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Magnetoresistance of amorphous ferromagnetic metals

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Amorphous ferromagnetic films of NiAu and and Co are prepared by quenched condensation onto a substrate at He temperature. Amorphous ferromagnetic metals are particularly well suited to investigate the influence of an external magnetic field in the ferromagnetic state on the transport properties. The small mean free path of the conduction electrons suppresses the complications of the band structure and its consequences on the transport properties. The magnetoresistance is measured in an external field up to 80 kG. $d\rho/dB$ is negative and takes values up to 2×10^{-9} m^3/Asec. The results are discussed within the band model of ferromagnetism. They contradict the rigid-band model for NiAu alloys. Within the Heisenberg model the influence of anisotropy fields on the resistivity again yields values for $d\rho/dB$ which are too small.

I. INTRODUCTION

The transport properties of ferromagnetic metals are barely treated in most text books of ferromagnetism. This is astonishing and regrettable because the ferromagnetic metals show particularly interesting electronic transport properties. They have an additional scattering mechanism, the electron-magnon interaction, an anisotropy of the resistivity with respect to the direction of magnetization, a spontaneous Hall effect, and a linear magnetoresistance in a magnetic field. The investigation of the latter ferromagnetic effect is often complicated because it is superimposed on electronic magnetic effects. Also nonmagnetic metals often show in a magnetic field a complicated behavior because the electrons which are responsible for the conductivity belong to different parts of the Fermi surface and have different effective masses and mean free paths. In a perfect crystal one finds in addition the transition from the low-field case ($\omega\tau \ll 1, \omega = eB/m$) to the high-field case ($\omega\tau \gg 1$). Here the Hall effect as well as the resistivity change drastically with magnetic field. These complications due to the electronic magnetic effects can be avoided by using amorphous ferromagnetic metals. Owing to their extremely small mean free path the conduction electrons do not show any band-structure effects. In the absence of ferromagnetism these amorphous metals behave regularly and generally obey the free-electron model. In this paper we investigate the magnetoresistivity of ferromagnetic amorphous alloys in a perpendicular and parallel magnetic field. We find a linear decrease of the resistivity with increasing magnetic field. The results are discussed within the band model and the Heisenberg model of ferromagnetism.

II. EXPERIMENT AND RESULTS

We investigate the transport properties of amorphous NiAu alloys and Co. The films with a thickness of about 1500 Å are prepared by quenched condensation onto a substrate at He temperature. The alloy is evaporated from a crucible of Be oxide. The concentration of the two components in the film is different from the original concentration in the alloy. The final concentration is obtained by x-ray fluorescence analysis with an accuracy of about 5%.[1] The film thicknesses are measured by means of an optical interference method and a surface profile measuring system. The amorphous structure of quench condensed Co has been investigated by Leung and Wright[2] who determined the structure factor of amorphous Co by an electron diffraction experiment. Quench condensed NiAu films show a sharp decrease of the resistivity during annealing, a behavior which is typical for the recrystallization of amorphous metals. However, for Ni_cAu_{1-c} films in the concentration range $0.5 < c < 0.7$ the transformation temperature lies above room temperature. We investigate a $Ni_{0.65}Au_{0.35}$ film at room temperature by an electron diffraction experiment and find the broad smeared pattern which is typical for an amorphous metal.

After the condensation, the evaporation cryostat is inserted into a superconducting magnet. Details of the apparatus and the procedure are described in Ref. 3. A magnetic field up to 80 kG can be applied perpendicular to the film. For a comparison between the perpendicular and the parallel magnetoresistance we use in addition a cryostat in which the substrate can be twisted by 90°. The magnetoresistivity $d\rho/dB$ in perpendicular and parallel magnetic fields agree within the experi-

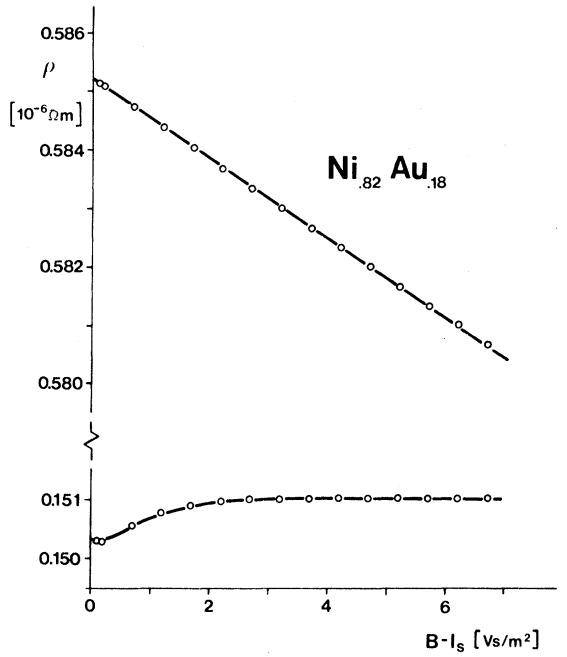

FIG. 1. Magnetoresistance of amorphous (upper curve) and crystalline (lower curve) $Ni_{0.82}Au_{0.18}$ at He temperature.

