the focusing collisions first suggested by Silsbee,[12] and strong interstitial clustering as suggested by Ward and Kauffman.[3]

The fact that the annealing spectrum is qualitatively different in gold than in copper and silver is of great importance since any scheme of imperfections used for interpretation must enable one to understand the fact that the noble metals can behave so differently. Leibfried suggested that the interstitial configuration may be different in gold from its configuration in copper and silver. This possibility will be examined in a later paper.

## VI. CONCLUSION

The low-temperature annealing spectrum of gold consists of at least 9 peaks ranging in activation energy from 0.02 to 0.15 eV for a frequency factor of $5.3\times 10^{11}$ $\mathrm{sec}^{-1}$. The total percentage of the original damage annealed out up to 51°K is 22.5%. The annealing behavior of cadmium is in qualitative agreement with earlier measurements.

The following conclusions can be made: (1) The amount of close pair recovery in gold is substantially smaller than in copper; (2) the close pair recovery spectrum in gold is more complex than that of copper. In the three experiments done on gold it is not possible to locate an annealing peak which is dose-dependent. It would be valuable to do careful isochronal annealing experiments aimed at determining whether any of the annealing peaks observed shift in temperature when the amount of damage is altered.

## ACKNOWLEDGMENTS

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[12] R. H. Silsbee, J. Appl. Phys. 28, 1246 (1957).

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# Longitudinal Nuclear Magnetic Relaxation in Ferromagnetic Iron, Cobalt, and Nickel*

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The longitudinal nuclear relaxation time $T_1$ has been measured in ferromagnetic iron, cobalt, and nickel. A model is presented to account for the measured values. In the Bloch walls, the relaxation is due mainly to domain wall fluctuations. In the domains, the relaxation is due to interaction of the conduction electrons with nuclei via spin waves. The expression for $T_1$ due to this process is $1/T_1=(kT/h)(\omega/\omega_d)^2a^2\Sigma/(32\pi^3S^2)$, where $\omega$ is the nuclear resonant frequency, $a$ is the lattice constant, $\omega_d$ is the parameter describing the spin wave spectrum $E(k)=\hbar\omega_d a^2k^2$, $S$ is the average spin per atom, and $\Sigma$ is the area of the Fermi surface per cubic unit cell. If the experimental value of $T_1$ is used in this formula to determine $\Sigma$, then in cobalt, $\Sigma$ will agree closely with the area of a spherical surface containing about one electron per atom. In iron and nickel, $\Sigma$ will be about three times larger.

## INTRODUCTION

NUCLEAR magnetic resonance (NMR) in a ferromagnetic metal was first observed by Gossard and Portis[1] in metallic cobalt. These authors investigated the resonance and described its salient features: the resonant frequency and its temperature dependence, the linewidth, the spin-spin and spin-lattice relaxation processes. However, the experimental apparatus employed in these experiments was designed for the interpretation of equilibrium spectra, and the purpose of the work reported in this paper is to study relaxation processes which can be investigated more directly by use of NMR pulse techniques.[2]

The experiments described in the present paper indicate that the longitudinal relaxation process can be understood in terms of two mechanisms: a nonexponential relaxation observed at low rf power levels which is believed to be due to thermal fluctuations of the domain walls, and an exponential relaxation observed at high rf power levels which is believed to be due to an interaction of the conduction electrons with the nuclei via spin waves. This model is used to determine experimentally the area of the Fermi surface. In cobalt the area determined this way is consistent with a model in

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* Supported by the Office of Naval Research.

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[1] C. Gossard and A. M. Portis, Phys. Rev. Letters 3, 164 (1959); J. Appl. Phys. 31, 205S (1960).

[2] M. Weger, A. M. Portis, and E. L. Hahn, J. Appl. Phys. 32, 124S (1961).

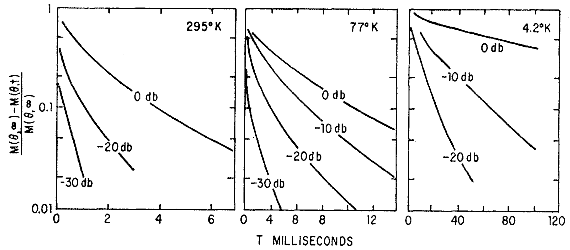

FIG. 1. Longitudinal relaxation of $Fe^{57}$ at various temperatures and rf power levels. The relative power level is given in decibels. A power of $-30$ dB corresponds roughly to saturating pulses of such length $t_w$ that $\pi/\gamma_n H_{rf} t_w \approx 4000$. The relaxation process is not exponential, and $T_1$ increases with the relaxation time $t$ and with the rf power level.

which the itinerant nature of the $d$ electrons is ignored, and the $s$ electrons behave almost like a free Fermi gas situated in the exchange field due to the $d$ electrons. In iron and nickel, the model can account for the observed relaxation. An instantaneous relaxation rate $1/T_1(t,\theta)$ is assumed.

