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Anisotropic magnetoresistance and weak spin-orbital coupling in doped ZnO thin films

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Both out-of-plane and in-plane anisotropic magnetoresistance (AMR) of Cu-doped ZnO thin films with different crystalline orientations are studied. Comparative data of angular dependent AMR suggest that the out-of-plane AMR comes from the geometric effect, while the in-plane AMR can be attributed to the field-dependent path-length effect. Moreover, the small magnitude of AMR and the negligible magnetocrystalline anisotropy suggest that the spin-orbit coupling in Cu-doped ZnO is relatively weak. © *2012 American Institute of Physics*. [doi:10.1063/1.3681795]

Anisotropic magnetoresistance (AMR) is a defining property of magnetic materials in which the resistivity depends on the magnetization orientation with respect to either the electrical current and/or the crystalline axis.[1] AMR in thin films has important applications in technologies such as magnetic field detection and data storage, and the related research often yields important insights on the intricate coupling between charge, spin, and orbital in the materials. In the "ordinary" situation, the dependence of resistance on the relative orientation between the magnetic field and the electrical current is known as the Lorentzian magnetoresistance (MR), which is a result of the Lorentz force acting on the conducting carriers. In some more profound cases, the dependence on the crystalline axis can provide valuable information regarding spin-orbit and magneto-elastic couplings in some materials.

Large AMR has been extensively reported in perovskite manganite and III-V magnetic semiconductors.[2–8] Recently, there has been interest in exploring the magnetic orders in transition metal (TM) doped wide band gap oxides like ZnO,[9–15] but little work has focused on the anisotropic magneto-transport properties of TM-doped ZnO.[16–20] Furthermore, previous results from different groups contradict each other. For example, some researchers found that MR is independent on the orientation of the applied field with respect to the current and/or the crystalline axis, while others claimed that MR is sensitively dependent on the field direction and/or the ZnO *c-axis*.

In this work, we focus on the angular-dependent MR effect in Cu-doped ZnO films. Cu doping does not introduce magnetic impurities or clusters,[21–26] thus helping to eliminate experimental artifacts. So far, however, there has been no report on the AMR effect in Cu-doped ZnO thin films. We carried out a systematic comparative study by growing films on sapphire substrate with different orientations with the *c-axis* of ZnO lying either parallel or perpendicular to the substrate plane. We found that the large out-of-plane AMR sensitively depends on the measurement configuration, while the in-plane AMR could be understood as a Lorentz force deduced path-length effect.

The studied films were grown using pulsed laser deposition at 400 °C on sapphire substrates. The film thickness of 90 nm was determined by using x-ray diffraction (XRD) and reflectivity (Smartlab, Rigaku, Japan). A nominal 5% Cu doping concentration was confirmed by using energy dispersive x-ray spectroscopy (EDS). Two different orientated sapphire substrates (*a-cut* and *m-cut*) were chosen to control the epitaxial growth direction of ZnO, which enabled the study of the transport properties with respect to the *c-axis*.

All the diffraction peaks in the XRD $\theta-2\theta$ and the reciprocal space mapping (RSM) data (Fig. 1) correspond to the ZnO phase with a wurtzite structure. The epitaxial relationship has been confirmed to be (001)ZnO//(110)Al_2O_3, (110)ZnO//(001)Al_2O_3 for films grown on *a-cut* (110) Al_2O_3 and (100)ZnO//(100)Al_2O_3, (001)ZnO//(110)Al_2O_3 for films grown on *m-cut* (100) Al_2O_3. It should be pointed out that the ZnO films grown on *m-cut* Al_2O_3 have a non-polar surface with *c-axis* aligned parallel to the substrate surface, while the ZnO films grown on *a-cut* Al_2O_3 have a polar surface with *c-axis* along the normal direction.

The transport properties of patterned Cu-doped ZnO films were measured in a physical properties measurement system (PPMS). All the studied samples maintain a semiconductor-like transport behavior (Fig. 2(a)). The measurement configuration for the out-of-plane AMR is shown in the inset of Fig. 2(a). As the field rotates in the plane perpendicular to the current direction, the magnitude of the conventional Lorentz force remains constant. In a previous report,[19] the AMR in Co-doped ZnO was linked to the relative orientation between the magnetic field and the *c-axis* of ZnO. However, as can be seen from Figs. 2(b) and 2(c), the two-fold symmetric angular dependence of the out-of-plane AMR for both films with different crystalline orientations is identical. Since the *c-axis* of ZnO is parallel to the substrate surface for the film grown on *m-cut* sapphire while it is perpendicular to the substrate surface for the film grown on *a-cut* sapphire, we can conclude that the large out-of-plane AMR more likely originates from the geometric effect instead of the magneto-crystalline anisotropy. Furthermore, we have patterned the Cu-doped ZnO film grown on *m-cut*

