Temperature dependence of the resistivity in amorphous ferromagnetic alloys: Spin-wave contribution and resistivity minimum

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The resistivities in amorphous ferromagnetic alloys of Ni_cAu_{1-c}, Co_cAu_{1-c}, and Fe_cAu_{1-c} are measured as a function of temperature. The amorphous alloys are purely metallic and obtained by quenched condensation. The resistivity shows a T^2 temperature dependence which is due to electron-magnon interaction. From a theroretical point of view, the temperature dependence of the resistivity in amorphous metals provides an interesting problem because the lifetime of the conduction electrons is much shorter than the oscillation period of the magnons and phonons. Therefore, the elastic and inelastic scattering processes cannot be treated independently and the Ziman-Baym formula fails. Only exceptionally in Fe-rich Fe_cAu_{1-c} alloys ($c > 0.9$) we found the strong increase of the resistivity towards low temperature which has been observed by others in metallic glasses. This behavior is correlated with the sudden drop of the magnetic moment of the Fe atoms in this concentration region.

I. INTRODUCTION

In this paper we present some accurate resistivity measurements in amorphous ferromagnetic alloys. The resistivity measurements have been performed for two reasons: (i) to look for the influence of the electron-magnon scattering on the resistivity and (ii) to check the supposition by Cochrane et al.[1] that amorphous metals have a resistivity minimum.

Measurements of the specific heat in amorphous metals[2-4] prove the existance of an atomic vibration spectrum in the amorphous state. Nevertheless there is no direct experimental evidence of a phonon-induced resistivity in these materials. Korn et al.,[5] Bergmann,[6] and Collver[7] found only a small linear dependence of the resistivity on the temperature. Therefore it is interesting to examine whether the scattering of conduction electrons by thermally excited spin waves in amorphous ferromagnets yields a contribution to the resistivity, and if so, which temperature dependence the resistivity shows. From the theoretical point of view this problem is quite interesting because in amorphous metals the lifetime of a conduction electron in a momentum state is so small that it is much less then the oscillation period of thermal excitations. Therefore it is not clear whether the elastic and the inelastic scattering can be separated and treated independently in the Boltzmann equation.

Cochrane et al.[1] tried to find an indication of the amorphous state in the transport properties. Experimentally they found a resistance minimum in a large number of metallic glasses.[8] Similar results have been

reported by Lin,[9] Hasegawa and co-workers,[10-12] Liang and Tsuei,[13] and Poon and Durand.[14] This minimum occurred as well in ferromagnetic alloys as in the non-ferromagnetic alloy $Ni_{0.75}P_{0.25}$. They interpreted the resistance minimum as originated from the amorphous structure and explained it with the same model which Anderson et al.[15] suggested for the interpretation of the linear T dependence of the specific heat in glasses. We extend their investigations to amorphous alloys which do not contain glass formers and are purely metallic.

II. EXPERIMENTAL RESULTS

We investigate the amorphous alloy system Ni_cAu_{1-c}, Co_cAu_{1-c}, and Fe_cAu_{1-c}. These amorphous alloys are prepared by quenched condensation onto a substrate at He temperature, the most effective method to obtain amorphous metals. The amorphous structure of the investigated alloys has been proved previously by x-ray or electron diffraction.[16-18] The composition of the evaporated films is analyzed by x-ray fluorescence. For the Co_cAu_{1-c} system we use in addition a He-backscattering analysis. The agreement between the two determinations of c is better than 2%.

Since the amorphous state is metastable many alloys transform during annealing to room temperature into the crystalline state. This transformation is accompanied with a steep decrease of the resistivity. The resistivity measurements have been performed sufficiently below the crystallization temperature and after a thermal cycling. The resistivity hysteresis is of

the order of 0.01%.

