Investigation of the spontaneous resistive anisotropy in Cr-based alloys with Fe and Co

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(Received 9 September 1996)

While both Fe and Co impurities carry localized moments in Cr, the behavior of these moments in the spin-density-wave state below T_N is quite different; the Co moments are strongly coupled to the spin-density wave, whereas the Fe moments are not. We have investigated the possibility that such differences *might* arise from the presence of an orbital component in the total impurity moment by examining the anisotropy in the magnetoresistance of several CrFe and CrCo alloys in fields up to 10 T at 4.2 and 2 K. Despite marked differences in this anisotropy in these two systems, the available data indicate that the origin of such behavior does *not* appear to result from the presence of such a component. [S0163-1829(97)03010-5]

The behavior of transition-metal impurities, in general, and Fe and Co, in particular, in a variety of hosts has been the subject of intensive study for several decades. Such studies have investigated the appearance of a stable magnetic moment or long-lived spin fluctuations at the transition-metal site and the interaction between both the impurity moment and the host's conduction electrons (leading to the Kondo effect) and interimpurity coupling resulting in a variety of ordered ground states, displaying predominantly ferromagnetic or spin-glass spin configurations. In those specific cases where the impurity moments have been judged to be stable, efforts have also been made to investigate the presence of an orbital component in the total impurity moment; such investigations have been based primarily on measurements of the *anisotropy* in the magnetoresistance and the anomalous Hall effect.[1] In addition to the well-documented studies of transition-metal impurities in, for example, nonmagnetic noble metals, both Fe and Co are also known to carry localized magnetic moments in the spin-density-wave (SDW) host Cr. This is demonstrated clearly by the observation[2] of a Curie-Weiss contribution to the susceptibility of the corresponding alloys in the paramagnetic phase above the Néel temperature ($T>T_N$), and the presence of such moments in this phase has been invoked to explain the slow depression of T_N on alloying in these systems (a depression which is attributed to local-moment-induced depairing effects, as changes in the electron/atom ratio are expected to increase T_N). By contrast, these local moments exhibit intriguingly different properties in the SDW state below T_N; the Fe moments (or at least some component) remain decoupled from the SDW, exhibiting a Curie-Weiss law for $T<T_N$, a significant field-dependent magnetization $M(H)$, and a marked negative magnetoresistance at 4.2 K, whereas the Co moments appear strongly coupled to the host's spin polarization, yielding a weaker field-dependent magnetization and magnetoresistance at helium temperatures.[2] While models which utilize an effective spin Hamiltonian can predict a weak coupling between a localized impurity moment and the SDW host[3] (which would give rise to a Curie-Weiss contribution to the susceptibility in the ordered phase), the marked difference between the behavior of Fe and Co moments remains difficult to explain. Nevertheless, even in $Cr_{1-x}Fe_x$ ($x\leqslant 0.1$), the low-temperature magnetization remains unsaturated in fields for which B/T exceeds 7 T/K, achieving a value of typically only 10% of that expected from the Curie-law slope.[2,4,5] The magnetoresistance behaves similarly. Models incorporating differences in the coupling strengths of isolated and paired Fe moments with the SDW have been invoked as an explanation of such behavior,[6] but several elements in this model have been judged to be unsatisfactory.[2] Here we investigate the possibility that elements of the behavior mentioned above *might* originate from the presence of an orbital component in the total Fe or Co local moment in this environment. This has been done by examining the magnetoresistance in both the longitudinal and transverse configurations, from which the ''spontaneous'' resistive anisotropy (SRA), that is, the difference between the longitudinal and transverse magnetoresistance extrapolated to zero induction, can be defined via[7]

$$\frac{\Delta\rho(0)}{\rho_0}=\left[\frac{\rho_{\parallel}(B)-\rho_{\perp}(B)}{\rho_0}\right]_{B\to 0}.$$

As has been established previously,[7] the occurrence of a nonzero SRA, whether interpreted in terms of localized or itinerant models, relies on the presence of two essential ingredients: (i) the existence of an orbital component (and associated spin-orbit coupling) in the total moment at the scattering site and (ii) a polarizing field to provide a preferred orientation for such moments; in the present experiment, this is provided by externally applied fields up to 10 T.

