC}_0/\kappa\theta \ll 1$, but we shall show later that the results of interest here remain unaltered if this approximation is not used. The variation of D with time, (A31), was found to be

$$(dD/dt)' = (D_0 - D)/T_1' + G(\Delta_0 - \Delta)/T_n'. \tag{9}$$

The "prime" is used here to denote quantities that are associated with the hyperfine interaction. Thus, T_1' and T_n' are the electronic and nuclear relaxation times due to this interaction. The difference in population between adjacent nuclear magnetic levels is Δ, whose equilibrium value in the field $\mathfrak{IC}_0$ is

$$\Delta_0 = Nh\gamma_n \mathfrak{IC}_0/(2I + 1)\kappa\theta. \tag{10}$$

The quantity G is the constant of proportionality between the total excess z component of nuclear spin and $\frac{1}{2}\Delta$, and is

$$G = \tfrac{2}{3}I(I+1)(2I+1).$$

Since Eq. (1) conserves the total spin angular momentum, we have

$$(dD/dt)' = G(d\Delta/dt)'.$$

Equation (9) can then be written

$$(d\Delta/dt)' = (\Delta_0 - \Delta)/T_n' + (D_0 - D)/GT_1'. \tag{11}$$

If we assume, temporarily, that the only interaction

through which Δ is allowed to change is Eq. (1), the steady-state condition corresponds to Eq. (11) being zero. The steady-state value of Δ, with the use of Eq. (7), is then

$$\Delta = \Delta_0 + sD_0 T_n'/T_1'G. \tag{12}$$

Now, the ratio T_n'/T_1' from (A34) is

$$T_n'/T_1' = 8I(I+1)\epsilon_f/9\kappa\theta.$$

On using this result together with Eqs. (3) and (10), Eq. (12) becomes

$$\Delta = \frac{N\hbar\mathcal{3C}_0}{(2I+1)\kappa\theta}(\gamma_n + s|\gamma_e|). \tag{13}$$

We see, therefore, that the steady-state nuclear polarization is larger by an amount that would be expected if the nuclear gyromagnetic ratio were increased by $s|\gamma_e|$. If s is near unity, an enhanced polarization by a factor of about 2000 should result.

If there are interactions other than Eq. (1) which also produce nuclear magnetic relaxation, so that the total nuclear relaxation time is T_n, we must consider the total variation of Δ with time:

$$d\Delta/dt = (\Delta_0 - \Delta)/T_n + (D_0 - D)/GT_1'.$$

This equation must be set equal to zero instead of Eq. (11), and we find, similarly, that the polarization is as though the nuclei had an effective gyromagnetic ratio

$$\gamma_{\rm eff} = \gamma_n + s(T_n/T_n')|\gamma_e|. \tag{14}$$

This equation expresses the general result, which has been reported earlier.[6] It should be appreciated that other electronic relaxation processes, of which there
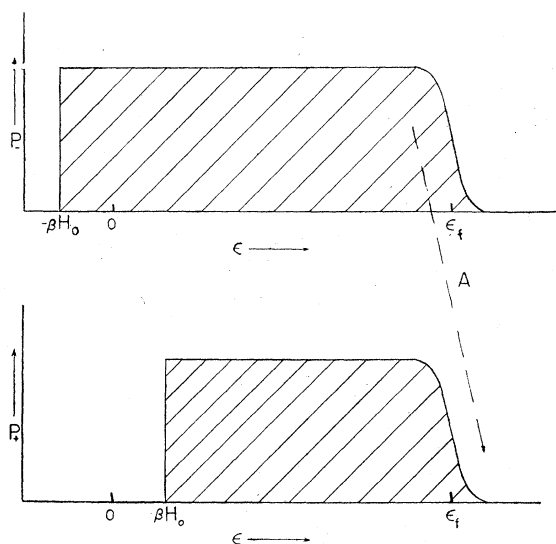