Figure 1 shows the resistivity of two amorphous Ni_cAu_{1-c} alloys. These alloys are ferromagnetic and their Curie temperatures have been determined by the anomalous Hall effect.[19] It is obvious that the strong temperature dependence occurs below T_C and is correlated with the loss of magnetic order. Above T_C there is only a small negative linear temperature dependence. At low temperature the thermal part of the resistivity is proportional to T^2. This is demonstrated in Fig. 2 for amorphous ferromagnetic Co_cAu_{1-c} alloys. Here we plot $\rho(T) - \rho(4.2)$ as a function of T^2. In this plot the usual spin-wave contribution to the resistivity would be a straight line. The T^2 law is well obeyed over a large temperature region except for the very dilute ferromagnet $Co_{0.31}Au_{0.61}$. For low temperatures the curves flatten. The increase of the resistitivity towards low temperatures as described in Ref. 1 corresponds to one-third of the ordinate in Fig. 2. We do not find such an increase of the resistivity. For Ni_cAu_{1-c} alloys a possible raising of the resistivity is not larger than the accuracy of the measurement of 0.01% of $\rho(4.2)$. In amorphous Co_cAu_{1-c} alloys a small minimum with a depth below 0.03% of $\rho(4.2)$ is observed.

Figure 3 shows ρ/ρ_{min} for pure amorphous Co at low temperatures. The accuracy of 0.01% of $\rho(4.2)$ is not sufficient to study this temperature dependence systematically. The experimental results for the amorphous system Fe_cAu_{1-c} are plotted in Fig. 4. In the concentration range $0.9 < c < 1$ we find a clear minimum at low temperatures. The resistivities are of the order of $(0.5-1.0) \times 10^{-6}$ Ω m, depending on the composition. Magnetic fields of the order of 75 kG do not change the shape of the curve. [Only $\rho(4.2)$ is decreased because the alloys show a negative magnetoresistance.] For Fe concentrations below 0.9, the depth of the minimum reduces to the accuracy of the experiment. There is again a T^2 law in a certain temperature range. The coefficient $\alpha = d\rho/dT^2$ increases

FIG. 2. Resistivity of the alloy series Co_cAu_{1-c} as a function of T^2.

drastically with decreasing concentration of the magnetic atoms for all three investigated alloy systems. In Fig. 5, α is plotted as a function of the composition for Co_cAu_{1-c} and Fe_cAu_{1-c}. A comparison between $\rho(T)$ in the amorphous and $\rho(T)$ in the crystalline state is given in Fig. 6. An amorphous $Fe_{0.70}Au_{0.30}$ alloy is annealed up to 100 K without crystallization. Then the reversible temperature dependence of the resistivity is measured and plotted in the upper part of Fig. 6. Afterwards the alloy is annealed up to 300 K. It transforms at about 270 K into the crystalline state. Finally, the low-temperature dependence of the resistivity is measured and plotted in the upper part of Fig. 6. The Curie temperature of the investigated Co_cAu_{1-c} and Fe_cAu_{1-c} alloys lies always above room temperature.
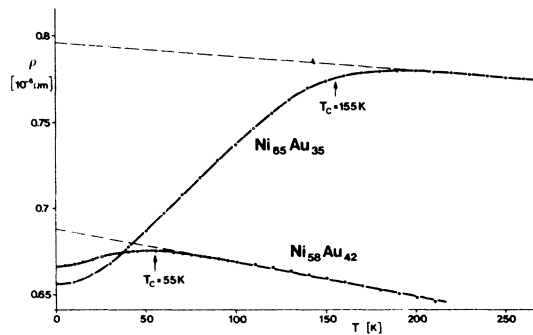