Samples of nominal composition $Cr_{1-x}Fe_x$ ($x=0.1$, 0.05, 0.025, and 0.0125), for which the ground-state ordering appears to be the least complicated, and $Cr_{1-x}Co_x$ ($x=0.08$, 0.05, and 0.025) were prepared by successive dilution from the most concentrated specimen in a conventional argon arc furnace using high-purity ($\geqslant$99.99%) starting materials. Each sample was inverted and remelted 6 times to ensure homogeneity. Specimens of approximate dimensions ($0.5\times 2\times 39$) mm^3 were spark cut from the arc-melted buttons, after which they were annealed for 48 h at 900 °C in an argon atmosphere. A low-frequency (37 Hz) differential ratio technique[8] (using a 40 mA exciting current applied along the largest sample dimension) was used to measure the longitu-

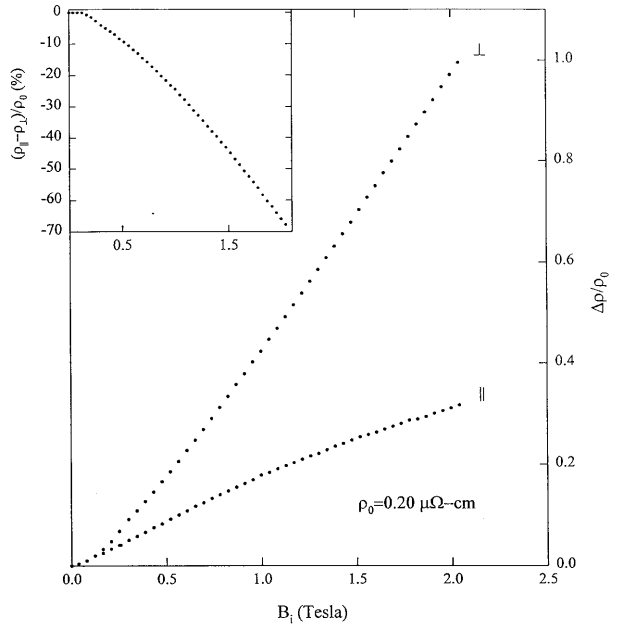

FIG. 1. Longitudinal ($\rho_\parallel$) and transverse ($\rho_\perp$) magnetoresistance of the pure Cr sample at 4.2 K, plotted as a function of internal field (in tesla). The inset shows the anisotropy $(\rho_\parallel-\rho_\perp)/\rho_0$ expressed in percent.

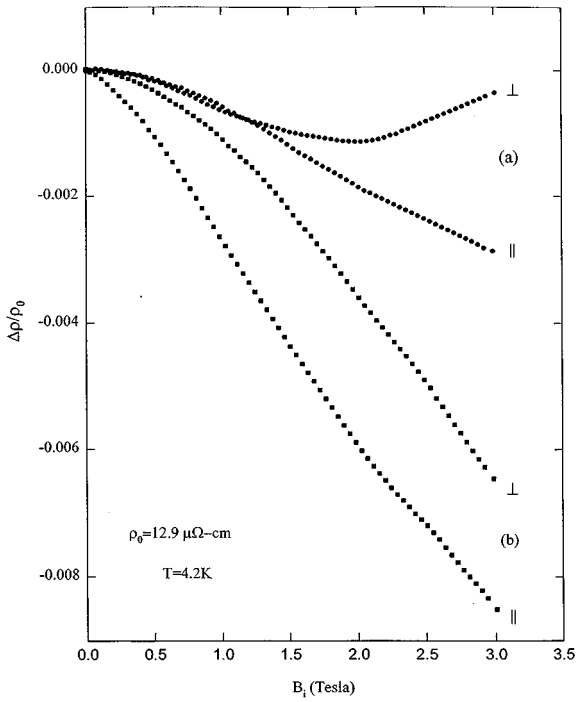

FIG. 2. (a) Measured $\rho_\parallel$ and $\rho_\perp$ of the 2.5 at. % Fe sample at 4.2 K as a function of internal field (in tesla). (b) Magnetoresistance in (a) corrected for the orbital contribution $\Delta\rho_K(B)$ using Kohler's rule. The residual resistivity ρ_0 agrees well with previous measurements (Ref. 11).

dinal and transverse magnetoresistance at 4.2 K and below in applied fields up to 3 T, extended in some cases to 10 T in a second solenoid.

