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Magnetic field effects in the Anderson model of dilute magnetic alloys. III. Transport properties

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An extension of previous studies is made on the magnetic field effects of the infinite-U Anderson Hamiltonian. The magnetothermopower, the thermal magnetoresistivity, and the Lorenz number are calculated. We find that the thermopower decreases monotonically as a function of the applied field. In low fields, it varies linearly with the square of the impurity magnetization as well as with the negative electrical magnetoresistivity. In high fields, it follows a $1/H - 1/H_0$ behavior. These features are in good qualitative agreement with the experiment of Berman and Kopp on Au-Fe alloys and with calculations based on the s-d model. The field and temperature dependence of the negative thermal magnetoresistivity closely resemble those of the electrical one. A close parallelism is therefore established between the two quantities. The Lorenz number is found to be practically independent of the external magnetic field.

I. INTRODUCTION

Recently, the author undertook the study of the effect of an external magnetic field on the Anderson model of dilute magnetic alloys. These have been reported in two previous papers,[1] hereafter referred to as I and II. In these papers, we have treated the field-dependent infinite-U Anderson Hamiltonian self-consistently by means of the double-time Green's-function method. Then, using the derived analytic solutions, we made a numerical study on the magnetic field effects of the local moments, the impurity magnetization, the electrical magnetoresistivity, and the Hall coefficient. The results obtained show remarkable agreement with experiments as well as other theoretical calculations based on the s-d model.

The present paper extends previous investigations to the magnetothermopower, the thermal magnetoresistivity, and the Lorenz number which are the transport properties of interest apart from the electrical magnetoresistivity in the dilute magnetic alloy system of Kondo[2] type. This is a somewhat more tedious calculation than the previous ones since it involves the transport integrals of higher moment K_n with $n>0$. The range of the outer integration in the twofold integral becomes extended. Consequently, much effort has been expended in acquiring the appropriate subdivisions in order to secure a smooth and efficient computation.

Experimental observations on the magnetic-field dependence of the thermopower have been made by Huntley and Walker,[3] and Berman and Kopp.[4] However, their low-field data are complicated by the presence of the internal fields due to interaction effects between the impurities. Only the most dilute samples (0.5 ppm of Ref. 3 and 10 ppm of Ref. 4) exhibit the dominant influence of the single-impurity effect. Theoretical studies on the magnetothermopower have been made by Weiner and Beal-Monod,[5] and Kozarzewski,[6] both based on the s-d model. The approach of the former is perturbative and the latter is exact but seminumerical, using the S-matrix theory.

As regards the magnetic field effects of the thermal resistivity and the Lorenz number, little work has been reported lately. Some of the old experimental data are collected in the review article of Van den Berg.[7] The data on the dilute Ag-Mn alloys indicate that the external magnetic field produces little effect on the Lorenz parameter of the alloy.

The outline of the paper is as follows. In Sec. II, the transport coefficients are formulated in terms of the t-matrix so that numerical computations can be performed. In Sec. III, the computed results are analyzed and compared with experiments as well as other theoretical calculations. A brief summary of the results and conclusions is made in Sec. IV.

II. TRANSPORT COEFFICIENTS

For a magnetic alloy, the influence of a magnetic field on the transport properties is exhibited in the spin-dependent scattering of conduction electron off the impurities. In the limit of dilute concentration, it is assumed that the usual Boltzmann transport theory applies in the alloy system. In terms of the usual relaxation time approximation, the Boltzmann equation can be solved with the assumption of a spherical Fermi surface. The solution gives rise to the following transport integrals[8]

$$K_n = -\int_{-\infty}^{\infty} dw\, \rho(w) v^2(w) w^n \sum_{\sigma} \tau_{\sigma}(w - \sigma H) \frac{\partial f(w)}{\partial w}, \tag{2.1}$$

where $\rho(w)$ denotes the density of states of the host metal, $v(w)$ the electronic velocity, τ_σ the relaxation time for an electron of spin σ, and $f(w)$ is the usual Fermi-distribution function. Here, the magnetic field has been fixed to be parallel to the z axis and we consider only the longitudinal effects. In terms of the integrals K_n, the longitudinal electrical resistivity ρ_L, and the thermoelectric power S are given by

