

# Extrinsic and intrinsic magnetoresistance contributions of CrO<sub>2</sub> thin films

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The growth of (010)-oriented CrO<sub>2</sub> thin films on Al<sub>2</sub>O<sub>3</sub>(0001) substrates leads to a higher grain boundary density than the growth of (100)-oriented CrO<sub>2</sub> thin films on isostructural TiO<sub>2</sub>(100) substrates. For both types of films an intrinsic linear contribution to the high field magnetoresistance (MR) due to spin disorder has been determined at  $T=300$  K. This contribution does not depend on the crystalline quality of the films and supports the suggested intrinsic double exchange mechanism for CrO<sub>2</sub>. At low temperature ( $T=10$  K) intergrain tunneling MR and Lorentz MR appear, which strongly depend on the crystalline properties of the CrO<sub>2</sub> films.

Strong efforts have been undertaken on the controlled growth of textured and epitaxial thin films of the halfmetallic ferromagnetic CrO<sub>2</sub> since there is an intensive demand of highly spin-polarized ferromagnetic materials for magneto-electronic devices.<sup>1,2</sup> According to Jullière's model<sup>3</sup> the theoretically predicted 100% spin polarization of half-metallic ferromagnetic CrO<sub>2</sub><sup>4</sup> makes it a potential material for high performance magnetic tunnel junctions. Nevertheless, tunnel junctions on the basis of CrO<sub>2</sub> electrodes have not yet been realized. This may be due to the generally observed surface degradation of CrO<sub>2</sub> to the more stable Cr<sub>2</sub>O<sub>3</sub> oxide phase<sup>5</sup> or due to crystal growth limitations of smooth single phase CrO<sub>2</sub> thin films.

In this article the growth of highly textured CrO<sub>2</sub> films on Al<sub>2</sub>O<sub>3</sub>(0001) and TiO<sub>2</sub>(100) substrates will be compared and relevant contributions to the magnetoresistance (MR) will be discussed.

Textured and epitaxial CrO<sub>2</sub>(100) thin films have been grown on Al<sub>2</sub>O<sub>3</sub>(0001) and TiO<sub>2</sub>(100) substrates, respectively, applying a chemical vapor deposition technique.<sup>6</sup> Transmission electron microscopy has been used to investigate the growth modes of CrO<sub>2</sub>(010) and (100) thin films on these substrates.<sup>7</sup> On Al<sub>2</sub>O<sub>3</sub>(0001) substrates the six-fold in-plane symmetry of an initial (0001)-oriented Cr<sub>2</sub>O<sub>3</sub> layer leads to three equivalent in-plane orientations for the {011} plane of the CrO<sub>2</sub> unit cell. This artificial in-plane symmetry of the CrO<sub>2</sub> unit cell leads to a well defined twin (grain boundary) formation as shown in the scanning electron microscopy image in Fig. 1(a).<sup>7</sup> The preferred growth direction of CrO<sub>2</sub> crystallites is the crystallographic  $c$  axis, which lies in-plane and results in a needle-like morphology of the CrO<sub>2</sub> crystallites. In contrast, the growth of CrO<sub>2</sub> on isostructural TiO<sub>2</sub>(100) leads to epitaxial CrO<sub>2</sub>(100) films. The atomic force microscopy image in Fig. 1(b) shows extended CrO<sub>2</sub> crystallites with an overall two-fold symmetry as expected

on isostructural TiO<sub>2</sub>(100) substrates. The in-plane  $c$  axis of the CrO<sub>2</sub> crystallites is indicated in Fig. 1(b).

For investigating the magnetotransport properties of CrO<sub>2</sub> thin films well defined four-probe transport structures have been microfabricated using electron-beam lithography techniques in conjunction with ion milling. Magnetotransport studies have been performed from 4.2 to 300 K in all field

