High field magnetoresistance of silver containing rare-earth impurities

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Résumé. — Nous présentons des mesures de magnétorésistance en champ pulsé (300 kG) sur des alliages Ag : Dy, Ag : Ho et Ag : Tm ainsi que l'analyse des contributions dues à la diffusion quadripolaire et à la diffusion d'échange.

Abstract. — We present magnetoresistance measurements in pulsed fields (300 kG) on Ag : Dy, Ag : Ho and Ag : Tm alloys together with the analysis of the contributions from quadrupole and exchange scattering.

1. **General.** — Rare-earth impurities in metals give rise to an anisotropic magnetoresistance (anisotropic meaning different for transverse and longitudinal fields). Studies of this effect in gold and silver base alloys [1, 2] and in magnesium based alloys [3] have shown that the anisotropic magnetoresistance disappears for Gd impurities ($L = 0$) and changes its sign half-way in the heavy rare-earth series, between Ho and Er. This demonstrates that the resistivity anisotropy is due to aspherical Coulomb scattering and the change of sing indicates that the main contribution is from quadrupole scattering.

The experimental results on gold based alloys have been first analysed in a model based on a phenomenological expression of the k-f quadrupolar interaction [2]. In addition the model assumes a spherical potential attracting s and d screening electrons and finds for the resistivity anisotropy

$$\frac{\rho_{\parallel} - \rho_{\perp}}{\rho_0} = 6\,\pi n(E_F)\, D^{(2)} \frac{\sin \eta_2 \cos \eta_2}{\sin^2 \eta_0 + 5 \sin^2 \eta_2} \times \times \left[\langle\langle J_z^2 \rangle\rangle - \frac{J(J+1)}{3} \right] \quad (1)$$

where $D^{(2)}$ is the coefficient of the quadrupole interaction and η_0, η_2 the phase shifts associated with the s and d screening.

Alternatively the resistivity anisotropy in noble metals can be calculated *ab initio*. Fert and Levy [4] have shown that the k-f quadrupole interaction mainly arises via the 5d screening electrons admixed in the conduction band. For example, in the case of a t_{2g} virtual bound state (vbs) they find

$$\frac{\rho_{\parallel} - \rho_{\perp}}{\rho_0} = \frac{18}{7} \sqrt{\frac{3}{35}} \frac{L(S - \frac{7}{4})}{J(J - \frac{1}{2})} \frac{\sin^3 \eta_{2t} \cos \eta_{2t}}{\sin^2 \eta_0 + 3 \sin^2 \eta_{2t}} \times \times \frac{A_2}{\Delta} \left[\langle J_z^2 \rangle - \frac{J(J+1)}{3} \right] \quad (2)$$

where A_2 is related to 4f-5d Coulomb integrals and Δ is the half-width of the vbs.

In addition to the quadrupole contribution to the magnetoresistance there is also an isotropic exchange contribution which can be isolated by considering $\Delta\rho_{Is} = \Delta\rho_{\parallel}/3 + 2\,\Delta\rho_{\perp}/3$. By considering again k-f exchange via a t_{2g} vbs, one finds [4]

$$\frac{\Delta\rho_{Is}}{\rho_0} = \frac{\sin^4 \eta_{2t}}{\sin^2 \eta_0 + 3 \sin^2 \eta_{2t}} \times \times \left\{ \frac{C_1^2}{\Delta^2} [\langle \mathbf{J}.w\mathbf{J} \rangle_H - \langle \mathbf{J}.w\mathbf{J} \rangle_{H=0}] - \frac{C_2^2}{\Delta^2} \frac{\sin^2 2\,\eta_t}{\sin^2 \eta_0 + 3 \sin^2 \eta_t} (\langle J_z^2 \rangle)^2 \right\} \quad (3)$$

where

$$\langle \mathbf{J}.w\mathbf{J} \rangle = \sum_{\mathrm{II}'} P(\mathrm{I}) \frac{\Delta_{\mathrm{I'I}}/k'T}{e^{\Delta_{\mathrm{I'I}}/kT} - 1} \langle \mathrm{I}' | \mathbf{J} | \mathrm{I} \rangle . \langle \mathrm{I} | \mathbf{J} | \mathrm{I}' \rangle$$

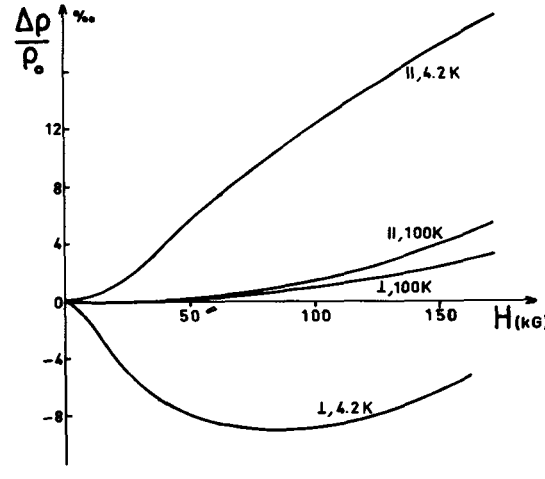

Fig. 1. — Longitudinal and transverse magnetoresistance of 1-at % Ag : Dy alloy at 4.2 K and 100 K.

