

plot proved to be too insensitive to represent clearly the small differences in magnetoresistive behavior found in these specimens. Accepting the apparent validity of Eq. (2) for Mg(Fe) specimen, Fig. 1 exhibits the data for both our specimens in the form of a modified Kohler plot in which  $\log(\Delta R/R_{0T}) - 1.326 \log(HR_{0^\circ C}/R_{0T})$  is given as a function of  $\log(HR_{0^\circ C}/R_{0T})$ . In this figure, the solid horizontal line is given by Eq. (2) and the dashed lines represent deviations of  $\pm 10\%$  in  $\Delta R/R_{0T}$ . The agreement with Kohler's rule in the measurements on the Mg(Fe) specimen, as well as the deviations from this rule found in the Mg(Mn) specimen, are quite evident.

Also shown in Fig. 1 are the results of measurements of the magnetoresistance of magnesium reported by Thomas and Mendoza<sup>11</sup> and by Yntema.<sup>12</sup> The curve "T & M" represents the average arrived at by Thomas and Mendoza from measurements on four specimens. Except for a few points taken at low magnetic fields, these data and also the results for Yntema's specimen Mg 2 are in excellent agreement with Eqs. (2) and (3). Yntema's specimen Mg 5 apparently forms an exception to the more common behavior. It should be noted that our first specimen, Mg(Fe), came from the same source (Johnson-Matthey, laboratory No. 1848) as did the specimens measured by Yntema and by Thomas. However, the mechanical preparation of these specimens was quite different in that our Mg(Fe) specimen was severely cold-worked, whereas the previously reported specimens had been drawn into wires and (except for one of the four specimens investigated by Thomas) subsequently reannealed.

## CONCLUSIONS

1. The magnetoresistance at liquid helium temperatures of a magnesium specimen containing 0.013% iron, Mg(Fe), agrees both with Kohler's rule and with the majority of previous measurements on specimens of similar composition.

2. In a second specimen, Mg(Mn), containing 0.043% manganese, the magnetoresistance at 4.2°K was in fair agreement with the magnetoresistance of the Mg(Fe) specimen. At 1.3°K, however, the magnetoresistance falls about 10% below the values prescribed by the Kohler rule.

3. In terms of the Gerritsen classification (see Introduction), it seems probable that the Mg(Fe) specimen falls into group 2 (metals showing a resistance minimum, but no maximum) and that the Mg(Mn) specimen may be an extreme case of group 3 (metals showing both a resistance maximum and minimum). Since the deviation of Mg(Mn) from Kohler's rule is still quite small even at one-tenth the temperature of the resistance minimum (14.5°K), it seems reasonable that the resistance maximum, if it occurs at all, may be found at a temperature very close to 0°K. These conclusions are consistent with the fact (see II) that the electrical resistance of both the Mg(Fe) and Mg(Mn) specimens increases monotonically for temperatures down to 0.2°K.

## ACKNOWLEDGMENT

The author is indebted to the members of the Cryogenics Branch for their encouragement and assistance, and especially to Dr. T. G. Berlincourt for providing the magnet calibration and some of the apparatus used in this research.

## Ferromagnetic Relaxation by the Exchange Interaction between Ferromagnetic Electrons and Conduction Electrons\*

A. H. MITCHELL

*Department of Physics, University of California, Berkeley, California*

(Received November 19, 1956)

The Hamiltonian for the exchange interaction between the ferromagnetic *d* electrons and the conduction *s* electrons is derived. The ferromagnetic relaxation time caused by the *s-d* exchange interaction is calculated in a spin wave approximation. When one uses a screened value of the exchange integral ( $J_{\text{atomic}}/30$ ), the calculated relaxation time for nickel at room temperature is  $5 \times 10^{-9}$  sec as compared to the time  $2.5 \times 10^{-10}$  sec needed to account for the experimental line width. The exchange relaxation may be dominant in materials such as alloys which have narrower lines than nickel.

### I. INTRODUCTION

THE line widths observed in electron spin-resonance experiments on ferromagnetic single metal crystals are of the order of several hundred oersteds,<sup>1</sup> cor-

responding to relaxation times of the order of  $10^{-9}$  to  $10^{-10}$  sec. The widths in several metals appear to be temperature-independent at low temperatures.<sup>1</sup> If we neglect all interactions except the isotropic exchange and Zeeman interactions, the Hamiltonian of the system of ferromagnetic electrons is

$$\mathfrak{H}_0 = -2 \sum_{i < j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - g \mu_B \mathbf{H} \cdot \sum_i \mathbf{S}_i. \quad (1)$$

\* Supported in part by the Office of Naval Research, U. S. Signal Corps, and the U. S. Air Force Office of Scientific Research.

<sup>1</sup> K. H. Reich, Phys. Rev. **101**, 1647 (1956); N. Bloembergen, Phys. Rev. **78**, 572 (1950); a list of line widths is given in C. Kittel and E. Abrahams, Revs. Modern Phys. **25**, 233 (1953).

