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Authors

Moetakef, Pouya
Jackson, Clayton A.
Hwang, Jinwoo
[et al.](#)

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Toward an artificial Mott insulator: Correlations in confined high-density electron liquids in SrTiO₃

Pouya Moetakef,¹ Clayton A. Jackson,¹ Jinwoo Hwang,¹ Leon Balents,² S. James Allen,³ and Susanne Stemmer¹

¹Materials Department, University of California, Santa Barbara, California 93106-5050, USA

²Kavli Institute of Theoretical Physics, Santa Barbara, CA 93106-4030, USA

³Department of Physics, University of California, Santa Barbara, California 93106-9530, USA

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We investigate correlation physics in high-density, two-dimensional electron liquids that reside in narrow SrTiO₃ quantum wells. The quantum wells are remotely doped via an interfacial polar discontinuity and the three-dimensional (3D) carrier density is modulated by changing the width of the quantum well. It is shown that even at 3D densities well below one electron per site, short-range Coulomb interactions become apparent in transport, and an insulating state emerges at a critical density. We also discuss the role of disorder in the insulating state.

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Strongly correlated (“Mott”) insulators defy simple models that predict metallic transport. An early triumph of theory was to recognize that Coulomb repulsion can localize the conduction electrons.¹ *Long-range* Coulomb interactions are important at low carrier densities typical for many semiconductors. *Short-range* interactions dominate if the probability for two electrons to occupy a single site becomes significant, which requires high electron densities,^{2–4} and result in metal-to-insulator transitions in materials with narrow bands. The pathways between this insulating state and the conducting Fermi liquid, and associated phenomena, such as high- T_c superconductivity, are subjects of intense study in condensed matter physics. In real materials, lattice distortions, and charge and magnetic ordering often accompany the transition, making it difficult to isolate effects from electron correlations.

Oxide interfaces that exhibit a polar discontinuity allow for two-dimensional electron liquids (2DELs) with unprecedented charge densities.^{5,6} For example, the GdTiO₃/SrTiO₃ interfaces shown in Fig. 1 produce a 2DEL with 1/2 electron per interface unit cell, and a sheet density of $\sim 3.3 \times 10^{14} \text{ cm}^{-2}$ per interface.⁷ Here, such extreme electron densities are introduced into the d bands of SrTiO₃ (see Ref. 7), a material that is a band insulator in bulk. In heterostructures that contain two such interfaces, carrier concentrations in sufficiently narrow SrTiO₃ quantum wells may approach one electron per site and the conditions for Hubbard-type strong correlation physics. Because the interface dopes the quantum well,⁸ high electron densities are obtained without introducing dopant atoms, which would cause disorder and lattice distortions—a situation much closer to the ideal Hubbard-like models of many-body theory. By changing the width of the quantum well, the three-dimensional (3D) electron density can be varied, which allows for a systematic study of interaction effects, without changing other parameters, and for addressing a number of scientific questions: (i) Do confined, high-density electron gases in SrTiO₃ exhibit signatures of short-range Coulomb interactions? For example, we recently reported weak ferromagnetism (magnetoresistance hysteresis) in thin, high-density SrTiO₃ quantum wells,⁹ indicating correlation effects; however, because GdTiO₃ is ferrimagnetic, proximity effects could not be ruled out as the source of the magnetism.⁹ (ii) Can a low-dimensional, artificial Mott insulator (AMI) with

a correlation-induced gap be created by purely electrostatic doping of a band insulator? This Rapid Communication addresses both questions and also discusses the role of disorder in the insulating state.

All samples were grown by molecular beam epitaxy, as described elsewhere.^{7,10} Structures included GdTiO₃/SrTiO₃ grown on (001) (LaAlO₃)_{0.3}(Sr₂AlTaO₆)_{0.7} (LSAT) substrates, containing either a single electrically active interface (GdTiO₃/SrTiO₃/LSAT) or two active interfaces (GdTiO₃/SrTiO₃/GdTiO₃/LSAT), as well as Gd _{x} Sr_{1- x} TiO₃ films grown on LSAT or SrTiO₃ single crystals, respectively. Details of the growth conditions and methods, as well as structural, electrical, and magnetic characterization, can be found elsewhere.^{7,9,10} Ohmic contacts for LSAT/GdTiO₃/SrTiO₃/GdTiO₃ and LSAT/SrTiO₃/GdTiO₃ were 50-nm Ti/300-nm Au, while for Gd _{x} Sr_{1- x} TiO₃ structures the contacts were 40-nm Al/20-nm Ni/300-nm Au (with Au being the top layer). Temperature-dependent electrical and magnetotransport properties were measured using van der Pauw geometry in a physical properties measurement system (Quantum Design PPMS), as described in detail elsewhere.^{7,9,10}

