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Thermoelectric Power of a Two-Band Ferromagnet*

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Recently it has been proposed that "magnon drag" is responsible for the very large peak in the thermopower of iron near 200 K. We report here the results of calculations which, while they do not discount the possibility of magnon-drag contributions, do indicate that the peak could be accounted for empirically by the transport process occurring in two overlapping bands under the influence of a very large internal magnetic field. It is speculated that the spin-orbit interaction may be the real source of the large peak in the thermoelectric power.

Recently it has been proposed that "magnon drag" is responsible for the very large peak in the thermoelectric power of iron near 200 K. We report here the results of calculations which, while they do not discount the possibility of magnon-drag contributions, do indicate that the peak could be accounted for empirically by the transport process occurring in two overlapping bands under the influence of a very large internal magnetic field simulating the effect of the spin-orbit interaction on the thermoelectric power.

Bailyn[1] suggested that the absolute thermoelectric power or the Seebeck coefficient S of ferromagnetic metals may exhibit a magnon-drag effect similar to the phonon-drag effect[2] found in pure elements and in dilute alloys. These drag effects are assumed to be responsible for large deviations from the expected linear temperature dependence of S due to the "diffusion" term,[3]

$$S_{\text{diff}} = -\left(\frac{\pi^2 k^2 T}{3|e|}\right)\left(\frac{\partial \ln\sigma(E)}{\partial E}\right)_{\text{Fermi level}}, \qquad (1)$$

with the temperature dependence of the term in the second large parentheses assumed to be small.

The general temperature dependence of the phonon drag and the proposed magnon-drag contributions are rather similar.[1] However, Blatt *et al.*[4] argued that the very large peak in S of iron is due to a magnon-drag and not a phonon-drag term, since it is found also in dilute alloys and is not removed by cold work, as phonon-drag contributions are. The first argument may be questionable. If phonon-drag and magnon-drag effects are similar, one would expect that impurities would affect these contributions similarly. Furthermore, Farrell and Grieg[5] have shown that changes in the phonon-drag contributions due to alloying can be accounted for using the concept of "spin mixing" and a two-band description of the transport process. Thus it seems reasonable that the peak in S of iron may be partly due to the transport process occurring in several energy bands.

The nature of transport processes in ferromagnets with their so-called "spontaneous" components in a magnetic field has led us to propose that the diffusion thermoelectric power is also augmented by the presence of anisotropic electron scattering resulting from the spin-orbit interaction.

The spin-orbit interaction is generally believed to account for the large electric Hall field E_H that occurs when the applied field H is strong enough to align all the magnetic domains parallel with one another. The observed Hall field is very much larger than that necessary to oppose the sideways deflection of the electrons due to the Lorentz force. The additional Hall field strength is needed to oppose the transverse current arising from the anisotropic electron scattering induced by the spin-orbit interaction. This anisotropic scattering may be simulated by a magnetic field, H_{so}, which acts on the electrons like an applied field. The "effective" magnetic field, $H_{\text{eff}} = H_{\text{applied}} + H_{so} \gg H_{\text{applied}}$, thereby gives a much larger Lorentz force and Hall field, $\vec{E}_H = R\vec{H}_{\text{eff}} \times \vec{j}$, where R is the Hall coefficient and $\vec{j}$ is the external current, as is observed in ferromagnets.

The effect of the spin-orbit interaction in the diffusion thermoelectric power may also be simulated by a large effective magnetic field interacting with the diffusing electrons via the Lorentz force. This empirical simulation seems justified as the more rigorous calculation of the "skew" scattering contribution to the Hall effect by Fivaz[6] has the same structure as the Boltzmann equation in a magnetic field.

To quantify this simulation of the proposed effect of the spin-orbit interaction and the possibility of the transport process occurring in more than one band, we have used the equations developed by Sondheimer and Wilson[7] for the thermoelectric power in a magnetic field. They originally set up simple interpolation formulas for the thermoelectric power and the adiabatic Ettingshausen-Nernst coefficient for a single band. The equations made it possible to obtain the generalized formulas for the discussion of the two-band model. The equations seem reasonable since they reduce to the exact expressions in the limiting cases of low and high temperatures and strong magnetic fields.

