SOVIET PHYSICS JETP VOLUME 33, NUMBER 4 OCTOBER, 1971

THERMAL EMF AND THERMAL RESISTANCE OF FERROMAGNETIC METALS WITH IMPURITIES

I. Ya. KORENBLIT and Yu. P. LAZARENKO

A. F. Ioffe Physico-technical Institute, U.S.S.R. Academy of Sciences

Submitted November 17, 1970

Zh. Eksp. Teor. Fiz. 60, 1548–1559 (April, 1971)

The joint scattering of electrons by magnons and impurities leads to a number of anomalies in the kinetic properties of ferromagnetic metals. If the impurities are magnetic, then the thermal emf depends on temperature in a nonmonotonic fashion for a given impurity concentration and on impurity concentration at a given temperature. At the peak, the thermal emf may have values comparable with k/e and its sign is determined by the nature of the impurity. As the impurity concentration increases, the thermal emf peak shifts towards higher temperatures; scattering of electrons by phonons decreases the thermal emf and shifts the peak towards lower temperatures. The theoretical results for the thermal emf are in good agreement with experiment. It is found from comparison of the theoretical with the experimental thermal emf that the characteristic temperature for single-magnon scattering in Ni is $T_0 \approx 25°K$. The effect on the thermal emf of the scattering of electrons from the s- to the d-band by magnons and phonons is discussed. It is shown that Mathiessen's rule for thermal resistance is violated in the scattering of electrons by magnetic as well as nonmagnetic impurities.

INTRODUCTION

THE scattering of conduction electrons by spin waves leads to interesting peculiarities in the kinetic properties of ferromagnetic metals. In ordinary metals, the thermal emf is $\alpha \approx k^2T/e\epsilon_F$, since the thermal flux due to the "cold" electrons and the flux due to the "hot" electrons cancel each other to within the temperature width of the Fermi distribution[1] (ϵ_F is the Fermi energy, T—the temperature, e—the electron charge, and k—the Boltzmann constant). This is not so with ferromagnetic metals. The scattering of electrons by magnons is asymmetric in character: an electron with spin $\uparrow$ can only absorb a magnon, while an electron with spin $\downarrow$ only emits a magnon. Consequently, the thermal flux of the "cold" electrons does not fully cancel out that of the "hot" electrons, and at temperatures that are small compared to the Curie temperature T_C the thermal emf in pure ferromagnetic metals is of the order of $kIT/e\epsilon_F T_C$[2,3] (I is the s-d exchange energy, $I \ll \epsilon_F$), whereas in metals with nonmagnetic impurities, it may have values of the order of $kI/e\epsilon_F$[3]. It is assumed here that the s-s electron scattering predominates over the s-d electron scattering. In both cases the thermal emf contains small parameters, since the dynamical properties of electrons with spins $\uparrow$ and $\downarrow$ differ by a small quantity of the order of I/ϵ_F. If, however, apart from the electron-magnon scattering, there exists another scattering mechanism, the effectiveness of which is different for electrons with different spin directions, then the thermal emf may not contain a small parameter.

Precisely such a situation arises when electrons are scattered by magnons and by those impurities for which the relaxation times for electrons with spins $\uparrow$ and $\downarrow$ ($\tau_\uparrow$, $\tau_\downarrow$) are not equal (we shall henceforth call these impurities magnetic). The situation is, in many ways, analogous to the case of inelastic scattering of electrons by magnetic impurities in nonferromagnetic metals in a magnetic field (see[4] and the papers cited therein). The large magnitude and other anomalies of the thermal emf of ferromagnetic metals with magnetic impurities were predicted by us in[5].

In the present paper we calculate the thermal emf and the thermal resistance of ferromagnetic metals for electron scattering by magnons, impurities, and phonons. The thermal emf depends in a nonmonotonic fashion on temperature and impurity concentration, may have large values and strongly depends on the nature and concentration of the magnetic impurities. The results obtained here and in[5] are in good agreement with the recently published experimental data on the thermal emf of Ni with magnetic impurities[6].

The most interesting feature of thermal resistance is the considerable deviation from the Mathiessen rule. This is not only the case in electron scattering by magnons and magnetic impurities, but also in electron scattering by magnons and nonmagnetic impurities.

1. THERMAL EMF

A. Let us first consider the thermal emf in s-s electron scattering by spin waves, magnetic impurities, as well as by phonons. We shall assume that the concentration of impurities is sufficiently large so that relaxation of the electrons with respect to momentum occurs on the impurities, while only energy relaxation occurs on the magnons and phonons. Accordingly, we retain in the kinetic equations for electrons with spin $\uparrow$ and $\downarrow$ only terms of lowest order in the small parameters T/T_C and T/T_D (T_D is the Debye temperature). We recall that the ratio of the momentum relaxation time to the energy relaxation time in electron scattering by magnons and phonons is T_C/T for the case of scattering by magnons and $(T_D/T)^2$[3,7] for phonons. We also neglect terms of the order of I/ϵ_F and kT/ϵ_F.

The spectrum of the s—electrons is assumed isotropic. Such an assumption is satisfactory, for example, for Ni[8]. Since we consider in this paper an interval of temperatures which are low compared to T_C, a quadratic spectrum may be assumed for the magnons; $\hbar\omega_q = q^2/2\mu$, where μ is the magnon mass.