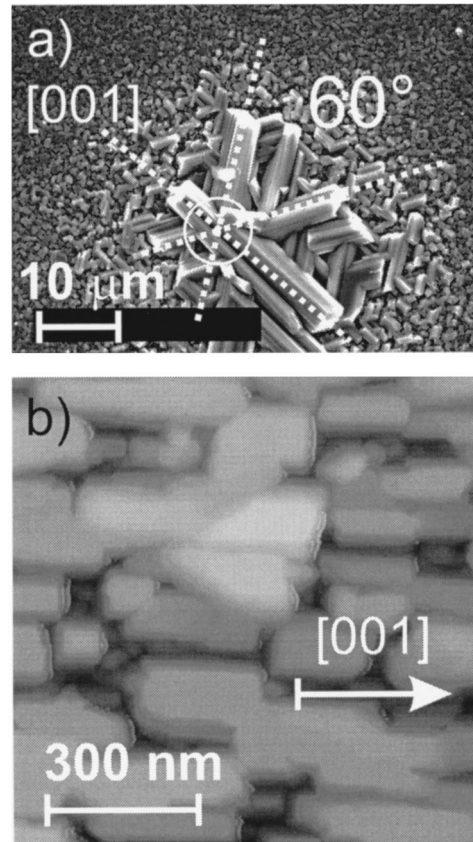


FIG. 1. (a) Scanning electron microscopy image of (010)-oriented CrO<sub>2</sub> crystallites on a initial Cr<sub>2</sub>O<sub>3</sub>(0001) layer (grown on an Al<sub>2</sub>O<sub>3</sub>(0001) substrate). (b) Atomic force microscopy image of (100)-oriented CrO<sub>2</sub> crystallites grown on isostructural TiO<sub>2</sub>(100) substrates.

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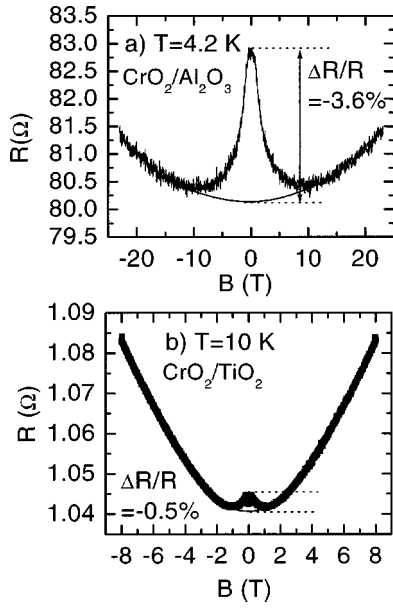


FIG. 2. High field magnetoresistance at of (a) a (010)-oriented  $\text{CrO}_2$  film at  $T=4.2$  K grown on an  $\text{Al}_2\text{O}_3(0001)$  substrate and (b) a (100)-oriented  $\text{CrO}_2$  film at  $T=10$  K grown on a  $\text{TiO}_2(100)$  substrate.

geometries for magnetic fields up to  $\pm 23$  T, allowing the separation of different characteristic contributions to the MR.

A clear indicator of the better crystalline quality of  $\text{CrO}_2$  films grown on  $\text{TiO}_2(100)$  substrates compared to  $\text{Al}_2\text{O}_3(0001)$  is the residual resistivity ratio (RRR) defined as  $R_{300\text{K}}/R_{4.2\text{K}}$ . The RRR of  $\text{CrO}_2$  thin films on  $\text{Al}_2\text{O}_3(0001)$  and  $\text{TiO}_2(100)$  substrates have been determined to 5.9 and 18, respectively.

In magnetic thin films the magnitude of the Lorentz MR can directly be related to the crystalline quality of the films investigated, because it depends on the mean free path.<sup>8,9</sup> In first approximation the field dependent resistivity with the magnetic field perpendicular to the current increases quadratic with the applied magnetic field ( $\rho \sim B^2$ ). Figure 2(a) shows the field dependent resistance of a 400-nm-thick (010)-oriented  $\text{CrO}_2$  film on  $\text{Al}_2\text{O}_3(0001)$  in the perpendicular field geometry ( $I \perp B$ ) at  $T=4.2$  K. Above  $\pm 10$  T a quadratic increase of the resistance with  $B$  appears. This field dependence is extrapolated back to zero external magnetic field (solid line). In comparison with the 400-nm-thick (100)-oriented  $\text{CrO}_2$  film on  $\text{TiO}_2(100)$  [see Fig. 2(b)] the Lorentz contribution to the MR of the (010)-oriented  $\text{CrO}_2$  film on  $\text{Al}_2\text{O}_3(0001)$  is negligible below  $\pm 10$  T. This observation is consistent with the longer mean free path of the (100)-oriented  $\text{CrO}_2$  film on  $\text{TiO}_2(100)$  in the low temperature range. With increasing temperature the Lorentz MR contribution decreases and plays no role for temperatures higher than 100 K.

