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Space-charge limited conduction in epitaxial chromia films grown on elemental and oxide-based metallic substrates

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ABSTRACT
We study temperature dependent (200 – 400 K) dielectric current leakage in high-quality, epitaxial chromia films, synthesized on various conductive substrates (Pd, Pt and V$_2$O$_3$). We find that trap-assisted space-charge limited conduction is the dominant source of electrical leakage in the films, and that the density and distribution of charge traps within them is strongly dependent upon the choice of the underlying substrate. Pd-based chromia is found to exhibit leakage consistent with the presence of deep, discrete traps, a characteristic that is related to the known properties of twinning defects in the material. The Pt- and V$_2$O$_3$-based films, in contrast, show behavior typical of insulators with shallow, exponentially-distributed traps. The highest resistivity is obtained for chromia fabricated on V$_2$O$_3$ substrates, consistent with a lower total trap density in these films. Our studies suggest that chromia thin films formed on V$_2$O$_3$ substrates are a promising candidate for next-generation spintronics.

I. INTRODUCTION
The magnetoelectric (ME) chromia (Cr$_2$O$_3$) is an antiferromagnetic oxide, whose bulk order parameter may be manipulated by the application of an electric field. At the same time, this dielectric is known to exhibit a robust boundary magnetization (at its (0001) crystalline surface), the direction of which may be controlled by the application of a suitable electric field. Recently, this boundary magnetization has been exploited to implement an all electrical scheme for the exchange biasing of a nearby magnetic multilayer, a result that represents a critical step for attempts to develop electrical control of magnetism for spintronics. This approach of voltage control of magnetism (VCM) has the potential to replace existing approaches to spintronics (such as spin transfer torque), which typically rely on the use of energetically-costly electrical currents for magnetic switching. In contrast, the use of even large electric fields to switch the boundary magnetization of chromia should (ideally) arise without any associated current, allowing VCM to be achieved at low power levels (the reader is referred to Ref. 7 for a recent benchmarking of chromia-based spintronics.)

Chromia is a linear ME that exhibits a transition from the paramagnetic to the antiferromagnetic phase at a Néel temperature of $T_N = 307$ K, making it one of the few materials to exhibit magnetoelectricity at room temperature. Its boundary magnetization onsets with the appearance of the antiferromagnetic order, and is found to be insensitive to the presence of roughness. Dependent upon the oxygen partial pressure utilized during the growth process, and on the impurities present within the material, chromia can be either a $p$-type or an $n$-type semiconductor. In bulk form, at least, its large bandgap of ~3 eV results in high intrinsic resistivity ($\rho \sim 10^{16}$ $\Omega$cm.
at 300 K, a key requirement for the implementation of low-power schemes for VCM. For practical devices, however, it is necessary to realize chromia in thin-film form, and a key challenge here is to meet this objective while maintaining the dielectric character of the material. Previously, thin-film growth has been implemented by a number of methods, with significant variations being reported in the resistivity of the resulting films. Published values vary from $10^2$ to $10^7$ Ω cm, with resistivities at the lower end of this range being obtained by sputter deposition, while those at the upper end are synthesized via pulsed-laser deposition. Most importantly, there have recently been successful reports of VCM for Cr$_2$O$_3$ films with resistivity values as low as $10^6$ Ω cm. This places a useful lower constraint for the purpose of future device development.

While the discussion above makes clear the motivation for utilizing chromia in future spintronics, this field nonetheless remains at a nascent stage, with a strong need to optimize the electrical properties of this material. This is particularly true for the thin-film form of chromia, whose properties (as noted already) are already known to be strongly sensitive to the details of preparation. Motivated by this, we have undertaken a detailed electrical characterization of thin chromia films, deposited on various substrate materials (Pd, Pt and V$_2$O$_5$). These substrates were chosen due to their wide use in oxide-film synthesis, and exhibit different lattice mismatch (3.9% for Pd, 3.1% for Pt and 0.1% for V$_2$O$_5$) with chromia. Consequently, we are concerned with understanding the manner in which this influences the resulting dielectric properties of the chromia films. We find clear evidence for space-charge limited conduction (SCLC) in the films, and show that the details of this are sensitive to the choice of the underlying substrate. Most notably, the highest resistivity is obtained for chromia films fabricated on V$_2$O$_5$ substrates, consistent with the improved lattice matching to this material. We furthermore calculate various electrical properties for chromia, including its effective mobility, its total trap density, and its substrate-dependent trap distributions and energies. Based on these studies, we suggest that chromia thin films formed on V$_2$O$_5$ substrates are a promising candidate for next-generation ME spintronics.