Representing the nonequilibrium correction to the electron distribution function in the form

$$n_{p\downarrow,\uparrow} = -(\mathbf{u}_{\downarrow,\uparrow}(\varepsilon), \mathbf{p})\frac{\partial n_p}{\partial \varepsilon_p}, \quad (1)$$

where p is the electron momentum and n_p is the Fermi distribution function, we write the system of linearized kinetic equations for electrons in an external electric field E in the form

$$\frac{\Theta}{t_{s0}}\int_{1/\Theta}^{\infty} d\xi\Phi^+(x,\xi)\,[\mathbf{u}_\downarrow(x+\xi)-\mathbf{u}_\uparrow(x)] + \frac{\Theta^3}{t_{\text{pho}}}\int_0^{\Theta_D/\Theta} d\xi\,\xi^2\Big\{\Phi^+(x,\xi)\,[\mathbf{u}_\uparrow(x+\xi)-\mathbf{u}_\uparrow(x)] + \Phi^-(x,\xi)\,[\mathbf{u}_\uparrow(x-\xi)-\mathbf{u}_\uparrow(x)]\Big\} + \frac{1}{\tau_\uparrow}\frac{\partial n}{\partial x}\mathbf{u}_\uparrow(x) = \frac{e\mathbf{E}v}{p}\frac{\partial n}{\partial x}, \quad (2a)$$

$$\frac{\Theta}{t_{s0}}\int_{1/\Theta}^{\infty} d\xi\,\Phi^-(x,\xi)\,[\mathbf{u}_\uparrow(x-\xi)-\mathbf{u}_\downarrow(x)] + \frac{\Theta^3}{t_{\text{pho}}}\int_0^{\Theta_D/\Theta} d\xi\,\xi^2\Big\{\Phi^+(x,\xi)\,[\mathbf{u}_\downarrow(x+\xi)-\mathbf{u}_\downarrow(x)] + \Phi^-(x,\xi)\,[\mathbf{u}_\downarrow(x-\xi)-\mathbf{u}_\downarrow(x)]\Big\} + \frac{1}{\tau_\downarrow}\frac{\partial n}{\partial x}\mathbf{u}_\downarrow(x) = \frac{e\mathbf{E}v}{p}\frac{\partial n}{\partial x}. \quad (2b)$$

Here $x = (\epsilon - \epsilon_F)/kT$, ϵ is the electron energy, v its velocity, and

$$\Phi^+(x,\xi) = \Phi^-(-x,\xi) = \frac{e^{x+\xi}}{(e^x+1)(e^{x+\xi}+1)(e^\xi-1)}. \quad (3)$$

The energy kT_0 is the minimum magnon energy that may be absorbed or emitted by an electron in the single-magnon process. It is determined from the condition $kT_0 = (\Delta p)^2/2\mu$, where $\Delta p \ll P_F$ is the minimum change in electron momentum in a scattering with spin flip at the Fermi surface. The dimensionless temperature is $\Theta = T/T_0$. The quantity t_{s0} has the meaning of energy relaxation time for electrons on magnons at the temperature $\Theta = 1$. If we consider the electron-magnon interaction in the framework of the customary model of s-d exchange, then we obtain for t_{s0} an expression which coincides with the expression for τ_S in[3] at $T = T_0$. The quantity t_{pho} has the meaning of energy relaxation time for electrons on magnons at $\theta = 1$. We shall not need the explicit form of t_{pho} (see, for example,[7]). In the collision integrals of the system (2), which describe the scattering of electrons by phonons, the upper limit of the temperature region considered is much larger than unity. Nevertheless, we shall not replace it by ∞, since, as we shall see, the dependence of these integrals on the upper limit can be significant.

For $\tau_\uparrow \neq \tau_\downarrow$ the scattering of electrons by magnons, described by the first term of each equation of the system (2), influences the transfer of momentum by the electrons to the impurities since flipping of the electron spin occurs in such scattering. There is, however, no momentum transfer from the electrons to the magnon system itself.

It follows from (1) and the Onsager relations that the thermal emf can be expressed in the following manner in terms of the drift velocities $u_\uparrow(x)$ and $u_\downarrow(x)$ given by the system (2):

$$\alpha = \frac{k}{e}\int_{-\infty}^{+\infty} dx\,x\frac{\partial n}{\partial x}[u_\uparrow(x)+u_\downarrow(x)] \Big/ \int_{-\infty}^{+\infty} dx\frac{\partial n}{\partial x}[u_\uparrow(x)+u_\downarrow(x)]. \quad (4)$$

We see from this that the thermal emf differs from zero only in the case when the function $u_\uparrow(x) + u_\downarrow(x)$ is not an even function of x[3]. The symmetry properties of the function $u_\uparrow(x) + u_\downarrow(x)$ can be seen directly from the system (2).