Figure 1 summarizes the magnetoresistance measured in the longitudinal ($\rho_\parallel$) and transverse ($\rho_\perp$) configurations at 4.2 K on the nominally pure Cr sample, plotted as a function of the estimated internal field B ($B=\mu_0(H_a+M)-NM$, M being the magnetization measured[4,5] in an applied field H_a and N being the appropriate demagnetization factor, found by evaluating the appropriate elliptic integral using the measured specimen dimensions;[9] typically, $N_\parallel \approx 3\times10^{-3} \approx 10^{-2}N_\perp$). Data acquired at 2 K are very similar. These magnetoresistivities are strong and positive, amounting to some 30% and 100%, respectively, in the longitudinal and transverse geometries in 2 T, in agreement with previous work;[10] the corresponding anisotropy $(\rho_\parallel-\rho_\perp)$ is displayed in the inset. This strong variation of $\rho_\parallel-\rho_\perp$ with B complicates the analysis of the alloy data, as the following discussion demonstrates.

Figure 2(a) reproduces the measured $\rho_\parallel$ and $\rho_\perp$ in the 2.5 at. % Fe sample at 4.2 K; these data appear to indicate that the magnetic contribution (which gives rise to a negative magnetoresistance[11]) in the transverse configuration saturates in fields exceeding ~1 T. This feature, however, simply reflects the dominant role played by the Cr host (particularly for $\rho_\perp$); these same data, corrected for the orbital term $\Delta\rho_K(B)$ scaled from the Cr magnetoresistivities using Kohler's rule

$$\Delta\rho_K(B)/\rho_0 = f(B/\rho_0),$$

are displayed in Fig. 2(b); this correction amounts simply to scaling both the measured magnetoresistance of the Cr specimen (in the appropriate orientation) and the induction at which it occurs, by the ratio of the zero-field resistivity of the particular alloy to that of the Cr host to generate the corresponding alloy data. From Fig. 2(b) it can be seen that the corrected $\rho_\parallel$ and $\rho_\perp$ remain unsaturated in available fields, in agreement with previous measurements[11] on $\rho_\parallel$ alone. These pseudosaturation effects are not seen in the measured $\rho_\perp$ for the 5 or 10 at. % Fe samples because the orbital contribution (scaled via ρ_0) is much less significant in them. Despite the result, clearly evident in Fig. 2(b), that the corrected $\rho_\parallel$ and $\rho_\perp$ do not saturate in available fields in any of the above samples, we find that the anisotropy in the magnetoresistance $\Delta\rho(B)=\rho_\parallel(B)-\rho_\perp(B)$, though small, does appear to approach a ''limiting'' dependence; in these latter respects, the present system displays several similarities to Pd(FeMn).[12]

To illustrate this point, the differences $\Delta\rho(B)/\rho_0$ acquired at the lowest measuring temperature are plotted as a function of B in Fig. 3; the error bars shown arise principally from uncertainties in $\Delta\rho_K(B)$ through scaling the magnetoresistance of the host. In all the Fe-doped samples, the magnitude of the SRA—the ratio $\Delta\rho(B)/\rho_0$ extrapolated to $B=0$ using principally the high-field data (as in ferromagnetic systems[7])—is quite small, typically $\pm0.1\%$ or less. While there are differences in the detailed behavior near 2 K and at 4.2 K, particularly at low field [as in the 2.5 at. % Fe sample at 4.2 K, Fig. 2(b), and near 2 K, Fig. 3], the extrapolated SRA remains of comparable magnitude at 4.2 K. The SRA estimates from Fig. 3 are plotted against the dopant concentration in Fig. 4 with attendant uncertainty estimates. This figure also includes comparable data from the PdCo, PdNi, PtCo, and PtFe systems to emphasize the objectives of the present investigation. In these latter systems, the SRA estimates reach a nonzero plateau as the dopant concentration is

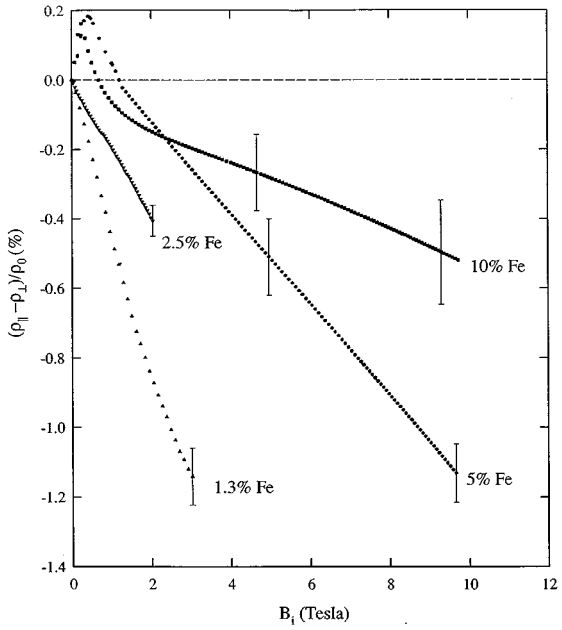

FIG. 3. Anisotropies $(\rho_\parallel - \rho_\perp)/\rho_0$, in percent, plotted as a function of internal field (in tesla), for the 1.3, 2.5, 5, and 10 at. % Fe samples at the lowest measuring temperature (approximately 1.9 K).