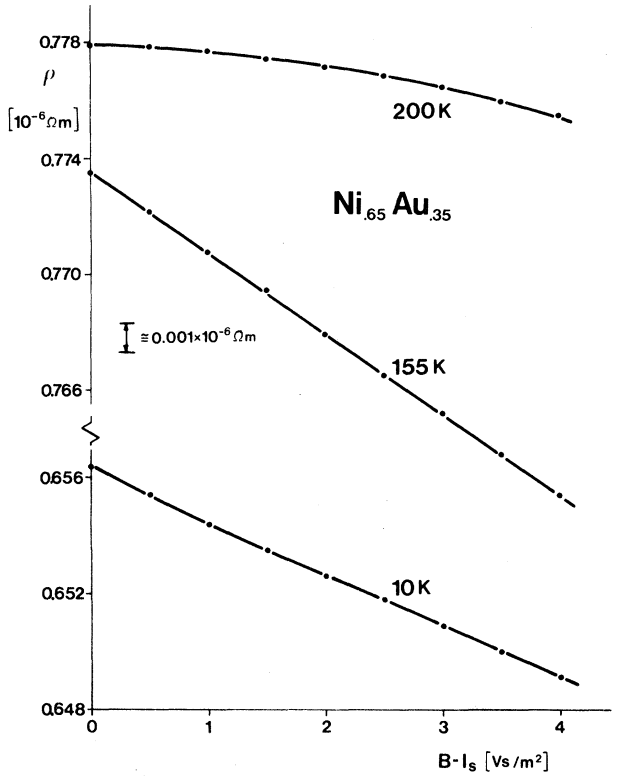

FIG. 2. Magnetoresistance of amorphous $Ni_{0.65}Au_{0.35}$ at $T < T_c$, $T = T_c$, and $T > T_c$.

mental error. If the magnetic field is applied perpendicular to the film, one needs, because of the high demagnetization factor, an external field equal to the magnetization J_s to orient the magnetization parallel to the external field direction.[4] (We use the mks system with $B = \mu_0 H + J$.) Only above J_s does the external magnetic field penetrate the film. Therefore the effective field in the perpendicular case is equal to $B - J_s$ which we use as the abcissa in Figs. 1 and 2. In Fig. 1 the field dependence of the resistivity of $Ni_{0.82}Au_{0.18}$ is plotted as a function of the effective field. The upper part belongs to the amorphous alloy whereas the lower part is measured after the crystallization in the crystalline state. Obviously the magnetoresistance is much larger in the amorphous state. The magnetoresistance is temperature dependent and decreases above the Curie temperature. In Fig. 2 the resistivity of an amorphous $Ni_{0.65}Au_{0.35}$ alloy is plotted as a function of the field for three different temperatures, $T = 10$ K, $T = T_c = 155$ K, and $T = 200$ K $> T_c$. The shape of the curves and the slope change with increasing temperature. For $T < T_c$ the curves intercept the ordinate axis with finite slope whereas for $T > T_c$ the slope of the tangent approaches zero. In Fig. 3 the slopes at a finite field of 40 kG = 4 V sec/m^2 are plotted as a function of the temperature.

The magnetoresistance is essentially restricted to the magnetic state. For small Ni concentrations the ferromagnetism vanishes. At the same time $d\rho/dB\,|_{B=0}$ approaches zero. In Fig. 4 $d\rho/dB$ is plotted as a function of the Ni concentration. The triangles give the Curie temperatures of the amorphous alloys which are measured by the anomalous Hall effect.[4] For $c < 0.5$, the alloys are no longer ferromagnetic and $d\rho/dB\,|_{B=0}$ vanishes. In the vicinity of the critical concentration the ferromagnetic alloys show, however, a particularly

