Interaction between Spin Waves and Conduction Electrons in Ferromagnetic Metals*

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With a view toward understanding ferromagnetic resonance line widths in metals, the relaxation time due to the spin wave-conduction electron interaction has been calculated. The effect due to the interaction of ferromagnetic spins with conduction electron currents is much more important than that due to the interaction of ferromagnetic spins with conduction electron spins. In nickel, for temperatures between 4°K and 300°K, the former process yields a relaxation time $10^{-8}-10^{-7}$ sec, while the latter yields $10^{-3}-10^{-4}$ sec. The interactions do not appear to be strong enough to account for the observed line widths.

I. INTRODUCTION

THE role of relaxation processes in determining the line width in ferromagnetic spin resonance absorption[1] has recently been discussed by several authors.[2–4] According to the discussion of KA,[4] for temperatures less than about one-half the Curie temperature, the line width is determined by spin-spin interactions of magnetic dipolar and pseudodipolar origin. On this view, a calculation of the spin-lattice relaxation involves the assumption that the ferromagnetic spin system is in thermal equilibrium at a definable spin temperature which is higher than the lattice temperature. This is an attractive idea and it greatly simplifies the calculation of spin-lattice relaxation times; furthermore, the experimental evidence lends some degree of support. However, this interpretation leaves the line width still unexplained, for it has not yet been demonstrated quantitatively that spin-spin interactions can lead to the large temperature independent line widths that have been observed at low temperatures.[5] Whether or not spin-spin interactions can be responsible for the line widths at low temperatures must therefore remain an open question. In the calculation which follows, we ascertain that the interaction of the magnetization (ferromagnetic spin system) with the conduction electrons cannot account for the major part of the line width in ferromagnetic metals at low temperatures. The line width must then be due to another mechanism which, according to KA, is the spin-spin interaction.

The direct spin-lattice relaxation time has previously been calculated using a macroscopic interaction between spin-wave and phonon fields.[6] In the present work, a calculation is made, by similar techniques, of the relaxation processes which transfer energy from the ferromagnetic spin system to the lattice by way of the conduction electrons (s-electrons). Since the conduction electron-lattice interaction is so strong (with a characteristic time of the order of 10^{-13} sec),[7] we may consider the s-electrons to be part of the lattice and we then treat the energy transfer as a "spin-electron relaxation." The conduction electrons will be treated as a degenerate electron gas by means of the single particle model in the plane wave approximation. The spin waves will be treated by methods discussed by KA.

It has been shown[8] that in a ferromagnetic conductor, the resonance frequency for the absorption of microwave power is shifted by an amount inversely proportional to the square of the eddy current skin depth δ. In spin-wave language, this means that the only spin waves excited by the rf field have wave vectors $\boldsymbol{\kappa}$ whose magnitudes lie close to the value $1/\delta$. This is to be compared with the situation in insulators where the microwave excitation is limited to those spin waves having wave numbers κ near zero. In the case of the direct spin-lattice interaction, the difference is insignificant as the phonons of interest have wave numbers very much larger than $1/\delta$, and the previous calculation[9] for the direct interaction which scatters $\kappa=0$ spin waves remains essentially unchanged in the case of metals. In order to determine whether the interaction of spin waves with conduction electrons can account for the line width, it is necessary to evaluate the time constant (relaxation time) of the decay of an excess number of spin waves of wave number $1/\delta$. If this interaction is to account for the width, the relaxation time must be as short as 10^{-9}–10^{-10} sec.

Previous work on spin-wave-electron interactions has been reported by Samoilovich and Yokovleff[10] in connection with electrical resistivity. Kondoh[11] has published a report on a calculation using the dipolar interaction between s- and d-electrons in the spin-wave

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[1] Ferromagnetic resonance is reviewed by C. Kittel, J. phys. radium **12**, 291 (1951); J. H. Van Vleck, Physica **17**, 234 (1951).

[2] P. W. Anderson, Phys. Rev. **88**, 1213 (1952).

[3] N. Bloembergen and S. Wang, Phys. Rev. **93**, 72 (1954).

[4] C. Kittel and E. Abrahams, Revs. Modern Phys. **25**, 233 (1953); henceforth denoted by KA.