The thickness of the SrTiO₃ quantum wells was varied from a few nm to one unit cell (u.c.), which changes the 3D density. For Mott transitions, the electron count per Ti site is important, thus the quantum well thickness notation used here should be clarified. Growth of the GdTiO₃ layers was started and ended with a TiO₂ layer. The SrTiO₃ thicknesses given here are those derived from the growth rate of the SrTiO₃ layers, and do not include the interfacial TiO₂ layers. The reported 3D carrier concentrations (n) are calculated by dividing the sheet carrier density by that thickness. For example, a quantum well of 1 u.c. (or 0.4 nm thickness), shown in Fig. 1(a), consisted of two SrO layers bound by two TiO₂ layers, each of which is shared with the adjacent GdTiO₃. Quantum wells labeled 0.8 nm consisted (on average) of three SrO layers (2 u.c.), bound by two TiO₂ layers shared with the GdTiO₃, and so forth. Furthermore, for quantum wells thicker than 4 nm it was assumed that most of the electrons in the 2DEL were confined to the first 4 nm. Support for this assumption comes from the fact that the mobility increased only moderately for wells thicker than 4 nm, and, as a result, the sheet resistance barely changed,¹¹ and from the coefficient for the quadratic temperature dependence of the

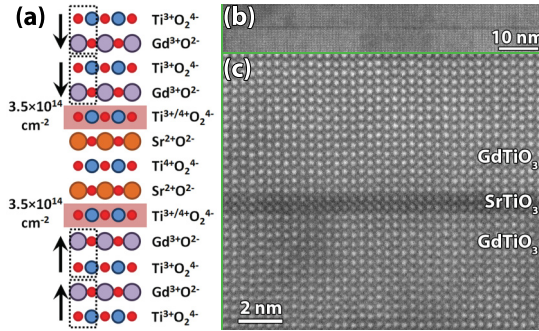


FIG. 1. (Color online) (a) Schematic showing the origin of the fixed polar charge at SrTiO₃/GdTiO₃ interfaces, compensated by $\sim 3.5 \times 10^{14} \text{ cm}^{-2}$ mobile electrons at each interface. (b), (c) HAADF-STEM images of a unit cell of SrTiO₃ embedded in GdTiO₃. The image in (b) shows a larger field of view, confirming the uniform thickness of the SrTiO₃ quantum well (thin dark region). The image in (c) is a magnified section of the atomic resolution image. The SrTiO₃ appears darker in these images because of the lower atomic number of Sr relative to Gd.

resistivity, discussed below. Theoretical calculations indicate that for 2DELs in SrTiO₃, containing $3.5 \times 10^{14} \text{ cm}^{-2}$ carriers, about 75% of the carriers are confined within the first three layers.¹² Returning to the question of electron count per Ti site, the thinnest quantum well studied here, a single u.c. of SrTiO₃ embedded in GdTiO₃, accommodate $\sim 7 \times 10^{14} \text{ cm}^{-2}$ carriers,¹¹ or about one electron shared between three TiO₂ layers, if the two interfacial TiO₂ layers are included in the electron count. However, the staggered band alignment⁷ favors charge transfer into the SrTiO₃ well, and the precise electron distribution over the three TiO₂ layers likely will depend on a number of atomistic parameters, including atomic relaxations.^{13,14}

High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images [Fig. 1(b)] confirmed that continuous, SrTiO₃ quantum wells down to 1 u.c. in thickness were obtained. The excellent control over quantum well thicknesses afforded by molecular beam epitaxy (MBE) is also apparent in the sheet resistance data of these samples.¹¹ For example, changing the thickness by one monolayer produced detectable decreases in the sheet resistance,¹¹ due to the increase in mobility (interface roughness scattering in thin quantum wells scales with $\sim a^6$, where a is the quantum well thickness¹⁵). Hall and Seebeck measurements indicated that transport occurred in the SrTiO₃, consistent with the band alignments in this system.⁷ In particular, even for the thinnest quantum wells, the Hall and Seebeck coefficients were n type, characteristic for SrTiO₃. In the Mott insulator GdTiO₃ these coefficients are p type and positive.¹⁶ Furthermore, mobilities were higher than those of doped GdTiO₃ for all samples.

