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Magnetic Field Effect on the Thermoelectric Power of a Dilute AuFe Alloy

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Abstract. Low temperature measurements are presented on the Seebeck coefficient of an alloy Au + 0,03 at.% Fe in presence of a longitudinal or transversal magnetic field. The effect observed is explained by a theory using the first Born approximation. The *s-d* exchange integral is estimated: $J \approx -0{,}35$ eV.

Introduction

Noble metals having magnetic impurities usually give a negative magnetoresistivity at low temperature. Such a behaviour is more easily explained when electron-impurity scattering is described by a model using first Born approximation. This can be done only if $T \ll T_K$, where T_K is the Kondo temperature, because higher order terms begin to be important when T approaches T_K. In parallel, the same approximation when applied to the calculation of the thermoelectric power S shows that S can reach a great value (much greater than predicted by the ordinary diffusion theories) when $g\,\mu\,H \approx k\,T$ [1, 2]. The purpose of this work is to experimentally observe such a phenomenon. We choose AuFe alloys because their behaviour at zero magnetic field is well explained by Kondo's theory [3]. In our study, the Fe concentration is identical to that of Berman et al. [4], who measured S at low transverse magnetic field.

Theoretical

A general expression for the thermoelectric power in metals is [5]:

$$S = \frac{1}{e\,T} \frac{\int_{-\infty}^{+\infty} \varepsilon\,\tau(\varepsilon) \frac{\partial f}{\partial \varepsilon}\,d\varepsilon}{\int_{-\infty}^{+\infty} \tau(\varepsilon) \frac{\partial f}{\partial \varepsilon}\,d\varepsilon} \tag{1}$$

where f is the Fermi function, $\varepsilon = E - E_F$ is the difference between the electron energy and the Fermi energy, and $\tau(\varepsilon)$ is the relaxation time of the electron. The denominator of expression (1) is directly proportional to the conductivity σ of the metal.

In the case of elastic electron-ion scattering, the expression (1) can be further simplified [6]:

$$S = \frac{\pi^2 k^2 T}{3 e} \left(\frac{d \ln N(E)}{dE} + \frac{d \ln v^2(E)}{dE} + \frac{d \ln \tau(E)}{dE} \right) \tag{2}$$

if the density of states $N(E)$, the electron speed $v(E)$ and $\tau(E)$ do not vary too strongly with energy.

With magnetic impurities however, equation (2) cannot be applied. When the calculation is restricted to first Born approximation, the relaxation times for the two electron spin orientation + and − relatively to the spin of the ion have been given by several authors [1, 7]:

$$(\tau_{\pm})^{-1} \div V^2 + J^2 \overline{M^2} \mp 2 V J \overline{M} + \frac{J^2 \overline{M} \operatorname{csch} (x/2)}{(1-f) \exp (\pm x/2) + f \exp (\mp x/2)} \tag{3}$$

where $x = (g \mu H)/k T$, and $\overline{M^2}$ and $\overline{M}$ are statistical averages of M^2 and M. J and V are respectively the s–d exchange integral and the static potential of the impurity.

When $\tau = \tau_+ + \tau_-$ is substituted in equation (1), the complicated expression obtained is difficult to use for comparison with experiment. De Vroomen and Potters [1] made a computer analysis with, however, values of J and V of the same order whereas in AuFe they are presumably of much different magnitude. Gurevich and Yassievich [2] only studied the behaviour of S in a magnetic field from the qualitative point of view. We supposed $J \ll V$ as Kondo's work [3] suggests. After simplification of equation (1) (for instance the even part of $\tau(\varepsilon)$ when integrated in the numerator gives a zero contribution) the thermoelectric power is given by:

$$S = \left(\frac{k}{e}\right) \left(\frac{J}{V}\right)^3 (\overline{M})^2 \left(1 - \operatorname{th}^2 \frac{x}{2}\right) \int_{-\infty}^{+\infty} \frac{\operatorname{th} \eta/2 \, (1 - \operatorname{th}^2 \eta/2)}{\operatorname{th}^2 \eta/2 \operatorname{th}^2 x/2 - 1} \eta \, d\eta \tag{4}$$

with

$$\eta = \frac{\varepsilon}{k T} \quad \text{and} \quad (\overline{M}) = -\frac{2\Gamma + 1}{2} \operatorname{cth} \left(\frac{2\Gamma + 1}{2} x \right) + \frac{1}{2} \operatorname{cth} \frac{x}{2}.$$

Γ is the spin of the impurity ion.

The integral is then independent of J and V and can be analyzed with a computer. The thermoelectric power at a given field depends only of J/V and Γ. S is zero at zero and infinite magnetic field but goes through a maximum when $x \cong 1$.

Experimental

The alloy has been melted in an arc furnace with an argon atmosphere. Wire of 0,1 mm diameter was drawn from the ingot for the measurements. Magnetic fields up to 50 kG were provided by a superconducting solenoid. Part of the wire was coiled around a teflon cylinder, another part was placed parallel to the axis of this cylinder (varnish was used for good thermal contact). This geometry allows one to measure transverse and longitudinal effects in the same conditions. The cylinder ends can be heated to maintain the desired temperature gradient. The system is evacuated and

placed in the magnet such that the coiled sample is submitted to a transverse magnetic field. The thermoelectric voltage is taken by copper wires (whose Seebeck coefficient is small) and measured with a Honeywell 2783 potentiometer followed by a Keithley 147 manovoltmeter. Temperatures are measured with help of AuFe-Cu thermocouples at zero magnetic field.