[5] N. Bloembergen, Phys. Rev. **78**, 572 (1950). A. F. Kip (private communication).

[6] E. Abrahams and C. Kittel, Phys. Rev. **88**, 1200 (1952). See also reference 3.

[7] It should be mentioned that the relaxation of s-electron spins to the lattice is also expected to be very fast due to the large spin-orbit coupling. See R. J. Elliott, Phys. Rev. **96**, 266 (1954).

[8] C. Kittel and C. Herring, Phys. Rev. **77**, 725 (1950).

[9] J. M. Luttinger and C. Kittel (unpublished). The present author has improved the calculation of Luttinger and Kittel and finds that the spin-lattice relaxation time for $\kappa=0$ spin waves in nickel is of the order of 10^{-5} sec at room temperature.

[10] A. G. Samoilovich and V. A. Yokovleff, Zhur. Eksptl. i. Teoret. Fiz. **22**, 350 (1952).

[11] A. Kondoh, Progr. Theoret. Phys. Japan **10**, 117 (1953).

approximation of Holstein and Primakoff.[12] There is essential agreement between his results and those of a similar calculation which forms Sec. IV of this paper.

II. INTERACTIONS

The interaction operator which we adopt is

$$H_{\text{int}}=H_1+H_2, \quad (1)$$

$$H_1=\frac{e}{mc}\int\frac{\mathbf{M}(\mathbf{r})\cdot(\mathbf{r}-\mathbf{r}_e)\times\mathbf{p}}{|\mathbf{r}-\mathbf{r}_e|^3}e^{-q|\mathbf{r}-\mathbf{r}_e|}d\mathbf{r}, \quad (2)$$

$$H_2=\beta\boldsymbol{\sigma}\cdot\mathbf{H}_s. \quad (3)$$

where $\mathbf{M}(\mathbf{r})$ is the magnetization at the point $\mathbf{r}$, $\mathbf{r}_e$ is the position of a conduction electron, $\mathbf{p}$ its momentum and $\boldsymbol{\sigma}$ its spin. $\mathbf{H}_s$ is the magnetic field due to the ferromagnetic spins and has been given by Herring and Kittel.[13] In Eq. (3), β is the Bohr magneton. H_1 represents an interaction between the ferromagnetic spins and the currents due to the motion of the s-electrons. Furthermore $1/q$ is the screening radius[14] for the magnetic field due to the s-electron currents and is given by $(mc^2/4\pi n_0e^2)^{-\frac{1}{2}}$ where n_0 is the s-electron density. This screening arises from the collective influence of the set of s-electrons and is discussed fully in reference 14. H_2 is the ferromagnetic spin—s-electron spin interaction in the Herring-Kittel approximation. The two interactions H_1 and H_2 will be treated separately since they do not interfere.

The system of conduction electrons is treated as a quantized free electron field with the following convention for the field operators:

$$\begin{aligned}\text{spin up:}\quad &\varphi=b_Ke^{i\mathbf{K}\cdot\mathbf{r}},\\ \text{spin down:}\quad &\psi=d_Ke^{i\mathbf{K}\cdot\mathbf{r}},\end{aligned} \quad (4)$$

where $\mathbf{K}$ is the electron wave vector. Following KA, we have for the spin-wave field:

$$\begin{aligned}M^+(\mathbf{r})&=M_x(\mathbf{r})+iM_y(\mathbf{r})=(2g\beta M_s)^{\frac{1}{2}}\textstyle\sum_\kappa\alpha_\kappa e^{i\boldsymbol{\kappa}\cdot\mathbf{r}},\\ M^-(\mathbf{r})&=M_x(\mathbf{r})-iM_y(\mathbf{r})=(2g\beta M_s)^{\frac{1}{2}}\textstyle\sum_\kappa\alpha_\kappa^*e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}},\end{aligned} \quad (5)$$

where α_κ, b_K, d_K, and their conjugates are annihilation and creation operators and M_s is the saturation magnetization. We work throughout in Gaussian units with a sample of unit volume.