I, I' are ion states ; C_1 and C_2 are related to 4f-5d Coulomb and exchange integrals. If the moment is quenched by the crystal field and progressively developed by H, $\langle \mathbf{J}.w\mathbf{J} \rangle$ increases with H and $\Delta\rho_{ls}$ can be positive in high fields.

2. **Experimental results.** — The magnetoresistance of silver containing Dy, Ho and Tm impurities ($C \sim 1$ %) has been measured in pulsed fields up to 300 kG at 4.2 K and about 100 K. At 4.2 K $\Delta\rho_{\parallel}$ is positive and $\Delta\rho_{\perp}$ is negative, which characterizes the quadrupole scattering. At 100 K the quadrupole contribution vanishes while the normal magnetoresistance (positive, weakly anisotropic and small) remains. By comparing the results at 4.2 K and 100 K and by using Kohler's rule we have separated the magnetic and normal contributions and finally we have extracted the quadrupole and exchange contributions (figures 2 and 3).

$\langle\langle J_z^2 \rangle\rangle$ has been computed for several values of the crystal-field parameters x and C_4 and the best fit of Eq. (1) to the experimental anisotropy curves is shown on figure 2. From the values of $D^{(2)}$ used for the fit we have determined for each rare-earth the energy E_{qd} characteristic of the quadrupole interaction ($E_{qd} = 5\,J(J - 1/2)\,D^{(2)}$ as in ref. [2]). One obtains 0.31 eV for Dy, 0.13 eV for Ho, − 0.91 eV for Tm. E_{qd} of Dy and Ho ions is about three times smaller than their exchange characteristic energy [5]. E_{qd} of Tm ions is larger than expected by a Stevens scaling and larger than their exchange energy.

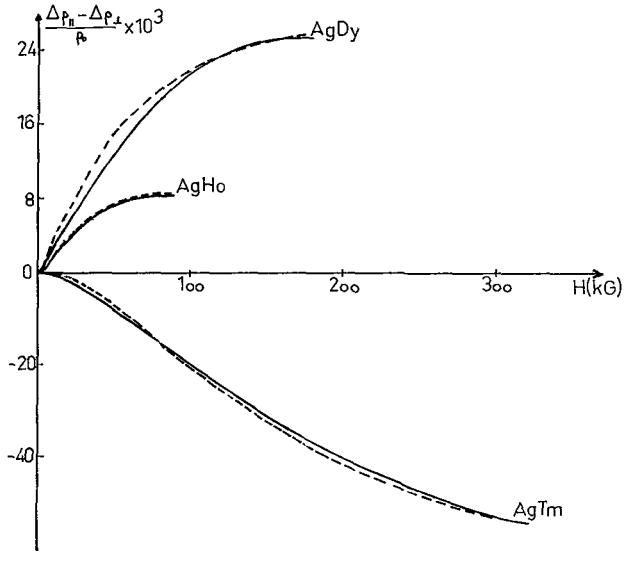

Fig. 2. — Anisotropic magnetoresistance (quadrupole contribution) of several 1 at % alloys at 4.2 K. Solid curves : experiment. Dashed curves : calculation with $x = 0.5$, $C_4 = -50$ K for Dy, $x = -0.37$, $C_4 = -70$ K for Ho, $x = 0.56$, $C_4 = -70$ K for Tm.

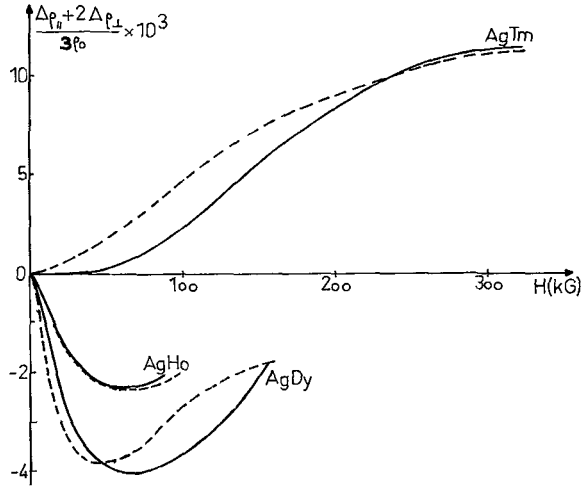

Fig. 3. — Isotropic magnetoresistance (exchange contribution) of several alloys at 4.2 K. Solid curves : experiment. Dashed curves : calculation.

We have also compared our results with the predictions of the *ab initio* calculation of Fert and Levy [4], Eq. (2). We have taken the value of A deduced from atomic Slater integrals, $A = 0.5$ eV and the values of the phase shift derived from the residual resistivity and the thermoelectric power [6]. For Dy and Ho, the calculated resistivity anisotropy is in reasonable agreement with the experimental anisotropy.

Finally, we have analysed the isotropic part of the magnetoresistance (figure 3). The upturn at high field cannot be obtained with a standard exchange interaction but appears in the vbs model, Eq. (3). The fit of figure 3 is obtained with the parameters already used for the resistivity anisotropy and with 4f-5d exchange integrals equal to half their atomic values. This reduction is not surprising owing to the wider extension of the 5d vbs states.

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