FIG. 1. Resistivity as a function of temperature for two amorphous ferromagnetic Ni-Au alloys. The corresponding Curie temperatures are marked with an arrow.
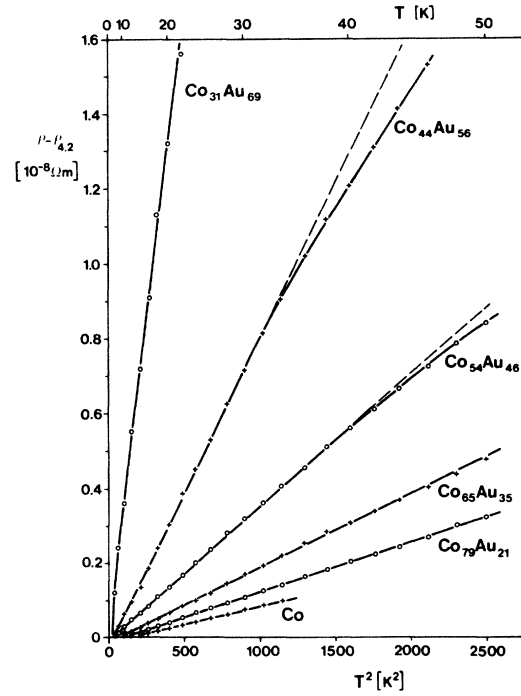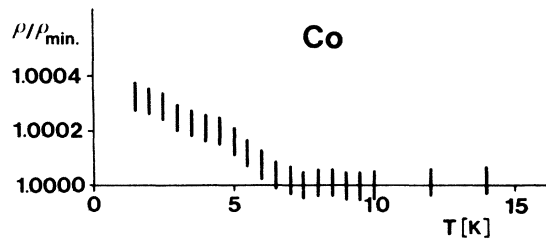

FIG. 3. Low-temperature resistivity of pure amorphous Co.
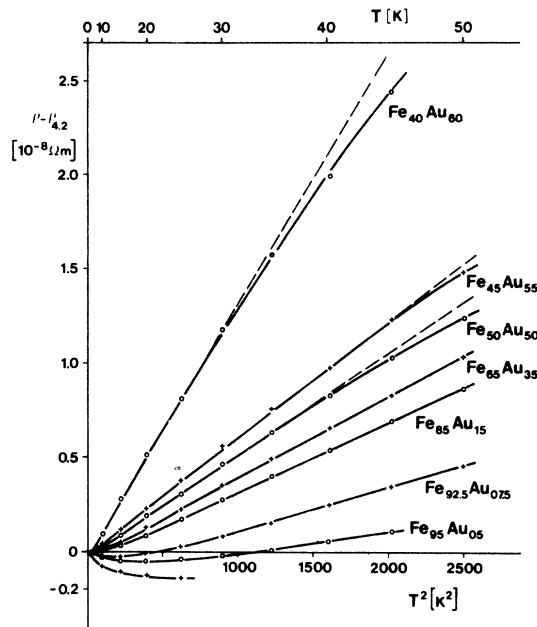
FIG. 4. Resistivity of the alloy series Fe_cAu_{1-c} as a function of T^2.

FIG. 6. (a) Comparison between the temperature dependent resistivity in the amorphous and the crystalline phase of $Fe_{0.70}Au_{0.30}$. (b) The annealing curve of the resistivity of $Fe_{0.70}Au_{0.30}$. The crystallization occurs at 270 K.

III. DISCUSSION

For the amorphous Ni_cAu_{1-c} and Co_cAu_{1-c}, the low-temperature resistivity obeys a T^2 law. The same applies for the Fe_cAu_{1-c} alloys with $c < 0.9$. Above

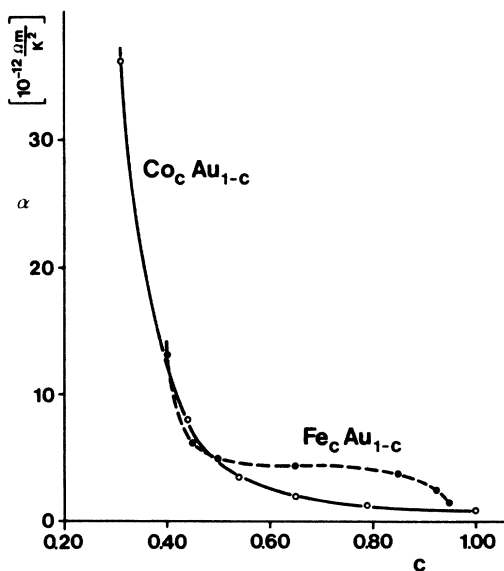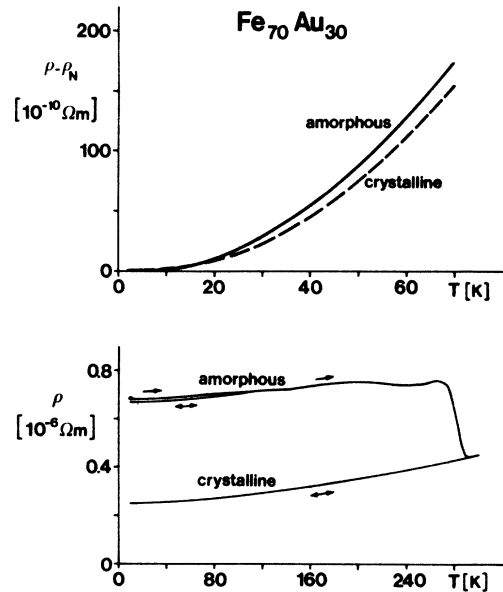FIG. 5. Coefficient $\alpha = d\rho/dT^2$ for amorphous Co_cAu_{1-c} and Fe_cAu_{1-c} alloys as a function of the composition.