Let us, following[3], introduce the function

$$w_\pm(x) = u_\uparrow(x) \pm u_\downarrow(-x), \quad (5)$$

so that

$$u_\uparrow(x) + u_\downarrow(x) = {}^1\!/_2[w_+(x) + w_+(-x) + w_-(x) - w_-(-x)]. \quad (6)$$

The system (2) may be rewritten in the form

$$\frac{\Theta}{t_{s0}}\int_{1/\Theta}^{\infty} d\xi\,\Phi^+(x,\xi)\,[w_+(-x-\xi)-w_+(x)] + \frac{\Theta^3}{t_{\text{pho}}}\int_0^{\Theta_D/\Theta} d\xi\xi^2\{\Phi^+(x,\xi)\,[w_+(x+\xi)-w_+(x)] + \Phi^-(x,\xi)\,[w_+(x-\xi)-w_+(x)]\} + \frac{1}{2}\frac{\partial n}{\partial x}\Big\{w_+(x)\left(\frac{1}{\tau_\uparrow}+\frac{1}{\tau_\downarrow}\right) + w_-(x)\left(\frac{1}{\tau_\uparrow}-\frac{1}{\tau_\downarrow}\right)\Big\} = \frac{2eEv}{p}\frac{\partial n}{\partial x}, \quad (7a)$$

$$\frac{\Theta}{t_{s0}}\int_{1/\Theta}^{\infty} d\xi\,\Phi^+(x,\xi)\,[w_-(-x-\xi)+w_-(x)] + \frac{\Theta^3}{t_{\text{pho}}}\int_0^{\Theta_D/\Theta} d\xi\,\xi^2\{\Phi^+(x,\xi)\,[-w_-(x+\xi)+w_-(x)] + \Phi^-(x,\xi)\,[-w_-(x-\xi)+w_-(x)]\} + \frac{1}{2}\frac{\partial n}{\partial x}\Big\{w_+(x)\left(\frac{1}{\tau_\downarrow}-\frac{1}{\tau_\uparrow}\right) - w_-(x)\left(\frac{1}{\tau_\downarrow}+\frac{1}{\tau_\uparrow}\right)\Big\} = 0. \quad (7b)$$

When $\tau_\uparrow = \tau_\downarrow$ the system (7) separates into two independent equations for $w_+(x)$ and $w_-(x)$, and the equation for $w_-(x)$ has only a trivial solution, i.e., according to (6), the function $u_\uparrow(x) + u_\downarrow(x)$ is even. If electron scattering by magnons is insignificant ($t_{s0} \to \infty$ in (7)), for an arbitrary relationship between $\tau_\uparrow$ and $\tau_\downarrow$, the solution of the system (7) has the properties $w_+(x) = w_+(-x)$, $w_-(x) = w_-(-x)$, i.e., $u_\uparrow(x) + u_\downarrow(x)$ is again an even function. In both cases, the thermal emf vanishes to within the previously neglected terms of order I/ϵ_F and kT/ϵ_F.

If, on the other hand, $\tau_\uparrow \neq \tau_\downarrow$, and the scattering of electrons by magnons is considerable, then, as can be seen from (6) and (7), $u_\uparrow(x) + u_\downarrow(x)$ ceases to be an even function of x and the thermal emf becomes anomalously large and comparable with k/e.

We shall seek the solution of the system (7) in the form

$$w_\pm(x) = w_\pm^{(1)} + w_\pm^{(2)}x,$$

where $w_\pm^{(1)}$ and $w_\pm^{(2)}$ do not depend on x. Then the system of integral equations (7) reduces to a system of four algebraic equations for $w_\pm^{(1)}$ and $w_\pm^{(2)}$. Solving this system, we find with the aid of (4) and (6) the thermal emf:

$$\alpha=\frac{\pi^2}{6}\frac{k}{e}(\tau_\uparrow-\tau_\downarrow)\left\{t_s\left(1+\frac{1}{2}\frac{\tau_\uparrow+\tau_\downarrow}{t_{ph}}\right)+A(\Theta)(\tau_\uparrow+\tau_\downarrow)+\tau_\uparrow\tau_\downarrow\left(B(\Theta)t_s^{-1}+C(\Theta)t_{ph}^{-1}\right)\right\}^{-1}. \tag{8}$$

Here,

$$t_s^{-1}=\frac{3}{\pi^2}\Theta K_2(\Theta)t_{s0}^{-1},\qquad t_{ph}^{-1}=\Theta^3 Q\left(\frac{\Theta}{\Theta_D}\right)t_{ph_0}^{-1},$$

$$A(\Theta)=\frac{4\pi^2K_1(\Theta)+3K_3(\Theta)}{12K_2(\Theta)},\qquad B(\Theta)=\frac{\pi^2}{3}\frac{K_1(\Theta)K_3(\Theta)-K_2^2(\Theta)}{K_2^2(\Theta)}$$

$$C(\Theta)=\frac{2\pi^2}{3}\frac{K_1(\Theta)}{K(\Theta)},\qquad K_n(\Theta)=\int_{1/\Theta}^{\infty}\frac{\xi^n e^\xi}{(e^\xi-1)^2}d\xi,$$

$$Q(z)=\frac{3}{\pi^2}\int_0^{1/z}\frac{\xi^5e^\xi}{(e^\xi-1)^2}d\xi. \tag{9}$$

We have again left out in the denominator of (8) one addend which, as an estimate of it showed, is considerably smaller than the remaining terms at all temperatures.

At low temperatures $\Theta \ll 1$ we have

$$t_s=\frac{\pi^2}{3}\Theta e^{1/\Theta}t_{s0},$$

$$A(\Theta)=\frac{1}{4\Theta},\qquad B(\Theta)=\frac{\pi^2}{3}\Theta^2,\qquad C(\Theta)=\frac{2\pi^2}{3}\Theta. \tag{10}$$

The exponential increase of t_s as Θ decreases at low temperatures is connected with the "freezing" of magnons with momentum larger than Δp which are responsible for electron scattering with spin flip at the Fermi surface.