$$\rho_L(H, T) = 3/e^2 K_0, \tag{2.2}$$

$$S(H, T) = K_1/eTK_0, \tag{2.3}$$

where the Fermi level is taken as the energy zero. Similarly, the thermal resistivity W and the Lorenz number L yield

$$W(H, T) = 3TK_0/(K_2K_0 - K_1^2), \tag{2.4}$$

$$L(H, T) = \rho_L/WT = (K_2K_0 - K_1^2)/(eTK_0)^2. \tag{2.5}$$

The relaxation time appearing in the integrals K_n is given by the imaginary part of the scattering amplitude

$$\tau_\sigma^{-1}(w) = \mp \frac{2\Delta C_i}{\pi\rho_0} \mathrm{Im} t^\sigma(w \pm i\delta), \tag{2.6}$$

where C_i denotes the impurity concentration and t^σ is related to ψ^σ as

$$t^\sigma(w \pm i\delta) = \mp (i/2\Delta)[1 - \psi^\sigma(w \pm i\delta)]. \tag{2.7}$$

In Paper I we derived and solved the singular integral equation for t^σ. The solutions are summarized in Paper II [Eqs. (II 1.2) to (II 1.23)] for the purpose of numerical computation. For transport properties at low T_K, $w \ll D$, we can approximate $v^2(w) \approx v_F^2$. The density of states $\rho(w)$ is given by ρ_0 for a square band. As a result, the transport integrals K_n in (2.1) can be simplified to yield

$$K_n = (3k^n/e^2\rho_L^0)K_n', \tag{2.8}$$

where

$$K_n' = -\int_{-\infty}^{\infty} dw\, w^n \frac{\partial f(w)}{\partial w} \{[1 - \mathrm{Re}\psi^\dagger(w - H + i\delta)]^{-1} + [1 - \mathrm{Re}\psi^\dagger(w + H - i\delta)]^{-1}\} \tag{2.9}$$

and

$$\rho_L^0 = 2m^* C_i/\mathrm{Ne}^2\pi\rho_0. \tag{2.10}$$

We note that the energy averaging of τ_σ has been preserved in (2.9). As discussed in Paper II, the energy averaging of the lifetimes is not small and it is not too good an approximation to discard it even in the evaluation of the electrical resistivity and the Hall coefficient which involve only the zero moment K_0. For other transport coefficients which involve higher moments K_n with $n > 0$, it becomes essential to retain such an averaging effect.

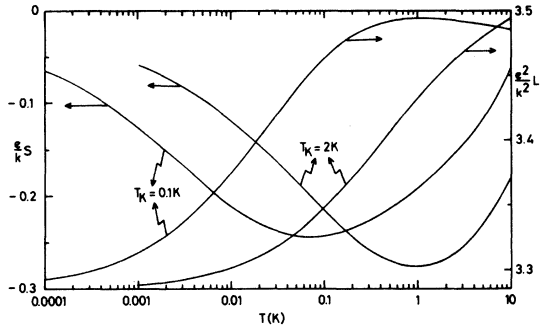

FIG. 1. Temperature variations of the thermopower S and the Lorenz number L at zero field for two values of T_K. The vertical scales are in reduced units.

Using the general expressions (2.3)–(2.9) and the equations [(II 1.2)–(II 1.23)] of Paper II, we have calculated the magnetothermopower, the thermal magnetoresistivity and the Lorenz number by means of numerical computation. The computing technique has been described in Paper II. Readers are referred to that paper for the details. In order to facilitate the comparisons with the results acquired previously, we adopt the same parameters $D = 2\times10^4$ K, $\epsilon_d = -0.05D$ and $T_K = 2$ K in the present computation. However, an additional value of T_K i.e. $T_K = 0.1$ K is also employed in the zero field calculation. The final results are made certain to be accurate to better than 1%.