the Curie temperature the resistivity is almost constant. This favors the explanation that the leading mechanism for the thermal part of the resistivity is the electron-magnon interaction and not the scattering of conduction electrons into the d states (Mott[20]) which also yields a T^2 law.

In amorphous ferromagnets one expects that electron-magnon processes without momentum conservation play an important role. The inclusion of these processes into the calculation of the resistivity of magnetic metals has been performed by a Lang and Turner[21] and Mills et al.[22] for dilute ferromagnets. They obtained—within a free-electron model—an additional $T^{3/2}$ dependence for the resistivity. The extension of this theory to amorphous metals corresponds to the Ziman-Baym theory.[23,24] However, the application of these theories to small momentum transfer appear to be problematic. The conduction electrons have quite a small mean free path of the order of 10 Å. Therefore, they should not feel the variation of the potential over much larger distances. For the calculation of the residual resistivity this restriction is not important because the contributions for small q are weighted with the factor $(q/2k_F)^3$. This is one of the reasons why the Ziman formula works surprisingly well. For the calculation of the thermal part of the low-temperature resistivity the situation is opposite. At low temperature, magnons (and pho-

nons) with small q are excited and the potential is mainly changed on a scale of large distances. In addition a similar problem occurs because of the small lifetime of the conduction electrons in a momentum state which is of the order of 10^{-15} sec. This time is much shorter than the oscillation period of the magnons. Therefore the conduction electrons should not feel the low-energy oscillation of the excitations during one scattering process.

Heuristically we sketch the scattering in Fig. 7. The conduction electron is scattered in a series of scattering processes. During each process the scattering is quasielastic because the lifetime τ of the conduction electron in the state k'' is much smaller than the period of the oscillating magnon (or phonon). The next scattering process is again quasielastic but the phase of the magnon has changed by $\omega\tau$ (ω is the magnon frequency) with respect to the first process. After many scattering processes the conduction electron has probed the oscillatorial behavior of the magnon and has to fulfill the energy conservation between electron and magnon energy. The mathematical problem is that it is not sufficient to describe the elementary processes alone but one has to follow the long sequence of quasielastic processes which are enveloped by the energy conservation. In addition, the conduction electron has probed the spatial phase variation of the magnon over a distance of the diffusion length.

We cannot give the mathematical treatment of this problem nor its solution. However, we want to show firstly that the Ziman-Baym theory gives a wrong temperature dependence of the resistivity, and secondly that the inclusion of magnons into the quasielastic scattering essentially corrects this fault.

In the Appendix we perform a calculation of the thermal part of the resistivity in amorphous ferromagnets within the classical framework of the Ziman-Baym theory. We use the magnon dispersion relation $\hbar\omega_q = Dq^2$. This relation has been experimentally obtained by Mook et al.[25] for amorphous ferromagnetic Co_4P. We obtain three different contributions due to

FIG. 7. Series of quasielastic scattering processes which as a whole emit inelastically a magnon.

the electron-magnon interaction: (i) the contribution of electron-magnon processes which conserve momentum (coherent scattering); it obeys a T^2 law as in the crystalline state[26-28]; (ii) the contribution of electron-magnon processes which do not conserve momentum (incoherent scattering); it obeys a $T^{3/2}$ law; (iii) the contribution of the elastic scattering which is temperature dependent because of the temperature dependence of $\langle S_z \rangle$ and $\langle S_z^2 \rangle$; it is negative and obeys a $T^{3/2}$ law.