FIG. 1. Schematic diagram of the Fermi distributions of the two spin states in equilibrium.
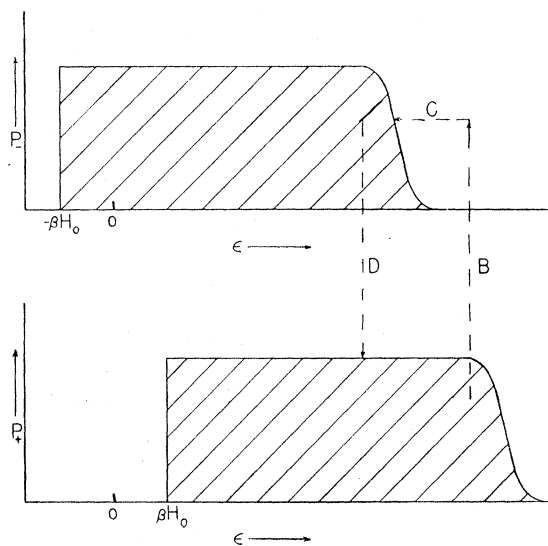
[6] A. W. Overhauser, Phys. Rev. 91, 476 (1953).

FIG. 2. Schematic diagram of the Fermi distributions of the two spin states for complete saturation.

are many,[4] do not affect the polarizing action other than in making the electron resonance more difficult to saturate. The nuclear relaxation processes have been studied theoretically by Korringa[7] and experimentally by a number of workers. The results indicate that Eq. (1) is the important relaxation interaction, so that T_n/T_n' is close to unity.

The foregoing derivation gives little insight into the physical reasons why such a polarizing effect takes place. To understand this action one may consider the conduction band as consisting of two Fermi distributions, one for the electrons with spin up and the other for the electrons with spin down. Two such distributions are shown schematically in Fig. 1, where the occupation probability is plotted as a function of the energy of the electronic state, including the magnetic energy in the field $\mathcal{3C}_0$. The equilibrium distributions are shown in Fig. 1, so that the Fermi energies are equal, and there is an excess number of electrons in the spin down distribution, corresponding to a net magnetization in the field direction. If the oscillating field is turned on, transitions represented by the arrow A will predominate, so that the number of electrons in each distribution will tend to be equalized. If the electron resonance is completely saturated, the two distributions will appear as in Fig. 2 and will have equal numbers of electrons. We need consider now only those relaxation processes due to the hyperfine interaction and which flip over nuclear spins. Such a process is represented by the vertical arrow B in Fig. 2, which corresponds to an electron spin being flipped down, so that a nuclear spin is flipped upwards. As was shown in A, the characteristic time of such processes is about 10^{-4} sec. As soon as the transition B has taken place, the electron will

[7] J. Korringa, Physica 16, 601 (1950).

undergo transitions into the surface of the Fermi distribution such as is represented by the arrow C. Such transitions are due to interactions with the lattice, familiar in the theory of electrical conductivity, and have a characteristic time of 10^{-13} sec. Transitions that flip nuclear spins in the reverse direction are represented by the arrow D in Fig. 2 and are forbidden because the final states are already occupied. The polarizing action takes place, therefore, because nuclear spins are flipped over in predominantly one direction by the relaxation processes due to Eq. (1) and cannot return. Actually, the reverse processes occur to some extent because the surfaces of the Fermi distributions are finite in width and have exponential tails. As can be seen from Eq. (14), the rates in the two directions must be so balanced that the excess polarization obeys Curie's law if $s(T_n/T_n')$ is kept constant.

A convenient definition for the degree of nuclear polarization $\mathcal{P}$ is the quotient of the z component of nuclear spin and the maximum possible value. Thus

$$\mathcal{P}= \left| \sum_{m=-I}^{I} m e^{mu} \right| \Big/ \left(I \sum_{m=-I}^{I} e^{mu} \right), \qquad (15)$$

where the ratio of the populations of adjacent magnetic states is

$$M_m/M_{m-1}=e^{\mu}. \qquad (16)$$

We have shown that μ is given by

$$\mu=[\gamma_n+s(T_n/T_n')|\gamma_e|]\hbar\mathfrak{K}\mathcal{C}_0/\kappa\theta, \qquad (17)$$

but only if $\beta_e\mathfrak{K}\mathcal{C}_0 \ll \kappa\theta$ (so that $\mu \ll 1$), since this approximation was used in the derivation of Eq. (9).

