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$LaAlO_3/SrTiO_3$ interfaces doped with rare-earth ions

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Doping the $LaAlO_3$ side of the $LaAlO_3/SrTiO_3$ interface with 2% Tm or Lu does not significantly affect its electron transport. Also, at low temperatures, carrier mobility is steeply anticorrelated with carrier concentration, for both doped and undoped interfaces. This relationship cannot be explained by ionized impurities alone but may be driven by positive charge in or on the LAO film.

From the quantum Hall effect to the field-effect transistor, two-dimensional electron gases (2DEGs) have been of both fundamental and technological interest. Historically, 2DEGs have been studied in covalent sp^3-band semiconductors. With advances in complex oxide heteroepitaxy, 2DEGs have also been created at interfaces between d-band oxides, such as the $LaAlO_3/SrTiO_3$ (LAO/STO) interface.[1] Because of the d band's relatively narrow bandwidth, the 2DEG at the LAO/STO interface exhibits strong electron-electron correlation effects not observed in conventional semiconductors.[2–6] Unfortunately, LAO/STO's low-temperature carrier mobilities (10 to 10^4 cm^2/Vs) are still far inferior to those of the best covalent semiconductor 2DEGs (as high as $4 \times 10^7 cm^2/Vs$).[8,9] A central puzzle for the field is to understand precisely why carrier mobility is so low in LAO/STO and how it can be improved.

Over the last decade, a number of research groups have managed to boost carrier mobility in LAO/STO by a variety of methods, such as lower growth temperatures,[10] capping layers,[11] polar solvents,[12] high substrate miscut angles,[13] and conducting force microscope.[12] Yet despite these improvements, no one has conclusively identified the primary defect or mechanism limiting carrier mobility.

To investigate how defects limit carrier mobility in LAO/STO, some researchers have experimented with purposely doping chemical impurities at and near the interface.[14–21] Most doping studies have doped the STO side of the interface with transition-metal dopants. Unfortunately, the results are difficult to interpret because the dopants trapped carriers, thus altering carrier concentration at the same time as they altered carrier mobility. More recently, transition metals have also been doped into the LAO side. These studies show little effect for low doping concentrations, but when the doping concentrations were high enough to distort the LAO lattice, electrical conductivity was suppressed.[21]

In our experiment, we attempted to modify carrier mobility by doping the LAO side of the interface with isovalent rare earth ions. In particular, we chose to dope 2% of the La sites with the rare-earth elements Tm or Lu. First, because Tm and Lu are isovalent with La^{3+}, they should not donate or accept carriers. Second, because Tm and Lu have more protons than La (and hence larger atomic spin-orbit coupling), they may enhance spin-orbit scattering. And third, because Tm^{3+} has a magnetic moment of 2 μ_B, it may affect magnetic scattering.

Our main result is that doping the LAO side of the LAO/STO interface with 2% Tm or Lu has little effect on electron transport, even at low temperatures and in high magnetic fields. This result places upper limits on some types of interface scattering in LAO/STO. Our secondary result is that at low temperatures carrier concentration is anticorrelated with mobility, for both doped and undoped interfaces. This anticorrelation is too steep to be explained by ionized impurity scattering alone but may be explained by charged defects in or on the LAO film. Together these results show that electron transport in LAO/STO interfaces is robust against doping the La site with rare-earth ions, up to the 2% level.

We synthesized LAO/STO interfaces using pulsed laser deposition. LAO films were deposited onto (001) TiO_2-terminated STO substrates at 700°C and in 3×10^{-5} Torr of O_2. To deposit the films, we focused a KrF laser at 2 Hz with an average energy density of 1.3 J/cm^2 onto sintered ceramic targets of the following compositions: $LaAlO_3$, $Tm_{0.02}La_{0.98}AlO_3$, or $Lu_{0.02}La_{0.98}AlO_3$. The target-heater distance was 76 mm. Our previous growth study of these doped samples indicated that the films had excellent crystallinity.[20] X-ray absorption and Rutherford backscattering spectroscopy confirmed the presence of Tm and Lu respectively in the doped samples. Film thicknesses ranged from 1 nm to 20 nm. Atomic force microscopy verified that the surfaces exhibited unit-cell steps. Magnetotransport measurements were performed in a Quantum Design Physical Property Measurement System in magnetic fields of up to 7 T and at temperatures down to 2 K. Samples were ultrasonically wire-bonded with Al wires in the van der Pauw configuration. Carrier concentrations and mobilities were calculated using the Hall slope at low magnetic fields.