## EXPERIMENTAL

The present experiments were carried out on finely divided powders (approximately 1–30 $\mu$) of iron, cobalt, and nickel. Some of the iron and nickel samples were isotopically enriched. The experiments were carried out over a wide temperature range (1.5–800°K). The longitudinal relaxation time was measured by the spin-echo technique.[3] In this method, $m$, the component of the nuclear magnetization in the direction of the internal field, is changed from its equilibrium value by applying one or more intense rf pulses, and measured at a later time $t$ by applying a pair of closely spaced rf pulses and measuring the amplitude of the resulting spin echo. The reason for using the spin echo rather than the free induction decay tail is that it allows the receiver to recover from the overload it suffers during the intense rf pulse. As the time $t$ is increased, the amplitude of the spin echo increases as a result of the longitudinal relaxation. An instantaneous relaxation rate $1/T_1(t,\theta)$ is defined as

$$1/T_1(t,\theta) = -[\partial m(t,\theta)/\partial t]/[m(t,\theta)-m(\infty,\theta)],$$

where $\theta = \int H_1 dt$ is the integrated intensity of the saturating pulses and $m(t,\theta)$ is the amplitude of the spin echo. Typical relaxation curves are shown in Fig. 1 and measured values of $T_1$ are given in Table I.

The experiments indicate the following features: (a) $T_1(t,\theta)$ increases with $t$ and $\theta$, varying over a wide range. (b) At large values of $t$ and $\theta$, $T_1(t,\theta)$ approaches an upper limit $T_{1\infty}$. (c) $T_{1\infty}$ is inversely proportional to the temperature over the whole investigated temperature range as shown in Fig. 2.

## INTERPRETATION

Our first task is to understand why the relaxation process is nonexponential and is dependent upon the rf power level. When this phenomenon was first observed in cobalt it was believed to be due to spin diffusion out of the domain walls[1]; but subsequently the same phenomenon was observed in iron and nickel in which spin diffusion is negligible because of the small nuclear moment and low abundance of the active isotope. Simple theoretical considerations[1] suggest that the signals observed at low rf power levels are due to nuclei situated in the domain walls while those observed at high power levels are due to nuclei situated in the domains. Therefore, the nonexponential relaxation is apparently associated with the domain walls, while the exponential relaxation observed at high rf power levels is an intrinsic property of the domains. To confirm this assumption, the following experiment was performed. Between the saturating and sampling rf pulses, a magnetic field of a few hundred oersteds was applied shifting the domain walls by a distance which was large compared with their width. Thus, if the nuclei saturated by the first rf pulse were initially situated in the domain walls, they are displaced by the magnetic field into the domains, relaxing there. When this experiment was performed at low rf power levels, $T_1$ was found to increase, indicating that the relaxation rate in the domains is slower than in the walls. Detailed description of the experiments involving pulsed magnetic fields will be given in a future paper.[2]

## THEORY

We have to account for a fast, nonexponential relaxation in the domain walls and for a slower exponential relaxation in the domains proper.

A possible relaxation mechanism in the domain walls

TABLE I. The longitudinal relaxation time $T_1$ of $Fe^{57}$, $Co^{59}$, and $Ni^{61}$ in the respective metals. The relaxation is not exponential, resulting in a distribution of relaxation times. In cobalt at 700 and 800°K, and in nickel at 1.5°K, the experimental accuracy was not good enough to determine upper and lower limits, and only representative values are given. ($T_1$ is given in msec.)

| Temperature (°K) | 1.5 | 1.85 | 4.2 | 77 | 295 | 700 | 800 |
|---|---|---|---|---|---|---|---|
| $Fe^{57}$ | | ~24–130 | 10 –500 | 0.7–11.2 | 0.9 –6.5 | | |
| $Co^{59}$ | | ~30 | 0.2–17 | 0.2– 1.2 | 0.1 –0.5 | 0.06–0.12 | 0.05–0.1 |
| $Ni^{61}$ | 50–70 | | 15 –25 | 1.5– 2.5 | ~0.35 | | |

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[3] E. L. Hahn, Phys. Rev. **80**, 580 (1950).