As known from the analysis of the MR in highly spin-polarized half-metallic manganites,<sup>10,11</sup> intergrain tunneling MR (ITMR) exhibits a low and high field regime. This phenomenon is characterized by an initial drop of the resistivity with applied magnetic field and a following slower decrease at higher fields. In the low field regime the MR is hysteretic and shows its maximum at the coercive field.<sup>12-14</sup>

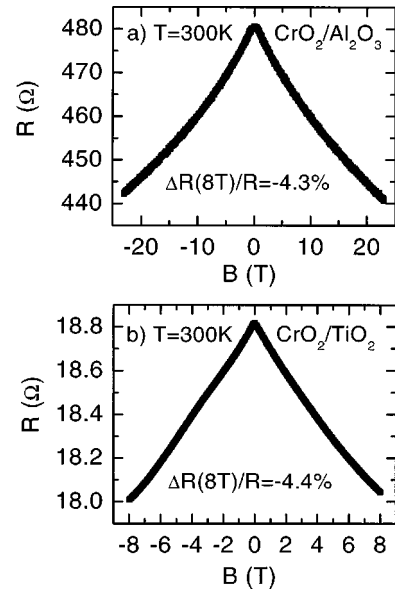


FIG. 3. Linear high field magnetoresistance at  $T=300$  K of (a) a (010)-oriented  $\text{CrO}_2$  film grown on an  $\text{Al}_2\text{O}_3(0001)$  substrate and (b) a (100)-oriented  $\text{CrO}_2$  film grown on a  $\text{TiO}_2(100)$  substrate.

In the low field regime of  $R$  intergrain tunneling MR has been analyzed and correlated with the grain boundary density of the  $\text{CrO}_2$  films.  $\text{CrO}_2$  films on  $\text{Al}_2\text{O}_3$  substrates with a higher grain boundary density show an ITMR of maximum  $-3.6\%$  at  $T=4.2$  K [see Fig. 2(a)], which rapidly decreases with increasing temperatures. Above  $T=150$  K the ITMR is less than  $-0.2\%$ . In contrast, the  $\text{CrO}_2$  films on  $\text{TiO}_2$  exhibit a significantly smaller ITMR contribution of approximately  $-0.5\%$  at  $T=10$  K [see Fig. 2(b)] and is negligible at higher temperatures. The better crystalline property (lower grain boundary density) of  $\text{CrO}_2$  films grown on  $\text{TiO}_2(100)$  results in a significantly smaller MR contribution due to spin-dependent tunneling across grain boundaries.

At 300 K the MR of  $\text{CrO}_2$  films on both types of substrates shows a linear decrease with increasing magnetic field [see Figs. 3(a) and 3(b)]. In order to explain the observed negative MR, the strong coupling between conduction electrons and localized core spins has to be taken into account.<sup>15,16</sup> An increasing magnetic field aligns adjacent spins which leads to an enhancement to the carrier mobility and, therefore, decreases the resistance. This MR contribution is due to the presence of a double exchange mechanism<sup>16</sup> and decreases with decreasing temperature, where spins are more perfectly aligned and spin fluctuations are suppressed. This also explains the presence of this linear and negative MR contribution only at higher temperatures. In case of the (010)-oriented  $\text{CrO}_2$  film on  $\text{Al}_2\text{O}_3(0001)$  [see Fig. 3(a)] at  $T=300$  K magnetic fields of  $\pm 23$  T are not sufficient to suppress these spin fluctuations, i.e., saturating the negative MR. The same observation has been made on the  $\text{CrO}_2$  film grown on  $\text{TiO}_2(100)$  [see Fig. 3(b)]. The magnitude of the observed negative MR [ $R(8\text{T}) - R(0\text{T})/R(8\text{T})$ ] is  $-4.3\%$  for the  $\text{CrO}_2$  film on  $\text{Al}_2\text{O}_3(0001)$  and  $-4.4\%$  for the  $\text{CrO}_2$  film on  $\text{TiO}_2(100)$ . This negative MR contribution seems to be independent of the crystalline properties (grain boundary density) of the

CrO<sub>2</sub> films and supports the suggested double exchange mechanism for CrO<sub>2</sub>.<sup>16</sup>

In summary, extrinsic and intrinsic contributions to the MR of (010)-oriented CrO<sub>2</sub> thin films grown on Al<sub>2</sub>O<sub>3</sub>(0001) substrates and of (100)-oriented CrO<sub>2</sub> thin films grown on isostructural TiO<sub>2</sub>(100) substrates have been determined between 4.2 and 300 K in magnetic fields up to  $\pm 23$  T. The magnitude of the Lorentz MR and the ITMR depends strongly on the defect density of the films. The observed linear and negative MR at higher temperatures ( $T=300$  K) of the same magnitude for films with strongly differing grain boundary density supports the assumption of an intrinsic source of the negative MR related to the double exchange of CrO<sub>2</sub>.

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