III. RESULTS AND DISCUSSIONS

A. Zero-field results

First, the transport properties are analyzed in zero field. The temperature variations of the thermopower and the Lorenz number have been computed numerically for five decades of temperatures from 10^{-4} to 10 K. Two values of Kondo temperatures are used, namely, $T_K = 2$ and 0.1 K. The outcomes are displayed in Fig. 1. The result on the thermopower indicates a giant dip of S at a temperature slightly below T_K. In the temperature range between $0.1T_K$ and $0.01T_K$ below T_K, the temperature dependence of S can be approximated by lnT. These features are in good qualitative agreement with various experimental observations.[4,9,10]

As for the Lorenz number, the variation is rather rapid at low temperatures. However, a well-defined broad peak is obtained at a temperature higher than T_K. This agrees with the theoretical calculations of Suhl and Wong,[11] Murata and Wilkin,[12] and Nam and Fullenbaum,[13] all based on the s-d model. The recent experiment of Sharma and Chari[14] on the dilute Cu-Cr alloys confirmed that the electronic Lorenz number increases with the

decrease of temperatures. A low-temperature maximum was conjectured to occur in the vicinity of T_K, in complete accord with the theoretical predictions.

It should be noted also that the positions of the minimum and maximum of S and L in Fig. 1 depend sensitively on the choice of the parameter ϵ_d. As illustrated in an earlier communication,[15] these positions can be made very close to T_K by shifting ϵ_d towards the bottom of the conduction band.

B. Magnetothermopower

Figure 2 shows the results for the magnetothermopower in the form of $S(H, T)/S(0, T)$ as a function of H for various values of T around T_K which is fixed at 2 K. The thermopower decreases monotonically with the increase of magnetic fields even at temperatures above T_K. The lower-temperature curves show a sharp decline initially and then level off at high fields, tending asymptotically to zero. Also in Fig. 2, we have displayed an independent plot of the data of Berman and Kopp[4] for the most dilute Au-Fe alloy at 0.44 K. We then compare the experimental curve with our theoretical one at 0.4 K. The agreement, though qualitative, is surprisingly remarkable indeed, considering that no fitting has been attempted between the theory and the experiment. Of course, the close agreement may be fortuitous.

We have in Paper II interpreted the magnetoresistivity phenomena in terms of two competing mechanisms: the spin-flip scattering process and the "freezing out" mechanism. The argument in fact applies generally to all transport properties. For thermopower, the spin-flip scattering process

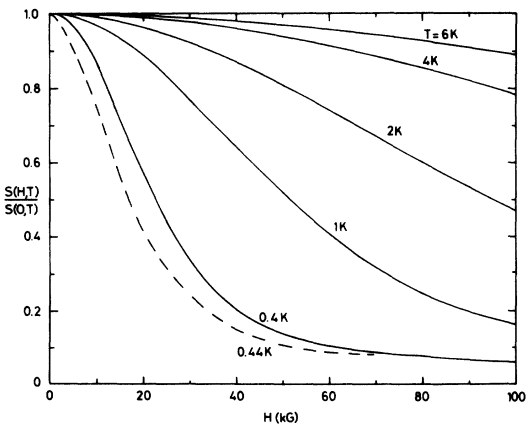

FIG. 2. Relative magnetothermopower $S(H, T)/S(0, T)$, as a function of external magnetic field for a number of temperatures. The solid lines represent our calculations and the single-dashed line is the experimental curve of Berman and Kopp (Ref. 4) for a 10-ppm Au-Fe alloy.

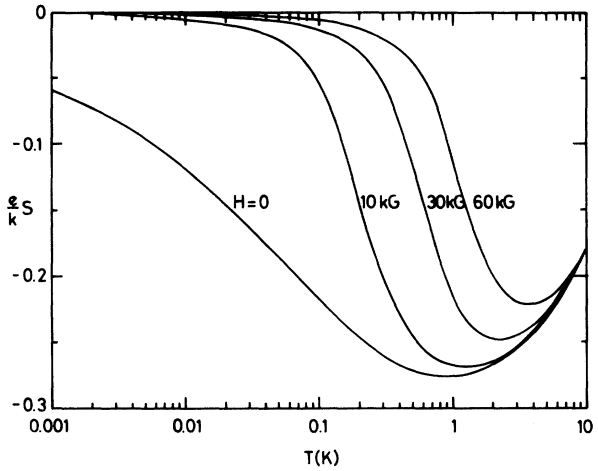

FIG. 3. Magnetothermopower S, in reduced units, as a function of temperature for a number of external magnetic fields.

is responsible for the giant minimum exhibited in Fig. 1 as well as the initial decay of the low-field magnetothermopower. When the value of H/T gets sufficiently large, the "freezing out" mechanism becomes dominant and the spin-flip scattering process becomes inhibited. This results in the leveling off of the thermopower to yield an asymptotic tail in the high-field region.