By ferromagnetic electrons we mean the  $3d$  electrons in metals and alloys of the first transition group. In the spin-wave approximation the energy of the low-lying states of the ferromagnet (neglecting shape and crystal anisotropy) is given by

$$\epsilon_0 = \sum_{\mathbf{k}} (n_{\mathbf{k}} + \frac{1}{2}) (2SJ\mathbf{k}^2 a^2 - g\mu_B H), \quad (2)$$

where  $n_{\mathbf{k}}$  is the number of spin waves of wave vector  $\mathbf{k}$ .

In an insulator transitions will occur only among the  $\mathbf{k}=0$  levels so that the resonance frequency is  $\omega = g\mu_B H/\hbar$ . It has been shown<sup>2</sup> that in a ferromagnetic conductor the resonance frequency for the absorption of microwave power is shifted in first approximation by an amount inversely proportional to the square of the eddy current skin depth  $\delta$ . In other words, the only spin waves excited by the rf field have wave vectors  $\mathbf{k}$  whose magnitudes lie close to the value  $1/\delta$ .

In order to obtain a relaxation effect we must introduce a perturbation into the Hamiltonian that will provide for the decay of any excess over the thermal equilibrium number of spin waves with  $|\mathbf{k}|=1/\delta$ . Several mechanisms<sup>3</sup> involving magnetic interactions have been suggested to explain the line widths, but they appear to yield relaxation times several orders of magnitude too long.<sup>4</sup> Bloembergen<sup>5</sup> has suggested that the magnetic interaction of the ferromagnetic electrons with the conduction electrons should be considered. Abrahams<sup>6</sup> has treated in detail the relaxation time due to the magnetic interaction of ferromagnetic spins with conduction electron currents and spins, using a spin-wave description of the ferromagnetic spins. These interactions do not appear strong enough to explain the experimental widths.

It is our purpose here to calculate the ferromagnetic relaxation time caused by the exchange interaction between the ferromagnetic and conduction electrons as suggested briefly by Kittel and Mitchell.<sup>7</sup> We consider specifically pure Fe, Co, and Ni; here the magnetization is associated with  $3d$  electrons in a sea of  $4s$  conduction electrons. In the free atoms of the transition metals the  $s$ - $d$  exchange energy<sup>8</sup> is of the order of 0.5 eV. However, we have made estimates suggesting that if screening by  $3d$  electrons is effective the  $s$ - $d$  interaction might be reduced to only 1 to 10% of the free-atom value. It is not obvious that the screening esti-

mates are correct. In this connection we might mention that in Cu-Mn alloys it has also been found necessary to consider values of the  $s$ - $d$  exchange energy 1/10 to 1/50 of the free-atom value.<sup>9</sup>

Before calculating the relaxation time we repeat the objections<sup>7</sup> to the argument that an interaction of the form  $\mathbf{S} \cdot \mathbf{s}$ , where  $\hbar\mathbf{S}$  is the spin of a  $3d$  ion core and  $\hbar\mathbf{s}$  is the spin of a  $4s$  conduction electron, cannot contribute to relaxation. The argument is that  $\mathbf{S} \cdot \mathbf{s}$  commutes with the total spin  $\mathbf{S} + \mathbf{s}$  and thus cannot affect the time dependence of the total magnetization. If, however, the conduction electron spin is relaxed independently by a rapid mechanism,<sup>10</sup> then in a ferromagnetic resonance experiment we are observing the resonance of  $\mathbf{S}$  alone and the above objection is no longer pertinent.

We can estimate how rapidly the conduction electron spin must be relaxed by an extension of the results of Solomon<sup>11</sup> on relaxation processes in a system of two spins. If we take a perturbation  $-2J\mathbf{S} \cdot \mathbf{s}$  giving rise to a transition probability per unit time  $w$  between states connected by the off-diagonal components of the perturbation and add a relaxation process characterized by a relaxation frequency  $\rho$  for the conduction electron spin  $\mathbf{s}$  alone, then Solomon's Eq. (9) becomes

$$\begin{aligned} d\bar{s}_z/dt &= -(w+\rho)(\bar{s}_z - s_0) + w(\bar{S}_z - S_0), \\ d\bar{S}_z/dt &= w(\bar{s}_z - s_0) - w(\bar{S}_z - S_0), \end{aligned} \quad (3)$$

where  $s_0$  and  $S_0$  refer to thermal equilibrium values. If the  $4s$  relaxation time is much shorter than the exchange relaxation time so that  $w/\rho \ll 1$ , then these equations have the solution  $(\bar{s}_z - s_0) \propto e^{-\rho t}$  and  $(\bar{S}_z - S_0) \propto e^{-wt}$  showing that the conduction electron spin is essentially in thermal equilibrium while the relaxation time of the  $3d$  electrons is governed by the  $s$ - $d$  exchange interaction. From the experimental line widths we know the relaxation time of the ferromagnetic spins is in the range  $10^{-9}$  to  $10^{-10}$  sec. If the mechanism relaxing the conduction electron spins is that proposed by Elliott,<sup>10</sup> the spin relaxation time is  $\tau_s \sim \tau_R/10(\Delta g)^2$ , where  $\tau_R$  is the relaxation time characteristic of electrical conductivity. In the transition elements the  $s$  and  $d$  bands overlap so that  $\Delta g$  is expected to be relatively large, say  $>10^{-2}$ . In Ni,  $\tau_R \sim 2 \times 10^{-15}$  sec at room temperature and  $\tau_R \sim 4 \times 10^{-14}$  at  $14^\circ\text{K}$  if we use the experimental values of the resistivity obtained by Jan and Gijsman.<sup>12</sup> With these values we obtain a  $\tau_s$  in the range  $10^{-13}$ – $10^{-10}$  sec, the longer time applying at low temperatures. Thus in the transition metals the relaxation times can be such that the exchange interaction is effective in relaxing the ferromagnetic spins. The failure thus far to detect conduction electron spin

<sup>2</sup> C. Kittel and C. Herring, Phys. Rev. **77**, 725 (1950).