In the Appendix we show that the sum of the second and third term should be much larger than the contribution of the coherent T^2 term. This contradicts the experimental results. First we found a T^2 law in the amorphous state. Secondly, the temperature dependence in the amorphous and the crystalline state do not differ drastically. We believe, that this deviation between experiment and Ziman-Baym theory is due to the inappropriate treatment of processes with small momentum and energy transfer.

In a second calculation we take the opposite point of view. We consider the conduction electrons as plane waves within volume of the order of l^3 (l is the mean free path). Within this volume the magnetization S^z does not show a spatial variation. In addition we consider S^z as quasistatic because the time scale of its oscillation is much larger than the lifetime of the conduction electrons. Then we obtain essentially two contributions of the thermal part of the resistivity: (i) a quasielastic contribution due to spin-flip processes by the "static" magnons; its contribution is one-third of the incoherent electron-magnon contribution; (ii) the unchanged elastic contribution due to the temperature dependence of S^z.

If we restrict ourselves to a rough calculation— where we do not distinguish between $\langle S^z \rangle^2$ and $\langle (S^z)^2 \rangle$—both contributions cancel quantitatively and we obtain a temperature-independent resistivity at low temperatures. We believe that the correct treatment of the thermal part of the resistivity lies somewhere between these two extremes and presents an interesting challenge in the transport theory.

With regard to the resistance minimum we summarize the experimental situation. A pronounced minimum occurs in many metallic glasses. In amorphous metals with purely metallic binding the resistance minimum is two orders of magnitude smaller than in the metallic glasses. Therefore it is difficult to believe that only the amorphous structure is responsible for the minimim. One may suspect that the high concentration of glass formers is involved in this phenomenon. On the other hand the amorphous structure is of importance since the flat minimum disappears after the crystallization. The occurrence of the more-pronounced resistance minimum for $Fe_c Au_{1-c}$ in the concentration range $c > 0.9$ is rather interesting because it coincides with the concentration range in which the Fe atom changes its magnetic mo-
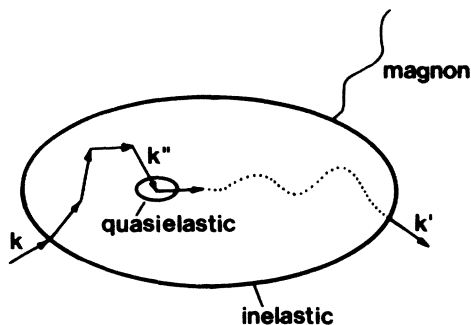
ment.[12] Within this concentration range we found several peculiarities in further transport properties.[29]

APPENDIX

We calculate the thermal part of the resistivity for an amorphous Heisenberg ferromagnet. The exchange interaction between the conduction electrons and the localized magnetic moments shall be

$$\sum_l 2w(r-r_l)\,\vec{s}\vec{S}_l = \sum_l w(r-r_l)$$

$$\times (s^+S_l^- + s^-S_l^+ + 2s^zS_l^z) \ ,$$

$$w_P = \frac{1}{\Omega_0}\int d^3r\, w(r)e^{-ipr} \ ,$$

where Ω_0 is the atomic volume. For the sake of simplicity we neglect the normal potential of the magnetic moments. Then the Fermi function f_k in the presence of an electrical field is the same for spin-up and spin-down electrons and given by

$$f_k = f_k^0 + \left(-\frac{\partial f}{\partial \epsilon}\right)\phi_k \ .$$

The resulting resistivity formula is

$$\rho^{11} = \frac{1}{2}\beta\Omega\sum_k\sum_{k'}\sum_q(\phi_k - \phi_{k'})^2 P_{kq}^k$$

$$\times \left[\left|\left|\sum_k e\vec{v}_k\phi_k\left(\frac{\partial f}{\partial\epsilon_k}\right)\right|^2\right|\right]^{-1} \ , \quad (1)$$

where $\beta = 1/k_BT$, Ω is the volume, P_{kq}^k is the transition probability for an electron $k\downarrow$ to absorb a magnon q and to go into the state $k'\uparrow$.