Experimental results on more concentrated Au-Fe alloys[3,4] indicate an initial rise in the relative thermopower, yielding a low-field maximum in the $S(H, T)/S(0, T)$ curves. This has been interpreted by Huntley and Walker[3] in terms of internal fields due to interactions between the impurities. This phenomena is however, beyond the scope of present investigations in which only the single impurity effect is studied. The one impurity phenomena is exhibited in the most dilute samples measured (0.5 ppm of Ref. 3 and 10 ppm of Ref. 4). There, the interaction effects are negligible and the relative thermopower decreases monotonically with increasing field, in excellent qualitative agreement with our theory as illustrated in Fig. 2.

In this connection, we wish to comment on the work of Kozarzewski[6] who has calculated the magnetothermopower using s-d model and compared with the 300-ppm sample of Berman and Kopp.[4] We feel that the comparison he made (see Fig. 2 of Ref. 6) is rather inappropriate as his calculation is based on one impurity theory which should be compared with the most dilute 10-ppm sample instead of the 300-ppm one. The increase of the relative thermopower in the 300-ppm sample is likely a consequence of the impurity interaction effect which is not included in his one impurity theory.

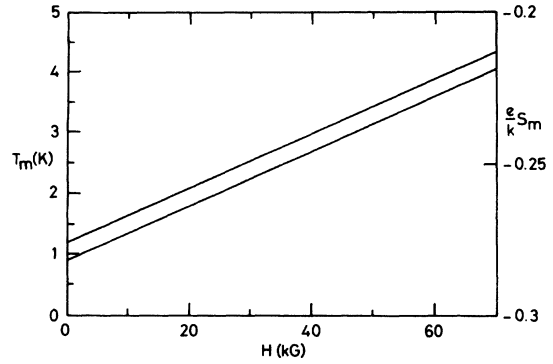

FIG. 4. Field dependence of T_m (the lower curve) and S_m (the upper curve), the latter is in reduced units.

It is of interest to study the influence of magnetic field on the low-temperature minimum of the thermopower. This is shown in Fig. 3 in which the thermopower is plotted as a function of temperature for a number of applied fields. The thermopower, taking its absolute value, decreases rapidly in the presence of the field, especially so at low temperatures. The application of the field not only reduces the value of the minimum S_m, but also shifts the temperature of the minimum T_m to higher temperature. The variation of S_m as well as T_m with H are shown in Fig. 4. Linear relations are obtained for both, yielding $S_m \propto H$ and $T_m \propto H$.

As mentioned earlier, the reduction of the thermopower is a consequence of the gradual "freezing out" of the local magnetic moment. The same mechanism has been held responsible for the depreciation of the electrical magnetoresistivity and the increase of the Hall coefficient. Naturally,

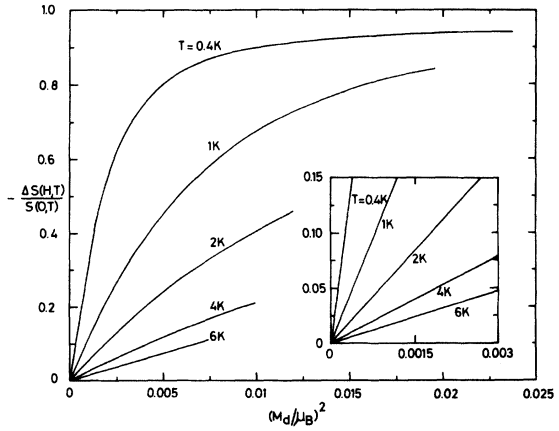

FIG. 5. Negative magnetothermopower, $-\Delta S(H,T)/S(0,T)$, vs the square of the impurity magnetization, $(M_d/\mu_B)^2$, for a number of temperatures around T_K. The low-field curves are enlarged and shown in the insert.