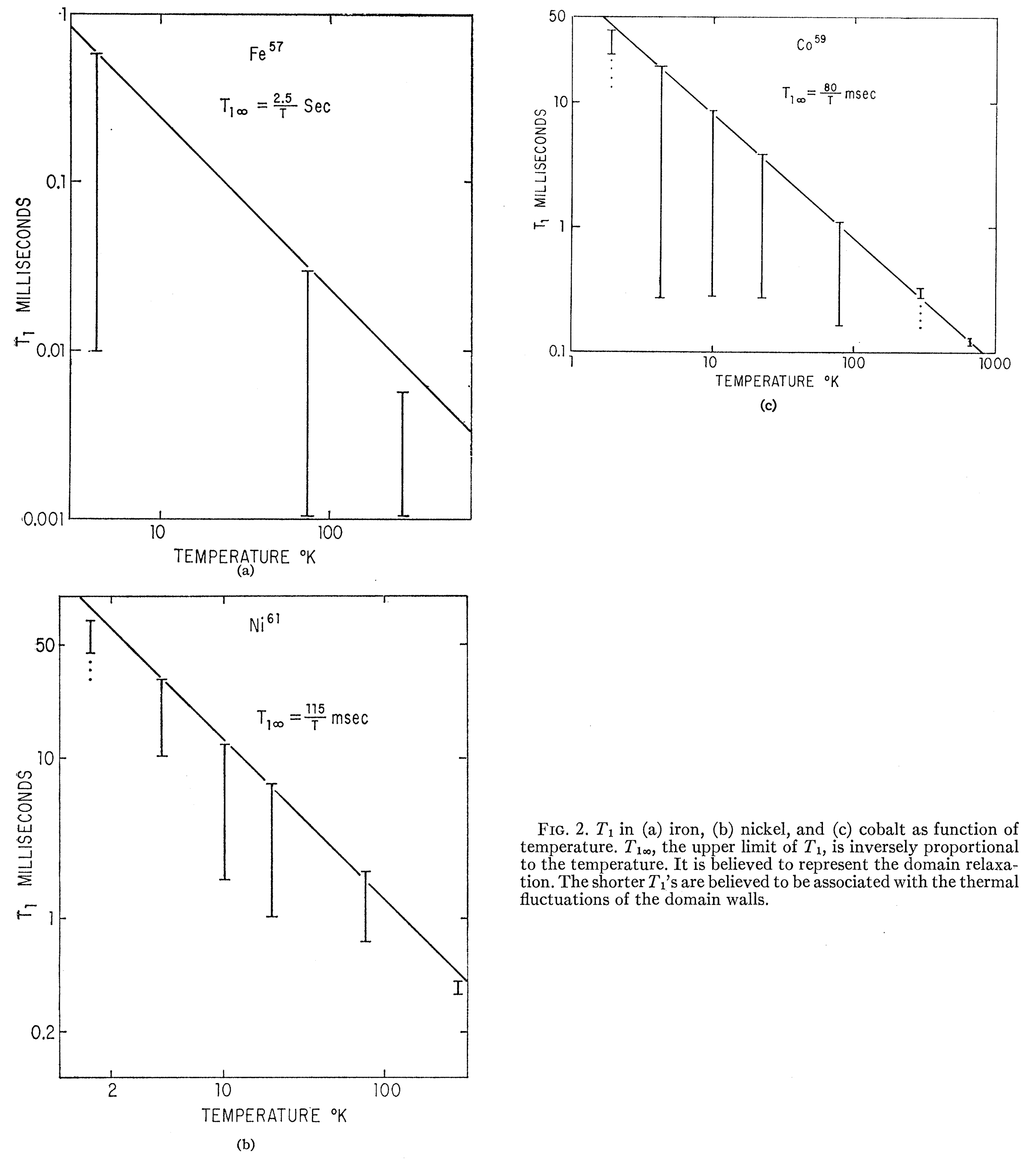

FIG. 2. $T_1$ in (a) iron, (b) nickel, and (c) cobalt as function of temperature. $T_{1\infty}$, the upper limit of $T_1$, is inversely proportional to the temperature. It is believed to represent the domain relaxation. The shorter $T_1$'s are believed to be associated with the thermal fluctuations of the domain walls.

is provided by the thermal fluctuations of the walls.[4] The order of magnitude of the relaxation rate due to these fluctuations can be estimated as follows. Consider a small sphere of diameter $d$ consisting of two equal oppositely directed domains, and a domain wall at the center. We imagine that the only low-lying wall excitation is a uniform displacement. When the domain wall shifts by a distance $x$, a net magnetization $M=3M_s x/d$ is created where $M_s$ is the saturation magnetization. The demagnetizing energy resulting from this magnetization is $E=(N/2)M^2(\pi d^3/6)$, where $N$ is a demagnetizing factor of order $4\pi/3$. By the theorem of equipartition of energy the average energy associated with this degree of freedom is given by $\langle E\rangle=\frac{1}{2}kT$. Thus, $\langle x^2\rangle=kT/2\pi^2M_s^2d$. The component of the internal field

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[4] M. Weger, thesis, University of California, June, 1961 (unpublished); E. Simanek and Z. Sroubek, Czech. J. of Physics **B11**, 764 (1961); J. M. Winter, Phys. Rev. **124**, 452 (1961).

perpendicular to the equilibrium direction is given by[1] $H_x = H_{\mathrm{int}}\, x/\delta$, where $H_{\mathrm{int}}$ is the internal field and $\delta$ is the domain wall thickness. Thus, $H_x^2 = H_{\mathrm{int}}^2 kT/2M_s^2 d\delta^2$. Assuming that the fluctuations are associated with a Lorentzian correlation spectrum $P(\omega) = 2\tau_c/\pi(1+\omega^2\tau_c^2)$, where $\tau_c$ is the correlation time, the relaxation rate caused by these fluctuations is given by[5]