At high temperatures $\Theta \gg 1$

$$t_s=\frac{1}{\Theta}t_{s0},\qquad A(\Theta)=1+\frac{9\zeta(3)}{2\pi^2}+\ln\Theta,$$

$$B(\Theta)=\frac{18\zeta(3)}{\pi^2}-\frac{\pi^2}{3}+\frac{18\zeta(3)}{\pi^2}\ln\Theta,\qquad C(\Theta)=2+2\ln\Theta, \tag{11}$$

where $\zeta(3)$ is the Riemann Zeta function.

Thus, the functions[1)] $A(\Theta)$, $B(\Theta)$ and $C(\Theta)$ vary more slowly with changing Θ than the "relaxation time" $t_s(\Theta)$. The function $Q(z)$ entering into the "relaxation time" (9) for electrons on phonons is tabulated in[9]. Its dependence on z becomes significant when $z \gg 1/4$. For $z < 1/4$ we have $Q(z) \approx Q(0) = 5!\zeta(5)$. It can be seen from (8) that the sign of the thermal emf is determined by the sign of the difference $(\tau_\uparrow - \tau_\downarrow)$ and, consequently, depends on the nature of the impurity.

1. At sufficiently low temperatures, when $t_s \gg \tau_\downarrow$ and $\tau_\uparrow$, the first addend in the denominator of (8) is the most important and the thermal emf has the form

$$\alpha=\frac{\pi^2}{6}\frac{k}{e}\frac{\tau_\uparrow-\tau_\downarrow}{t_s(1+{}^1/_2(\tau_\uparrow+\tau_\downarrow)/t_\Phi)}. \tag{12}$$

If, moreover, $t_{ph} \gg \tau_\uparrow$ and $\tau_\uparrow$, then the thermal emf

$$\alpha=\frac{\pi^2}{6}\frac{k}{e}\frac{\tau_\uparrow-\tau_\downarrow}{t_s} \tag{13}$$

increases with temperature

$$\alpha\sim\begin{cases}\Theta^{-1}e^{-1/\Theta}, & \Theta\ll 1\\ \Theta & \Theta\gg 1\end{cases} \tag{14}$$

and is inversely proportional to the impurity concentration c. We may also obtain formula (13) by solving the system (7) by perturbation theory.

Scattering by phonons weakens the concentration and temperature dependences of the thermal emf.

If $t_{ph} \ll \tau_\uparrow$ and $\tau_\downarrow$, then

$$\alpha=\frac{\pi^2}{3}\frac{k}{e}\frac{\tau_\uparrow-\tau_\downarrow}{\tau_\uparrow+\tau_\downarrow}\frac{t_{ph}}{t_s}. \tag{15}$$

The thermal emf does not depend on the impurity concentration and varies with Θ as

$$\alpha\sim\begin{cases}\Theta^{-4}e^{-1/\Theta}, & \Theta\ll 1\\ \Theta^{-2}Q^{-1}(\Theta/\Theta_D), & \Theta\gg 1\end{cases}. \tag{16}$$

2. At high temperatures, when $t_s \ll \tau_\uparrow$ and $\tau_\downarrow$,

$$\alpha=\frac{\pi^2}{6}\frac{k}{e}\frac{\tau_\uparrow-\tau_\downarrow}{\tau_\uparrow\tau_\downarrow}\left(\frac{B(\Theta)}{t_s}+\frac{C(\Theta)}{t_{ph}}\right)^{-1}, \tag{17}$$

i.e., the thermal emf decreases as Θ increases and is proportional to c. If, at the same time, scattering by magnons predominates over scattering by phonons, $t_s \ll t_{ph}$, then

$$\alpha\sim B^{-1}(\Theta)t_s\sim\begin{cases}\Theta^{-1}e^{1/\Theta}, & \Theta\ll 1\\ \Theta^{-1}, & \Theta\gg 1\end{cases}. \tag{18}$$

For the opposite inequality $t_s \gg t_{ph}$, when scattering by phonons predominates,

$$\alpha\sim C^{-1}(\Theta)t_\Phi\sim\begin{cases}\Theta^{-4}, & \Theta\ll 1\\ \Theta^{-3}Q^{-1}(\Theta/\Theta_D), & \Theta\gg 1\end{cases}. \tag{19}$$

Thus, when electron scattering by magnons predominates over scattering by phonons, the thermal emf as a function of temperature has a maximum at $t_s \approx \sqrt{\tau_\uparrow\tau_\downarrow}$. Scattering by phonons shifts the peak towards lower temperatures. As the concentration of magnetic impurities increase, $\tau_\uparrow$ and $\tau_\downarrow$ decreases, so that the peak shifts towards higher temperatures. The thermal emf at the peak may be comparable with k/e.

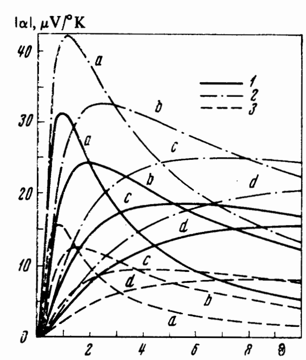

FIG. 1. Thermal emf as a function of the dimensionless temperature $\Theta = T/T_0$ for $\gamma = 0$. For the curves 1, 2 and 3, r is respectively equal to 5, 15 and 2. The curves a, b, c and d correspond to β equal to 0.5, 2, 8 and 20.