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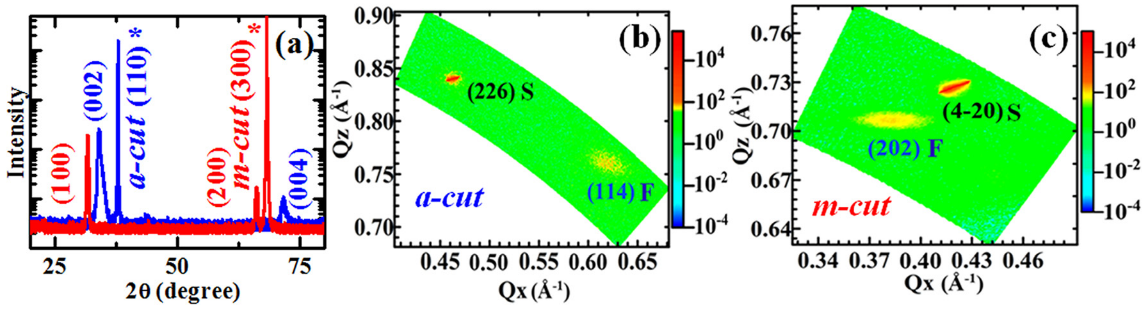

FIG. 1. (Color online) XRD *θ-2θ* (a) and the corresponding RSM results for the Cu-doped ZnO films grown on *a-cut* (b) and *m-cut* (c) sapphire substrates. "S" and "F" stand for substrate and film, respectively.

Al_2O_3 into a cross structure using lithography, which enables the current flow either along or perpendicular to the *c-axis* of ZnO. The orientation was confirmed by XRD in-plane phi scan in prior to the patterning. We did not observe any notable difference (Fig. 2(a)), which further confirms that the magneto-transport has no dependence on the current flow direction with respect to the ZnO *c-axis*.

In the scenario of geometric effect, the resistance is higher (lower) when the applied field is parallel (perpendicular) to the film surface. The angular dependence can be well explained by a phenomenological uniaxial anisotropic model,[27] which gives $\rho(\theta)/\rho(0) = \alpha(\alpha^2\cos^2\theta + \sin^2\theta)^{-1/2}$. Here, $\alpha \equiv \rho_{\perp}^{out}/\rho_{//}^{out}$ represents the uniaxial resistivity ratio, where $\rho_{//}^{out} \equiv \rho(\theta = 0)$ ($\rho_{\perp}^{out} \equiv \rho(\theta = 90)$) denotes the resistivity for field parallel (perpendicular) to the substrate normal direction. In line with the previous works, we believe that the observed AMR between $\rho_{//}^{out}$ and $\rho_{\perp}^{out}$ mainly reflects the difference in current path through the sample, which leads to variations in the effective localization and scattering of free carriers.[19,28] In particular, when the applied field is in plane, the Lorentz force is out of plane, thus there is more surface and interface scattering, which results into a higher resistance state. On the other hand, when the applied field is out of plane, the force is in plane, thus the surface and interface scattering is less pronounced, which leads to a lower resistance state. A larger magnetic field increases the local magnetization, boosting the AMR (Fig. 3(a)), while a higher temperature makes the film more uniform, leading to a decrease of spin-dependent scattering and AMR (Fig. 3(b)). The good agreement between the theoretical fitting and the experimental results indicates that the underling physics has been adequately captured: the geometric effect can explain the AMR in doped ZnO films without invoking the magneto-crystalline effect.

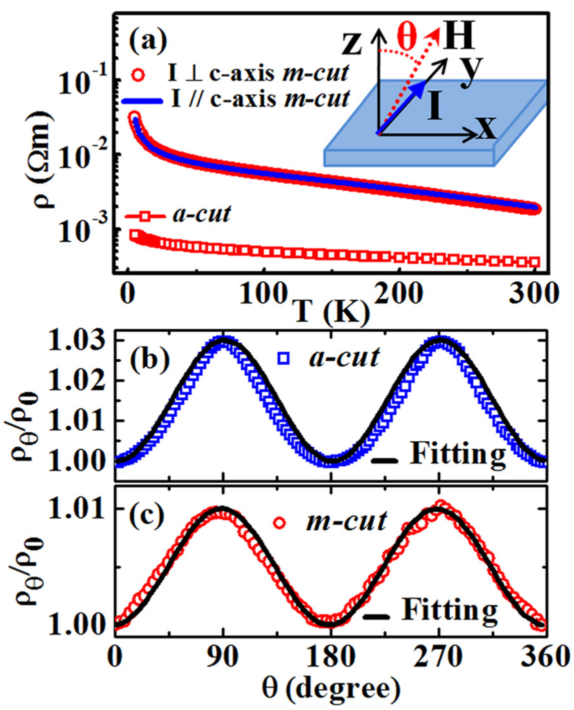

FIG. 2. (Color online) (a) Resistivity versus temperature curves for the Cu-doped ZnO films grown on *m-cut* and *a-cut* sapphire substrates. Inset shows the schematic of the measurement configuration for the out-of-plane AMR, where the magnetic field rotates in the xz plane while the current flows in the y direction. (b) and (c) show the normalized out-of-plane MR, ρ_θ/ρ_0, measured at 5 K under 1 T for films grown on *a-cut* and *m-cut* sapphires, respectively. Solid lines are the theoretical fittings as discussed in the main text.