All quantum well structures with SrTiO₃ thicknesses greater or equal to 2 u.c. (“0.8 nm”) were metallic (Fig. 2). The upturn in resistivity at very low temperatures coincides with the appearance of negative magnetoresistance,¹¹ indicative of weak localization.¹⁷ As shown in Fig. 2(a), at temperatures above the onset of weak localization the temperature dependence of the resistivity $\rho(T)$ can be described by Fermi-liquid

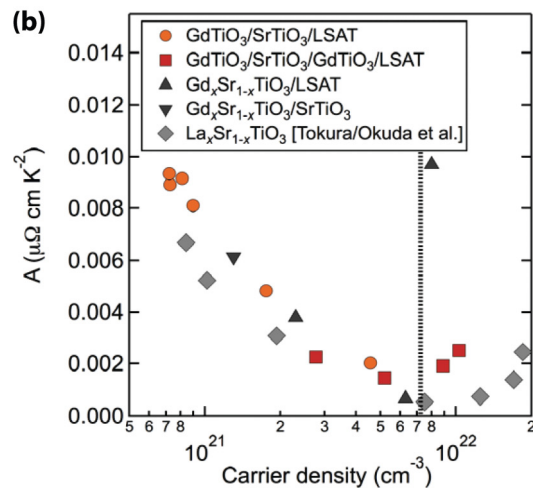
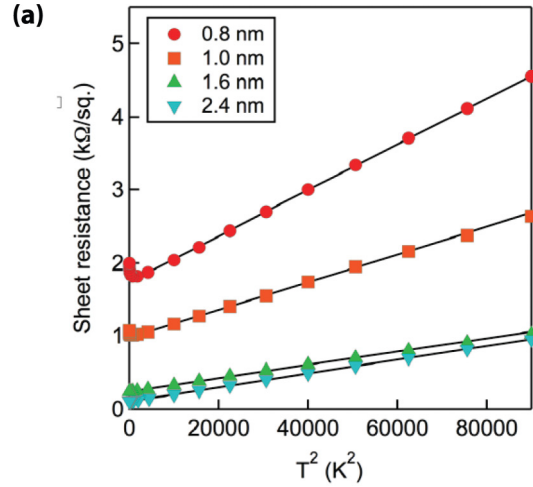


FIG. 2. (Color online) (a) Sheet resistance as a function of T^2 for quantum wells of SrTiO₃ embedded in GdTiO₃, with different SrTiO₃ quantum well thicknesses. The solid black lines are fits to Eq. (1). (b) Temperature coefficient A as a function of carrier density for the samples investigated in this study and bulk La_xSr_{1-x}TiO₃ from the literature [Tokura *et al.* (Ref. 25) and Okuda *et al.* (Ref. 24)], respectively. A was obtained from fits to Eq. (1) between ~ 40 and 300 K, except for Gd_{0.285}Sr_{0.715}TiO₃ and Gd_{0.56}Sr_{0.44}TiO₃, for which it was obtained from ~ 10 to 250 K. The thickness of the SrTiO₃ quantum wells for the GdTiO₃/SrTiO₃/LSAT samples was between 0.4 and 8.6 nm [see Supplemental Material (Ref. 11)] and the 3D carrier concentrations were calculated from the sheet densities as described in the text. The vertical line is a guide to the eye showing the critical carrier density for which A increases as the Mott transition is approached.

theory as

$$\rho(T) = \rho_0 + AT^2, \quad (1)$$

where ρ_0 is the residual resistivity due to impurity scattering, and T the temperature. The quadratic temperature dependence indicates electron-electron scattering as the dominant scattering mechanism¹⁸ and was found for all samples investigated here, up to room temperature,¹¹ except for the Gd_{0.285}Sr_{0.715}TiO₃ and Gd_{0.56}Sr_{0.44}TiO₃ films, which showed the quadratic temperature dependence only below 250 K.

The A coefficient in Eq. (1) is proportional to the mass enhancement due to electron correlations,^{18,19} and other materials parameters, including the carrier concentration and band structure.^{20–22} The quadratic dependence of A on the mass holds in both two dimensions (2D) and 3D.²³ In general, A decreases with increasing carrier concentration.¹⁸ Figure 2(b) shows A as a function of n for quantum wells, $\text{Gd}_x\text{Sr}_{1-x}\text{TiO}_3$ thin films, and $\text{La}_x\text{Sr}_{1-x}\text{TiO}_3$ bulk samples from the literature.^{24,25} For all samples, A decreases with increasing n for $n \lesssim 7 \times 10^{21} \text{ cm}^{-3}$, consistent with Fermi-liquid behavior. A was constant for the quantum wells thicker than 4 nm ($\text{GdTiO}_3/\text{SrTiO}_3/\text{LSAT}$ samples), consistent with the tight confinement of the 2DEL, discussed above. However, for the thinnest, still metallic quantum wells with the highest densities (0.8 and 1 nm thickness sandwiched between two GdTiO_3 layers), the behavior changes, and A increases with n . This indicates that electron correlations are enhanced in the extreme 3D carrier concentration regime. This mass enhancement is also found in bulk $\text{La}_x\text{Sr}_{1-x}\text{TiO}_3$ (Ref. 25) and $\text{Gd}_x\text{Sr}_{1-x}\text{TiO}_3$ films (this study) on the verge to the transition to the Mott insulating state, i.e., at large filling factors x . A mass enhancement near the Mott metal-insulator transition, as a half-filled band is approached, is predicted from Mott-Hubbard theory.⁴ Theory also predicts A to diverge near half filling.²⁶ The similarity in the behavior of A as a function of filling between bulk rare earth titanates near the filling-induced Mott metal-insulator transition, and the high-carrier-density quantum wells of SrTiO_3 is remarkable.