Because the rare-earth dopants Tm and Lu might increase magnetic scattering or spin-orbit scattering, we looked first for their effects on magnetoresistance (MR). We measured the sheet resistance and carrier concentration of doped LAO/STO samples and found little difference compared to undoped LAO/STO samples. The

MR of a typical Lu-doped sample is shown in Figure 1 (Tm-doped and undoped samples are very similar). At low magnetic fields, the MR is parabolic with respect to magnetic field, consistent with ordinary magnetoresistance. However, at high magnetic fields, the MR appears to shift to a linear regime. Interestingly, this linear MR appears to be orbital in nature: when plotted as a function of mobility times magnetic field, the MR curves collapse onto a single function, in accordance with Kohler's rule[22] (see the lower plot of Figure 1). This high field linear MR is in agreement with measurements from other groups[23] and has also been seen in gated and ion irradiated STO,[24–26] but lacks explanation as far as we know. Theories of linear MR such as quantum linear MR,[27] classical linear MR,[28,29] topological linear MR,[30] berry-phase linear MR,[31] phase-coherence linear MR,[32] and anisotropic-layered-metal linear MR[33] do not seem relevant to the LAO/STO interface.

In addition to measuring MR with the magnetic field directed out-of-plane, we also measured MR with the magnetic field directed in-plane along the <110> direction. As with the out-of-plane MR, the in-plane MR of our doped LAO/STO is very similar to that of undoped LAO/STO. At low temperatures, the in-plane MR is negative, in the range of -10% to -50% at 7 T. The temperature dependence of resistance and Hall curves is also similar among doped and undoped samples.[20]

When the Hall curves are nonlinear (ignored by some papers but addressed by others), calculating carrier concentrations and carrier mobilities is not straightforward (see Figure 2). Assuming only a single carrier type, we calculated carrier concentration from the Hall slope at low magnetic fields. Although simple, this assumption seems justified because using it produces agreement with Kohler's rule, as shown in the lower plot of Figure 1. Even if there are multiple carrier types in reality, the agreement with Kohler's rule implies that the MR is dominated by just a single carrier type.

Using this procedure for calculating carrier concentration and mobility, we can compare our doped LAO/STO with undoped LAO/STO. Figure 3 shows the carrier mobilities and carrier concentrations of our Tm-doped and Lu-doped LAO/STO interfaces (measured at 2 K), plotted alongside undoped interfaces from our group and others (measured between 0.5 K and 15 K). Although our doped interfaces have slightly higher carrier mobility and lower carrier concentration than our undoped interfaces, the difference is smaller than the typical variance of undoped LAO/STO interfaces, making it difficult to attribute the difference to the dopants rather than target variability. Overall Figure 3 makes clear that the rare-earth dopants in the LAO did not have a large effect on low-temperature carrier mobility or carrier concentration.

One of the most interesting relationships in our data is the steep anticorrelation between carrier mobility and carrier concentration at low temperatures (noted before by Wong et. al.[34] and Xie et. al.[12] for undoped