<sup>3</sup> N. Bloembergen, Phys. Rev. **78**, 572 (1950); C. Kittel and E. Abrahams, Revs. Modern Phys. **25**, 233 (1953); A. Kondoh, Progr. Theoret. Phys. Japan **10**, 117 (1953); T. Kasuya, Progr. Theoret. Phys. Japan **12**, 803 (1954); E. Abrahams, Phys. Rev. **98**, 387 (1954).

<sup>4</sup> In regard to the relaxation time calculated by T. Kasuya, Progr. Theoret. Phys. Japan **12**, 803 (1954); Abrahams, Phys. Rev. **98**, 387 (1954) has shown that inclusion of screening increases the relaxation time by two orders of magnitude.

<sup>5</sup> N. Bloembergen, Phys. Rev. **78**, 572 (1950).

<sup>6</sup> E. Abrahams, Phys. Rev. **98**, 387 (1954).

<sup>7</sup> C. Kittel and A. H. Mitchell, Phys. Rev. **101**, 1611 (1956).

<sup>8</sup> C. Zener and R. R. Heikes, Revs. Modern Phys. **25**, 191 (1953).

<sup>9</sup> Owen, Browne, Knight, and Kittel, Phys. Rev. **102**, 1501 (1956); Owen, Browne, Arp, and Kip (to be published).

<sup>10</sup> The spin-orbit mechanism considered by R. J. Elliott, Phys. Rev. **96**, 266 (1954), is a very effective relaxation mechanism.

<sup>11</sup> I. Solomon, Phys. Rev. **99**, 559 (1955).

<sup>12</sup> J. P. Jan and J. M. Gijsman, Physica **18**, 339 (1952).

resonance in pure copper is not incompatible with the above estimates of  $\tau_s$ .

## II. SPIN HAMILTONIAN

In deriving the form of the Hamiltonian describing the exchange interaction between the ferromagnetic electrons and the conduction electrons, we consider a periodicity volume of a cubic crystal containing  $N$  atoms and we shall assume that there are  $N$  ferromagnetic electrons forming a half-filled band. Of course, in Fe, Co, and Ni the  $d$  band is actually more than half full and there are five orbital states per atom, not one. The assumption of a half-filled band is made in order that we may make use of the equivalence of the running wave and the localized descriptions of the ferromagnetic electrons. The assumption of one orbital state per atom is made for the sake of simplicity as we are interested mainly in finding the form of the interaction; we shall later generalize our result in the obvious way to take approximate account of the actual spin on each atomic site. These assumptions are equivalent to those made in developing the usual simplified spin-wave model of a ferromagnet.<sup>13</sup>

Because the conduction electron spin is relaxed independently by a rapid mechanism, we can compute the relaxation time of the ferromagnetic spin system by considering the scattering of a single conduction electron by the system of  $N$  ferromagnetic electrons. We shall describe the one-electron states of the ferromagnetic electrons by localized Wannier functions  $a_s^\pm(\mathbf{r})$  which are defined in terms of running wave Bloch functions  $b_{\mathbf{k}}^\pm(\mathbf{r})$  according to

$$a_s^\pm(\mathbf{r}) = a^\pm(\mathbf{r} - \mathbf{R}_s) = (N)^{-\frac{1}{2}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_s} b_{\mathbf{k}}^\pm(\mathbf{r}), \quad (4)$$

where the superscript  $\pm$  refers to the spin state of the electron. The functions defined in this way are localized about the lattice site  $\mathbf{R}_s$  and are orthogonal and normalized in the periodicity volume  $V = N\Omega$ ,  $\Omega$  being the atomic volume. Because the band is half full, the description in terms of localized functions is equivalent to the running-wave description. The conduction electron is specified by a Bloch function  $\psi_{\mathbf{k}}^\pm(\mathbf{r})$ , but in certain instances it will be convenient to express it in terms of Wannier functions  $\phi_s(\mathbf{r})$  as follows:

$$\psi_{\mathbf{k}}^\pm(\mathbf{r}) = (N)^{-\frac{1}{2}} \sum_s e^{i\mathbf{k} \cdot \mathbf{R}_s} \phi_s(\mathbf{r}). \quad (5)$$

The total wave function of the  $N+1$  electrons will be written as an antisymmetric linear combination of these one-electron functions. We will limit the number of possible states by considering only the atomic states in which one ferromagnetic electron is on each lattice site. We can place one electron on each atomic site with either spin up or spin down and we have the same choice for the conduction electron; thus there are  $2^{N+1}$  states. Using Slater determinants, we can write the

