Magnetotransport properties of polycrystalline and epitaxial chromium dioxide nanowires

Xiaojing Zou,[1] Gang Xiao,[1,a)] Sunxiang Huang,[2] Tingyong Chen,[2] and Chia-Ling Chien[2]
[1]*Department of Physics, Brown University, Providence, Rhode Island 02912, USA*
[2]*Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218, USA*

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Temperature dependent magnetotransport measurements were performed on polycrystalline and epitaxial chromium dioxide (CrO_2) nanowires fabricated using the selective-area growth technique. Polycrystalline nanowires showed a negative temperature coefficient of resistivity at low temperatures because of strong grain boundary scattering. The magnetoresistance (MR) value exhibited a width dependence, reaching a maximum of 20% for a 150 nm wide wire. In contrast, the MR response of single crystal CrO_2 wires was mainly determined by magnetocrystalline and shape anisotropy. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2836800]

A newly developed form of electronics, known as spintronics, has attracted much attention recently. It uses electron's spin degree of freedom to encode and process data, rather than the electrical charge in the conventional way.[1] Among the magnetic materials being actively investigated in spintronics, chromium dioxide is one of the leading contenders, which is classified as being half-metallic for exhibiting complete spin polarization at the Fermi level.[2] The experimental result from point-contact Andreev reflection has also validated the nearly 100% spin polarization for this material.[3,4]

Furthermore, there is a considerable interest in the fabrication and characterization of magnetic nanostructures. This is motivated by the constant demand for miniaturization in the field of spintronics. The magnetic properties of small structures can differ significantly from the bulk behavior as the sample size becomes comparable to some characteristic length scale, such as the domain wall width or the grain size.[5] Hence, the study of nanosized half-metallic ferromagnets is of great importance to both fundamental spin-related physics and applications in future spintronics devices. In this paper, we report our temperature dependent magnetotransport measurements on polycrystalline and epitaxial CrO_2 nanowires with different linewidths. Different physical mechanisms will be discussed.

In our experiments, epitaxial CrO_2 nanowires were deposited on the (100)-oriented single crystal TiO_2 substrates (obtained from CrysTec at Germany), while the polycrystalline CrO_2 wires were grown using a polycrystalline TiO_2 film, which was made by oxidizing at 800 °C the Ti film sputtered on the SiO_2-covered silicon wafer.

Chromium dioxide grows on rutile-phased TiO_2 substrates with chemical vapor deposition, but not on amorphous SiO_2.[6] Since this material is thermodynamically unstable at atmosphere and easily decomposes into Cr_2O_3, we adopt a so-called selective-area growth technique to fabricate CrO_2 nanowires. This way we avoid the degradation of the CrO_2 quality by various postdeposition etching methods. The detailed fabrication process we used has been reported previously.[7] To summarize, (1) the TiO_2 substrate was first covered by a layer of amorphous SiO_2 (~100 nm) using rf sputtering, and then spin coated with e-beam resist. (2) After e-beam writing and subsequent development, the patterned resist was used as an etching mask for reactive ion etching of the underlying SiO_2 layer in a CHF_3 atmosphere. (3) Finally, the sample was carefully cleaned in acetone and de-ionized water before loading into oxidation furnace to deposit CrO_2. Both polycrystalline and epitaxial CrO_2 nanowires with sub-100-nm width can be obtained with this method.

Figure 1 displays the scanning electron microscopy (SEM) images (accelerating voltage 5 kV) of a polycrystalline and an epitaxial CrO_2 nanowires. As evident, the polycrystalline wire is composed of many CrO_2 grains with different orientations, and neighboring grains are separated by very thin grain boundaries. Since the nanowire was grown using the selective-area growth technique, no postdeposition damage existed, as indicated by the clear crystal border and grain face in Fig. 1(a). However, no obvious grain boundaries are present in the wire shown in Fig. 1(b), which was grown on the (100)-TiO_2 substrate, showing a good single crystal structure.