In order to obtain a large degree of polarization, $\mathcal{P}$ near 1, μ must be of the order of magnitude of unity or greater, and we must now establish that Eq. (16) remains valid for larger μ. To do so we shall assume that interactions of the form (1) are the only source of nuclear relaxation, and hence that $T_n/T_n'=1$. We consider first the special case $I=\frac{1}{2}$ and calculate the number of nuclei per second $W_{+,-}$ flipped from the state $m=-\frac{1}{2}$ to the state $m=+\frac{1}{2}$. This quantity is given by (A29), and the indicated integration can be performed by elementary methods and yields

$$W_{+,-}=M_{-1/2}\mu/[2(1-e^{-\mu})T_n'].$$

Similarly, the number of nuclei flipped per second from $m=+\frac{1}{2}$ to $m=-\frac{1}{2}$ can be calculated and is

$$W_{-,+}=M_{+1/2}\mu/[2(e^{\mu}-1)T_n'].$$

When a steady state is established, $W_{+,-}$ and $W_{-,+}$ must be equal, and we find

$$M_{+1/2}/M_{-1/2}=e^{\mu}.$$

Thus, Eq. (16) is obtained as an exact result, with

$$\mu=(\gamma_n+s|\gamma_e|)\hbar\mathfrak{K}\mathcal{C}_0/\kappa\theta.$$

The generalization required for $I>\frac{1}{2}$ is trivial. It is

sufficient to calculate $W_{m,m-1}$ and $W_{m-1,m}$ and to observe that Eq. (16) is the condition that they be equal. If there are other nuclear relaxation processes, one can show that Eq. (16) is a good approximation, with μ given by Eq. (17), if T_n/T_n' is not too much smaller than 1.

In order to illustrate the degree of polarization that should be possible by this technique, consider the following example: $I=\frac{3}{2}$, $\mathfrak{K}\mathcal{C}_0=10^4$ oersteds, $\theta=2°$K, and $s=0.8$. One finds $\mu=0.54$, and Eq. (15) gives a value of 42 percent for the polarization. The electron resonance for this case would require microwaves in the 1-cm region.

It may be asked why the effect which we have described can occur only in metals, since the hyperfine interaction that is responsible certainly exists in other paramagnetic materials. The basic reason is that electrons in an unfilled band can change energy by small amounts, $\sim\kappa\theta$, by small changes in momentum or wave number. This freedom is necessary in order that energy be conserved during the simultaneous spin flip of an electron and a nucleus. In nonmetals, where the valence bands are full or, alternatively, where bound state wave functions are more appropriate, small energy changes necessary to compensate for jumps in magnetic energy can be taken up only by the emission, absorption, or scattering of phonons, and these processes do not couple electron and nuclear spins.

IV. ELECTRON RESONANCE SHIFT

The hyperfine interaction gives rise to a nuclear resonance shift in metals. This shift, first observed by Knight,[8] can be described as the effect of an extra magnetic field at the nucleus due to the feeble spin paramagnetism of the conduction electrons, which interact with the nucleus according to Eq. (1). A similar change in the electron-spin resonance will occur if there is an appreciable degree of nuclear polarization. Thus, the energy of an electron spin in the presence of a field $\mathfrak{K}\mathcal{C}_0$ and the N nuclei per cc of metal is

$$H=-\beta_e\cdot[\mathfrak{K}\mathcal{C}_0+(8\pi/3)|\psi(0)|^2\sum\beta_n].$$

Here, $|\psi(0)|$ is the magnitude of the wave function at the nucleus, normalized in 1 cc, of a conduction electron. Since the nuclear spins will be polarized in the direction of $\mathfrak{K}\mathcal{C}_0$, the second term in Eq. (18) can be represented as an extra magnetic field parallel to $\mathfrak{K}\mathcal{C}_0$ and of magnitude

$$\Delta\mathfrak{K}\mathcal{C}=\mathcal{P}(\Delta\mathfrak{K}\mathcal{C})_0, \qquad (18)$$

where $(\Delta\mathfrak{K}\mathcal{C})_0$ is the maximum value, for 100 percent nuclear polarization, and is given by

$$(\Delta\mathfrak{K}\mathcal{C})_0=(8\pi/3)|\psi(0)|^2N\beta_n. \qquad (19)$$

This shift can be relatively large, and the value of Eq. (19) for Li[7] is 217 oersteds. $|\psi(0)|^2$ was taken to be 34, a value based on the calculations of Kohn and

[8] W. D. Knight, Phys. Rev. 76, 1259 (1949).