$(N+1)$ -electron wave function as

$$A(N^+, n_i, \mathbf{k}^\pm) = [(N+1)!]^{-\frac{1}{2}} \times \det | a_1^-(\mathbf{r}_1) \cdots a_{n_1}^+(\mathbf{r}_{n_1}) \cdots a_{N^-}^-(\mathbf{r}_N) \psi_{\mathbf{k}}^\pm(\mathbf{r}_{N+1}) |. \quad (6)$$

This wave function describes the state in which the conduction electron has wave vector  $\mathbf{k}$  and spin  $\pm$ . If the conduction electron has spin  $-$ , then there are  $N^+$  ferromagnetic electrons with spin  $+$  (located on sites  $n_1, n_2, \cdots, n_{N^+}$ ); if the  $s$  electron has spin  $+$ , there are  $(N^+ - 1)$   $d$  electrons with spin  $+$  (located on sites  $n_1, n_2, \cdots, n_{N^+ - 1}$ ). These states are eigenfunctions of

$$\frac{1}{2}(N+1) \pm \left( \sum_{j=1}^N S_j^z + s^z \right),$$

with eigenvalues  $N^\pm$ , the number of up and down spins. The number of states with a given value of  $N^+$  is  $(N+1)! / (N^+)!(N^-)!$ ; the ground state  $N^+ = 0$  is non-degenerate. The matrix elements of the interaction Hamiltonian,

$$\mathcal{H} = \sum_{i < j}^{N+1} e^2 / r_{ij},$$

between these Slater determinants vanish if the determinants do not have the same  $N^+$  value. If the initial and final states have the same number of up spins, there are three cases in which the matrix element is nonzero:

*Case 1.*—The same set of electrons have spin up in the initial and final states.

$$\begin{aligned} \langle A(N^+, n_i, \mathbf{k}'^\pm) | \mathcal{H} | A(N^+, n_i, \mathbf{k}^\pm) \rangle \\ = \left( \sum_{i < j}^N K_{ij} - \sum_{i < j}^N J_{ij} \right) \delta_{\mathbf{k}', \mathbf{k}} \\ + \sum_{j=1}^N K_j(\mathbf{k}', \mathbf{k}) - \sum_{j=1}^N J_j(\mathbf{k}', \mathbf{k}), \quad (7) \end{aligned}$$

where the  $J$ 's are summed only over parallel spins and

$$K_{ij} = \int d\tau_{12} \frac{e^2}{r_{12}} a_i^*(1) a_i(1) a_j^*(2) a_j(2),$$

$$J_{ij} = \int d\tau_{12} \frac{e^2}{r_{12}} a_i^*(1) a_j(1) a_j^*(2) a_i(2),$$

$$K_j(\mathbf{k}', \mathbf{k}) = \int d\tau_{12} \frac{e^2}{r_{12}} \psi_{\mathbf{k}'}^*(1) \psi_{\mathbf{k}}(1) a_j^*(2) a_j(2),$$

$$J_j(\mathbf{k}', \mathbf{k}) = \int d\tau_{12} \frac{e^2}{r_{12}} \psi_{\mathbf{k}'}^*(1) a_j(1) a_j^*(2) \psi_{\mathbf{k}}(2).$$

*Case 2.*—Same as Case 1 except that in the initial state there is a pair of electrons with opposite spins on

<sup>13</sup> F. Bloch, Z. Physik **61**, 206 (1930); T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940); F. J. Dyson, Phys. Rev. **102**, 1217 (1956).

sites  $i$  and  $j$ , while in the final state the spins on these sites are reversed.

$$\langle A(N^+) | \mathfrak{H} | A(N^+) \rangle = -J_{ij} \delta_{\mathbf{k}', \mathbf{k}}. \quad (8)$$

*Case 3.*—Same as Case 1 except that in the initial state the electron on site  $j$  and the conduction electron have opposite spins while in the final state these spins are reversed.

$$\langle A(N^+) | \mathfrak{H} | A(N^+) \rangle = -J_j(\mathbf{k}', \mathbf{k}). \quad (9)$$

Now we note that the matrix elements of the following spin Hamiltonian  $\mathfrak{H}(\mathbf{k}', \mathbf{k})$  computed using simple products of spin functions are identical with the preceding results and hence  $\mathfrak{H}(\mathbf{k}', \mathbf{k})$  must be the correct spin Hamiltonian for the exchange interaction:

$$\mathfrak{H}(\mathbf{k}', \mathbf{k}) = \sum_{i < j}^N [K_{ij} - \frac{1}{2}(1 + 4\mathbf{S}_i \cdot \mathbf{S}_j) J_{ij}] \delta_{\mathbf{k}', \mathbf{k}} + \sum_{j=1}^N [K_j(\mathbf{k}', \mathbf{k}) - \frac{1}{2}(1 + 4\mathbf{S}_j \cdot \mathbf{s}) J_j(\mathbf{k}', \mathbf{k})], \quad (10)$$

where  $\hbar \mathbf{S}_j$  is the spin of the ferromagnetic electron on site  $j$  and  $\hbar \mathbf{s}$  is the spin of the conduction electron.