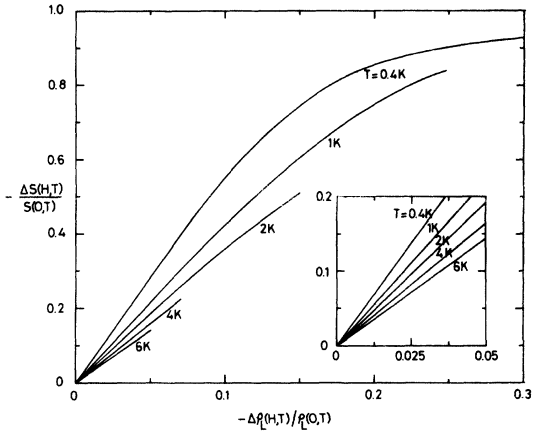

FIG. 6. Relation between the negative magnetothermopower, $-\Delta S(H,T)/S(0,T)$ and the negative electrical magnetoresistivity, $-\Delta\rho_L(H,T)/\rho_L(0,T)$, for a number of temperatures around T_K. The low-field results are enlarged and shown in the insert.

all these quantities are interrelated. The variation of the thermopower with the square of the d magnetization is shown in Fig. 5. In the low-field region, i.e., $H/T<2$, the variation is linear with slopes strongly temperature dependent. The same proportionality to M_d^2 was observed in the low-field behavior of the negative electrical magnetoresistivity and the Hall coefficient. With increasing magnetic field, the curves give rise to large curvatures and tend to saturate at high values of H/T. The relation between the thermopower and the electrical magnetoresistivity is shown in Fig. 6. Linear variations are obtained for $H/T<2$ and the slopes are weakly temperature dependent. The high-field curves tend to saturate just as the case in Fig. 5. The low-field results so acquired in both Fig. 5 and 6 confirm the perturbation calculation of Weiner and Béal-Monod[5] that in the region $H/T<2$,

$$-\Delta S(H,T)/S(0,T) \propto -\Delta\rho_L(H,T)/\rho_L(0,T) \propto M_d^2, \tag{3.1}$$

where $\Delta S(H,T)=S(H,T)-S(0,T)$.

The behavior of the thermopower in the high-field region is also investigated. In Figs. 7 and 8, the thermopower is plotted as a function of $1/H$ for various values of temperatures. The variation is nonlinear in the low-(H/T) region. However, when the values of H/T become sufficiently large, perfect straight lines are obtained. This is clearly illustrated in Fig. 8. By extrapolating the lines to the horizontal axis, we obtain a common intercept, H_0^{-1}, which is finite and of the order of 0.0014 kG^{-1}. This leads to the relation

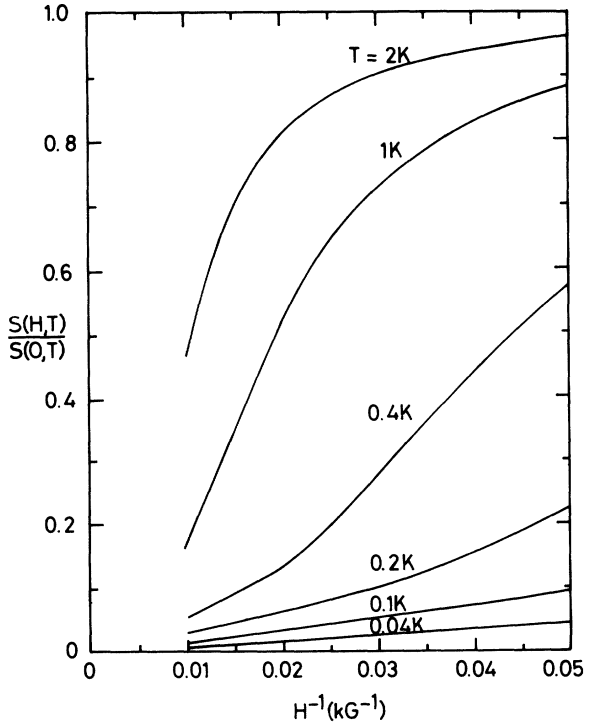

FIG. 7. Relative magnetothermopower, $S(H,T)/S(0,T)$, plotted against $1/H$ for a number of temperatures.