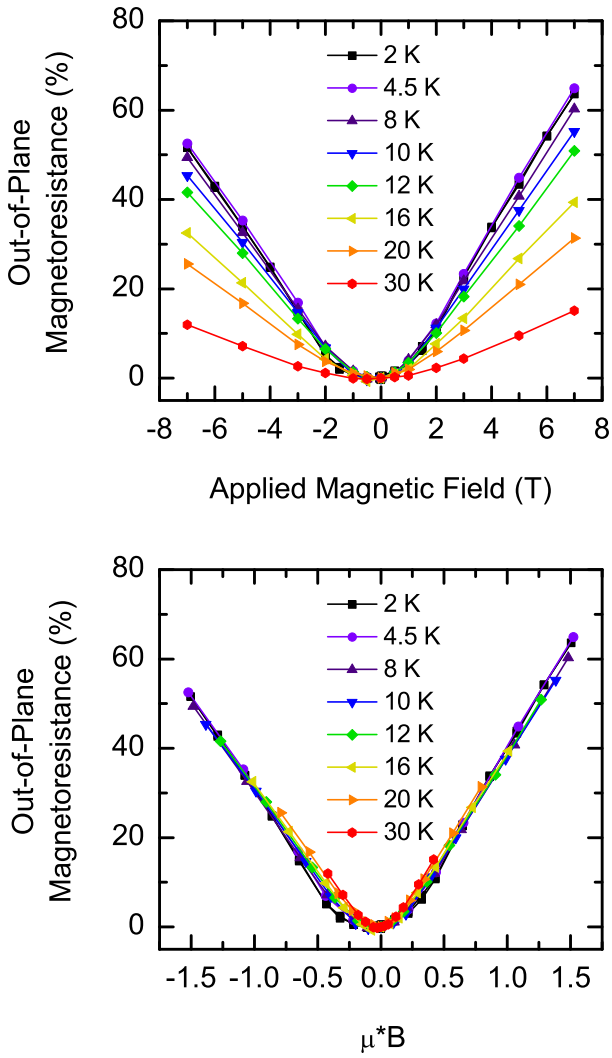

FIG. 1. Upper: The out-of-plane magnetoresistance of a typical Lu-doped LAO/STO interface is parabolic at low field and linear at higher field, similar to undoped LAO/STO. Lower: Plotting the magnetoresistance against mobility times magnetic field ($\mu * B$) instead of B mostly collapses the magnetoresistance curves onto a single universal curve, implying that the magnetoresistance is orbital in nature and mostly due to a single carrier type.

LAO/STO). The anticorrelation is robust: we see it in both our doped and undoped samples, as well as in samples from most other groups, who use a variety of growth temperatures (600 °C to 850 °C), oxygen pressures (10^{-5} Torr to 10^{-2} mbar), oxygen post-annealing procedures, LAO thicknesses (4 unit cells to 37 unit cells), laser energy densities (0.6 J/cm^2 to 2 J/cm^2), laser pulse frequencies (1 Hz to 10 Hz), laser spot sizes (2 mm^2 to 10 mm^2), and electrical measurement techniques.[12,13,34–42]

The steepness of the anticorrelation between carrier mobility and carrier concentration gives clues about its cause. Fitting a simple power law to our data shows that carrier mobility roughly varies as carrier concentration to

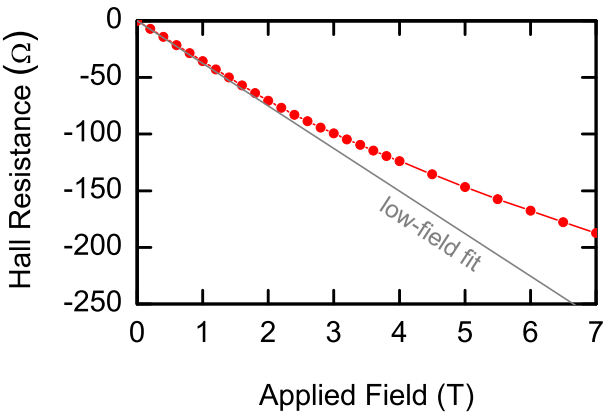

FIG. 2. The Hall resistance of a typical Lu-doped LAO/STO interface at 2 K (Tm-doped and undoped interfaces are very similar). Although its nonlinearity suggests multiple carrier types, magnetoresistance measurements suggest that conductivity is dominated by the high mobility carriers.