II. ELECTRICAL CONDUCTION IN DIELECTRICS

To aid the analysis and understanding of our experimental results, in this section we give a brief overview of electrical-conduction mechanisms in thin dielectrics. SCLC is widely exhibited by such materials, and arises when current through the insulator becomes limited by the build-up of charge injected from the source electrode. This mechanism dominates once the applied voltage ($V$) reaches a characteristic value ($V_0$), at which point the number of injected charge carriers in the vicinity of the source becomes equivalent to the number of thermally-generated ones. At smaller voltages ($V < V_0$), in contrast, thermal carriers provide the dominant contribution to the current, and ohmic conduction is consequently obtained. In the presence of these two mechanisms, the total current density may follow the functional form:

$$ J = aV + \beta V^2, $$

where the parameters $a$ and $\beta$ are associated with ohmic transport and SCLC, respectively. The defining equations for these parameters can be expressed as:

$$ a = \frac{q\mu}{d}, $$

$$ \beta = \frac{9\mu e^2\varepsilon_0}{8d^2}. $$

In these expressions, $n$ is the density of thermally generated charge carriers, $q$ is the electronic charge, $\mu$ is the carrier mobility, $d$ is the film thickness, $\varepsilon_0$ is the dielectric constant and $\varepsilon_0$ is the permittivity of free space. $\Theta$ is the ratio of free carriers to the total carrier concentration (i.e. free plus trapped charges). Assuming $n$-type material, this ratio can be expressed as:

$$ \Theta = \frac{N_t}{g_nN_t}\exp\left(\frac{E_t - E_c}{k_BT}\right), $$

where $g_n$ denotes the degeneracy of energy states in the conduction band, $N_t$ is the total trap density, $N_c$ is the effective density of states in the conduction band, $E_t$ is the trap energy, $E_c$ is the energy at the conduction band edge, $k_B$ is the Boltzmann constant and $T$ is the temperature. In samples free of traps, the parameter $\Theta = 1$. In the presence of traps, however, the value of $\Theta$ is voltage dependent, ultimately reaching unity once the last trap is filled.

Quite generally, the interplay of ohmic transport with SCLC leads to a four-stage conduction mechanism in dielectrics, as illustrated schematically in Fig. 1(a). Starting from the situation at...
low bias, these mechanisms are as follows: (1) ohmic-like conduction ($I \propto V^{\alpha}$, $\alpha = 1$); (2) trap-limited SCLC ($\alpha = 2$); (3) the trap-filled limit (reached at voltage $V_{TFL}$, $m > 2$); and (4) trap-free SCLC ($m = 2$). Dependent upon the nature of the defects in the material, a variety of $I-V$ curves may therefore be exhibited. In an insulator free of traps, for example, only stages (1) and (4) will be observed. With discrete traps present (with energy $E_T$, see Fig. 1(b)), on the other hand, charge will begin to fill the traps at $V_T$ (see Fig. 1(a)), an estimate for which can be derived by equating both terms on the right side of Eq. (1) and determining the $\Theta$ term from Eq. (4). In this way it can be shown that the resulting value for $V_T$ can be expressed as:

$$V_T = \frac{8\pi d^2 \varepsilon_0 \varepsilon_{rel} N_t}{9 \varepsilon_r \varepsilon_0} \exp \left( - \frac{E_T - E_F}{k_B T} \right).$$

where $E_F$ denotes the Fermi level. From this relation it is apparent that $V_T$ increases with increasing temperature. At voltages larger than this value, the second ($\beta$) term on the right-hand side of Eq. (1) dominates the current, as injected carriers overwhelm thermally-generated ones. However, once all traps are completely filled, implying that the quasi-static Fermi level is precisely aligned with the generated ones. However, once all traps are completely filled, implying that the quasi-static Fermi level is precisely aligned with the generated ones. However, once all traps are completely filled, implying that the quasi-static Fermi level is precisely aligned with the generated ones. However, once all traps are completely filled, implying that the quasi-static Fermi level is precisely aligned with the generated ones. However, once all traps are completely filled, implying that the quasi-static Fermi level is precisely aligned with the generated ones. 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FIG. 2. (a) The main panel plots the temperature-dependent $I$-$V$ characteristics of epitaxial chromia grown on Pd. Open circles are experimental data points (only 10% of which are shown) and solid lines are fits to the form of Eq. (1). The inset shows the same data, with current plotted on a logarithmic scale. (b) The main panel plots the temperature-dependent $I$-$V$ characteristics of epitaxial chromia grown on Pt. The inset shows the same data, with current plotted on a logarithmic scale. (c) The main panel plots the temperature-dependent $I$-$V$ characteristics of epitaxial chromia grown on $V_2O_3$. The upper-left inset shows the temperature-dependent resistivity of chromia on Pd, Pt and $V_2O_3$, calculated at an electric field of 33 kV/cm. The lower-right inset shows the data of the main panel, with current plotted on a logarithmic scale.