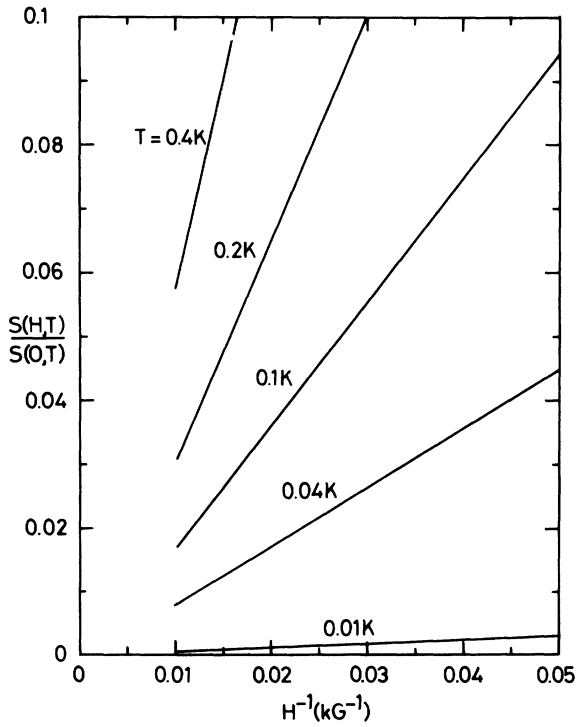

FIG. 8. High-field behavior of the relative magnetothermopower as a function of $1/H$.

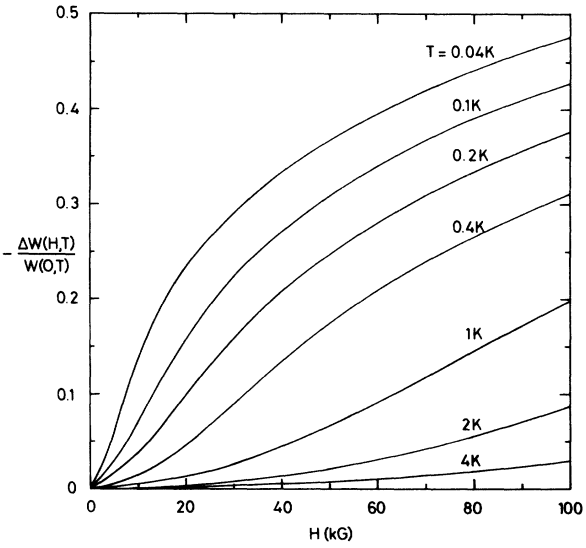

FIG. 9. Negative thermal magnetoresistivity, $-\Delta W(H,T)/W(0,T)$, as a function of external magnetic field for a number of temperatures.

$$S(H,T)/S(0,T) \propto (1/H - 1/H_0), \tag{3.2}$$

for the thermopower in the high-field region. Such a high-field behavior of S has been observed experimentally on Au-Fe alloys (cf Fig. 6 of Ref. 4). Our result thus, confirms the perturbation calculation of Weiner and Béal-Monod[5] on the $1/H$ law of S at high fields. However, their prediction that all lines of the $S(H,T)/S(0,T)$-vs-$1/H$ curve should pass through the origin is not observed by us, neither is it detected by experiment.[4]

C. Thermal magnetoresistivity and Lorenz number

The thermal resistivity due to the scattering of the electrons off the impurities has similar field and temperature anomalies as in the electrical resistivity. Accordingly, the negative thermal magnetoresistivity is defined as

$$-\frac{\Delta W(H,T)}{W(0,T)} = \frac{W(0,T) - W(H,T)}{W(0,T)}, \tag{3.3}$$

the field dependence of which is shown in Fig. 9 for a number of temperatures. The same results are plotted in Fig. 10 as a function of temperature for a number of external magnetic fields. The field and temperature dependence of the negative thermal magnetoresistivity closely resemble those of the electrical one if one compares Fig. 9 with the corresponding Fig. 9 of Paper II. The similarity between the two implies that the resistivities are dominated by the field and temperature dependence of the imaginary part of the transition matrix t^o.