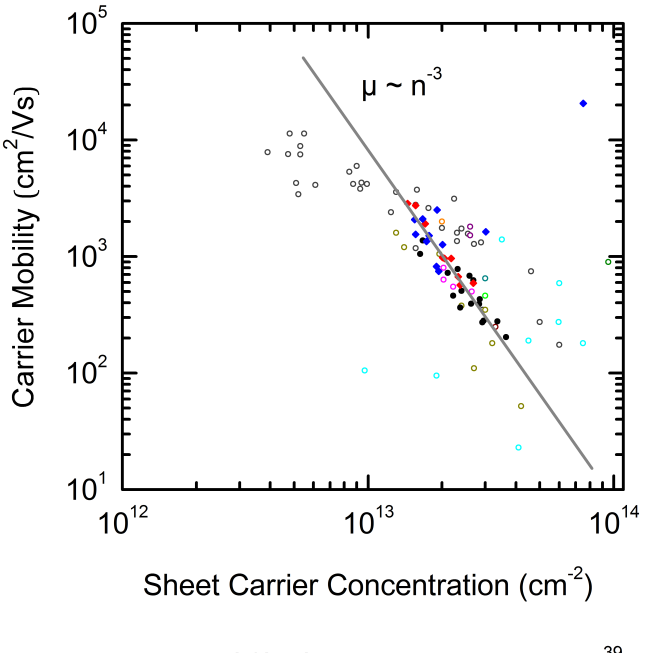

FIG. 3. At low temperatures, carrier mobility μ is steeply anticorrelated with carrier concentration n. This is true of both our doped and undoped LAO/STO, as well as for undoped LAO/STO from other groups.[12,13,34–42] A rough power law fit to our own data shows that $\mu \propto n^{-3}$.

the power of -3, a power so negative that it rules out otherwise plausible explanations such as ionized impurities. Ionized impurities, which both donate and scatter carriers, are often responsible for anticorrelations between carrier mobility and carrier concentration. However, ionized impurities normally result in a power law exponent of only -1, because changing the density of ionized impurities affects both carrier concentration and scattering proportionally. And in reality, the slope from ionized impurities is even shallower because additional carriers will screen the scatterers (as is the case in bulk STO, which has a power of about -0.8).[7,46,47]

Another plausible explanation for the anticorrelation is the two-band (or multi-band) model.[48] The key idea is that as more carriers are added by some unspecified means, they fill a second, low-mobility band, bringing down the average mobility of the carriers. However, by itself, this explanation cannot explain the power of -3, because no matter how low the mobility of the second band, in an independent-electron band picture adding additional carriers ought to increase conductivity rather than reduce it. (Of course, if the two-band model is combined with strong electron-electron scattering then adding carriers would reduce the conductivity, in agreement with our data.)

In general, any explanation that relies only on static defects is likely incorrect, given that other groups have been able to *dynamically* tune LAO/STO samples along this anticorrelation curve. These dynamic techniques include using a conducting atomic force microscope,[12] top gating through the LAO film,[43,44] and using polar solvents.[12,45] One simple fact links these dynamic techniques: they work by modifying the electrostatic potential from the LAO side of the interface. (Also, we emphasize that backgating through the STO substrate, which modifies the electrostatic potential from the STO side, does not tune samples along this anticorrelation curve. In fact, backgating results in a positive correlation between carrier mobility and carrier concentration.[49] For this reason, backgated samples have not been included in Figure 3.)

This observation supports the notion that the anticorrelation between carrier mobility and carrier concentration is driven by the amount of positive charge in or on the LAO film (possibly due to oxygen vacancies or other defects). One possible explanation is the previously discussed two-band model combined with strong electron-electron scattering. In this model, positive charge attracts low-mobility carriers, which then strongly scatter the high-mobility carriers, strongly dropping their mobility. A second possibility relies on interface scattering. As more positive charge in the LAO attracts more carriers, it also causes a larger interfacial electric field, pulling carriers closer to the interface where they experience more scattering and a consequently lower mobility. This phenomenon may be strongly amplified by the dielectric sat-

uration of STO,[50] perhaps explaining why mobility varies so dramatically relative to the carrier concentration.[51]

Lastly, we note that these hypotheses, which rely on positive charge in or on the LAO, are consistent with our result that rare-earth dopants in the LAO have little effect on electron transport. If mobility is indeed determined by charged defects in or on the LAO film, then isovalent dopants would be expected to have no effect on carrier mobility, as our measurements confirm. Lastly, some theoretical papers have suggested that Ti-La hopping may be important for binding the 2DEG to the LAO/STO interface.[52,53] However, our null results imply that Ti-La hopping is negligible, at least for electron scattering.

In conclusion, doping 2% of the La sites with Tm or Lu does not significantly alter the electron transport of the LAO/STO interface. This result is consistent with the idea that carrier mobility is determined by the positive charge in or on the LAO film. Also, as with undoped LAO/STO, we observe a steep anticorrelation between carrier mobility and carrier concentration at low temperatures. This steep anticorrelation cannot be explained by ionized impurities but seems to be driven by positive charge in or on the LAO film.

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