The transition probability is given by

$$P_{kq}^k = (2\pi/\hbar)|g_{kk'q}|^2\delta(\epsilon_k + \hbar\omega_q - \epsilon_{k'})$$

$$\times n_q^0 f_k^0(1-f_{k'}^0) \ , \quad (2)$$

where $g_{kk'q}$ is the electron-magnon matrix element, n_q^0 is the boson occupation number, and f_k^0 is the Fermi occupation number in equilibrium.

For the calculation of the electron-magnon matrix element one replaces S^+,S^- by magnon operators and obtains

$$|g_{qq'}|^2 = (2S/N)|w_{q'}|^2\delta_{q'+q}$$

$$+ (2S/N^2)|w_{q'}|^2 a(q'+q) \ .$$

$\delta_{q'+q} = 1$ when $q'+q=0$ and otherwise it is zero, $a(p)$ is the structure factor of the alloy.

The first term describes the coherent electron-magnon processes and yields the T^2 law for the resistivity[26-28]:

$$\rho_c^{11} = \frac{\Omega_0\pi^3 S}{2\,\hbar v_F^2 e^2}|w_0|^2\left(\frac{k_BT}{Dk_F^2}\right)^2 \ .$$

The second term describes the incoherent electron-magnon processes and yields a $T^{3/2}$ law for the resistivity

$$\rho_{in}^{11} = \Omega_0^2\frac{24S(\frac{3}{2})!\zeta(\frac{3}{2})}{\pi\,\hbar v_F^2 e^2}$$

$$\times \int_0^1 d\left[\frac{q'}{2k_F}\right]\left[\frac{q'}{2k_F}\right]^3|w_{q'}|^2 a(q')\left[\frac{k_BT}{D}\right]^{3/2} \ ,$$

$(\frac{3}{2})!\zeta(\frac{3}{2}) \approx 3.472$, where $(\frac{1}{2})! = \Gamma(\frac{5}{2})$. In addition the elastic scattering due to $2s^zS^z$ is temperature dependent. Its contribution to the thermal part is

$$\rho_z^{11} = -\Omega_0^2\frac{4(2S-1)(\frac{3}{2})!\zeta(\frac{3}{2})}{\pi\,\hbar e^2 v_F^2}$$

$$\times \int_0^1 d\left[\frac{q'}{2k_F}\right]\left[\frac{q'}{2k_F}\right]^3 a(q')|w_{q'}|^2\left[\frac{k_BT}{D}\right]^{3/2} \ .$$

This negative contribution to the resistivity has the same temperature dependence ($T^{3/2}$ law) as the resistivity due to incoherent electron-magnon processes. Therefore we have a partial compensation. A quantitative comparison between ρ_z and $(\rho_{in}+\rho_z)$ is difficult because the q' dependence of the exchange potential is hardly known. For the ratio we find

$$\frac{\rho_c}{\rho_{in}+\rho_z} \approx \frac{0.118}{z}$$

$$\times \left[|w_0|^2\bigg/4\int_0^1 d\left[\frac{q'}{2k_F}\right]\left[\frac{q'}{2k_F}\right]^3|w_{q'}|^2 a(q')\right]$$

$$\times \left[\frac{k_BT}{Dk_F^2}\right]^{1/2} \ ,$$

where z is the total number of conduction electrons per atom.

To get an estimate of the ratio we set $z=1$, $a(q')=1$, and $w_q = w_0 = \text{const}$. Furthermore, for D we take the value by Mook et al., $D = 0.185$ eV Å^2, and for k_F the value for Co, $k_F = 1.36$ Å^{-1}. Under these conditions the ratio takes the value $0.118\times(8.6\times10^{-5}T/0.34)^{1/2}$. For $T=30$ K we obtain for the ratio a value of about 10^{-2}. Although the q dependence of w_q and $a(q')$ may alter this ratio, one would expect from this calculation that the $T^{3/2}$ law overpowers the T^2 law.

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