$$1/T_1 = kT\tau_c/\pi\delta^2 M_s^2(1/\omega^2+\tau_c^2). \qquad (1)$$

For cobalt at room temperature, we may assume $\delta \approx 10^{-6}$ cm, $d \approx 10^{-4}$ cm, $M_s = 1460$ G, $\omega = 1.36\times10^9$ sec$^{-1}$, $\tau_c \approx 2\times10^{-11}$ sec. Substituting these values in (1), we get $T_1 \approx 300\,\mu$sec. The shortest $T_1$ observed experimentally under these conditions is approximately 100 $\mu$sec. Similar agreement with experiment is obtained for iron and nickel. Winter[4] investigated this model very carefully and has shown that in addition to parallel translations of the domain wall, one must also consider modes in which the domain wall vibrates somewhat like a membrane, and also the relaxation caused by spin waves distorted in the domain walls. From the expression Winter[4] gives for the energy of nonuniform domain wall excitations (Eq. 12, reference 4), we see that when $k_{\min} \approx \pi/d \gg (K'/Ja^2)^{1/2}$, the energy of these modes is large compared with that of the uniform mode. $K'$ is a parameter characterizing the wall stiffness, and is of order 1–2 Mc/sec in iron and cobalt. $J$ is of order $4\times10^{12}$ Mc/sec, thus the inequality is satisfied for $d \ll 10^{-1}$ cm. Since the size of the particles did not exceed about $10^{-2}$ cm, the nonuniform domain modes require high excitation energies and the neglect of their contribution to the relaxation is justified for an order of magnitude estimate. Since $\tau_c$ is probably temperature dependent, increasing at lower temperatures, the relaxation rate due to this effect cannot be expected to be proportional to the temperature. As a matter of fact, if the damping of the domain wall motion is due to eddy currents induced by its motion,[6] and the electrical resistivity is proportional to the temperature, then the relaxation rate will be temperature independent as long as $\omega\tau_c \ll 1$. Thus, the experimentally observed weak temperature dependence of the domain wall relaxation times can be accounted for.

The relaxation rate in the domains themselves is exponential and proportional to the temperature over the whole investigated range to a high accuracy (Fig. 2). This rules out thermal fluctuations of the domain walls, or direct interaction with phonons, as major relaxation mechanisms, and suggests that conduction electrons are responsible for the relaxation. Several mechanisms for nuclear relaxation by conduction electrons have been suggested by Korringa[7] and Mitchel.[7] Korringa considered the relaxation due to the hyperfine interaction between the conduction electrons and the nuclei. His expression for the relaxation time is

$$1/T_1 = 2(2\pi/\hbar)[(8\pi/3)\gamma_e\gamma_n\hbar^2]^2 \\ \times|\psi_\uparrow(0)|^2|\psi_\downarrow(0)|^2 kTg_\uparrow(E_F)g_\downarrow(E_F).$$

The value of the wave function at the nucleus, and the density of states, are not known precisely in Fe, Co, and Ni; but if we assume that these quantities do not change much as we go along the periodic table from vanadium to nickel, we can estimate the relaxation rate due to the Korringa process in Fe, Co, and Ni. $T_1$ has been measured in vanadium by Butterworth[8] and found to be $790/T$ msec. $V^{51}$ has the same spin as $Co^{59}$, $I = 7/2$, and nearly the same nuclear moment, 5.1 nm for $V^{51}$ vs 4.6 nm for $Co^{59}$. It is, therefore, difficult to see why $T_1$ in cobalt should be an order of magnitude shorter than in vanadium. Similar considerations apply to iron and nickel. Mitchel considered the relaxation caused by the interaction of the nuclear quadrupole moment with fluctuating electric field gradients at the site of the nucleus. It is difficult to see, however, how such a mechanism could produce a much shorter $T_1$ in cobalt ($Q = 0.5$) than in vanadium ($Q = 0.3$). Furthermore, this mechanism cannot contribute to the relaxation in iron at all, since $I = 1/2$ for $Fe^{57}$. Consequently, the rapid relaxation rate in Fe, Co, and Ni is apparently associated with their ferromagnetic nature. In the models of Korringa and Mitchel the electrons are assumed to constitute a free Fermi gas. Admittedly, one cannot expect such a model to work for a ferromagnet.

The following mechanism is presented here in an attempt to account for the fast nuclear relaxation. Consider a model in which the electronic excitations in the ferromagnet can be divided into two groups: spin waves, and the excitations that can be attributed to independent conduction electrons, situated in an effective exchange field. Assume that this exchange field can be effectively described by a Hamiltonian

$$\mathcal{H} = (\hbar\omega_{sd}/S)(\mathbf{s}\cdot\mathbf{S}_j)f(\mathbf{r}_s-\mathbf{r}_j).$$

Here, $\mathbf{s}$ is the spin operator of the conduction electron, $\mathbf{S}_j$ is the spin operator of the "magnetic" electrons of the $j$th atom; (this operator can be formally defined as the Fourier transform of the creation operators of spin waves),[9] and $f(\mathbf{r}_s-\mathbf{r}_j)$ is a function of the position of a conduction electron $\mathbf{r}_s$, the function being normalized to unity. Then, our system can be described by an effective Hamiltonian,

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}',$$

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[5] N. Bloembergen, E. M. Purcell, and R. V. Pound, Phys. Rev. **73**, 679 (1948).

[6] C. Kittel and J. K. Galt, in *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 3, p. 437.

[7] J. Korringa, Physica **16**, 601 (1950); A. H. Mitchel, J. Chem. Phys. **26**, 1714 (1957).