With regard to the derivation of this Hamiltonian, it is important to note that use is made of the exact orthogonality of all the one-electron functions and therefore we could not have used simple atomic functions to describe the localized electrons. In order that the conduction electron wave function be orthogonal to all the wave functions of the ferromagnetic electrons it must be in a different band; hence the Hamiltonian does not describe the interaction of one extra electron in the ferromagnetic band with the other  $N$  ferromagnetic electrons. The final result does not depend on the fact that we described the ferromagnetic electrons in terms of Wannier rather than Bloch functions in setting up the Slater determinants since a ferromagnetic spin-wave state can be written equivalently in terms of localized or running wave functions, at least in the case of a half-filled band. The actual expression for  $J_{ij}$  that we obtained is always positive; as Slater<sup>14</sup> has pointed out, we should actually consider also configuration interactions with ionic states when we compute the term in the Hamiltonian multiplying  $\delta_{\mathbf{k}', \mathbf{k}}$  which is the  $d$ - $d$  exchange interaction. This modification will change the numerical value of  $J_{ij}$  but will not alter the form of the Hamiltonian.

Adding the Zeeman interaction, we write the Hamiltonian of Eq. (10) as the sum of two terms:  $\mathfrak{H}_0$  describing the system in the absence of the exchange interaction between the conduction and ferromagnetic electrons and  $\mathfrak{H}_1$  representing the perturbation caused by the  $s$ - $d$  exchange interaction. Keeping only terms

involving the spin, we write

$$\mathfrak{H}_0 = -2 \sum_{j=1}^N \mathbf{S}_j \cdot \mathbf{S}_j J_{ij} - g\mu_B \mathbf{H} \cdot \sum_{j=1}^N \mathbf{S}_j - 2\mu_B \mathbf{H} \cdot \mathbf{s}; \quad (11)$$

$$\mathfrak{H}_1 = -2 \sum_{j=1}^N \mathbf{S}_j \cdot \mathbf{s} J_j(\mathbf{k}', \mathbf{k}). \quad (12)$$

To take account of the fact that the  $3d$  ion cores of the transition metals have a spin greater than one-half, we shall in general interpret  $\mathbf{S}_j$  as a spin operator with magnitude  $[S(S+1)]^{\frac{1}{2}}$ .

### III. SPIN WAVE APPROXIMATION

The Hamiltonian  $\mathfrak{H}_0$  of the unperturbed system can be diagonalized in the spin-wave approximation by the method of Holstein and Primakoff.<sup>13</sup> If  $\chi(n_j)$  is the spin function for the  $j$ th lattice site,  $n_j$  being the number of electrons on site  $j$  with spin parallel to  $\mathbf{H}$ , then we have the relations

$$\begin{aligned} S_j^z \chi(n_j) &= (n_j - S) \chi(n_j), \quad 0 \leq n_j \leq 2S, \\ S_j^+ \chi(n_j) &= (2S)^{\frac{1}{2}} (n_j + 1)^{\frac{1}{2}} \left(1 - \frac{n_j}{2S}\right)^{\frac{1}{2}} \chi(n_j + 1), \\ S_j^- \chi(n_j) &= (2S)^{\frac{1}{2}} (n_j)^{\frac{1}{2}} \left(1 - \frac{n_j - 1}{2S}\right)^{\frac{1}{2}} \chi(n_j - 1). \end{aligned} \quad (13)$$

We define creation and destruction operators for  $n_j$  according to

$$\begin{aligned} a_j^* \chi(n_j) &= (n_j + 1)^{\frac{1}{2}} \chi(n_j + 1), \\ a_j \chi(n_j) &= (n_j)^{\frac{1}{2}} \chi(n_j - 1), \end{aligned} \quad (14)$$

so that the usual commutation relation for bosons,  $[a_j, a_j^*] = 1$ , is satisfied. In terms of these operators, we can write the spin operators in the form

$$\begin{aligned} S_j^z &= a_j^* a_j - S, \\ S_j^+ &= (2S)^{\frac{1}{2}} a_j^* \left(1 - \frac{a_j^* a_j}{2S}\right)^{\frac{1}{2}} \cong (2S)^{\frac{1}{2}} a_j^*, \\ S_j^- &= (2S)^{\frac{1}{2}} \left(1 - \frac{a_j^* a_j}{2S}\right)^{\frac{1}{2}} a_j \cong (2S)^{\frac{1}{2}} a_j, \end{aligned} \quad (15)$$

where we expand to second order in the  $a$ 's. In terms of creation and annihilation operators, the Hamiltonian of the unperturbed system becomes, to second order in the  $a$ 's,

$$\begin{aligned} \mathfrak{H}_0 &= g\mu_B S N H - 2\mu_B H S^z - S^2 \sum_{i,j} J_{ij} \\ &\quad - 2S \sum_{i,j} J_{ij} (a_i^* a_j - a_j^* a_i) - g\mu_B H \sum_j a_j^* a_j. \end{aligned} \quad (16)$$

By making the transformation<sup>13</sup> to the spin-wave representation,

$$a_j = (N)^{-\frac{1}{2}} \sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{R}_j) a_{\mathbf{k}}, \quad (17)$$

<sup>14</sup> J. C. Slater, *Revs. Modern Phys.* **25**, 199 (1953).

the Hamiltonian  $\mathcal{H}_0$  can be brought into diagonal form

$$\mathcal{H}_0 = -NS^2zJ_{dd} + g\mu_B NSH - 2\mu_B HS^z + \sum_{\mathbf{k}} n_{\mathbf{k}} [2SJ_{dd}k^2a^2 - g\mu_B H]. \quad (18)$$

The energy of the unperturbed system is determined by the number of spin waves with various  $\mathbf{k}$  values and by the conduction electron wave vector and spin.