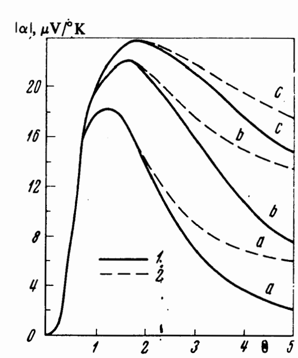

FIG. 2. Thermal emf as a function of the dimensionless temperature $\Theta = T_D/T_0$ for r = 5 and $\beta = 2$. The curves 1 and 2 correspond to $T_D/T_0 = 40$ and 20; a, b and c correspond to γ equal to 0.63, 0.13 and 0.03, respectively.

1)The functions $A(\Theta)$ and $B(\Theta)$, which were introduced by us in [5], are larger by a factor of two than the functions $A(\Theta)$ and $B(\Theta)$ in the present paper.

Figures 1 and 2 show the dependence of $|\alpha|$ on temperature for different values of the parameters $r = \tau_\uparrow/\tau_\downarrow$, $\beta = t_{s0}/\tau_\uparrow$ for $\tau_\uparrow > \tau_\downarrow$ (or $r = \tau_\downarrow/\tau_\uparrow$, $\beta = t_{s0}/\tau_\downarrow$ for $\tau_\uparrow < \tau_\downarrow$) and $\gamma = Q(0)t_{s0}/t_{ph\,0}$. The nature of the curves in Fig. 1 is determined by the values for β. We see that the smaller β is and the larger r is, the higher and sharper the peak of the thermal emf. Since at low temperatures the thermal emf decreases with increase of the impurity concentration while at high temperatures it increases, the curves $\alpha(\Theta)$ intersect each other (for a given r).

The thermal emf, as a function of the concentration at constant temperature, has maximum values at concentrations c_0 given by the relation

$$\tau_\uparrow\tau_\downarrow = t_s/[B(\Theta)t_s^{-1} + C(\Theta)t_{ph}^{-1}]. \tag{20}$$

B. Apart from s-s scattering, s-d scattering by magnons or phonons can be important in ferromagnetic metals. Let us consider the effect of this process on the thermal emf.

Let us, as is customary, suppose that the mobility of the d-electrons is considerably smaller than the mobility of the s-electrons; hence we assume the d-electrons to be in equilibrium. This permits us to describe the scattering of s-electrons into the d-band with the aid of the relaxation times $t_d^{\uparrow,\downarrow}(x)$:

$$\frac{1}{t_d^{\uparrow,\downarrow}(x)} = \frac{1}{t_M^{\uparrow,\downarrow}(x)} + \frac{1}{t_{ph}^{\uparrow,\downarrow}(x)}, \tag{21}$$

where the relaxation times on magnons are given by

$$\frac{1}{t_M^{\uparrow}(x)} = \frac{1}{t_1}\frac{T}{T_1^{\uparrow}}\ln\frac{e^{T_1^{\uparrow}/T}+e^{-x}}{e^{T_1^{\uparrow}/T}-1},$$

$$\frac{1}{t_M^{\downarrow}(x)} = \frac{1}{t_1}\frac{T}{T_1^{\uparrow}}\ln\frac{e^{T_1^{\downarrow}/T}+e^{x}}{e^{T_1^{\downarrow}/T}-1}, \tag{22}$$

while on phonons, the relaxation times are given by

$$\frac{1}{t_{\Phi}^{\uparrow,\downarrow}(x)} = \frac{1}{t_2}\left(\frac{T}{T_2^{\uparrow}}\right)^3 (e^x+1)\int\limits_{T_2^{\uparrow,\downarrow}/T}^{\infty}\frac{d\xi\,\xi^2}{(e^\xi-1)}\left\{\frac{e^\xi}{e^{x+\xi}+1}+\frac{1}{e^{x-\xi}+1}\right\}. \tag{23}$$

The temperatures $T_1^\uparrow$, $T_1^\downarrow$ ($T_2^\uparrow$, $T_2^\downarrow$) are analogous to the temperature T_0 in the case of s-d scattering and are determined by the minimum magnon (phonon) energy capable of transferring an electron from the s- to the d-band at the Fermi surface. The times t_1 and t_2 are constants characterizing respectively the magnitudes of the electron-magnon and electron-phonon interactions in s-d scattering.

The time $t_{ph}^\uparrow(x)$ and $t_{ph}^\downarrow(x)$ are even functions of x. Therefore, s-d scattering by magnons alone, as can be seen from (4), leads to a vanishing thermal emf. In the case, when, apart from this mechanism, s-s scattering by magnons is important, s-d scattering plays a role similar to scattering by magnetic impurities if $T_2^\downarrow \neq T_2^\downarrow$, i.e., $t_{ph}^\uparrow \neq t_{ph}^\downarrow$. Then the thermal emf is given by an expression similar to (8). Since now the effective times $\tau_\uparrow$ and $\tau_\downarrow$ depend on temperatures, the temperature dependence of the thermal emf, generally speaking, differs from the dependence that obtains in the case of magnetic impurities. For s-d scattering by magnons, the sum $u_\uparrow(x) + u_\downarrow(x)$ is not an even function of x if $T_1^\uparrow \neq T_1^\downarrow$. Consequently, in this case, s-d scattering by magnons alone can lead to a large thermal emf[2)].

