

# Lorenz Numbers of Dilute Ag Alloys

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The Lorenz numbers of some dilute Ag alloys have been measured and found equal within experimental error to the Sommerfeld value, in disagreement with the perturbation theory of the Kondo effect.

SOME recent measurements<sup>1</sup> of the electrical and thermal conductivities of 99.999%-pure Ag have indicated the Lorenz number for this material at temperatures from 0.3 to 1.2°K to be constant and equal to a value some 3% greater than the Sommerfeld prediction of 24.5 nΩW(°K)<sup>-2</sup>. Deviations of this kind are commonly due to a strong energy dependence of the scattering of the conduction electrons. Since it is recognized<sup>2,3</sup> that the presence of magnetic impurities may give rise to such effects, it was thought of interest to investigate the conductivities of samples of Ag alloys in which the nature of the impurity was known. The results we report are in disagreement with the predictions of perturbation theory.

It is a result of the perturbation-theory approach<sup>2</sup> that the probability of scattering of a conduction electron can be written in the form

$$1/\tau(\mathcal{E}, T) = g_0 + \alpha g(\mathcal{E}, T),$$

where  $g_0$  and  $\alpha$  are constants,  $\mathcal{E}$  is the energy of the electron measured relative to the chemical potential,  $T$  is the temperature, and

$$g(\mathcal{E}, T) = \int_{-D}^D \frac{\frac{1}{2} - f(\mathcal{E}')}{\mathcal{E}' - \mathcal{E}} d\mathcal{E}',$$

with  $f$  as the Fermi-Dirac function and  $D$  as a cutoff parameter much larger than  $kT$ . This integral is related to the digamma function.<sup>4</sup> When  $\alpha$  is small, one may then express the relaxation time  $\tau$  as

$$\tau(\mathcal{E}, T) = \tau_0 + \tau_1 [\ln T + g_2(a)],$$

with  $\tau_0$  and  $\tau_1$  as constants,  $T$  measured in Kelvins,  $a$  equal to  $\mathcal{E}/2kT$ , and

$$g_2(a) = a^2 \sum_{n=0}^{\infty} \left\{ \left( n + \frac{1}{2} \right) \left[ a^2 + \left( n + \frac{1}{2} \right)^2 \pi^2 \right] \right\}^{-1}.$$

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<sup>1</sup> A. C. Anderson, R. E. Peterson, and J. E. Robichaux, *Phys. Rev. Letters* **20**, 459 (1968).

<sup>2</sup> J. Kondo, *Progr. Theoret. Phys. (Kyoto)* **32**, 37 (1964).

<sup>3</sup> H. Suhl and D. Wong, *Physics* **3**, 17 (1967).

<sup>4</sup> P. J. Davis, in *Handbook of Mathematical Functions* (U. S. Government Printing Office, Washington D. C., 1964), p. 253.

The ratio of the Lorenz number to the Sommerfeld value  $L_0$  is then

$$\frac{L}{L_0} = \left[ \tau_0 + \tau_1 \left( \ln T + \frac{12}{\pi^2} \int_{-\infty}^{\infty} \frac{a^2 g_2(a)}{2 \cosh^2 a} da \right) \right] / \left[ \tau_0 + \tau_1 \left( \ln T + \int_{-\infty}^{\infty} \frac{g_2(a)}{2 \cosh^2 a} da \right) \right].$$

Since

$$\int_{-\infty}^{\infty} \frac{g_2(a)}{2 \cosh^2 a} da = 2 \ln 2 - 1$$

and

$$\frac{12}{\pi^2} \int_{-\infty}^{\infty} \frac{a^2 g_2(a)}{2 \cosh^2 a} da = 2 \ln 2 - \frac{1}{3},$$

we find

$$\frac{L}{L_0} \simeq 1 + \frac{2\tau_1}{3\tau_0} + O\left(\frac{\tau_1^2}{\tau_0^2}\right). \quad (1)$$