III. INTERACTION WITH S-ELECTRON CURRENTS

The interaction of spin waves with s-electron currents does not cause s-electron spin transitions so that we may consider interactions with s-electrons of one spin only. From Eq. (2), we get the perturbation operator $\mathfrak{H}_1=\int\varphi^*H_1\varphi d\mathbf{r}_e$:

$$\mathfrak{H}_1=(e\hbar/mc)\textstyle\sum_{KK'}b_K^*b_{K'}\displaystyle\int d\mathbf{r}e^{i(\mathbf{K}'-\mathbf{K})\cdot\mathbf{r}}$$
$$\times\mathbf{M}(\mathbf{r})\cdot\int(\mathbf{R}\times\mathbf{K}'/R^3)e^{i(\mathbf{K}'-\mathbf{K})\cdot\mathbf{R}-qR}d\mathbf{R}. \quad (6)$$

Here we have considered electrons with spin up. The evaluation of the integrals which appear here is facilitated by the expansion of $e^{i(\mathbf{K}'-\mathbf{K})\cdot\mathbf{R}}$ in spherical harmonics. The result is

$$\mathfrak{H}_1=(2\pi ie\hbar/mc)(2g\beta M_s)^{\frac{1}{2}}[1-(q/\kappa)\tan^{-1}(\kappa/q)]$$
$$\times\textstyle\sum_{K\kappa}b_K^*b_{K'}\kappa^{-2}[(\mathbf{K}\times\boldsymbol{\kappa})^+\alpha_{-\kappa}^*+(\mathbf{K}\times\boldsymbol{\kappa})^-\alpha_\kappa],$$
$$\boldsymbol{\kappa}=\mathbf{K}-\mathbf{K}'\neq0, \quad (7)$$

plus another term which does not contribute to the relaxation. The first term in square brackets creates a spin wave of wave vector $-\boldsymbol{\kappa}$ while the second destroys one of wave vector $\boldsymbol{\kappa}$. In order to evaluate the decay rate of spin waves of given wave number, we fix κ and consider the rate of change of the population in the state κ if there is a nonequilibrium excess of spin waves in that state. The nonequilibrium excess is due to the excitation by the microwave field.

The net number of collisions per unit time which transfer energy from the spin system to the s-electrons is given by the kinetic equation

$$\dot{N}_{\text{coll}}=(2\pi/\hbar)\textstyle\sum_{K\kappa}(|H_1^e|^2-|H_1^a|^2)\delta(\epsilon_\kappa+\epsilon_{K'}-\epsilon_K), \quad (8)$$

where H_1^e and H_1^a are those parts of Eq. (7) which emit and absorb, respectively, a spin wave of wave vector $\boldsymbol{\kappa}$, and ϵ_κ, ϵ_K are the energies of a spin wave and an electron of wave vectors $\boldsymbol{\kappa}$, $\mathbf{K}$. Here ϵ_κ is equal to $(2g\beta A/M_s)\kappa^2-g\beta H$ where A is the macroscopic exchange constant and H is the applied constant field.

To set up the relaxation time, we fix κ and identify $\dot{N}_{\text{coll}}$ with $\dot{n}_\kappa$, the rate of change of population in the state κ. Further, we take the matrix elements of the creation and destruction operators and average over the directions of the vector $\mathbf{K}\times\boldsymbol{\kappa}$. The result for $\dot{n}_\kappa$ is

$$\dot{n}_\kappa=(32\pi^3/3)(e^2\hbar g\beta M_s/m^2c^2)$$
$$\times[1-(q/\kappa)\tan^{-1}(\kappa/q)]^2\kappa^{-4}\textstyle\sum_K(\mathbf{K}\times\boldsymbol{\kappa})^2$$
$$\times[(n_\kappa+1)(1-f_{K'})f_K-n_\kappa(1-f_K)f_{K'}]$$
$$\times\delta(\epsilon_\kappa+\epsilon_{K'}-\epsilon_K), \quad (9)$$
$$\boldsymbol{\kappa}=\mathbf{K}-\mathbf{K}\neq0.$$

Here n_κ is the occupation number of the spin-wave state κ and f_K is the probability of finding an s-electron in the state K, that is, the Fermi distribution function.