reduced, a result indicative of an orbital component in the corresponding localized moment.[1] The uncertainties referred to above notwithstanding, the clear consensus emerging from the present data—if interpreted in a comparable manner—is that these magnetoresistance data appear to indicate that Fe does *not* carry an *orbital* contribution to its total moment in Cr, resembling[1] Fe in Pd (and Mn in Pt).

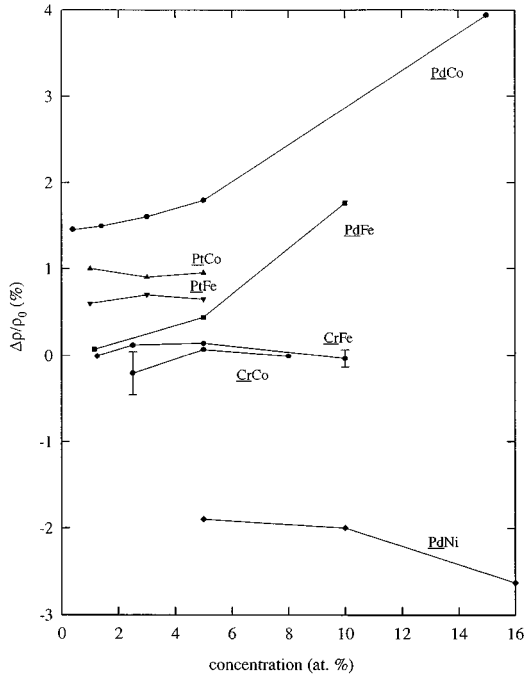

FIG. 4. Estimates of the spontaneous resistive anisotropy (the fractional magnetoresistive anisotropy extrapolated to zero induction) as a function of concentration (at. %) in the CrFe and CrCo, as well as values for PdCo, PdFe, PdNi, PtCo, and PtFe (Ref. 1).

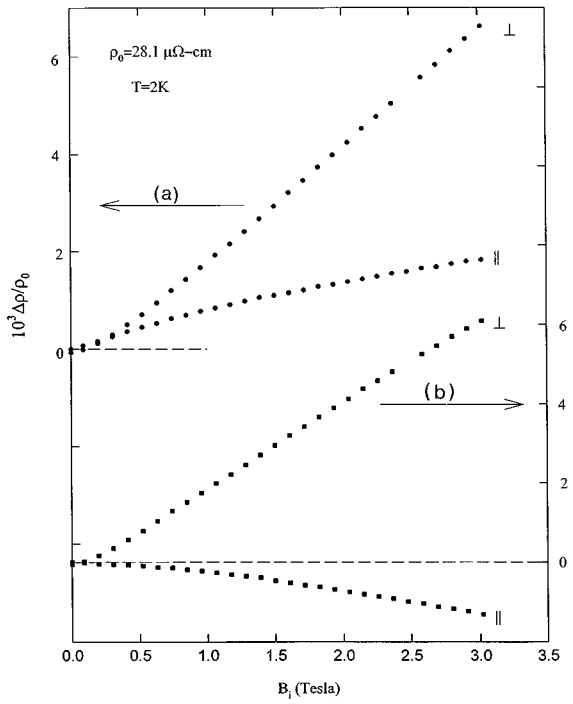

FIG. 5. (a) Measured $\rho_\parallel$ and $\rho_\perp$ of the 5 at. % Co sample at 2 K plotted as a function of internal field (in tesla). (b) Magnetoresistance in (a) corrected for the orbital contribution $\Delta\rho_K$. The residual resistivity is slightly lower than previous estimates (Ref. 10), indicating that the true composition may also be lower.