#### IV. RELAXATION TIME

The perturbation  $\mathcal{H}_1$  provides a mechanism by which the nonequilibrium number of spin waves of vector  $|\mathbf{k}|=1/\delta$  excited by the microwave power in a resonance experiment can return to its thermal equilibrium value.  $\mathcal{H}_1$  can be written in a more useful form if we utilize the translational property of the Bloch function,

$$\psi_{\mathbf{k}} = (N)^{-1/2} e^{i\mathbf{k} \cdot \mathbf{r}} u_{\mathbf{k}}(\mathbf{r})$$

(assumed normalized over an atomic volume  $\Omega$ ). Thus

$$\begin{aligned} J_j(\mathbf{k}', \mathbf{k}) &= \int d^3r_1 d^3r_2 \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_{\mathbf{k}'}^*(\mathbf{r}_1) a(\mathbf{r}_1 - \mathbf{R}_j) a^*(\mathbf{r}_2 - \mathbf{R}_j) \psi_{\mathbf{k}}(\mathbf{r}_2) \\ &= e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_j} \int d^3r_1 d^3r_2 \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_{\mathbf{k}'}^*(\mathbf{r}_1) a(\mathbf{r}_1) a^*(\mathbf{r}_2) \psi_{\mathbf{k}}(\mathbf{r}_2) \\ &\cong (N)^{-1} e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_j} \int d^3r_1 d^3r_2 \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \phi^*(\mathbf{r}_1) a(\mathbf{r}_1) a^*(\mathbf{r}_2) \phi(\mathbf{r}_2) \\ &= (N)^{-1} e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_j} J_{sd}, \end{aligned} \quad (19)$$

where  $J_{sd}$  is similar to an atomic  $s$ - $d$  exchange integral. This expression for  $J_{sd}$  takes no account of screening; we include the effect of screening by modifying the numerical value of  $J_{sd}$  which we treat as a parameter in that we do not calculate it from the preceding definition. With this substitution, the Hamiltonian<sup>15</sup> becomes

$$\mathcal{H}_1(\mathbf{k}', \mathbf{k}) = -2(N)^{-1} J_{sd} \mathbf{S} \cdot [\sum_j e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_j} \mathbf{S}_j]. \quad (20)$$

This Hamiltonian for  $\mathbf{k}' \neq \mathbf{k}$  does not commute with either  $\sum_{ij} S_i \cdot S_j$  or  $\sum_{j=1}^N S_j^z$ , so both the  $z$  component and the magnitude of the magnetization of the  $d$  electrons can be relaxed by this interaction. For  $\mathbf{k}' = \mathbf{k}$  only  $\sum_{j=1}^N S_j^z$  fails to commute with the Hamiltonian; the magnitude of the  $d$  electron spin in this case is not altered. It is easy to see that in general the total spin of the  $d$  electrons need not be conserved under the  $s$ - $d$  exchange interaction since this interaction need conserve only the total spin of the  $d$  and  $s$  electrons (we again stress that the  $s$  electron is relaxed rapidly by a separate mechanism). Thus the Hamiltonian  $\mathcal{H}_1$  can describe for instance a process in which a conduction electron is scattered with a spin flip from state  $\mathbf{k}$  to state  $\mathbf{k}' \neq \mathbf{k}$  and the ferromagnetic electron system changes from the state in which  $S = -S_{\max}$  (all  $d$  spins down) and spin wave vector  $\mathbf{k} = 0$  to the state  $S = -S_{\max} + 1$  (one  $d$  spin up) and spin wave vector  $\mathbf{k} = \mathbf{k} - \mathbf{k}'$ .

If we now transform to the spin wave representation by Eqs. (15) and (17), we obtain

$$\mathcal{H}_1(\mathbf{k}', \mathbf{k}) = - \left( \frac{2S}{N} \right)^{\frac{1}{2}} J_{sd} \sum_{\mathbf{k}} [s^+ a_{\mathbf{k}} \delta_{\mathbf{k}' - \mathbf{k}, \mathbf{k}} + s^- a_{\mathbf{k}}^* \delta_{\mathbf{k} - \mathbf{k}', \mathbf{k}}] - \frac{2J_{sd} S^z}{N} \sum_{\mathbf{k}', \mathbf{k}} a_{\mathbf{k}'}^* a_{\mathbf{k}} \delta_{\mathbf{k} - \mathbf{k}', \mathbf{k}' - \mathbf{k}}. \quad (21)$$