The effect of a magnetic field on the thermoelectric power of a single band of charge carriers is very small.[7] The effect on the thermoelectric power of a two-band model was found by Sondheimer[7] to be qualitatively the following: As the field increases, the thermoelectric power decreases in absolute value, eventually changing sign and saturating at a value $S(H=\infty)=-(L_n/L_o)\,S(H=0)$, which is generally larger than $S(H=0)$ in absolute value. L_n is the Sommerfeld value of the Lorenz number and $L_o=\kappa_o/\sigma_o T$ is the observed zero-field Lorenz number. The change in the thermoelectric power is small at low and high temperatures and passes through a maximum at intermediate temperatures. The temperature at which the maximum change occurs is proportional to H. For ordinary (nonferromagnetic) metals and laboratory fields this maximum should occur at 10–50 K. However, if we include the simulation of the effect of spin-orbit interaction-induced scattering, by means of a large internal effective field the temperature and magnitude of the maximum change are correspondingly higher.

Sondheimer's equation (22) gives the thermoelectric power S of a metal, with overlapping bands, in a magnetic field B and at temperature T, functionally as

$$S=S(S_s, S_d, B^a_{\mathrm{EN}s}, B^a_{\mathrm{EN}d}, \sigma_{0s}, \sigma_{0d}, \kappa_{0s}, \kappa_{0d}, n_s, n_d, B, T)\ . \tag{2}$$

The arguments are, respectively, the thermoelectric powers, adiabatic Ettingshausen-Nernst coefficients, electrical conductivities, thermal conductivities, and the number of electrons in the s (band with positive curvature) and d (band with negative curvature) bands. The subscript "0" indicates $B=0$ conditions.

Sondheimer states that in the s or d band, the variation of S_s, S_d, $B^a_{\mathrm{EN}s}$, and $B^a_{\mathrm{EN}d}$ with the magnetic field can be neglected compared with the effects due to the existence of overlapping bands. Therefore, we substitute S_{0s} for S_s and S_{0d} for S_d in Eq. (2). Our calculations have shown that the terms containing $B^a_{\mathrm{EN}s}$ and $B^a_{\mathrm{EN}d}$ make an exceedingly small contribution to S. We used the values for B_{EN} for copper as representative of the s band of iron and found negligible effects for all values of the ratio $B_{\mathrm{EN}d}/B_{\mathrm{EN}s}$ in the range $0.01\le B_{\mathrm{EN}d}/B_{\mathrm{EN}s}\le 100$ that we tried. In what follows, terms containing these coefficients were omitted. The remainder of the equation is parameterized as follows: $n_s=Zn$, $n_d=(1-Z)n$; $\sigma_{0d}=X\sigma_0$; $\sigma_{0s}=(1-X)\sigma_0$; $\kappa_{0d}=Y\kappa_0$; $\kappa_{0s}=(1-Y)\kappa_0$; $S_{0d}=-LS_{0s}$. Under zero-field conditions, Eq. (2) reduces to the usual formula for two bands:

$$S_0=(\sigma_{0s}S_{0s}+\sigma_{0d}S_{0d})/(\sigma_{0s}+\sigma_{0d})=S_{0s}(1-X(1+L))\ . \tag{3}$$

This result is used for S_{0s} in Eq. (2) and one obtains a parametrized equation relating the thermoelectric power in the magnetic field to that in zero field, which is of the form

$$S(B,T)=S_0(T)F(X,Y,Z,L,\sigma_0,\kappa_0,R,B,T)\ , \tag{4}$$

where R is the Hall coefficient. Sondheimer[7] has discussed qualitatively the case where $X=Y=Z=\frac{1}{2}$, as noted earlier.

We have used a computer[8] to evaluate Sondheimer's equation for a wide range of parameters involved at 50-K temperature intervals from 0 K to the Curie temperature. The observed values of σ and κ for iron[9,10] were used for σ_0 and κ_0. For a value of X, the fraction of the electrical current transported by the d carriers (holes), we have used the estimates of Markov[11] and Goodings[12] who have studied the effect of s-d transitions on the thermoelectric power. Their estimate of the ratio of the currents was $j_s/j_d=(v_s/v_d)(k_s/k_d)^2=(14.9)(1.22)^2=(1-X)/X$, where v_s, v_d, k_s, and k_d are the Fermi velocities and the wave vectors of the carriers in the s or d band, respectively. This gives $X=0.045$. Usually we set Y, the fraction of the thermal current carried by the d carriers, equal to X, implying that the Lorenz numbers of the s and d electrons are equal, and equal to the observed Lorenz number of iron. The saturation magnetization ($2.22\mu_B$ per atom) gives an estimate[3] of n_s/n_d as $n_s/n_d=(2.22-2.00)/2.22=Z/(1-Z)$, giving $Z=0.091$. For more mobile d electrons, a larger value of Z would perhaps be more appropriate, and results with $Z=0.3$ are also presented. The ratio $(-L)$ of the thermoelectric powers of the d and s carriers is expected to be large, as $S_d\simeq K(E_{0d}-E_F)^{-1}\gg S_s\simeq K(E_F-E_{0s})^{-1}$, where E_{0d} or E_{0s} is the energy of the d or s band edge, respectively. The parameter L and the zero-temperature value of the effective internal field, B, were usually left as free parameters in attempting to obtain agreement in the least-squares fit between the calculated $S(B,T)$ and the