Figure 2(a) plots the longitudinal resistivity of 150-nm-wide polycrystalline and epitaxial CrO_2 nanowire as a function of temperature, in the presence (4.8 T) and absence of a magnetic field. For the polycrystalline wires, the resistance reaches a minimum at about 215 K, and below this point, a high negative temperature coefficient of resistivity (TCR) is observed. Due to the sharp nonmetal-like increases in resistivity at low temperatures, the residual resistivity ratio, defined as $\rho(300\ \mathrm{K})/\rho(T)$, decreases to about 0.6 at $T=5$ K.

Since polycrystalline CrO_2 nanowire has numerous grains and boundaries in its structure, electron scattering from both inside the grains and across the grain boundaries will contribute to the resistance of the wire. However, the resistivity of epitaxial CrO_2 nanowire is very small at low

a)Electronic mail: Gang_Xiao@brown.edu.

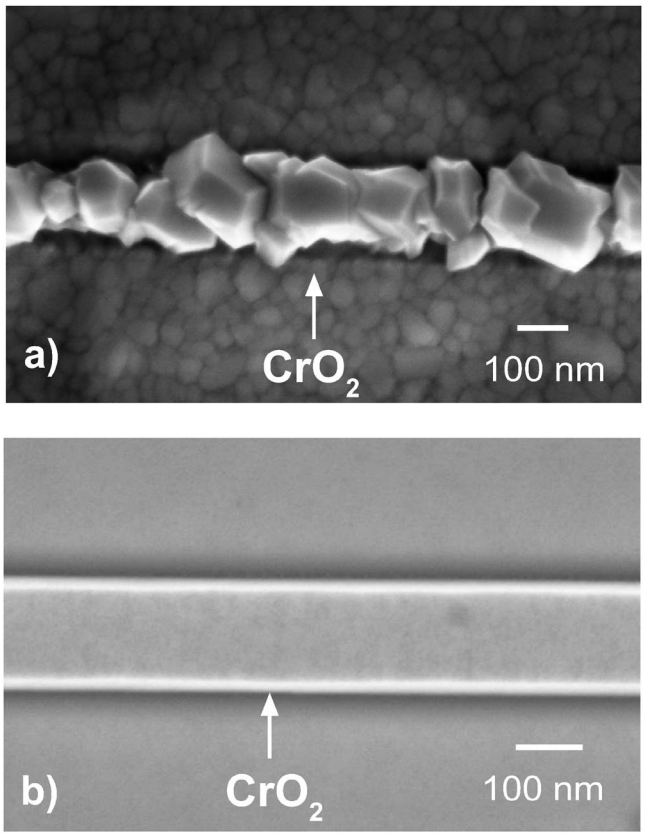

FIG. 1. SEM images of (a) 200 nm wide polycrystalline CrO_2 nanowire and (b) 150 nm wide epitaxial CrO_2 nanowire. These wires have a thickness of nearly 100 nm. The small patterns beside the CrO_2 wire in the upper image are the polycrystalline TiO_2 nanograins under the amorphous SiO_2.

temperature, as shown in Fig. 2(a), implying that the resistivity contribution from inside the grain is negligible. Thus, in our case, the intergrain region is the dominant contributor to the resistivity at low temperatures. Physically, when the electron mean free path (MFP) is comparable to the size of grains, every small grain acts like a potential well with grain boundary as an energy barrier. As a result, the effective conduction electrons are limited to those which tunnel through all the boundaries along the MFP, which leads to the decrease of effective density of conduction electrons. According to a theoretical model by Reiss *et al.*, the dc resistivity of a polycrystalline CrO_2 wire can be approximately written as:[9]

$$\rho = \frac{mv_F}{ne^2 l} P^{-l/D}, \tag{1}$$

where the mv_F/ne^2l term is the conventional Drude formula, where m is the mass of electron, v_F is the Fermi velocity, n is the electron density, and l is the innercrystalline MFP describing the volume scattering of the electrons. The factor $P^{-l/D}$ reflects the total influence of grain boundaries on the resistivity, with D being the mean grain size and P (<1) being the mean transmission probability of the electron through a boundary.