The results are an experimental demonstration that mass enhancement due to short-range, on-site Coulomb interactions (Mott-Hubbard physics) can be obtained in a two-dimensional band insulator, due to purely electronic effects, i.e., in the absence of chemical doping (alloying). Furthermore, the discontinuity in A appears for all three materials (SrTiO_3 quantum wells, $\text{La}_x\text{Sr}_{1-x}\text{TiO}_3$, and $\text{Gd}_x\text{Sr}_{1-x}\text{TiO}_3$), at approximately the same 3D carrier density, $n_c \approx 7 \times 10^{21} \text{ cm}^{-3}$. This indicates similarity in the basic correlation physics in all three samples, and lends credibility to the way in which the 3D carrier concentration of the quantum wells was calculated from the thickness and sheet density (see above). The results suggest that an increase in the A coefficient occurs when the probability of double occupancy (and thus Hubbard-type strong correlation physics) becomes non-negligible, and that this depends *only* on density and not on the specific material (within this series). Thus, the effects of electrostatic or chemical doping are remarkably similar. Above this critical density, the *rate* of increase in A with filling appears to depend on the specific material. The electron-correlation strength depends on the rare earth titanate, specifically their bandwidth.²⁷ The correlation strength is larger in GdTiO_3 than in LaTiO_3 ; the results here show that this is correlated with $\partial A/\partial n$. By this measure, the correlation strength in the confined quantum wells appears to be intermediate between $\text{La}_x\text{Sr}_{1-x}\text{TiO}_3$ and $\text{Gd}_x\text{Sr}_{1-x}\text{TiO}_3$.

Even thinner quantum wells (0.4 nm) were insulating, although Hall measurements showed no significant change in carrier density.¹¹ Figure 3 shows the sheet resistance as a function of temperature for the insulating quantum wells with 1 u.c. SrTiO_3 . The transition to an insulator for the high-density quantum well sandwiched between two GdTiO_3 layers appears to be consistent with a filling-controlled Mott

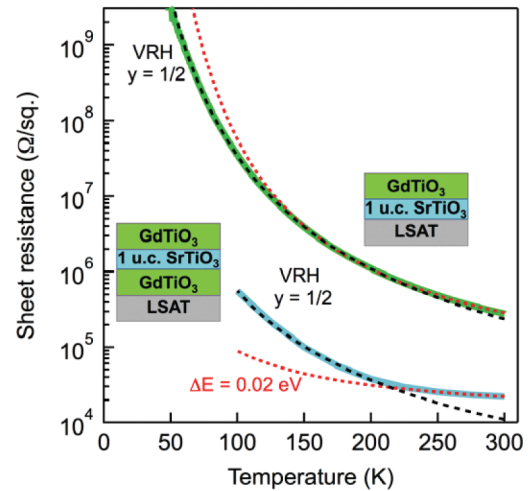


FIG. 3. (Color online) Sheet resistance as a function of temperature for insulating SrTiO_3 quantum wells. The solid lines are the experimental data, the red dotted lines fits to an Arrhenius law, and the black dashed lines are fits to the Efros-Shklovskii law ($y = 1/2$).