Furthermore, in conjunction with s-s scattering, s-d scattering by magnons as well as by phonons can play a role similar to magnetic impurities, which also leads to a large thermal emf with a temperature dependence different from the case of magnetic impurities.

2. COMPARISON WITH EXPERIMENT

In the paper by Farrel and Greig[6] (henceforth referred to as FG), results are given of the measurement of the thermal emf of pure Ni and Ni containing various magnetic impurities in the temperature range from liquid helium temperature to 100°K.

Let us now compare our results, obtained in[5] and in the preceding section of the present paper, with the experimental results of FG.

In the first place, it cannot but be noticed that the magnitude of the thermal emf measured by FG, assumes, in agreement with the theory, large values at comparatively low temperatures. For example, in Ni with 1 at.% of Co, the thermal emf at 50°K is equal to 16 μV/°K. The sign of the thermal emf in all the samples with impurities coincides with the sign given by formula (8)[3)].

The theoretical temperature dependence of the thermal emf (see Figs. 1 and 2) is similar to the experimental one, and in both cases the absolute value of the thermal emf, for not too large impurity concentrations, has a maximum in the considered temperature range; up to the peak, the thermal emf increases rapidly with temperature, while after the peak its rate of decrease depends on the impurity concentration—the higher the concentration the slower the rate of decrease. At low temperatures the experimental thermal emf, like the theoretical one, decreases as impurity concentration increases while at high temperatures it increases with concentration, so that the curves $\alpha(T)$ corresponding to different concentrations intersect.

Thus, all the main features of the thermal emf which follow from the theory are in complete qualitative agreement with experiment. For quantitative comparison of the results of the theory with experiment, it is necessary to know the parameters r, β, γ, and T_0.

The values of r obtained by Farrel and Greig for the same samples for which they measured the thermal

---

[2)] Attention was first called to this by Markov [10]. Markov took into consideration s-d as well as s-s scattering. However, he neglected the difference between electrons with spins ↑ and ↓; in this approximation, the contribution of s-s scattering by magnons to the thermal emf is only of the order of $(k/e)(kT/\epsilon_F)$. Moreover, since $T_1\uparrow$ is then equal to $T_1\downarrow$, s-d scattering gives a thermal emf of the same order. Markov obtained a large thermal emf in this situation because he calculated the contribution to the thermal emf connected with only one group of carriers (↑ or ↓); he did not take into account the contribution from the second group which is equal in magnitude but of opposite sign.

[3)] The designations ↑ and ↓ have in our work a meaning opposite to that in FG.

emf, are given in[6,11]. We use the quantities T_0, β and γ as fitting parameters for the experimental $\alpha(T)$ curve for the Ni + 5 at.% Fe sample which, in the FG experiments, had the highest residual resistance. This sample has been chosen by us for the following reasons. First, electron scattering by nonmagnetic impurities and into the d-band, which we did not consider, was least important for this sample. Secondly, scattering by magnons into the s-band can, for small $\tau_\uparrow$ and $\tau_\downarrow$, be considered in a wide range of temperatures with the aid of perturbation theory. Since, as we have already noted, the expression (13) obtained by us in this limiting case for the thermal emf coincides with the result obtained by perturbation theory, the errors in the determination of T_0, β and γ, connected with the approximate character of the method employed in this paper for the solution of the kinetic equation, are reduced.

For the curve 1a in Fig. 3, $\beta = 14$, $\gamma = 0$. The experimental points for the sample with 5 at.% of Fe lie on the theoretical curve for $T_0 = 25$. The theoretical curve 1 constructed for $\beta = 13$, $\gamma = 0.12$ shows the same good agreement with experiment. (The Debye temperature for Ni is $T_D = 410°K$[12].) The value $T_0 = 25°K$ is close to the theoretical estimate which may be obtained by relating Δp to the difference in the population of the sub-bands of the s-electrons with spins $\downarrow$ and $\uparrow$. Such an estimate is the most reliable since no concrete model for the electron spectrum is involved in its derivation. We have

$$T_0 = \frac{(6\pi^2)^{2/3}}{9}\frac{\hbar^2 n_\uparrow^{2/3}}{2\mu}\left(\frac{n_\uparrow - n_\downarrow}{n_\uparrow}\right)^2. \tag{24}$$

For Ni with $\mu = 0.8 \times 10^{-26}$ g[13], $n_\uparrow = 0.3$ electron/atom, $n_\uparrow - n_\downarrow = 0.05$ electron/atom[8], we obtain $T_0 \approx 24°K$. We note that according to[14], $T_0 \approx 22–27°K$ for Fe.

In order to determine β for the samples with 2 and 0.5 at.% Fe impurities, which were investigated by FG, we shall assume β to be proportional to the residual resistance. Then, according to FG, we obtain for the sample with 2 at.% Fe the value $\beta = 5.6$ when $\gamma = 0$; or $\beta = 5.2$ when $\gamma = 0.12$, while for the sample with 0.5 at.% Fe, $\beta = 2.3$ and $\beta = 2.2$ respectively. Like FG we suppose here that the parameter r is the same for all samples, although, judging by the experimental data of FG, r is apparently different in samples with different iron concentrations.