According to Eq. (1), the dc resistivity is highly dependent on the number of grains per mean free path (l/D). For epitaxial CrO_2 wires, which can be regarded as $D \gg l$ and

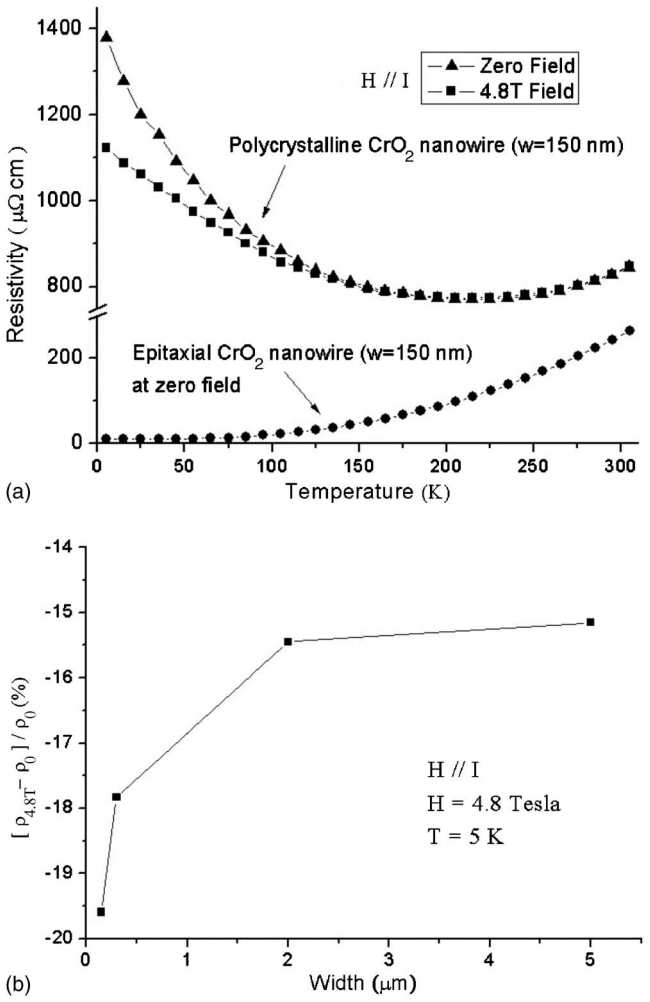

FIG. 2. (a) Temperature dependence of zero magnetic field and high magnetic field (4.8 T) resistivity of a 150 nm wide polycrystalline CrO_2 nanowire with magnetic field applied parallel to the current direction, and the resistivity of a 150 nm wide epitaxial CrO_2 wire with long axis parallel to the [010] direction. (b) Longitudinal magnetoresistance of polycrystalline CrO_2 wires, measured at 5 K and 4.8 T, as a function of linewidth.

$P \approx 1$, Eq. (1) reduces to the normal Drude term mv_F/ne^2l. Increase of the electron MFP at lower temperatures leads to a decrease in the total resistivity (normal metallic behavior). However, in the case of polycrystalline CrO_2 wires, where the mean grain size $(D \sim 100$ nm) is comparable to the electron MFP (70 nm at 5 K),[2] the $P^{-l/D}$ factor cannot be neglected. Furthermore, we can assume that the mean transmission probability P is weakly temperature dependent since it is more closely related to the structure of grain boundaries. Then, the increase in l at low temperature leads to a higher total transmission probability $P^{-l/D}$. The resultant decrease or increase of the resistance with temperature depends on which influence is dominant. In Fig. 2(a) a positive TCR is observed at temperatures above 215 K, which means the electron phonon scattering inside the grains is dominant in this range. Upon decreasing the temperature below 215 K, the grain boundary scattering gradually increases and dominates, giving the TCR a negative sign.

Figure 2(b) shows the longitudinal MR, defined as $\Delta\rho/\rho_0 = [\rho(H) - \rho_0]/\rho_0$, as a function of the linewidth of polycrystalline CrO_2 nanowires measured at 5 K and 4.8 T.