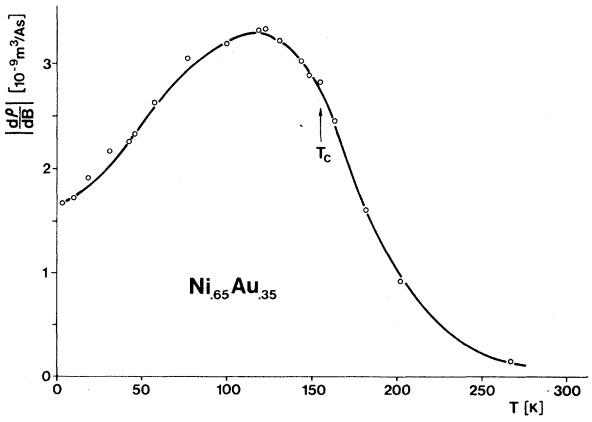

FIG. 3. $|d\rho/dB|$ of amorphous $Ni_{0.65}Au_{0.35}$ as a function of temperature at $B = 4$ V sec/m^2.

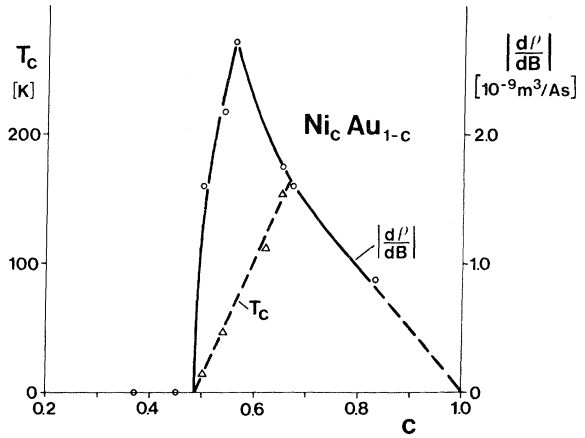

FIG. 4. $|d\rho/dB|_{B=0}$ of amorphous $Ni_c Au_{c-1}$ alloys as a function of the composition. The triangles give the corresponding Curie temperatures.

large magnetoresistance. If we extrapolate the curve to pure amorphous Ni we obtain a very small or a vanishing value for the magnetoresistance. Unfortunately pure Ni cannot be obtained in the amorphous state. However, Co can be forced into the amorphous state by quenched condensation without any additives. Indeed the magnetoresistance of pure amorphous Co is very small, -0.5×10^{-10} m^3/A sec.

III. DISCUSSION

It is characteristic of the present state of ferromagnetism that most of the magnetic phenomena can be discussed in the model of band magnetism as well as in the Heisenberg model. This applies also for the magnetoresistance. Mott[5] described the resistivity of a transition metal in particular by the scattering of the conduction or s electrons into the d band with its large density of states. (See also Blatt.[6]) Alternatively we find within the Heisenberg model a spin-dependent scattering which has been calculated, e.g., by Kasuya,[7] Mannari,[8] and Goodings.[9] In the following we are going to discuss how these different mechanisms yield a magnetoresistance. We begin with the band model.

A. Band model

The band model has, for example, been successfully applied by Chang *et al.*[10] to explain the magnetoresistance of Ni_3Al and Ni_3Ga. According to Mott the essential part of the resistivity is due to the scattering of s electrons into the d states. The probability for the transition into the d states is proportional to the density of states of the d bands. The two d subbands are occupied up to the energies ϵ_+ and ϵ_-. An applied magnetic field will shift the energies ϵ_+ and ϵ_-. The charge transfer from the d band into the s band can be neglected. In addition we assume that the matrix elements for the transition depend only weakly on the energy. If one neglects the orbital momentum of the d states one obtains for the shift of the energies

$$d\epsilon_\pm = \pm 2\mu_B B_0 N_\mp / (N_+ + N_- - U N_+ N_-) . \tag{1}$$

$N_\pm = N_d(\epsilon_\pm)$ are the densities of the d states, U is the exchange energy.

The part of the resistivity which is due to the s-d scattering experiences the relative change

$$\frac{\Delta\rho}{\rho_{sd}} = \left(\left.\frac{dN}{d\epsilon}\right|_{\epsilon_+} d\epsilon_+ + \left.\frac{dN}{d\epsilon}\right|_{\epsilon_-} d\epsilon_-\right)(N_+ + N_-)^{-1}$$