In Fig. 3 the curves 2a and 3a were plotted with $\gamma = 0$, $\beta = 5.6$, and $\beta = 2.3$ while for the curves 2 and 3, $\gamma = 0.12$, $\beta = 5.2$ and $\beta = 2.2$; the light and dark points are experimental points for $T_0 = 25°K$.

Figure 4 shows theoretical and experimental curves for a Ni sample with 0.3 at.% Cu. The parameter r = 3.68. The parameter β was found by recalculation from the residual resistance, $\beta = 2.5$.

The theoretical curves are similar to but lie beneath the experimental curves, i.e., the dependence of the thermal emf on the impurity concentration is weaker than that predicted by the theory. On the theoretical curves, in contrast to the experimental ones, the thermal emf at the peak increases as the impurity concentration decreases. The same is true for Ni samples with other magnetic impurities investigated by FG.

On the whole, considering the approximate character of the calculation and the unreliability of the parameters taken from experiment, we can regard the agreement of the theory with experiment as good.

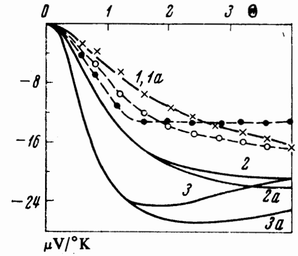

FIG. 3

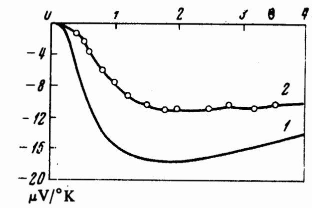

FIG. 4

FIG. 3. Theoretical and experimental thermal emf as a function of $(\Theta) = T/T_0$ for Ni with Fe impurities, r = 7.35. The theoretical curves 1, 2 and 3 correspond to $\beta = 13$, 5.2 and 2.2 respectively and were constructed for $\gamma = 0.12$; for the theoretical curves 1a, 2a and 3a, $\beta = 14$, 5.6 and 2.3 respectively, and $\gamma = 0$. The experimental curves for the samples Ni + 5% Fe (x), Ni + 2% Fe (0) and Ni + 0.5% Fe (O) were plotted with $T_0 = 25°K$.

FIG. 4. Theoretical and experimental thermal emf as a function of $(\Theta) = T/T_0$ for Ni + 0.3 at.% Cu. For the theoretical curve 1, r = 3.68; $\beta = 2.5$ and $\gamma = 0.12$. The experimental curve 2 was plotted with $T_0 = 25°K$.

The good agreement of the theory with experiment, obtained in the present paper, serves as a conclusive confirmation of Fert's view[14] that the main contribution to electron scattering with spin flip is made by magnons.

Let us point out a number of causes to which may be due the disagreement of the theory with experiment.

1. The kinetic equations were solved approximately. The largest error should be expected in the temperature range in which the thermal emf is a maximum.

2. The values for the parameter r taken from[11,6], were determined on the basis of a very crude theory of the electrical conductivity of ferromagnetic metals and, hence, are unreliable. Thus, for the alloys Ni–Fe and Ni–Co, according to[11,6], r = 7.35 and 13.2 respectively while according to[15], r = 20 and 30. Moreover, we have already mentioned that under the conditions of the experiment of FG, the quantity r apparently decreases with the concentration of magnetic impurities, which may be explained by, say, the presence of uncontrollable impurities. If such a decrease of r is taken into account in the construction of the theoretical curves in Fig. 3, then the agreement between theory and experiment improves considerably.

3. Formula (8) gives for pure Ni a vanishing thermal emf to within the small terms which were discarded in the kinetic equation. At the same time, the experimental values for the thermal emf in pure Ni are only 2–3 times smaller than in samples with Fe, Co, and Cu impurities. The relatively large thermal emf in pure Ni may be related to the scattering of electrons into the d-band by phonons or magnons discussed in the preceding section. This same mechanism may lead to a weaker—in comparison with (8)—dependence of the thermal emf on the concentration of the magnetic impurities.

3. THERMAL CONDUCTIVITY

To calculate the thermal conductivity, we must add a term proportional to ∇T to the right hand side of (2). Solving this system by the same method as was used in Sec. 1, we obtain for the thermal conductivity the following expression:

$$\begin{aligned}\varkappa^{-1} = \frac{\rho_0}{LT_0}\frac{1}{k\Theta}\Big\{ 1 &+ \frac{1}{2}\frac{B(\Theta)}{C(\Theta)}\frac{\tau_\uparrow+\tau_\downarrow}{t_s} + \frac{1}{2}\frac{\tau_\uparrow+\tau_\downarrow}{t_\Phi} \\ &+ \frac{\pi^2}{3}\frac{\tau_\uparrow+\tau_\downarrow}{[2t_s+C(\Theta)(\tau_\uparrow+\tau_\downarrow)]C(\Theta)} \\ &+ \frac{1}{4}(\tau_\uparrow-\tau_\downarrow)^2\frac{t_s+D(\Theta)t_\Phi}{[t_s\tau_\uparrow\tau_\downarrow + {}^1/_2 t_s t_\Phi(\tau_\uparrow+\tau_\downarrow)+D(\Theta)\tau_\uparrow\tau_\downarrow t_\Phi]}\Big\} \\ &\times\left\{1-\frac{e\alpha}{k}\frac{\tau_\uparrow-\tau_\downarrow}{2t_s+C(\Theta)(\tau_\uparrow+\tau_\downarrow)}\right\}^{-1}, \end{aligned} \tag{25}$$