Results and Discussion

Figure 1 shows the result of the measurements for three different temperatures and for longitudinal and transverse fields. The relative increase in absolute value of the thermoelectric power is plotted. At zero field the value of S is already large ($-7.8\,\mu V/°K$ at 2°K) although smaller than the value of Berman et al. ($-11.8\,\mu V/°K$ at 2°K). This difference, and the smaller relative increase observed in case of transverse field (Berman et al. did not study the effect of longitudinal field), probably come from differences in effective Fe concentration and from the fact that our sample was not annealed (the relative contribution of dislocations decreases the total value of the thermoelectric power at low Fe concentration). For the two directions of the magnetic field, the curves are almost identical when S is plotted in function of H/T, except that the maximum value of S decreases when temperature decreases.

We think the effect observed is a manifestation of electron-magnetic ion interaction at least qualitatively explainable by a first Born approximation model for the following reasons:

1) For dilute AuFe alloys: $T_K \approx 0.1\,°K$ [8], perturbation theory is consequently still valid in our studied temperature range. In fact, Kondo's second Born approximation theory well describes the behaviour of $S(T)$ at zero magnetic field down to 0.5°K [3]. If the magnetoresistivity can be explained using a first Born approximation theory, as in fact it can, then the thermoelectric power with the presence of a magnetic field can also, since S and ϱ are closely connected. Experimentally the magnetoresistivity shows a concentration effect indicating correlations between impurities [8]. At low Fe concentration however, such as used in this work, most of the variation of S is due to isolated Fe impurities.

2) ΔS is negative, as expected if J is negative and V positive.

3) The smaller values observed with transverse field are consistent with Gurevich and Yassievich's work. In transverse field, using first Born approximation, a reversal

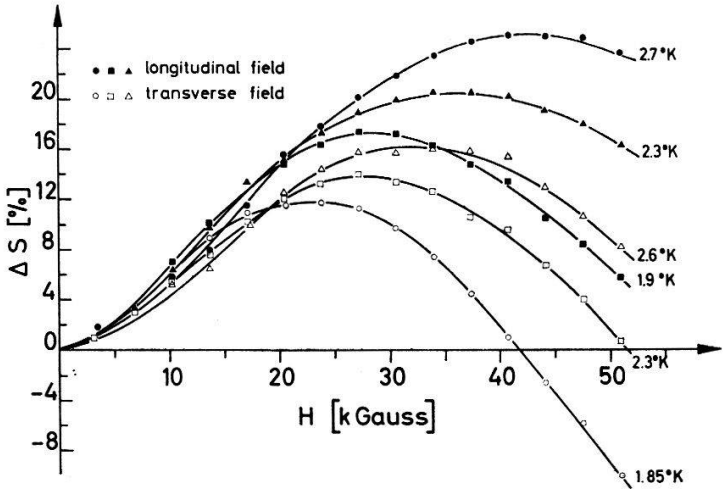

Figure 1
Relative increase of the thermoelectric power of an Au+0,03 at. % Fe alloy as a function of magnetic field.

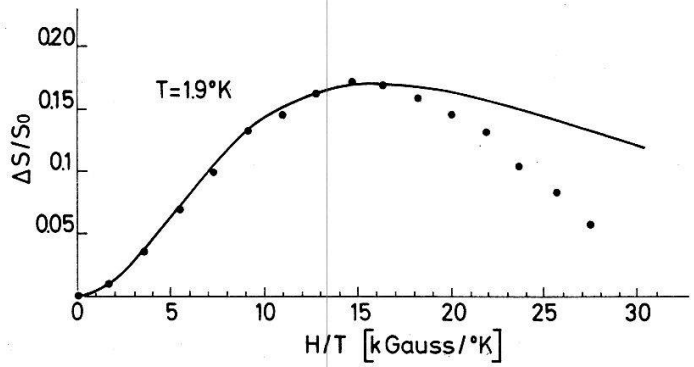

Figure 2
Comparison between experiment and first Born approximation theory (for longitudinal magnetic field).

of the sign of S is expected when $\Omega\tau \approx 1$, where Ω is the Larmor frequency of the electrons. Consequently, at low field S is smaller relative to longitudinal field, of a value depending of Ω and τ.

4) Good agreement (Fig. 2) is obtained when data are compared with equation (3) (with longitudinal field and $S = 5/2$, $g = 2$). Only one parameter has been chosen for adjustment: the ratio $J/v \approx -0.10$. If $V = 3.5$ eV from residual resistivity measurements [3], then $J \approx -0.35$ eV (from the logarithmic term in the expression for resistivity, Kondo [9] evaluated $J = -0.25$ eV). It is clear, however, that theory must include at least the second Born approximation for proper comparison with experiment, because of the already large value of S at zero magnetic field. This explains the divergence observed at high field on Figure 2. In a more recent paper, Berman et al. [10] have experimentally studied the decrease of S at high field and made comparisons with Kondo's expression with $H \neq 0$. Kondo, however, admitted a random distribution for the orientations of impurity spins, so that an effect of de Vroomen-Potters' type is not included in their theory. Considering this high field decrease, the effective value of the maximum can be somewhat larger than admitted here, however as J/V is proportional to the cubic root of the value of the maximum, the correction on J is not expected to be large.

Conclusions

Proper comparison of experimentally determined values of $S(H, T)$ with theory must take into account higher perturbation terms for calculating the relaxation time τ. However, good qualitative and even semi-quantitative agreement are obtained with a first Born approximation theory.

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