[8] J. Butterworth, Phys. Rev. Letters **5**, 305 (1960).

[9] H. Suhl, J. de Phys. **20**, 333 (1959).

where

$$\mathcal{H}_0=\textstyle\sum_{\mathbf{k}} E(\mathbf{k})+\sum_j AS_{z_j}I_{z_j}+\sum_{\mathbf{q}} E'(\mathbf{q})$$
$$+\textstyle\sum_{j,\mathbf{q}}(\hbar\omega_{sd}s_{zq}/S)S_{z_j}f(\mathbf{r}_s-\mathbf{r}_j),$$
$$\mathcal{H}'=\textstyle\sum_j A(S_j^+I_j^-+S_j^-I_j^+)$$
$$+\textstyle\sum_{j,\mathbf{q}}(\hbar\omega_{sd}/S)(s_q^+S_j^-+s_q^-S_j^+)f(\mathbf{r}_s-\mathbf{r}_j).$$

$E(\mathbf{k})$ is the energy of a spin wave with wave vector $\mathbf{k}$. The term $A(\mathbf{S}_j\cdot\mathbf{I}_j)$ describes the effective interaction between the nuclei and spin waves. The component $AS_zI_z$ is determined experimentally from the nuclear resonant frequency $\omega$, $\hbar\omega=AS$. Thus, the interaction via the polarization of the conduction electrons[10] is included in it. $E'(\mathbf{q})$ is the energy of a conduction electron with wave vector $\mathbf{q}$. The summation over $j$ extends over the nuclei and the electrons bound to them, and the summation over $\mathbf{q}$ extends over the occupied conduction electron states. By second-order perturbation theory, the matrix element for a transition involving a spin flip is

$$\langle I_{z_j}s_z\mathbf{q}|\mathcal{H}'|I_{z_j}'s_z'\mathbf{q}'\rangle=\sum_{\mathbf{k}}\frac{\langle 0I_{z_j}|A(I_j^+S_j^-+I_j^-S_j^+)|\mathbf{k}I_{z_j}'\rangle\langle\mathbf{k}s_z\mathbf{q}|(\hbar\omega_{sd}/S)(S_j^+s^-+S_j^-s^+)f(\mathbf{r}_s-\mathbf{r}_j)|0s_z'\mathbf{q}'\rangle}{E(\mathbf{k})+AS(I_{z_j}-I_{z_j}')}. \tag{3}$$

Using the relationship[9] $|\mathbf{k}\rangle=(\sqrt{2}S)^{-1}S^-(\mathbf{k})|0\rangle$ where $S^-(k)=N^{-1/2}\sum_j S_j^-\exp ik\mathbf{r}_j$, we have:

$$\langle I_{z_j}s_z\mathbf{q}|\mathcal{H}'|I_{z_j}'s_z'\mathbf{q}'\rangle=\frac{A\hbar\omega_{sd}}{2N}\sum_{\mathbf{k}}\frac{\langle I_{z_j}s_z|I_j^+s^-+I_j^-s^+|I_{z_j}'s_z'\rangle\langle\mathbf{q}|f(\mathbf{r}_s-\mathbf{r}_j)e^{ik(\mathbf{r}_s-\mathbf{r}_j)}|\mathbf{q}'\rangle}{E(\mathbf{k})+AS(I_{z_j}-I_{z_j}')}. \tag{4}$$

Assuming that the wave functions of the conduction electron are Bloch functions,

$$|\mathbf{q}\rangle=a^{3/2-}U_q(\mathbf{r}-\mathbf{r}_j)e^{i\mathbf{q}(\mathbf{r}-\mathbf{r}_j)},$$

where $a$ is the lattice constant, we have

$$\langle\mathbf{q}|f(\mathbf{r}_s-\mathbf{r}_j)e^{i\mathbf{k}\cdot(\mathbf{r}_s-\mathbf{r}_j)}|\mathbf{q}'\rangle$$
$$=a^{-3}\delta_3(\mathbf{q}+\mathbf{k}-\mathbf{q}')+\sum_{\mathbf{K}\neq 0}\delta_3(\mathbf{q}+\mathbf{k}-\mathbf{q}'-\mathbf{K})$$
$$\times\int d^3r\, U_q^\dagger(\mathbf{r})U_{q'}(\mathbf{r})f(\mathbf{r})e^{i\mathbf{K}\cdot\mathbf{r}}. \tag{5}$$

$\mathbf{K}$ is a vector of the reciprocal lattice. The term involving it represents the contribution due to umklapp processes. In a more sophisticated approximation one has to distinguish between the wave functions of conduction electrons with spins up and down. Substituting (5) in (4), we have

$$\langle I_{z_j}s_z\mathbf{q}|\mathcal{H}'|I_{z_j}'s_z'\mathbf{q}'\rangle$$
$$=\frac{A\hbar\omega_{sd}}{2}\langle I_{z_j}s_z|I_j^+s^-+I_j^-s^+|I_z's_z'\rangle$$
$$\times\left[\frac{1}{E(\mathbf{q}-\mathbf{q}')+AS(I_z-I_z')}+\sum_{\mathbf{K}\neq 0}\frac{f(\mathbf{q},\mathbf{q},'\mathbf{K})}{E(\mathbf{q}-\mathbf{q}'-\mathbf{K})+AS(I_z-I_z')}\right]. \tag{6}$$

If the Fermi surface does not approach the zone boundaries, the term representing umklapp processes can probably be neglected.