where ρ_0 is the residual resistance of the sample $L = \pi^2 k/3e$ is the Lorentz number, and

$$D(\Theta) = \frac{4\pi^2 K_1(\Theta)+K_3(\Theta)}{6K_2(\Theta)} = \begin{cases} \dfrac{1}{6\Theta}, & \Theta \ll 1 \\ 2+3\pi^{-2}\zeta(3)+2\ln\Theta, & \Theta \gg 1. \end{cases} \tag{26}$$

The thermal emf $\alpha(\Theta)$ and the functions $B(\Theta)$ and $C(\Theta)$ are given by the formulas (8) and (9).

The first three terms in the numerator of the right hand side of formula (25) are thermal resistances due, respectively, to scattering by impurities only, by magnons only[4)] and by phonons only. The rest of the terms as well as the denominator lead to deviations from Matthiessen's rule.

Let us emphasize that Matthiessen's rule is also violated in the case when the impurities are nonmagnetic, i.e., $\tau_\uparrow = \tau_\downarrow = \tau$. In that case

$$\varkappa^{-1} = \frac{\rho_0}{LT_0}\frac{1}{k\Theta}\left\{1+\frac{B(\Theta)}{C(\Theta)}\frac{\tau}{t_s}+\frac{\tau}{t_{ph}}+\frac{\pi^2}{3}\frac{\tau}{C(\Theta)[t_s+C(\Theta)\tau]}\right\}. \tag{27}$$

The deviation from Matthiessen's rule is due to the third term on the right hand side of this formula.

The violation of Matthiessen's rule in Ni containing magnetic impurities was observed by Farrel and Greig[17], who gave a formula for the deviation from Matthiessen's rule for thermal resistance in analogy with the deviation for electrical resistance, corresponding to the fourth term in the numerator of formula (25). This would have been correct if the collision integral for electrons with magnons could be represented in the form $(u_\uparrow - u_\downarrow)/t_{\uparrow\downarrow}$. Such an approximation is satisfactory for the calculation of electrical conductivity when the dependence on x of $u_\uparrow$ and $u_\downarrow$ may be neglected, but it is not suitable for the calculation of the thermal emf and thermal conductivity. Therefore, according to[17], the deviation from Matthiessen's rule for thermal conductivity is valid only in the case of magnetic impurities, whereas in fact, as can be seen from (27), Matthiessen's rule is not fulfilled in the case of nonmagnetic impurities as well.

Comparison of the expression (25) with experiment is difficult since the electron part was not separated from the total experimental thermal conductivity in[17].

The authors are grateful to V. P. Lazarenko for his help in the numerical computations.

4)Thermal conductivity due to s-s scattering of electrons by magnons have been calculated in [2,16].

1 J. M. Ziman, Electrons and Phonons, Clarendon Press, Oxford, 1960 (Russian Trans., 1962).
2 T. Kasuya, Progr. Theor. Phys., 22, 227 (1959).
3 L. É. Gurevich and G. M. Nedlin, Zh. Eksp. Teor. Fiz. 45, 576 (1963) (Sov. Phys.-JETP 18, 396 (1964)).
4 M. Bailyn, Adv. Phys., 15, 179 (1966).
5 I. Ya. Korenblit and Yu. P. Lazarenko, ZhETF Pis. Red. 11, 353 (1970) (JETP Lett. 11, 236 (1970)).
6 T. Farrel and D. Greig, J. Phys. C., 3, 138 (1970).
7 L. É. Gurevich and G. M. Nedlin, Zh. Eksp. Teor. Fiz. 37, 765 (1959) [Sov. Phys.-JETP 10, 546 (1960)].
8 J. C. Phillips, Phys. Rev., 133A, 1020 (1964).
9 E. H. Sondheimer, Proc. Roy. Soc., A203, 75 (1950).
10 P. A. Markov, FMM (Sov. Journal-Physics of Metals and Metallography) 25, 1043 (1968).
11 T. Farrel and D. Greig, J. Phys. C., 1, 1359 (1968).
12 M. A. Krivoglaz, Teoriya rasseyaniya rentgenovskikh lucheĭ i teplovykh neĭtronov real'nymi kristalami (The Theory of X-ray and Thermal Neutron Scattering by Real Crystals), Nauka, 1967.
13 V. J. Minkievicz, M. F. Collins, R. Nathans, and G. Shirane, Phys. Rev., 182, 624 (1969).
14 A. Fert, J. Phys. C., 2, 1784 (1969).
15 A. Fert, I. A. Campbell, Phys. Rev. Lett., 21, 1190 (1968). A. Fert, I. A. Campbell, and M. Ribault, J. Appl. Phys., 41, 1428 (1970).
16 Sh. Sh. Abel'skiĭ and E. A. Turov, FMM (Sov. Journal-Phys. of Metals and Metallography) 10, 801 (1960).
17 T. Farrel and D. Greig, J. Phys. C., 2, 1465 (1969).

Translated by A. K. Agyei
169