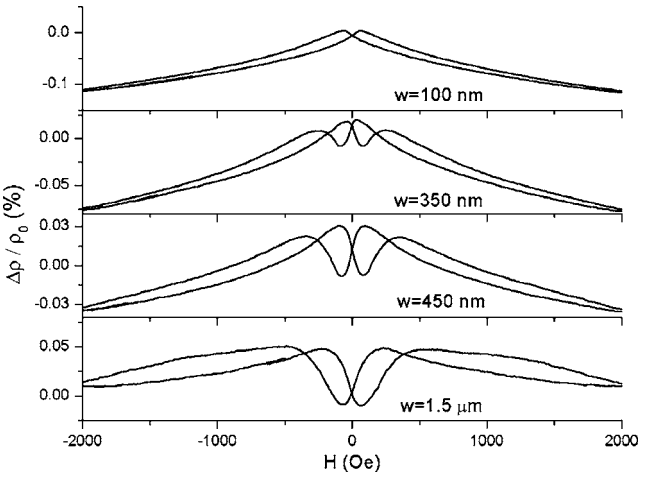

FIG. 3. Low-field transverse magnetoresistance curve for epitaxial CrO_2 nanowires oriented in the [010] direction with different linewidths. This measurement was done in 77 K.

Negative MR values are observed due to the suppression of the electron spin scattering at high field.[8] The magnitude of the MR effect increases upon reducing the linewidth and a maximum value of 19.6% was recorded for a nanowire with a width of 150 nm. This can be explained in the following way: in bulk polycrystalline CrO_2 films, each grain has several neighbors. When a current is applied, the conduction electrons take the path of least resistance by passing through a certain number of boundaries that have relatively high transmission probabilities. However, in the case of nanowires, the number of neighboring grains is significantly reduced, and electrons are forced to travel along the wire axis regardless of the conductivity of the grain boundaries. Since the grain boundaries (scattering centers) are effective spin dependent, the MR value would increase as the wire width decreases.[10,11]

Unlike polycrystalline CrO_2 nanowires, the reduction in the width of epitaxial nanowires has a completely different effect on their magnetotransport properties. Here, the MR behavior is controlled mainly by the magnetic domain wall resistance. The remanent magnetization state of an epitaxial CrO_2 wire grown on the (100)-TiO_2 substrate is determined by two effects: the magnetocrystalline anisotropy favors a magnetic easy axis oriented along the *c* axis ([001] direction),[12] while the shape induced anisotropy prefers the magnetic moment aligned along the axis of the wire. An interesting case is for wires aligned along the [010] direction, where the shape and magnetocrystalline anisotropy compete against each other.

Figure 3 displays the low-field transverse magnetoresistance behavior of epitaxial CrO_2 wires along the [010] direction. As can be seen, the MR response shows a strong linewidth dependence. For the widest wire ($w=1.5$ μm), the contribution from magnetocrystalline anisotropy is dominant which gives it a stripelike domain configuration, with each domain either magnetized parallel or antiparallel to the [001] direction at remanence.[13] At the largest magnetic field (2 kOe), nearly all the magnetic domains are aligned along the external field orientation ([001] direction). On decreasing the field value, some of the domains break into multiple ones with different magnetization directions in order to minimize the total magnetic energy. In this intermediate state, the increase of domain wall scattering leads to a higher wire resistance. As the field approaches zero, the broken multiple domains recombine and the whole wire tends to stabilize at the above mentioned stripelike domain structure, which causes the reduction of the domain walls and leads to the resistance valley near zero field shown in Fig. 3.

However, when the linewidth decreases, the stipelike domain structure (preferred by the magnetocrystalline anisotropy) becomes less stable and the remanent state has a tendency to have the magnetization aligned along the wire axis ([010] direction) due to the increase in the shape induced effect. As evidenced in Fig. 3, the resistance valley becomes less pronounced when reducing the width of the wire and finally disappears for the smallest one ($w=100$ nm). This indicates that the magnetization reversal in the 100 nm wide wire is mainly driven by coherent rotation process, instead of the domain break and recombination which happens in the wires with larger linewidth.

In conclusion, low temperature magnetotransport study of polycrystalline and single crystal chromium dioxide nanowires have been presented. A transformation of temperature coefficient of resistivity from positive to negative was observed for polycrystalline nanowires and their magnetoresistance values increased upon reducing the linewidth. Our results for epitaxial CrO_2 nanowires reflect the change of domain wall configuration due to increase of the shape anisotropy.

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