The behavior of $\rho_\parallel$ and $\rho_\perp$ for the Co-doped systems is generally quite different. The magnetoresistance is much weaker, so that the negative (magnetic) contribution does not dominate the measured response at any composition studied (in contrast to the Fe-doped samples). After correcting for the Kohler term (Fig. 5), a weak negative contribution does emerge in $\rho_\parallel$ for the 5 at. % Co system, in agreement with previous data,[2,11] which has been taken as indicating a strong Co coupling to the SDW. The magnetoresistive anisotropy is summarized in Fig. 6. Despite the differences referred to above, this anisotropy is markedly *larger* in the Co-doped system than in its Fe-doped counterpart; furthermore, it exhibits a different trend with increasing concentration. The magnetoresistive anisotropy appears to have saturated in the 2.5 at. % Co sample, whereas in the 8 at. % Co specimen it has not. However, the associated SRA estimates, as in the Fe substituted system, are quite small; when plotted in Fig. 4, these data also appear to indicate the absence of an orbital contribution to the total moment localized at the Co site.

In summary, measurements of $\rho_\parallel$ and $\rho_\perp$ in fields up to 10 T in Cr samples doped with Fe and Co confirm marked differences in the behavior of these systems, specifically the observation of a strong negative (magnetic) component in the former system, but not the latter, which has been attributed to a strong coupling of the Co moments to the SDW. Measurements of the SRA, despite showing a larger magnitude in the *latter* system, provide no direct support for the possibility that a strong coupling *might* be associated with an orbital component in the total moment localized at the Co site.

Nevertheless, uncertainties still remain over the influence

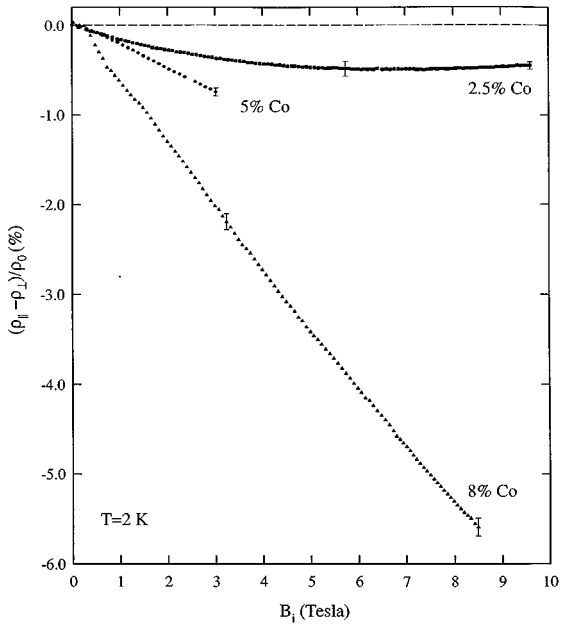

FIG. 6. Anisotropies $(\rho_\parallel - \rho_\perp)/\rho_0$ as a function of internal field for the 2.5, 5, and 8 at. % Co samples at 2 K.

of the polarizing field existing at scattering sites in these systems (the second essential ingredient for a nonzero SRA) and its implications in this context. In both systems studied, an essentially vanishing SRA *might* result even if there were an orbital contribution to the total moment at impurity sites, provided that this component remained locked in an orientation determined by the *local* SDW polarization; the polycrystalline nature of the present samples would then ensure the observed result, in that no unique SDW vector exists in them. This contrasts with the behavior reported for conventional polycrystalline and amorphous ferromagnets.[1,7] Work on single crystals would *not* resolve this situation due to intrinsic anisotropies from other sources that exist in them, as for ''conventional'' SRA.

Clearly the origin of such a strong coupling would need further elucidation, as crystal field and other anisotropy effects do *not* preclude the observation of a SRA in polycrystalline ferromagnetic systems. Furthermore, the differences between Fe and Co impurities would still need to be clarified. Within this admittedly tentative possibility, a strong spin-orbit coupling in addition to a large orbital-moment–SDW interaction would be consistent with the field-independent properties of the Co-substituted system, whereas for Fe impurities some weakening of the spin-orbit coupling would need to occur. The latter would enable the spin contribution (or some subcomponent of it)—but *not* the entire (paramagnetic) moment—to be oriented by external fields, leading to a field-dependent magnetization, a negative magnetoresistance, but no SRA. Such circumstances notwithstanding, the available data presented above do not indicate the occurrence of an orbital component in the total moment localized at Fe or Co sites in Cr, so that such a component does not appear to be the origin of differences in the magnetic behavior of these impurities in this host, viz., of the result, deduced from the temperature dependence of the susceptibility, that the Co moments couple strongly to the SDW, while the Fe moments (or some component of them) do not.

Support for this work from the Natural Sciences and Engineering Research Council of Canada is gratefully acknowledged.

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