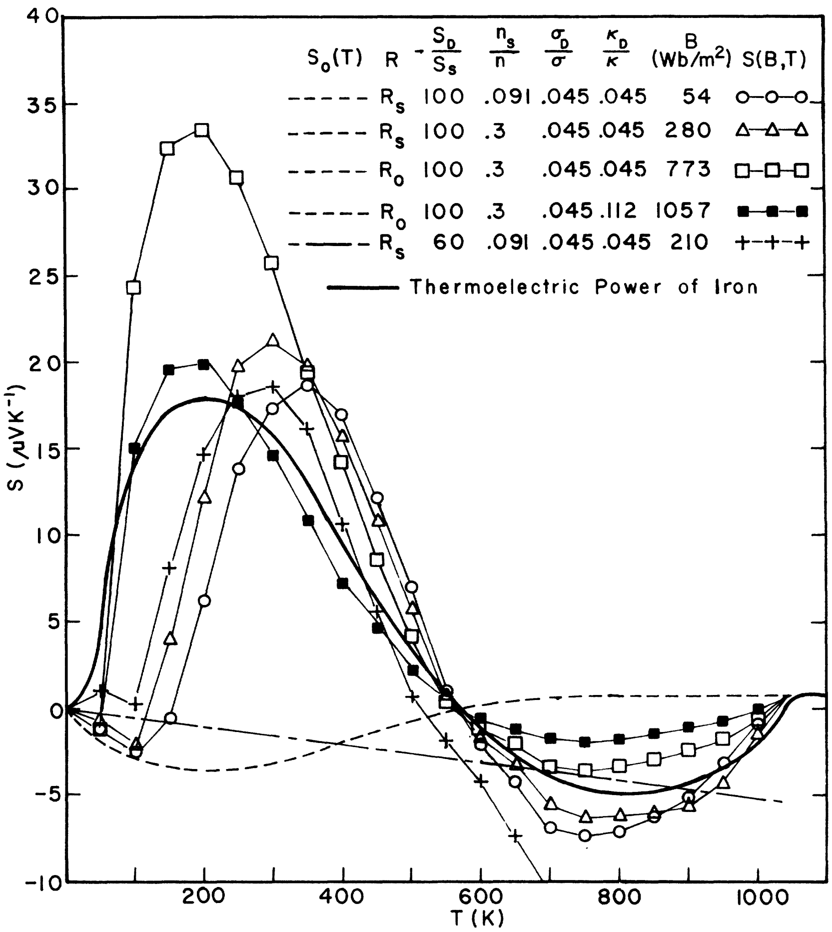

FIG. 1. Thermoelectric power of a two-band model for iron with large internal magnetic fields. $S_0(T)$ is the assumed thermoelectric power of nonmagnetic iron. R is the Hall coefficient, spontaneous (R_s), or ordinary (R_0). S_d/S_s is the ratio of the thermoelectric power of the s and d electrons, respectively. n_s/n is the ratio of the number of s electrons to the total number of electrons. σ_d/σ and κ_d/κ are the ratios of the electrical and thermal conductivities of the d electrons to the total conductivities, respectively. B is the magnetic field, and $S(B, T)$ is the calculated thermoelectric power that one obtains using Sondheimer's two-band formula and the values of the parameters listed.