Following Korringa,[7] the relaxation time resulting from this matrix element will be given by

$$1/T_1=2\frac{2\pi}{\hbar}\int\int\int\int\left(\frac{A\hbar\omega_{sd}}{2}\right)^2$$
$$\times\frac{(a/2\pi)^6d\Sigma d\Sigma'dEdE'}{[E(\mathbf{q}-\mathbf{q}')-AS]^2|\nabla_qE|\,|\nabla_{q'}E'|}$$
$$\times f(E)[1-f(E')]\delta(E-E'), \tag{7}$$

where $d\Sigma$ is an element of area in $q$ space, perpendicular to $\nabla_qE$, and $f(E)=\{\exp[(E-E_F)/kT]+1\}^{-1}$. $f(E)[1-f(E)]$ can be expanded in powers to $kT/E_F$.[11] The lowest order term is the $\delta$ function $\delta[(E-E_F)/kT]$. Let us neglect the higher order terms, which is equivalent to assuming that $T_1T=\text{const}$. Integration over $E$, $E'$ yields

$$1/T_1=2\frac{2\pi kT}{\hbar}\int\int\left(\frac{A\hbar\omega_{sd}}{2}\right)^2$$
$$\times\frac{(a/2\pi)^6d\Sigma d\Sigma'}{[E(\mathbf{q}-\mathbf{q}')-AS]^2|\nabla_qE|\,|\nabla_{q'}E|}, \tag{8}$$

where now the integration must be carried out over the Fermi surface.

Now let

$$E(\mathbf{q}-\mathbf{q}')=\hbar[\omega_{\text{loc}}+a^2\omega_d(\mathbf{q}-\mathbf{q}')^2]. \tag{9}$$

This relationship cannot be expected to hold for values of $\mathbf{q}-\mathbf{q}'$ near the zone boundary, but $E(\mathbf{q}-\mathbf{q}')$ there is large and the contribution to the integral is small. Furthermore, this relationship does not take into account the dependence of $E(\mathbf{k})$ upon the direction of $\mathbf{k}$; hence, if both the Fermi surface and the spin wave

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[10] W. Marshall, Phys. Rev. **110**,280 (1958).

[11] C. Kittel, *Introduction to Solid-State Physics* (John Wiley & Sons, Inc., New York, 1956), p. 256.

TABLE II. Area of Fermi surface calculated by use of formula 12. $\bar{\Sigma}=(4\pi/a)^2(9\pi/4)^{1/3}$ for an fcc lattice, and half this value for a bcc lattice, is the area of a spherical Fermi surface containing one electron per atom. In cobalt, $\omega_d$ and $S$ can be determined from the temperature-dependence of the NMR frequency[a] or from ferromagnetic resonance in thin films.[b] For iron and nickel, unfortunately, no film data were available.

| Quantity | Unit | $Fe^{57}$ | | $Co^{59}$ | | $Ni^{61}$ |
|---|---|---|---|---|---|---|
| $\omega$ (measured at 4.2°K) | rad/sec | $2.92\times10^8$ | | $1.36\times10^9$ | | $1.76\times10^8$ |
| $J_d$ ($d$-electron exchange integral) | $k$ (Boltzmann constant) | $205^c$ | | | | $380^c$ |
| $S$ | | $1.1^c$ | $0.85^a$ | | $0.772^b$ | $0.3^c$ |
| $\omega_d$ $(2J_dS/\hbar)$ | rad/sec | $5.3\times10^{13}$ | $3.94\times10^{13}$ | | $3.1\times10^{13b}$ | $3\times10^{13}$ |
| $T_1T$ (experimental) | sec-°K | 2.5 | | 0.08 | | 0.115 |
| $a$ | Å | 2.861 | | 3.554 | | 3.517 |
| $\Sigma$ | $cm^{-2}$ | $7.6\times10^{17}$ | $3\times10^{17}$ | | $1.5\times10^{17}$ | $8.9\times10^{17}$ |
| | $\bar{\Sigma}$ | 3.2 | 1.2 | | 0.6 | 3.5 |

[a] See reference 13. [b] See reference 15. [c] See reference 11.

spectrum are highly anisotropic, this approximation may not be very good. We may further simplify Eq. (8) by noting that in our case, $\hbar\omega_{\mathrm{loc}}\gg AS$, since $AS$ is at most $h$ 220 Mc/sec (in cobalt), while $\hbar\omega_{\mathrm{loc}}$ is of the order $\beta H_{\mathrm{anis}}\approx\beta$ 1000 Oe$\approx h$ 3000 Mc/sec ($H_{\mathrm{anis}}$ is the anisotropy field). If $AS$ could equal $\hbar\omega_{\mathrm{loc}}$, we would have to consider a first-order process consisting of radiation of spin waves by nuclei, in addition to the present second-order process. However, since $AS\ll\hbar\omega_{\mathrm{loc}}$, we can neglect $AS$ in the denominator. We may further note that $\omega_{\mathrm{loc}}\ll\omega_d$, while $aq_F$ is of the order unity. Hence, most of the contribution to the integral comes from regions in which the angle $\theta$ between $\mathbf{q}$ and $\mathbf{q}'$ is small. Therefore, let us neglect the $\theta$-dependence of $q'$ and of $\nabla_{q'}E$, and carry out the integration over $d\Sigma'$.