Turning to a more detailed analysis of the current variations exhibited by the different films, for $Cr_2O_3$ on Pd we find that the $I$-$V$ curves are well described by the form of Eq. (1). To illustrate this, in Fig. 2(a) we have used this relation to perform a two-parameter ($\alpha$ & $\beta$) fit to the measured currents. The fits are indicated by the solid lines and closely follow the experimental data, which are plotted using the open circles (for clarity, only 10% of data points are shown). The $\beta$ term enables an estimate of the effective mobility ($\mu\Theta$, see Eq. (3)), and in the main panel of Fig. 3 we plot the variation of this quantity as a function of temperature. Much like the behavior reported for bulk chromia, and for numerous other insulators (See Ref. 12 and references therein), we find that this mobility increases exponentially with increasing temperature. Another parameter that may be inferred from the fits is the voltage ($V_{tr}$) at which SCLC exceeds ohmic conduction (defined as $V_{tr} = \alpha/\beta$, see Eq. (1)). For these particular films, $V_{tr}$ increases with increasing temperature, a result that is consistent with the prediction of Eq. (5) for SCLC in the presence of discrete traps. To establish this connection explicitly, in the inset to Fig. 3 we plot $\ln(V_{tr})$ vs. $1/k_B T$ and find that the resulting data fall on a straight line. The slope of this line yields information on the trap energy ($E_T$), and from the data in the figure we obtain $E_T \sim \approx 50$ meV. While the value of the Fermi level in this material is not known a priori, its insulating character would tend to suggest a position somewhat close to midgap. Consequently, the inferred value of $E_T$ points to the presence of deep traps, located well away from the conduction band. This conclusion can be reconciled with the results of a recent structural study, which has demonstrated a preponderance of twinning defects in chromia when it is grown on Pd. Calculations, based on density functional theory, suggest that these defects may give rise to a local reduction of the bandgap (by as much as 65%), in those regions corresponding to the boundary between neighboring, twinned domains. Such a reduction could be considered to be consistent with the presence of deep traps.

To understand the nature of the current variations exhibited by the other two types of film, in Fig. 4 we replot their $I$-$V$ curves on double-logarithmic scales. In each of these panels, we observe a transition from ohmic conduction ($I \propto V^m$, $m \approx 1$) to SCLC ($m \geq 2$), as the magnitude of the applied voltage is increased. In contrast to the behavior exhibited by $Cr_2O_3$ on Pd, the voltage scale ($V_{TFL}$) associated with this transition is found to shift to smaller values with increasing temperature. Such behavior is consistent with expectations for materials whose traps are distributed exponentially within the gap (see Fig. 1(c) & Eq. (6)). Behavior of this kind has

FIG. 3. The main panel plots the temperature-dependent variation of the effective mobility ($\mu\Theta$, see Eq. (3)), for the Pd-based chromia system. The inset plots the transition voltage ($V_{tr}$) as a function of temperature for the same film.

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The I-V curves for (a) chromia on Pt and (b) chromia on V$_2$O$_3$, plotted on double-log scales to reveal the connection to Fig. 1(a). The different dotted lines are included as guides to the eye and denote different power-law variations of current (with index $m$) as a function of voltage. Left/right panels in both figures are for positive/negative voltage.

been reported previously for numerous dielectric materials, and is especially clear in the V$_2$O$_3$-based films (Fig. 4(b)), in which the power-law exponent ($m = l + 1$) associated with SCLC increases from around 3 to 7 as the temperature is lowered over the indicated range.