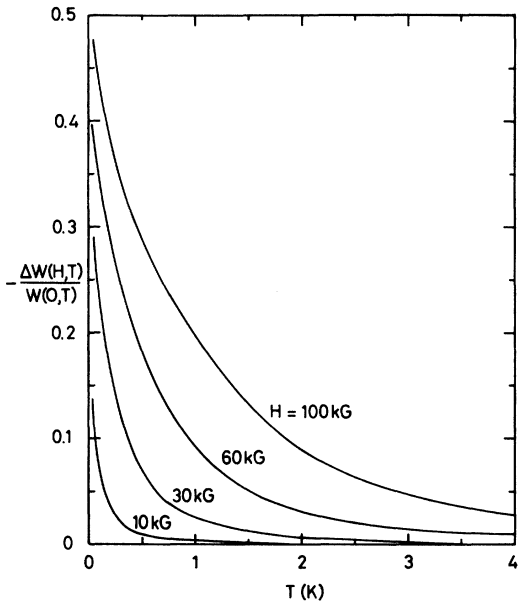

FIG. 10. Negative thermal magnetoresistivity, $-\Delta W(H,T)/W(0,T)$, as a function of temperatures for various applied fields.

Experimental data on the thermal magnetoresistivity are rather meager. The measured thermal conductivity includes not only the electronic part but also the lattice contribution. One has the problem of making the right separation of the two quantities. At low temperatures, the lattice contribution may be small but this depends on the relative scattering strength due to the impurities

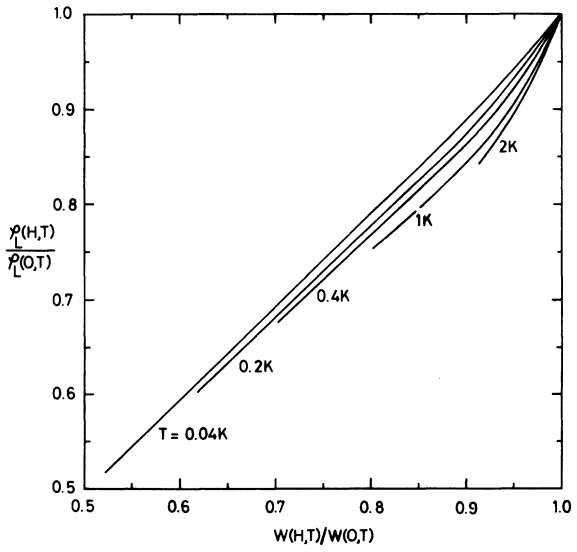

FIG. 11. Relative electrical magnetoresistivity, $\rho_L(H,T)/\rho_L(0,T)$, vs the relative thermal magnetoresistivity, $W(H,T)/W(0,T)$, for various values of temperatures.

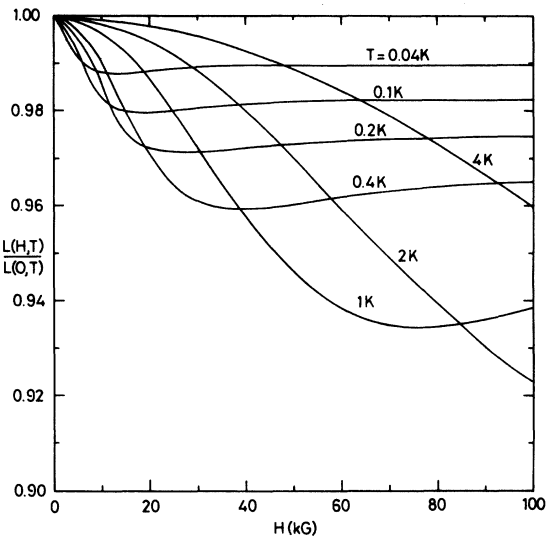

FIG. 12. Relative Lorenz number $L(H,T)/L(0,T)$, as a function of external magnetic field or a number of temperatures.

or crystal defects present.