Bloembergen.[9] This effect will be even larger for other metals and will complicate the procedure for producing large amounts of polarization.

A more favorable feature of the electron resonance shift is that it should provide, in principle, a direct measurement of the degree of polarization obtained. For example, if one were to apply the microwave power, the resonance line would be shifted towards higher frequencies, or for fixed frequency, towards lower magnetic fields. The field would then have to be decreased gradually to a critical value at which maximum polarization is reached. Beyond this value the resonance would be lost suddenly, and the line would snap back to its original, low microwave power position. The polarization $\mathscr{P}$ is related to the critical shift by Eq. (18) and could be maintained by operating on the high field side of the resonance. For nuclei having a negative gyromagnetic ratio this shift would be in the opposite direction, since these nuclei will at first be depolarized and then lined up in the opposite direction by the action of the relaxation processes.

V. CONCLUSION

Within the framework of the single particle model an enhanced nuclear polarization is predicted as the result

of relaxation processes that occur when the electron-spin resonance is saturated. The possibility of producing large amounts of polarization by this technique is of great experimental interest, although the problems that would have to be solved are by no means trivial. Microwaves in the 1-cm range are appropriate, and considerable power may be needed, depending upon the value of the electronic relaxation time at liquid helium temperature. In order to avoid eddy current losses as well as excessive breadth of the electron resonance,[2] it would be necessary to use finely divided metallic samples of particle size somewhat smaller than a micron. The use of chemically impure samples to increase the electrical resistivity at low temperatures would be helpful in this regard.

In nuclear resonance experiments the signal strength is proportional to the degree of nuclear polarization, which is ordinarily quite small. Such nuclear resonances would be enhanced by several orders of magnitude if the electron resonance were saturated simultaneously. This interdependence could be used as a means for detecting the electron resonance. Other effects would be a disappearance of the nuclear resonance shift at high saturation and a decrease in the dipolar broadening of the nuclear line for large polarizations.

[9] W. Kohn and N. Bloembergen, Phys. Rev. 82, 263 (1951).

Effects of the Geomagnetic Field on Solar Cosmic Rays

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(Received June 2, 1953)

The force acting on a charged solar cosmic ray due to the action of the geomagnetic field has been analyzed in terms of its components along the E–W, the N–S, and the vertical directions. With certain simplifying assumptions, the relative magnitudes of these components and their directions have been determined for specific values of the geographic latitude λ of the place of observation as well as the declination δ and the hour angle t of the sun. The deflection that can be produced in the trajectories of solar cosmic rays is qualitatively discussed in terms of the resulting changes which may be expected in the hour of maximum of the diurnal variation of cosmic rays assumed to be due to particles from the sun. It appears that the hour of maximum should become later with increase of latitude. In a northern latitude, a north-pointing cosmic-ray telescope should reveal a diurnal variation with an earlier maximum than a south-pointing telescope. There is a possibility therefore of being able to interpret the experimental results of directional studies of the diurnal variation of cosmic rays in terms of geomagnetic effects.

INTRODUCTION

EVIDENCE[1–5] has been accumulating recently indicating that the sun continuously emits cosmic rays. These rays are believed to be responsible for a daily variation in the pressure-corrected meson intensity

measured in the lower atmosphere. The variation has a predominant 24 hourly harmonic component whose amplitude may be designated by M^D. The hour of maximum of the diurnal component can be expressed in terms of the angle $M\phi^D$ made by the radius vector of M^D with respect to midnight on a 24 hourly harmonic dial representation. In general, $M\phi^D$ is different from π as the maximum does not coincide with the local noon of the place of observation. This is believed to be due to the effect of the geomagnetic field on charged solar cosmic rays.

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