observed S of iron. For simplicity, the temperature dependence of the internal field was assumed to be $1-(T/T_c)^2$. Two assumed temperature dependences of the thermoelectric power of hypothetical nonmagnetic iron, $S_0(T)$, were used. The first (the short-dashed curve in Fig. 1) represents an extrapolation from the high temperature S of nonmagnetic bcc iron (approximately $+1\ \mu\mathrm{V K}^{-1}$ in both the nonmagnetic α and δ phases[13]). This extrapolation was forced to pass through zero at $T = 575$ K, since $S(B, T) \propto S_0(T)$ and the observed S of iron goes through zero at approximately this temperature. In this two-band model such points are fixed points and the thermoelectric power there is unchanged by magnetic fields. The extrapolation below 575 K was made relative to that above 575 K to reflect the relative magnitude of the maximum and minimum in S of iron at 200 and 800 K, respectively. This estimate for $S_0(T)$ obtains an $S(B, T)$ (see Fig. 1) in qualitative agreement with S of iron. A second, less arbitrary temperature dependence was chosen to be $S_0(T) = -0.05T\ \mu\mathrm{V K}^{-1}$. This agrees with the negative linear contribution to S that was found to be needed by Blatt *et al.*[4] to fit the low-temperature data to a $T^{3/2}$ magnon-drag contribution to S. In Fig. 1 it can be seen that both estimates give calculated $S(B, T)$ curves that exhibit a large peak in the same temperature range where the peak in S of iron occurs. Selected results of the calculations are shown in Fig. 1. To obtain best agreement between the observed S of iron and $S(B, T)$ (in the least squares sense) unreasonably large values of L are required. Therefore, L was fixed at the value of 100 in most of the results shown in Fig. 1. With L and the other parameters fixed, B was varied to obtain the best fit. These values of B are listed in Fig. 1. Both the extraordinary (R_s) and the ordinary (R_o) Hall coefficients were used in the calculations, with only small qualitative changes in the results. Generally, the quantity that needs to be large is $(\sigma R B)^2$. This indicates why larger fields are needed when R_o is used instead of R_s. R_s would seem appropriate for zero external fields, as is assumed in these calculations. Values of B of the order of 100–1000 $\mathrm{Wb/m^2}$ are needed to obtain the large change in S_o of hypothetical nonmagnetic iron. These fields are of the same order of magnitude as required to simulate the effect of spin-orbit interaction induced scattering on the Hall electric field in iron. From this empirical simulation, it would seem that the large peak in the thermoelectric power may arise from the spin-orbit interaction, as does the "spontaneous" Hall field.

At higher temperatures, agreement between the calculated $S(B, T)$ and S of iron is not expected to be too good, as critical fluctuations in the magnetization or shifts in the Fermi energies of the two bands[14] affect the thermoelectric power as the temperature approaches the Curie temperature.

At low temperatures, $S(B, T)$ is not affected very much by the field until the temperature is greater than approximately 100 K. Markov[11] and Goodings[12] have found that s-d transitions contribute to the thermoelectric power at low temperatures, but this contribution is small at the temperature of the peak in S of iron. Similarly phonon-drag and magnon-drag contributions are to be expected at low temperatures.

More rigorous investigations of the effect of the spin-orbit interaction on thermoelectromagnetic transport processes are underway.

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Magneto-Elastic Dependence of the Propagation of Sound in Gadolinium at the Critical Point*

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The change in the attenuation of sound and in the elastic constant c_{33} of gadolinium has been measured at 5 MHz as a function of applied magnetic fields to 13 kOe in the temperature range 270–320 °K. The minimum in the elastic constant at the Curie point moved upwards in temperature and tended to diminish and broaden with increasing field. Studies of the change in attenuation due to magnetic field in the same region revealed sharp λ-shaped increases up to a field of 1.8 kOe. At higher fields this peak split into two diminishing maxima, one moving upwards and the other downwards with increasing field. The zero-field-attenuation data decreased as $\Delta\alpha \sim \epsilon^y$, where $\epsilon = |(T - T_c)/T_c|$ and $y = -1.8 \pm 0.2$ in the paramagnetic region. In the presence of a magnetic field, the changes in the velocity are attributed to changes in the spin-correlation function.

INTRODUCTION

Anomalies in the sound attenuation and velocity at second-order phase transitions have been observed in many ferromagnetic materials. Near the Curie point, λ-shaped increases in the specific heat are normal and are accompanied by similarly shaped increases in the attenuation of sound as well as sharply peaked decreases in the sound velocity. These phenomena have been reported in nickel,[1–3] iron,[4] and in most of the rare earths including gadolinium.[5] The characteristic λ shape which the specific-heat and attenuation-vs-temperature curves assume is common to most ferromagnetic materials although there are some exceptions (e.g., EuO[6,7]). The specific heat of gadolinium exhibits the typical