In the measurements of in-plane AMR, a magnetic field of 7 T was applied to ensure the orientation of magnetization is identical with that of the applied field. In this measurement configuration, both Lorentz force and spin-orbital coupling could contribute to the observed AMR. Experimentally, both Cu-doped ZnO films with different growth orientations show the same $\sin^2\varphi$ dependence, indicating that the in-plane AMR only depends on the angle between magnetization and current with $\rho_{\perp}^{in} > \rho_{//}^{in}$. This is consistent with the Lorentz force induced path-length effects. For field perpendicular to

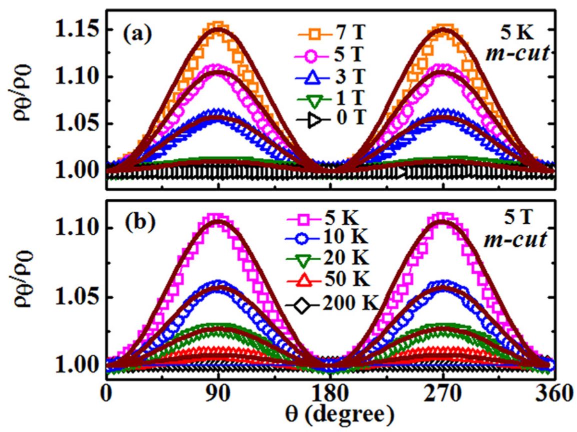

FIG. 3. (Color online) Angular dependence of the out-of-plane AMR measured at 5 K under different applied fields (a) and at different temperatures under a 5 T field (b). The Cu-doped ZnO film was grown on *m-cut* sapphire. Solid lines are the theoretical fittings.

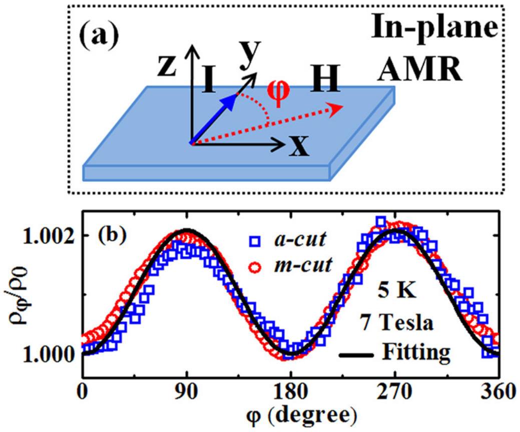

FIG. 4. (Color online) (a) Schematic of the experimental configuration for the measurement of in-plane AMR, where H rotates in the xy plane. (b) In-plane MR, ρ_φ/ρ_0, for the Cu-doped ZnO film grown on two different sapphire substrates. Solid line is the theoretical fitting.

current, the current path through sample is extended as a result of the circular movement of carriers between two scattering events and leads to a high resistance state.[28]

Beside the Lorentz force effect, scattering anisotropy due to the spin-orbit interactions can also lead to the $\sin^2\varphi$ dependence in the in-plane MR, which has been invoked to explain the AMR data of both manganites and ferromagnetic alloys.[4,29] According to the microscopic theory, the sign and the magnitude of AMR can be described by the formula: $(\rho^{in}_{//} - \rho^{in}_{\perp})/\rho_{average} = \gamma(\beta - 1)$, where γ is a spin-orbit coupling constant, and β is the ratio between the spin-up and the spin-down resistivities.[30] The positive AMR is generally regarded as a consequence of scattering of the majority-spin electrons, while the minority-spin electrons are responsible for the negative AMR.[30] However, the conduction band in *n*-type Cu-doped ZnO is formed from *s* electrons, which should lead to a much smaller spin-orbit coupling compared to that in other *p*-type materials like Mn-doped GaAs.[6,18] Furthermore, the very small in-plane AMR (0.2% at 5 K and 7 T) in our sample compared to that of *p-type* Mn-doped GaAs (10% at comparable measurement conditions) unambiguously indicates that the spin-orbit coupling in Cu-doped ZnO is rather weak.[2]

In summary, the angular dependence of both the out-of-plane and in-plane AMR of Cu-doped ZnO thin film with two different growth orientations was systematically studied. Clearly, MR in doped ZnO thin films depends on not only the microscopic interactions between the carriers and the host, but also the measurement geometry and the resulting current path through the samples. In particular, the AMR depends on the orientation of the applied magnetic field with respect to the film normal direction instead of the crystalline axis of ZnO, indicating that the magneto-crystalline anisotropy does not play a significant role in ZnO. Furthermore, we found the angular-dependent in-plane MR in ZnO is very small, indicating that the spin-orbital coupling in ZnO is rather weak and not capable to cause any notable anisotropy in magneto-transport.

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