$$\approx \frac{d}{d\epsilon}\frac{N_+ - N_-}{N_+ + N_-}\mu_B B_0 . \tag{2}$$

The right-hand side is obtained by neglecting the Stoner enhancement factor. For the evaluation the details of the band structure are required. Of particular interest is the case in which one of the d subbands is completely filled or empty. In this case $\Delta\rho/\rho_{sd}$ vanishes. Particulary for Ni and alloys of Ni with noble metals the rigid-band model is often used.[11] Here it is assumed that the up band is completely occupied and the down band has about 0.5 holes per atom in the d band which are successively filled by the s electrons of the noble metals. Within the framework of this model one d subband is always completely filled and the magnetoresistance must be negligibly small. Therefore, these measurements contradict the simple rigid-band model. An alternative description of the scattering by transition metal atoms is that of resonance scattering.[12] For liquid and amorphous metals the single-site approximation has been used[13] where each atom is considered as an independent scatterer. The resistivity is essentially proportional to $\sin^2\delta_2$, where δ_2 is the phase shift for the angular momentum $l=2$. In ferromagnetic metals the d-phase shift is spin dependent and for pure amorphous Ni we expect that $\delta_2^\dagger$ is close to π. An applied magnetic field shifts the resonance energies of the spin-up and -down electrons and therefore also the phase shifts. If we use the Breit-Wigner formula for δ_2, then the phase shift in a magnetic field is

$$\tan\delta_2^\pm = \Gamma/[E - (E^\pm \pm \Delta)] , \tag{3}$$

where Γ is half the line width of the resonance and Δ is the energy shift of spin-up or -down electrons and is given by $\mu_B B$ multiplied by the effective Stoner enhancement factor. The relative change in resistivity yields under favorable conditions values only of the order of $\mu_B B/\Gamma$. Assuming Γ of the order of 1 eV, one gets values for the magnetore-

sistance $d\rho/dB \approx 10^{-10}$ m^3/A sec, which is far too small to account for the experimental results.

B. Heisenberg model

Within the Heisenberg model the magnetic part of the scattering of the conduction electrons is essentially described by a term of the form $2w(\vec{r}-\vec{r}_i)\vec{s}\circ\vec{S}_i$. This scattering mechanism is superimposed on the normal potential scattering with the potential $v(\vec{r}-\vec{r}_i)$. The resistivity takes the smallest value when all spins have parallel alignment. If the spins are twisted or flipped the resistivity increases. A negative magnetoresistance can therefore be understood if the spins are initially not fully aligned. Then an additional external magnetic field increases the alignment and reduces therefore the resistivity. The reason for an incomplete alignment of the spins even at $T=0$ can be, for example, local anisotropy fields.

1. Anisotropy effects

Anisotropy fields are mainly due to a destruction of the spatial isotropy of the electron orbits in the presence of its neighboring atoms. The spin orbit interaction transfers the structural asymmetry to the spins.

The explicit calculation of the anisotropy fields for an amorphous transition metal is rather difficult. If the local anisotropy field $\vec{B}_i$ is given, we may determine its influence on the orientation of the spins. We assume that the spins feel a strong molecular field $\vec{B}_w$ in the z direction as well as a small applied magnetic field $\vec{B}_0$. These fields are superimposed on the internal anisotropy fields $\vec{B}_i$. The spin will precess around the resulting direction. The expectation value of S_z is

$$\frac{\langle S_z\rangle}{S} = \frac{B + B_i\cos\theta}{(B^2+B_i^2+2BB_i\cos\theta)^{1/2}}\,, \tag{4}$$

$$B = B_w + B_0\,.$$

Harris *et al.*[14] introduced a local easy direction for the magnetization. Such an easy axis cannot yield a local field opposite to the molecular field. Therefore we take as the local field $\vec{B}_i = B_r\hat{z} + \vec{B}_r$, with $B_r = \text{const}$. (See Fig. 5.) The integration over all directions of B_r yields

$$\frac{\langle S_z\rangle}{S} = 1 - \frac{1}{3}\frac{B_i^2}{(B_w+B_i)^2}\,, \quad \frac{d}{dB_0}\frac{\langle S_z\rangle}{S} = \frac{2}{3}\frac{B_i^2}{(B_w+B_i)^3}\,.$$