The electrical conductivity under these conditions would vary with temperature as

$$\sigma(T) = \sigma_0 [1 + (\tau_1/\tau_0)(\ln T + 0.39)]. \quad (2)$$

To test the above results, we have measured the thermal and electrical resistivity of dilute **AgIn** and **AgMn** alloys. The samples were prepared from 99.9999%-pure component materials and were in the form of strips approximately 10 cm long with a 1×0.2 mm cross section. They were subjected to an homogenization treatment at 700°C for about 24 h under high vacuum. The thermal conductivity was measured with an accuracy of about 1% by the steady-state method using 10-Ω, 0.1-W Allen-Bradley resistors as thermometers. The measuring technique was essentially that described by Jericho.<sup>5</sup> Between 4.2 and 0.6°K the absolute temperature was measured with the help of He<sup>4</sup> and He<sup>3</sup> vapor-pressure bulbs that were in direct contact with the specimen heat sink. For the resistivity measurements of the **AgIn** sample, this temperature range was extended down to about 0.3°K by extrapolating the calibration curves of the carbon thermometers to lower temperatures. The electrical resistivity was measured

<sup>5</sup> M. H. Jericho, *Phil. Trans. Roy. Soc. (London)* **257**, 385 (1965).

with an accuracy of 0.3%, using the relay modulator described by Jericho and March.<sup>6</sup> The resistivities of the samples at 4.0°K were 17.21 nΩ cm for the Ag-0.011 at.% Mn alloy and 26.55 nΩ cm for the Ag-0.015 at.% In sample.

Extrapolation of lattice thermal-conductivity data on silver alloys<sup>7</sup> to this purity range suggests that the phonon contribution to the heat transport in our samples is at most 1% at 4°K and about 0.1% at 1°K and can therefore be neglected.

The experimental results are shown in Fig. 1, where the electrical resistivity  $\rho$ , and the thermal resistivity  $W$  multiplied by  $L_0T$  are plotted as functions of  $\ln T$ . It will be seen that the electrical resistivity of the AgIn is independent of temperature over the range stated, while that of the AgMn can be expressed in the form given in Eq. (2) with  $\tau_1/\tau_0=0.038$ . The thermal resistivities of both samples were such that the Wiedemann-Franz law was obeyed to within 1% with the Lorenz number taking on the Sommerfeld value  $L_0$ .

The disagreement between theory and experiment is shown clearly when the above value of  $\tau_1/\tau_0$  for the AgMn alloy is substituted in Eq. (1), which then predicts a Lorenz number 2.6% greater than  $L_0$ . It is possible that the failure of the theory to describe correctly the Lorenz number in this case could be attributed to a concentration dependence of  $L$ ; if this were the case, however, it would be surprising that no deviations were detected from the linear dependence of  $\rho$  on  $\ln T$ .

<sup>6</sup> M. H. Jericho and R. H. March, Rev. Sci. Instr. **38**, 428 (1967).

<sup>7</sup> C. Van Baarle, F. W. Gorter, and P. Winsemius, Physica **35**, 223 (1967).

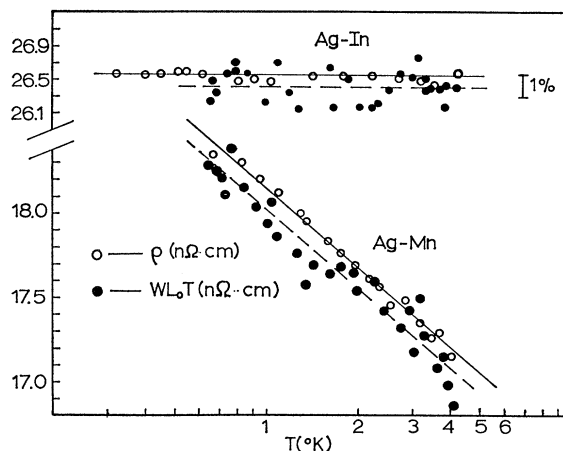


FIG. 1. Electrical resistivity  $\rho$  and thermal resistivity  $W$  multiplied by  $L_0T$  plotted as a function of  $\ln T$  for the two alloys investigated.

It is also interesting to note that the 3.2% enhancement of the Lorenz number observed<sup>1</sup> in "pure" Ag by Anderson *et al.* cannot be interpreted within the present theory as being due to the presence of magnetic impurities; if their value of  $L$  is fitted with Eq. (1), then Eq. (2) predicts that over the temperature range studied there should have been a 5% change in  $\rho$ , and no such variation was observed.

*Note added in proof.* The Lorenz number measurements for the AgMn alloy system have recently been extended over a wide range of Mn concentration, and a definite trend in the concentration dependence of  $L$  is discernible. For a 50 ppm. AgMn alloy, for example, theory and experiment are in good agreement.