metal-insulator transition. However, both disorder (Anderson) and on-site Coulomb interactions (Mott-Hubbard) can cause a transition to an insulating state. For a two-dimensional system with disorder and interacting electrons, theory predicts a universal crossover between the correlated metallic state and an Anderson insulator near the Mott minimum conductivity, $\sim (e^2/h)$ or $\sim 25 \text{ k}\Omega^{-1}$.²⁸ The conductance of both samples was below this value. For extremely narrow quantum wells, and high electron densities, surface roughness scattering becomes the dominant mechanism of electron localization, relative to those due to impurities and remote dopant scattering.¹⁵ Using the formalism developed in Ref. 15, the interfacial roughness parameters for strong localization can be estimated for a given carrier density and quantum well thickness. A SrTiO_3 quantum well [in-plane mass of $\sim 0.9m_e$, out-of-plane mass $\sim 8m_e$, where m_e is the free electron mass,²⁹ a dielectric constant (ϵ) of 25 (Ref. 30)], with a thickness of 0.4 nm, and roughness parameters of $\Lambda = 15 \text{ nm}$, $\Delta = 0.2 \text{ nm}$, where Δ is the height and Λ the length of the Gaussian-like interface roughness, and a carrier density of $7 \times 10^{14} \text{ cm}^{-2}$, is *still metallic*.¹¹ The particular choice for Δ is justified by the HAADF images, but Λ is more difficult to estimate. It is likely that both Anderson and Mott localization are at play here. In this case, the band tails are localized over a range of energy. Localized transport would occur by means of variable range hopping (VRH) in the band tail and thermal activation to a mobility edge. The resistivity in the VRH regime is described as $\rho(T) \sim \exp(T_0/T)^y$, where y is a parameter that has a value of 1/2 in case of strong electron interaction and the formation of a Coulomb gap in both 2D and 3D (Efros-Shklovskii law³¹). VRH gives way to Arrhenius behavior [$\rho(T) \sim \exp(\Delta E/kT)$, where ΔE is the activation energy and k the Boltzmann constant] at higher temperatures, when carriers above the mobility edge participate in transport.

To determine the respective roles of disorder versus electron correlations in the localization, data were fit to Arrhenius and VRH expressions, respectively. The $\text{GdTiO}_3/1 \text{ u.c. SrTiO}_3/\text{GdTiO}_3/\text{LSAT}$ sample, with a sheet carrier density of $\sim 8 \times 10^{14} \text{ cm}^{-2}$, exhibits two distinct regimes at high and

low temperatures, respectively. In particular, there is a clear crossover between Arrhenius ($\Delta E = 0.02\text{eV}$, red dotted line in Fig. 3) and VRH with a Coulomb gap, $\rho(T) \sim \exp(T_0/T)^{1/2}$ (dashed black line in Fig. 3) with a characteristic temperature T_0 of 8690 K. These results provide evidence of thermally activated transport to a mobility edge and band transport at high temperatures. The results support the interpretation of correlation modified density of states in which incoherent transport occurs, at the Fermi energy. At the same time, and similar to what has been reported for bulk titanates,²⁷ the small activation energy appears to be a signature of close proximity to the metal-insulator transition.

For comparison, the insulating GdTiO₃/1 u.c. SrTiO₃/LSAT sample could be described over a large temperature range by the Efros-Shklovskii law (see Fig. 3), with a T_0 of 13 750 K, indicating the importance of disorder. This quantum well suffers from more scattering due to the proximity to the substrate surface. Furthermore, as it contains half the 3D carrier density, the Fermi energy will be deeper in the band tail suppressing activation to the mobility edge.

The very large value for T_0 for both quantum wells gives further evidence for Hubbard physics. Efros-Shklovskii³¹ relate T_0 to the localization length ξ , assuming *only long-range Coulomb interactions*, $\xi = 6.5[e^2/(4\pi\epsilon_0\epsilon kT_0)]$, where e is the electron charge, ϵ_0 is the permittivity of free space, and the factor of 6.5 is for a 2D system.³² Using $\epsilon \approx 25$, we obtain $\xi = 0.5$ and 0.31 nm for the GdTiO₃/1 u.c. SrTiO₃/GdTiO₃/LSAT and GdTiO₃/1 u.c. SrTiO₃/LSAT, respectively, comparable to

or smaller than a single unit cell. This clearly points to the importance of strong on-site Coulomb repulsion.

In summary, we have shown evidence for on-site Mott-Hubbard-type correlation physics in thin, high-carrier-density quantum wells of SrTiO₃, a material that is a band insulator in bulk. With increasing 3D carrier densities in these wells we observe a correlation-induced mass enhancement, followed by a transition to a correlated insulator at the highest 3D densities. We have shown that polarization-induced mobile charges in oxide quantum wells provides for a tool to probe strong correlations associated with on-site Coulomb interactions. In particular, very large carrier densities—beyond what is possible with electric field gated structures and conventional dielectrics—can be introduced electrostatically, and without the chemical and lattice changes that accompany bulk alloying studies. Future experiments should address the precise nature (such as charge and magnetic ordering¹³) of the insulating ground state in these quantum wells, and compare the results with predictions by two-dimensional Hubbard models.

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