Owing to the lack of the latest available data, we can only compare our result with the old experiment of Chari and De Nobel[16] on dilute Ag-Mn alloys (cf. Figs. 17 and 18 of Ref. 7). The agreement is qualitative but good. We reach the same conclusion that there is a close parallelism between the relative thermal magnetoresistivity and the electrical one.

The thermal resistivity can also be analyzed in terms of the Lorenz number which is given by Eq. (2.5). The relative strength of the Lorenz number in a field can be expressed as

$$\frac{L(H,T)}{L(0,T)} = \frac{\rho_L(H,T)/\rho_L(0,T)}{W(H,T)/W(0,T)} . \tag{3.4}$$

In Fig. 11, a graph of $\rho_L(H,T)/\rho_L(0,T)$ vs $W(H,T)/W(0,T)$ is plotted for a number of temperatures in order to detect the field dependence of L. A linear dependence of $\rho_L(H,T)/\rho_L(0,T)$ on $W(H,T)/W(0,T)$ is obtained at high fields and low temperatures. This indicates that in the high-(H/T) region, the Lorenz number is independent of the external magnetic field. This agrees with the conclusion drawn by Chari[17] in analyzing the experimental data of Ag-Mn alloys.[7] However, a certain amount of nonlinearity is exhibited in the low field and high-temperature region. In this region, the Lorenz number seems to be weakly field dependent. To get a clearer picture, we make a direct plot on the field and temperature variation of L in Figs. 12 and 13 in an amplified scale. The variation appears rather rapid in the

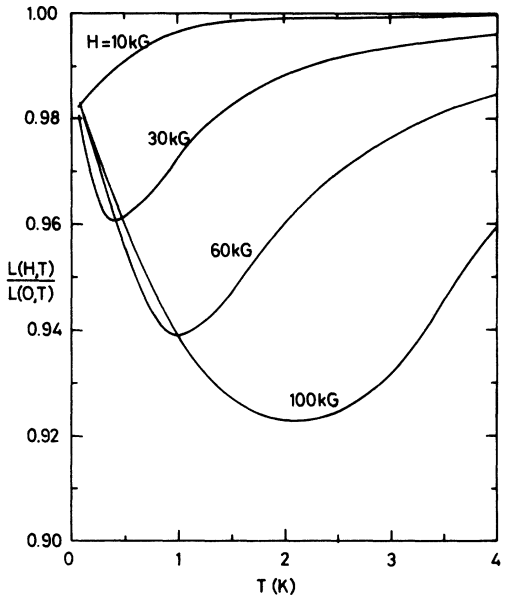

FIG. 13. Relative Lorenz number, $L(H,T)/L(0,T)$, as a function of temperature for a number of applied fields.

low-(H/T) region. Nevertheless, the actual change of the value of L in Figs. 12 and 13 is very small. For fields up to 100 kG, the change is only of the order of 8%. It is therefore doubtful whether such a small change of L can be detected experimentally.

IV. CONCLUSIONS

The main results and conclusions are summarized as follows:

(a) At zero field, the temperature variation of the thermopower and the Lorenz number yield a well-defined broad minimum and maximum respectively, at a temperature in the vicinity of T_K, in complete accord with experiments[4,9,10,14] and other theories.[11,12,13]

(b) The magnetothermopower shows a monotonical decrease as a function of the applied field even at temperatures above T_K. This agrees remarkably well with the experimental data of Berman and Kopp[4] on Au-Fe alloys.

(c) In the low-field region, i.e., $H/T<2$, we find that the negative magnetothermopower and the negative electrical magnetoresistivity are proportional to each other and to the square of the impurity magnetization. This confirms the perturbative result of Weiner and Béal-Monod.[5]

(d) In the high-field region, the relative thermopower follows a $1/H - 1/H_0$ behavior, in agreement with the experimental result on Au-Fe alloys.[4]

(e) The field and temperature dependence of the negative thermal magnetoresistivity closely resemble those of the electrical one. A close parallelism is therefore established between the two quantities.

(f) The Lorenz number is found to be practically independent of the external magnetic field. This agrees with the conclusion drawn by Chari[17] on Ag-Mn alloys.

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