The resistivity of an amorphous metal with local moments depends on the orientation of the magnetic moments. We recently calculated the resistivity within the Heisenberg model[15] for the paramagnetic state above the Curie temperature and for the ferromagnetic state where all spins are aligned parallel. The resistivity is increased if we disturb the parallel alignment of the magnetic moments. One obtains in good approximation

$$\frac{d\rho}{dB} = -\alpha\frac{\Delta\rho}{B_w}\,,$$

where $\Delta\rho$ is the measured resistivity difference between the paramagnetic and the ferromagnetic state and α depends on the model of the internal anisotropy field and its relative value compared with B_w. In the model discussed above one finds $\alpha < 0.23$ with the maximum value for $B_r \approx 2.5B_w$. We estimate the upper limit of $d\rho/dB$ for the alloy $Ni_{0.65}Au_{0.35}$ with the Curie temperature of 155 K and $\Delta\rho = 0.14\times10^{-6}\ \Omega$ m and setting $S=\frac{1}{2}$:

$$\left|\frac{d\rho}{dB}\right| = 1.4\times10^{-10}\ \text{m}^3/\text{A sec}.$$

This is about a factor of 10 smaller than the experimental value of 1.7×10^{-9} m^3/A sec. The above calculation yields only an estimate of the effect of local anisotropy and does not completely exclude this explanation. Nevertheless it appears to be rather unlikely, since the estimate is an upper bound and requires very large anisotropy fields.

Another mechanism for the magnetoresistance shall be suggested here, which, according to our knowledge, has not been discussed in the literature yet: the influence of the strong electron-magnon interaction which causes an electron-magnon mass enhancement.[16-18]

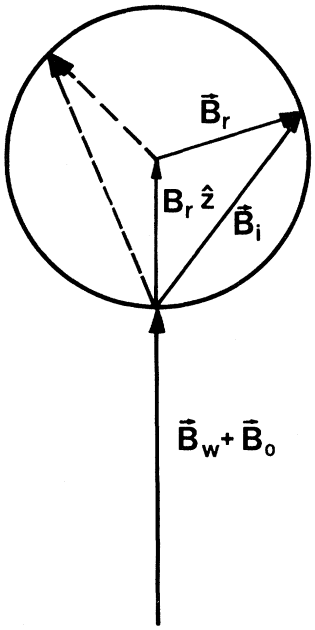

FIG. 5. Local anisotropy fields $\vec{B}_i = B_r\hat{z} + \vec{B}_r$, B_r = const, simulating random easy axis.

2. *Virtual electron-magnon processes*

In the ground state of a metallic Heisenberg ferromagnet all ion spins are aligned parallel and the electrons with spin up and down occupy all states within their Fermi spheres. Excited states can be obtained in many ways. We are going to discuss those excited states which are produced by a spin flip of a conduction electron and the creation of a spin wave. These excited states are particularly interesting because they can be produced by the interaction $w(\vec{r}-\vec{r}_l)\,|2s^z S_l^z + s^+ S_l^- + s^- S_l^+|$. In perturbation theory of second order one finds the new ground state as a superposition of the unperturbed ground state and these excited states. This changes both the number of spin-up and -down electrons and the expectation values of S_z. Both effects change the residual resistivity due to the interaction $2s^z S_l^z$.[19] By applying a magnetic field one can increase the magnon energy by $2\mu_B B_0$. This reduces the influence of the electron-magnon mass enhancement and $\langle S_z\rangle$ approaches S. The contribution of the electron-magnon interaction can be considerable.[20] We are not going to perform these calculations here because of their complexity although it would be very interesting to show whether or not the electron-magnon mass enhancement can be studied by simple magnetoresistance measurements.

IV. CONCLUSION

The magnetoresistance of amorphous ferromagnetic Co and NiAu alloys is negative, linear in the magnetic field, and much larger than in the crystalline phase. We discussed the experimental results within the rigid-band model, the resonance scattering picture, and the Heisenberg model, assuming anisotropy fields. None of the models appeared to be able to yield such a large magnetoresistance as was measured. Finally, we suggested that the electron-magnon interaction and its effect on the magnetization $\langle S_z\rangle$ may be responsible for the magnetoresistance.

ACKNOWLEDGMENT

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[19] This distinguishes the electron-magnon interaction from the electron-phonon interaction, where the renormalization of the density of states and the matrix elements compensate each other.
[20] Estimates yield, for Fe, Co, and Ni, a (50%–75)% admixture of the excited states to the ground state. The coefficient in the T^2 law of the resistivity which is an inverse measure for the stiffness D of the magnon spectrum ($E = Dq^2$) is considerable for the amorphous alloys and increases with increasing Au concentration. For $Ni_{0.65}Au_{0.35}$ we find for $\alpha = |\rho(T) - \rho(0)|/T^2$ the value $\alpha = 1.6\times 10^{-11}\ \Omega\,\mathrm{m\,K^{-2}}$ which is much greater than the value for crystalline pure Ni ($\alpha = 1.6\times 10^{-13}\ \Omega\,\mathrm{m\,K^{-2}}$). Therefore the electron-magnon interaction is rather strong in these amorphous ferromagnetic alloys.