The first term in the Hamiltonian corresponds to the scattering of a conduction electron from the state with vector  $\mathbf{k}$  and spin down to the state with vector  $\mathbf{k}'$  and spin up and the destruction of a spin wave of vector  $\mathbf{k} = \mathbf{k}' - \mathbf{k}$ . The second term scatters a conduction electron from the state  $\mathbf{k}$  to the state  $\mathbf{k}'$  and creates a spin wave of vector  $\mathbf{k} = \mathbf{k} - \mathbf{k}'$ . The third term in  $\mathcal{H}_1$  scatters a conduction electron from state  $\mathbf{k}$  to state  $\mathbf{k}'$  without flipping its spin, destroys a spin wave of vector  $\mathbf{k}$  and creates a spin wave of vector  $\mathbf{k}' = \mathbf{k} - \mathbf{k}' + \mathbf{k}$ .

The single spin-wave processes provide the most rapid method of relaxation for the spin waves of momentum  $|\mathbf{k}| \sim 1/\delta$  which are excited by the microwave field. The double spin-wave process does, however, provide a mechanism for relaxing spin waves of vector zero.

Consider a spin-wave state of vector  $\mathbf{k}$  ( $|\mathbf{k}| \sim 1/\delta$ ); by standard time-dependent perturbation theory we find for the direct-spin processes:

$$\begin{aligned} \dot{n}_{\mathbf{k}} &= \frac{2\pi}{\hbar} \left( \frac{2S}{N} \right) J_{sd}^2 \sum_{\mathbf{k}', \mathbf{k}} \delta_{\mathbf{k} - \mathbf{k}', \mathbf{k}} \delta(E_{\mathbf{k}'}^- + \epsilon_{\mathbf{k}} - E_{\mathbf{k}}^+) \\ &\quad \times [(1 - f_{\mathbf{k}'}) f_{\mathbf{k}}^+ (n_{\mathbf{k}} + 1) - (1 - f_{\mathbf{k}}^+) f_{\mathbf{k}'}^- n_{\mathbf{k}}]. \end{aligned} \quad (22)$$

Here  $E_{\mathbf{k}}^{\pm} = (\hbar^2 k^2 / 2m^*) \mp \mu_B H$ , the energy of the conduction electron of wave vector  $\mathbf{k}$  and spin  $\pm$ , and  $\epsilon_{\mathbf{k}} = 2SJ_{dd}k^2a^2 - g\mu_B H$ , the energy of a spin wave with vector  $\mathbf{k}$ . The Fermi function  $f_{\mathbf{k}}^{\pm} = \{1 + \exp[(E_{\mathbf{k}}^{\pm} - E_F) / k_B T]\}^{-1}$  is used as the density of states for the conduction electron, since in the introduction we showed that the conduction electrons are essentially in thermal equilibrium. To simplify the expression in the square brackets in Eq. (22), we set  $n_{\mathbf{k}} = n_{\mathbf{k}}^0 + \Delta n_{\mathbf{k}}$ , where  $n_{\mathbf{k}}^0 = [\exp(\epsilon_{\mathbf{k}} / k_B T) - 1]^{-1}$ . Making use of the energy

<sup>15</sup> Professor K. Yosida has pointed out that a Hamiltonian of this form has been obtained by T. Kasuya, Progr. Theoret. Phys. (Japan) 16, 45 (1956).

$\delta$  function and expanding the Fermi function near the top of the Fermi distribution, we obtain for the bracketed expression

$$\epsilon_{\kappa}(df_k/dE_k)\Delta n_{\kappa}.$$

After summing out  $\mathbf{k}'$ , we can write the following expression for the relaxation time  $T_1$ :

$$\begin{aligned} \frac{1}{T_1} &= -\frac{\Delta \dot{n}_{\kappa}}{\Delta n_{\kappa}} = \frac{4\pi S J_{sd}^2 \epsilon_{\kappa}}{N \hbar} \sum_{\mathbf{k}} \left( -\frac{df_k}{dE_k} \right) \\ &\times \delta(E_{|\mathbf{k}-\kappa|} - E_k + \epsilon_{\kappa} + 2\mu_B H) = \frac{S J_{sd}^2 \Omega \epsilon_{\kappa}}{\pi \hbar} \int_0^{\infty} \left( -\frac{df_k}{dE_k} \right) \\ &\times k^2 dk \int_0^{\pi} \frac{\sin \theta d\theta}{\hbar^2 \kappa k} \frac{m^*}{\hbar^2 \kappa k} \delta(x - \cos \theta), \quad (23) \end{aligned}$$

where

$$x = \left( \frac{\hbar^2 \kappa^2}{2m^*} + \epsilon_{\kappa} + 2\mu_B H \right) / (\hbar^2 \kappa k / m^*).$$