In order to evaluate the terms in square brackets, we set $n_\kappa=n_\kappa^0-\Delta n_\kappa$ where n_κ^0 is the value of n_κ when the spin-wave system is in equilibrium with the s-electrons and Δn_κ is the excess population in the state κ due to the resonance absorption. If $\Delta n_\kappa=0$, the terms

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[12] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).
[13] C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).
[14] D. Bohm and D. Pines, Phys. Rev. **82**, 625 (1951).

in square brackets vanish if one accounts for the energy δ function. Therefore, $\dot{n}_\kappa$ is proportional to Δn_κ and the terms in question become

$$-(df_K/d\epsilon_K)\epsilon_\kappa\Delta n_\kappa. \quad (10)$$

Here we have taken into account the energy δ functions of Eq. (9) and expanded the Fermi factors f near the top of the Fermi distribution.[15] The procedure is justified here since the maximum spin wave energy is of order one-fortieth of the Fermi energy.

The resulting expression for $\dot{n}_\kappa$ in which (10) replaces the square bracket of Eq. (9) is treated in the following way: The sum is transformed into an integral over $\mathbf{K}$ space with the introduction of a factor $2/(2\pi)^3$ where the 2 in the numerator accounts for two possible spin directions. The energy δ function is expressed in terms of $\mathbf{K}$, $\boldsymbol{\kappa}$ and the angle between them, θ, by the substitution $\mathbf{K}'=\mathbf{K}-\boldsymbol{\kappa}$.

$$\begin{aligned}\delta(\epsilon_\kappa+\epsilon_{K'}-\epsilon_K)&=(m/\hbar^2K\kappa)\delta[\cos\theta-F(\kappa,K)],\\ F(\kappa,K)&=[(\hbar^2\kappa^2/2m)+\epsilon_\kappa]/(\hbar^2K\kappa/m).\end{aligned} \quad (11)$$

The δ function is annihilated by the integration over θ and the remaining angular integration is trivial. The result is

$$\begin{aligned}\dot{n}_\kappa=&-(16\pi e^2g\beta M_s/3mc^2\hbar)\Delta n_\kappa\\ &\times[1-(q/\kappa)\tan^{-1}(\kappa/q)]^2(\epsilon_\kappa/\kappa^3)\\ &\times\int K^3dK(df_K/d\epsilon_K)[1-F^2(\kappa,K)],\\ &\qquad F^2(\kappa,K)\leq 1. \quad (12)\end{aligned}$$

The condition $F^2\leq 1$ arises from the integration over θ where we must have $\cos^2\theta\leq 1$. The factor $df_K/d\epsilon_K$ behaves as a δ function, $\delta(\epsilon_K-E_F)$ where E_F is the Fermi energy for the s-electrons. This and the fact that the wave numbers of interest for the spin waves are of order 10^5 assure the smallness of F and it will henceforth be neglected. The integral is easily evaluated with this δ-function approximation. The result is

$$\begin{aligned}1/\tau=-\dot{n}_\kappa/\Delta n_\kappa=&(32\pi me^2g\beta M_s/3c^2\hbar^5)\\ &\times[1-(q/\kappa)\tan^{-1}(\kappa/q)]^2(\epsilon_\kappa/\kappa^3)E_F. \quad (13)\end{aligned}$$

This result is the relaxation time for spin waves of wave number κ. In the case at hand we set $\kappa=1/\delta=(4\pi\omega\mu_2/\rho c^2)^{\frac{1}{2}}$ and find the relaxation time for those spin waves which are excited by the microwave field. Here ρ is the resistivity, ω is the angular frequency of the microwaves, and $i\mu_2$ is the permeability at resonance. For nickel, at 24 400 mc/sec, we have $\mu_2=17$,[5] $\rho=7.74$ micro-ohm cm,[16] and $\delta=1.5\times10^{-5}$ cm at room temperature. Thus, from Eq. (13), we find the relaxation time at room temperature in nickel

$$\tau=6\times10^{-8}\text{ sec},\quad T=293°\text{K}. \quad (14)$$

The temperature dependence below room temperature is quite insignificant. The reason for this is that as the temperature is lowered, the skin depth is reduced in magnitude as the resistivity decreases. This results in a decrease in the factor $\kappa^{-3}=\delta^3$ of Eq. (13) which is offset by a concomitant increase of the screening factor which appears in the square brackets. The experimental value of Jan and Gijsman[16] for the resistivity at 14°K is about 1/20 of the room temperature result. If we use this value in our formulas we obtain

$$\tau=2\times10^{-8}\text{ sec},\quad T=14°\text{K}. \quad (15)$$

One would ordinarily expect a somewhat stronger temperature dependence of the resistivity in a very pure sample. However, even if we assume that the resistivity has decreased by as much as a factor 100, the relaxation time at 14°K remains of the same order because of the compensation of the aforementioned screening and skin depth effects. At room temperature and above, the relaxation time increases with δ because of the increased effectiveness of the screening, although we must realize that the spin-wave approximation is of doubtful validity before these temperatures are reached.