$$\int\frac{d\Sigma'}{[\omega_{\mathrm{loc}}+a^2\omega_d(\mathbf{q}-\mathbf{q}')^2]^2}=\int\frac{q'^2 2\pi\sin\theta d\theta}{[\omega_{\mathrm{loc}}+a^2\omega_d(q^2+q'^2-2qq'\cos\theta)]^2}=\frac{\pi q'}{\omega_d a^2}\frac{q'}{q}\times\left[\frac{1}{\omega_{\mathrm{loc}}+a^2\omega_d(q-q')^2}-\frac{1}{\omega_{\mathrm{loc}}+a^2\omega_d(q+q')^2}\right].$$

Substituting in (8), we have

$$1/T_1=\frac{4\pi kT}{\hbar}\left(\frac{a}{2\pi}\right)^6\int\frac{q'}{q}\left(\frac{A\hbar\omega_{sd}}{2}\right)\frac{\pi}{\hbar\omega_d a^2}\frac{d\Sigma}{|\nabla_qE|\,|\nabla_{q'}E|\hbar}\times\left[\frac{1}{\omega_{\mathrm{loc}}+a^2\omega_d(q-q')^2}-\frac{1}{\omega_{\mathrm{loc}}+a^2\omega_d(q+q')^2}\right].\quad(9)$$

We may note further that $\omega_{\mathrm{loc}}\ll a^2\omega_d(q_\uparrow-q_\downarrow)^2$. This can be seen as follows. The difference in Zeeman energy between electrons pointing up and down is given by $\hbar\omega_{sd}$. Since over-all energy must be conserved in the relaxation process, this energy change must be accompanied by an equal and opposite change in the kinetic energy of the conduction electron. In the free electron model, $dE=(2E_F/m^*)^{1/2}\Delta q$. If we assume, following Marshall,[10] that $E_F=10^{-11}$ erg, $\omega_{sd}=4\times10^{14}$ sec$^{-1}$, and $m^*=m$, then $\Delta q=3\times10^6$ cm$^{-1}$. On the other hand, the inverse range of the Suhl interaction is

$$(\omega_d a^2/\omega_{\mathrm{loc}})^{-1/2}\approx0.7\times10^6\ \mathrm{cm}^{-1},$$

for

$$\omega_d/\omega_{\mathrm{loc}}\approx2000,\quad a=3\times10^{-8}\ \mathrm{cm}.$$

We can make the further approximation $(q_\uparrow+q_\downarrow)^2\gg(q_\uparrow-q_\downarrow)^2$. Thus, our expression for $T_1$ simplifies to

$$1/T_1=\frac{4\pi kT}{\hbar}\left(\frac{a}{2\pi}\right)^6\int\frac{q'}{q}\frac{(A\hbar\omega_{sd})^2}{2}\frac{\pi}{\hbar^2\omega_d q^2}\frac{d\Sigma}{|\nabla_qE|\,|\nabla_{q'}E|\,a^2\omega_d(q-q')^2}.\quad(10)$$

Now, making use of the assumption that the Fermi surfaces of electrons pointing up and down are not radically different, we may replace $\nabla_qE$ and $\nabla_{q'}E$ by $\Delta E/(q_\uparrow-q_\downarrow)$, where $\Delta E$ is the difference in Zeeman (or kinetic) energies between electrons pointing up and down at the Fermi surface. Thus $\nabla_qE=\hbar\omega_{sd}/(q_\uparrow-q_\downarrow)$. Furthermore, we may replace $q'/q$ by 1. Thus,

$$1/T_1=\frac{4\pi kT}{\hbar}\left(\frac{a}{2\pi}\right)^6\int\left(\frac{A}{2}\right)^2\frac{\pi}{\hbar^2\omega_d^2a^4}d\Sigma=\frac{\pi kT}{\hbar}\left(\frac{A}{\hbar\omega_d}\right)^2\frac{\pi a^2\Sigma}{(2\pi)^6}.\quad(11)$$

$\Sigma$ is the total area of the Fermi surface. The nuclear resonant frequency is given by $\hbar\omega=AS$. Hence, finally

$$1/T_1=\frac{kT}{\hbar}\left(\frac{\omega}{\omega_d}\right)^2\frac{a^2\Sigma}{32S^2\pi^3}.\quad(12)$$

In this expression, all quantities except $\Sigma$ are known experimentally. $T_1$ has been determined in the present work. $\omega$, the nuclear resonant frequency, is known from the NMR data.[1,12] The spin wave spectrum can be determined from the specific heat at low temperatures,[11] or more accurately from the temperature dependence of

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[12] E. L. Boyd, L. J. Bruner, J. I. Budnick, and R. J. Blume, Bull. Am. Phys. Soc. 6, 159 (1961).

the magnetization,[13] neutron diffraction,[14] or ferromagnetic resonance in thin films.[15] $S$ is determined from the saturation magnetization.[11] Thus, $\Sigma$ can be determined. The values of $\Sigma$ calculated by use of formula (12) are given in Table II.