Paying attention to the limits of integration, we get

$$\frac{1}{T_1} = \frac{S J_{sd}^2 m^{*2} \Omega \epsilon_{\kappa}}{\pi \hbar^5 \kappa} \left[ 1 + \exp\left(\frac{E_{\min} - E_F}{k_B T}\right) \right]^{-1}, \quad (24)$$

with

$$E_{\min} = (E_{\kappa} + \epsilon_{\kappa} + 2\mu_B H)^2 / 4E_{\kappa}.$$

For  $|\kappa| = 1/\delta$ ,  $E_{\min} \ll E_F$  so that

$$\frac{1}{T_1} = \frac{S J_{sd}^2 m^{*2} \Omega \epsilon_{\kappa}}{\pi \hbar^5 \kappa}. \quad (25)$$

However as  $\kappa \rightarrow 0$ ,  $E_{\min} \rightarrow \infty$  and thereby counteracts the explosive tendency of  $\epsilon_{\kappa}/\kappa$ ; since  $T_1 \rightarrow \infty$  as  $\kappa \rightarrow 0$ , we must consider the term in Eq. (21) quadratic in the  $a$ 's if we wish to calculate the relaxation time for  $\kappa = 0$  spin waves (we find that it is several orders of magnitude longer than the time for  $|\kappa| \sim 1/\delta$ ).

## V. COMPARISON WITH EXPERIMENT

For nickel,<sup>6</sup> at room temperature and at 24 400 Mc/sec,  $\mu_2 = 17$  and  $\rho = 7.74 \times 10^{-6}$  ohm cm so that  $\kappa = 1/\delta = (4\pi\omega\mu_2/\rho c^2)^{1/2} = 2 \times 10^5$  cm<sup>-1</sup>. We note that the experimental line width in nickel is temperature-independent<sup>1</sup> from  $T = 4^\circ\text{K}$  to  $T = 293^\circ\text{K}$ , but  $T_1$  as calculated from Eq. (25) has a slight temperature dependence owing to the temperature dependence of  $\kappa$

through  $\rho$ . However, the experimental value for the resistivity at  $4^\circ\text{K}$  quoted by Reich<sup>1</sup> is only about  $\frac{1}{8}$  of the room temperature value, so  $T_1$  is increased only by a factor of about 3 over the room temperature value. Since the spin-wave approximation is valid only at low temperatures, we can expect Eq. (25) to hold for  $T \ll T_c$  ( $631^\circ\text{K}$  in nickel) so that  $1/T_1$  will not show the large increase that is observed<sup>5</sup> in the line width at high temperatures. The calculated relaxation time is also field-dependent both through the explicit appearance of  $H$  in  $\epsilon_{\kappa} = 2S J_{sd} \kappa^2 a^2 - g\mu_B H$  and indirectly through the dependence of  $\kappa$  upon  $\omega$ . If we compare the relaxation times as calculated at, say, 3 cm and 0.6 cm, we find  $(T_1)_{0.6} \simeq \frac{1}{4} (T_1)_3$ . Bloembergen's measurements<sup>5</sup> on a polycrystalline nickel sample yield  $(\Delta H)_3 / (\Delta H)_{1.2} = \frac{2}{3}$  and Hoskins' results<sup>16</sup> for a nickel single crystal give  $(\Delta H)_{1.2} / (\Delta H)_{0.6} = \frac{2}{3}$ . The measurements of Hoskins and Wiener<sup>17</sup> on Fe-Ni alloys show  $(\Delta H)_3 / (\Delta H)_{0.6} = 0.36 - 0.48$ .

At 24 300 Mc/sec, with  $\kappa = 2 \times 10^5$ ,  $H = 5200$  oe,  $a = 3.52 \times 10^{-8}$  cm,  $J_{sd} = 230 k_B = 2 \times 10^{-2}$  ev, we find  $\epsilon_{\kappa} = 10^{-16}$  erg. If we take the screened exchange integral  $J_{sd} \simeq 10^{-2}$  ev, we obtain  $1/T_1 \simeq 2 \times 10^8$  sec<sup>-1</sup>; this value of  $T_1$  is an order of magnitude shorter than the shortest time calculated using magnetic interactions,<sup>3</sup> but is an order of magnitude longer than  $1/T_2 = 3.8 \times 10^9$  sec<sup>-1</sup> observed by Reich<sup>1</sup> in nickel. We cannot choose a larger value of  $J_{sd}$  to obtain agreement with experiment as this would result in a large line shift.<sup>7</sup> The line width in nickel is unusually large and the mechanism suggested by Stevens<sup>18</sup> may play an important role in explaining the observed width. In some materials, such as alloys, in which the lines are narrower, the exchange mechanism may play a dominant role in ferromagnetic relaxation.

## VI. ACKNOWLEDGMENTS

It is a pleasure for the author to express his thanks to Professor C. Kittel for suggesting this problem and for several helpful discussions. He would also like to thank Professor K. Yosida for reading the manuscript and discussing the relation of the magnitude of  $J_{sd}$  to the Zener theory<sup>8</sup> of ferromagnetism. This research was done during the tenure of a National Science Foundation Predoctoral Fellowship.

<sup>16</sup> R. Hoskins, thesis, University of California, 1954 (unpublished).

<sup>17</sup> R. Hoskins and G. Wiener, Phys. Rev. **96**, 1153 (1954).

<sup>18</sup> K. W. H. Stevens, Proc. Phys. Soc. (London) **A65**, 149 (1952).