IV. INTERACTION WITH S-ELECTRON SPINS

The perturbation operator for the interaction of spin waves with s-electron spins is given by

$$\mathcal{H}_2=\int\begin{pmatrix}\varphi\\ \psi\end{pmatrix}^*\beta\boldsymbol{\sigma}\cdot\mathbf{H}_s\begin{pmatrix}\varphi\\ \psi\end{pmatrix}d\mathbf{r},$$

where[13] $\mathbf{H}_s=-2\pi(2g\beta M_s)^{\frac{1}{2}}(\boldsymbol{\kappa}/\kappa^2)(\alpha_\kappa\kappa^-+\alpha_{-\kappa}^*\kappa^+)e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}$. Evaluation of this expression gives

$$\begin{aligned}\mathcal{H}_2=&-2\pi\beta(2g\beta M_s)^{\frac{1}{2}}\textstyle\sum_{K\kappa}(1/\kappa^2)(\alpha_\kappa\kappa^-+\alpha_{-\kappa}^*\kappa^+)\\ &\times[b_K^*d_{K'}\kappa^-+d_K^*b_{K'}\kappa^++(b_K^*b_{K'}-d_K^*d_{K'})\kappa_z]\\ &\times\delta(\boldsymbol{\kappa}+\mathbf{K}'-\mathbf{K}). \quad (16)\end{aligned}$$

The first two terms in square brackets involve s-electron spin flips while the third term does not. Consider first that process in which an s-electron flips from down to up and a spin wave is destroyed. This and its inverse process together give the collision rate by an expression exactly similar to Eq. (8) where, for example,

$$\begin{aligned}H_2{}^e=&-2\pi\beta(2g\beta M_s)^{\frac{1}{2}}(\kappa^+\kappa^-/\kappa^2)\\ &\times[(n_\kappa+1)(1-f_{K'}{}^-)f_K{}^+]^{\frac{1}{2}}\delta(\boldsymbol{\kappa}+\mathbf{K}'-\mathbf{K}).\end{aligned}$$

Here we have taken the matrix elements of the creation and destruction operators. $f_K{}^+$, for example, denotes the Fermi factor for wave vector $\mathbf{K}$ and spin up. For this interaction, the rate of energy transfer is given by

$$\begin{aligned}\dot{n}_\kappa=&(128\pi^3\beta^3gM_s/15\hbar)\textstyle\sum_{K\kappa}\epsilon_\kappa[(n_\kappa+1)(1-f_{K'}{}^-)f_K{}^+\\ &-n_\kappa(1-f_K{}^+)f_{K'}{}^-]\delta(\epsilon_{K'}{}^-+\epsilon_\kappa-\epsilon_K{}^+), \quad (17)\\ &\boldsymbol{\kappa}+\mathbf{K}'=\mathbf{K},\end{aligned}$$

where we have averaged over directions of $\boldsymbol{\kappa}$. Here

[15] The method used here is similar to that used in another connection by A. W. Overhauser, Phys. Rev. 89, 689 (1953).

[16] J. P. Jan and J. M. Gijsman, Physica 18, 339 (1952).

$\epsilon_K{}^{\pm}=(\hbar^2K^2/2m)\pm\beta H$. The terms in square brackets are handled in a way similar to the treatment of Eq. (9). We assume that there are equal numbers of s-electron spins up and down with small error since $\beta H/kT\ll 1$. With this assumption, as in the case of Eq. (9), the terms in square brackets reduce to Eq. (10) with the same notation.