In the insets Figs. 5(a) and 5(b), we plot the variation of the power-law index ($l$) as a function of $1/T$. The resulting data fall on a reasonable straight line, consistent with the definition of $l$ in Eq. (7). The slope of the resulting straight line allows an estimate of the characteristic range ($E_t$) of the exponential trap distribution, and from the insets in Fig. 5 we infer $E_t = 27$ & 90-meV for Cr$_2$O$_3$ on Pt and on V$_2$O$_3$, respectively. In contrast to the case of the Pd-based films, these values correspond to trap distributions that are relatively shallow, lying close to the conduction band.

With an exponential distribution of traps, it is possible to make an estimate of the total trap density ($N_t$), by extrapolating the space-charge limited component of the current back to larger voltages. As we demonstrate in the main panels of Fig. 5, this yields a temperature-independent fixed point ($V_c$) at which the various I-V curves intersect one another. To obtain an analytical solution of $V_c$, we follow the treatment in Ref. 47 by writing Eq. (7) in an Arrhenius form to isolate the temperature dependence of the current. After some algebraic calculations, Eq. (7) may then be expressed as:

$$f(l) = \left(\frac{2l+1}{l+1}\right)^{l+1} \left(\frac{l}{l+1}\right)^{\frac{l}{2l+1}}.$$  

(9)

At the voltage $V_c$, the space-charge limited current is temperature independent and the form of Eq. (8) implies that:

$$V_c = \frac{qN<td>^2}{2e<\epsilon_r_\epsilon_0}.$$  

(10)

By making use of Eq. (10), and the experimentally determined values of $V_c$ from Fig. 5, we obtain the total trap densities $N_t = 2.1 \times 10^{19}$ cm$^{-3}$ & $3.2 \times 10^{18}$ cm$^{-3}$ for Cr$_2$O$_3$ on Pt and on V$_2$O$_3$, respectively. These values fall within the range expected of high-quality insulating thin films. Interestingly, we note that the trap density is an order of magnitude smaller for Cr$_2$O$_3$ on V$_2$O$_3$, which might be related to the improved lattice matching (with just 0.1% mismatch) in this system.

V. DISCUSSION

In this report, we have demonstrated how it is possible to use temperature-dependent measurements of current leakage in epitaxial chromia films, grown on different conducting substrates, and to infer quantitative information on the microscopic characteristics of their defects. The studies here suggest a fundamental
difference between chromia films grown on Pd, and those formed on Pt and $V_2O_3$ substrates, with the former being dominated by deep, discrete traps while the latter are characterized by the presence of exponentially-distributed, fairly shallow traps. This difference is consistent with what we have learned previously, from structural investigations of these films. These have shown that growth on Pd results in films with a preponderance of twinning defects, which function as highly-effective leakage paths. Theoretically, the leakage has been attributed to a 65% reduction of the chromia bandgap, which arises at the boundaries between neighboring twinning domains. At the same time, our analysis has shown that the leakage in these particular films may be attributed to the presence of deep traps, a result that could be consistent with such bandgap narrowing at localized positions. Our earlier work showed that the twinning defects can be suppressed through the growth of chromia on either Pt or $V_2O_3$, consistent with which the microscopic nature of the defects in these films is found to be very different to that in $Cr_2O_3$ on Pd. The resulting traps are much shallower in energy, and distributed exponentially within the forbidden gap, in contrast to the discrete, deep traps inferred for $Cr_2O_3$ on Pd. The presence of such different trap distributions should, perhaps, not be altogether surprising; while the twinning defects are suppressed by growing chromia on either Pt or $V_2O_3$, these materials are nonetheless known to exhibit dislocations that may function as an alternative source of trap(s).

VI. CONCLUSIONS

In conclusion, we have measured the temperature dependent (200 – 400 K) dielectric breakdown of high-quality, epitaxial chromia films, synthesized on various conductive substrates (Pt, Pd and $V_2O_3$). We find evidence of SCLC in all of these materials, Pd-based chromia is found to exhibit behavior consistent with the presence of deep, discrete traps. The Pt- and $V_2O_3$-based films, in contrast, show behavior typical of insulators with shallow, exponentially-distributed traps. The total trap density is approximately an order of magnitude smaller in the $V_2O_3$-based films, a characteristic that appears consistent with the fact that the resistivity in this film system approaches that of bulk chromia. Overall, our results suggest that $V_2O_3$-based $Cr_2O_3$ is a promising candidate for the implementation of next-generation spintronic devices.

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