## DISCUSSION

The area of the Fermi surface in iron, cobalt, and nickel is not known. Hence, it is not possible to say just how well the present model accounts for the observed relaxation rates. However, it is seen that a rather simple surface—namely, a sphere—accounts very well for the observed relaxation rate in cobalt. Therefore, we have to account for two features: first, why does the vastly oversimplified theory work at all; and second, why does it fail to account for the fast relaxation rate in iron and nickel. Concerning the first point, it may be noted that the errors introduced by the various approximations have a tendency to cancel out. The *s-d* interaction is not known in the solid. However, for reasonable values of $\omega_{sd}$, $T_1$ is independent of $\omega_{sd}$ since both the strength of the interaction and its inhibition due to the difficulty of momentum transfer are proportional to $\omega_{sd}^2$, and, as a result, $\omega_{sd}$ cancels out. Therefore, even a vague concept of the *s-d* exchange interaction can do here. The same argument applies to the density of states. Both the rate of the relaxation and the polarization of the conduction electrons are proportional to $g(E_F)^2$, which, therefore, cancels out. Another feature which appears to be not too critical is the shape of the Fermi surface. $T_1$ depends only upon the total area of the Fermi surface, and not upon various subtle features of it (unless the Fermi surface is *very* badly distorted). Another simplifying feature is that $T_1$ does not depend much upon the details of the spin wave spectrum at very low or very high values of $k$. The spin wave spectrum at low values of $k$, which is very intricate, does not affect $T_1$ much because the conduction electrons must change their momentum considerably when flipping, and this momentum must be supplied by the spin waves. (Note that $\omega_{\text{loc}}$ does not appear in the final expression for $T_1$.) The spin wave spectrum at high values of $k$ does not affect $T_1$ much because the high virtual energy required to excite these spin waves limits their contribution. This is fortunate, since in metals the spin wave model breaks down at high values of $k$. As a result, only the spin waves of medium energy (such that their wave vector $\mathbf{k}$ is of the order of the difference in the wave number between electrons point "up" and "down" at the Fermi surface) are important. But the medium-energy spin waves are just those which contribute most to the specific heat and change in saturation magnetization at low temperatures. Therefore, a value of $\omega_d$ determined experimentally using these values, has a good chance to account well for the $T_1$ process. Another simplifying feature is, that the details of the conduction electron wave function do not affect the relaxation rate much. As mentioned above, the important spin waves are those with $k\approx q_\uparrow - q_\downarrow \approx 3\times 10^6$ cm$^{-1}$. Therefore, in order to calculate the interaction of the electronic wave function with such spin waves, we must average the electronic wave function over distances of order $1/k\approx 30$ Å. Averaging over such distances, the functions $u(\mathbf{r})\exp(i\mathbf{q}\cdot\mathbf{r})$ and $\exp(i\mathbf{q}\cdot\mathbf{r})$ are virtually indistinguishable. This gives further support to the neglect of umklapp processes. The area of the Fermi surface predicted by formula 12 for iron and nickel is about three times larger than that which would be expected for a spherical surface containing about one electron per atom. There may be various causes for this deviation. In cobalt, as shown in Table II, slightly different spin wave spectra result in values of $\Sigma$ differing by a factor of 2. This deviation is not surprising. The temperature dependence of the magnetization in iron and cobalt is given by the expression[16]

$$M_s = M_0(1 - aT^{3/2} - bT^3).$$

The $aT^{3/2}$ term is predicted by elementary spin wave theory,[11] and $\omega_d$ is determined from this term. However, the large $bT^3$ is not yet understood, and as long as we cannot account for it, the value of $\omega_d$ determined from the $aT^{3/2}$ term cannot be considered to be precise. In iron, the value of $a$ determined from NMR data[13] agrees with the older value, quoted by Kittel,[11] to within 10%. In nickel, it has not yet been possible to determine the $aT^{3/2}$ term from the temperature dependence of the NMR frequency.[12] Consequently, the ambiguity in the values of $\omega_d$ and $S$ in iron and nickel may easily result in errors or a factor of two or so in $\Sigma$. In nickel, a small change in $S$ will result in a particularly large change in $\Sigma$ because $S$ is so small. In iron, the experimental value quoted for $T_1T$ may be somewhat too low, because the extremely wide distribution of relaxation times in iron makes it difficult to verify that the quoted value of $T_1T$ is actually the upper limit. Consequently, the value of $\Sigma$ quoted for iron and nickel is reliable only within a factor of 3 or 4. Furthermore, it would be very surprising if the Fermi surface in iron, cobalt, and nickel were actually spherical.[17] A nonspherical surface enclosing a given volume possesses a larger area than a spherical one, thus the deviations are in the right direction. Thus, we must conclude that in light of the presently available data, the proposed model accounts for the observed relaxation rates surprisingly well.

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