We now proceed as before. The energy δ function becomes $(m/\hbar^2K\kappa)\delta(\cos\theta-F^-)$, where F^- is as in Eq. (11) save that $-2\beta H$ is added to the numerator. If we had considered instead the process in which the electron of wave vector $\mathbf{K}$ has down-spin and $\mathbf{K}'$ up-spin rather than the reverse, we would have gotten a factor F^+ with $+2\beta H$ added to the numerator. A detailed examination of the resulting expression for $\dot{Q}$ shows that we may neglect the distinction between F, F^+, and F^- since $\beta H\ll E_F$. We therefore account for the two kinds of processes mentioned here by the replacement of $F^{\pm}$ by F and the introduction of a factor 2. From Eq. (17) we obtain

$$\dot{n}_\kappa=-(64\pi m\beta^3 gM_s/15\hbar^3)\Delta n_\kappa(\epsilon_\kappa/\kappa)\times\int KdK(df_K/d\epsilon_K),\quad F^2(\kappa,K)\leq 1. \quad (18)$$

As was the case with Eq. (12), $F^2\leq 1$ for κ near $1/\delta$ and K at the top of the Fermi distribution. The integral then reduces immediately to $(m/\hbar^2)$.

If we had considered the third term in square brackets of Eq. (16), i.e., processes in which the s-electron spin does not flip, then we would have gotten the same expression as Eq. (18) but reduced by a factor 1/4. To take all processes into account we need only multiply Eq. (18) by 5/4. The relaxation time for the spin-wave —s-electron interaction is therefore given by

$$1/\tau=-\dot{n}_\kappa/\Delta n_\kappa=(8\pi/3)(m^2g\beta^3M_s/\hbar^5)(\epsilon_\kappa/\kappa). \quad (19)$$

We again set $\kappa=1/\delta$ and find for nickel,

$$\tau=2\times 10^{-9}/\delta=10^{-4}\text{ sec at }T=293°\text{K}$$
$$=10^{-3}\text{ sec at }T=\ 14°\text{K}.$$

V. CONCLUSION

The interaction considered in Sec. IV is negligible compared with the "spin-current" interaction of Sec. III. The latter results in a relaxation time whose temperature dependence does not contradict the experimental results for the line width between liquid hydrogen and room temperatures. However, the relaxation time appears to be at least one order of magnitude too large to account for the magnitudes of the observed widths.

The present calculation does not apply to low-resistivity ferrites[17] because the free electron model cannot work well for d-band semiconductors such as the ferrites.

It is of interest to discuss the relaxation time for the "spin-current" interaction in the case that interactions between ferromagnetic spins are so strong that they account for the line width. In this case, according to KA, the spin system is in an equilibrium state before other relaxation processes have progressed appreciably. The relaxation time for energy transfer to the lattice via the conduction electrons is then determined by the spin-spin relaxation time (line width) itself for the following reason: the spin-current relaxation time for spin waves having wave numbers near q is very short indeed due to the negligible screening and the smallness of q (10^5 cm^{-1}). In this connection see Eq. (13). The rate of energy transfer to the lattice is then limited by the rate at which energy can be transferred from other spin-wave states to those spin waves with wave numbers near q by means of spin-spin interactions. This is because the relaxation primarily takes place through the spin-wave states of wave number q and at a rate somewhat faster than the spin-spin relaxation time.

It should be noted that the calculation begins to lose validity because of the inadequacy of the spin-wave approximation as the temperature rises above about one tenth of the Curie temperature (631°K for nickel). At higher temperatures, one would expect that the relaxation times will become somewhat shorter than those predicted here. Within the limits of the spin-wave approximation, however, the interactions discussed here do not appear to be strong enough to account for the experimentally observed line widths. which correspond to relaxation times of $10^{-9}-10^{-10}$ sec. The interaction of spin waves and conduction electron currents is stronger, however, than the direct spin-lattice interaction calculated previously,[6] especially at low temperatures where it gives much the shorter relaxation time.

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Note added in proof.—A paper of T. Kasuya [Busseiron Kenkyu **74**, 1 (1954)], in which a calculation is made which is similar to the one reported here has come to the author's attention. Kasuya, however, fails to include the effect of screening of the magnetic field of the s-electron currents; nor does he attribute the breakdown to the $\kappa=0$ selection rule to the effect of the finite skin depth. The neglect of these effects yields a relaxation time an order of magnitude shorter than the result of the present work.

[17] Galt, Yager, and Merritt, Phys. Rev. **93**, 1119 (1954).