Magnetic-field induced resistivity minimum with in-plane linear magnetoresistance of the Fermi liquid in SrTiO$_{3-x}$ single crystals

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We report novel magnetotransport properties of the low temperature Fermi liquid in SrTiO$_{3-x}$ single crystals. The classical limit dominates the magnetotransport properties for a magnetic field perpendicular to the sample surface and consequently a magnetic-field-induced resistivity minimum emerges. While for the field applied in plane and normal to the current, the linear magnetoresistance (MR) starting from small fields (< 0.5 T) appears. The large anisotropy in the transverse MRs reveals the strong surface interlayer scattering due to the large gradient of oxygen vacancy concentration from the surface to the interior of SrTiO$_{3-x}$ single crystals. Moreover, the linear MR in our case was likely due to the inhomogeneity of oxygen vacancies and oxygen vacancy clusters, which could provide experimental evidences for the unusual quantum linear MR proposed by Abrikosov [A. A. Abrikosov, Phys. Rev. B 58, 2788 (1998)].

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I. INTRODUCTION

SrTiO$_3$ (STO) is one of the most important workhorses in oxide electronics. Recently, a two-dimensional electron gas [1,2] and electronic phase separation [3] have been demonstrated to emerge on the bare STO single crystal surface. Understanding the electronic properties of STO under different oxidation states is therefore crucial to reveal the origin of these novel phenomena and to use STO in electronic devices. Generally, STO is a nonpolar band insulator with an indirect bandgap of $\sim$3.27 eV [4] and a large dielectric constant $\varepsilon_r$ [5]. A semiconducting (or metallic) phase of STO can be obtained by reduction [6], chemical doping [7] or photo-carrier injection [8], with a high carrier mobility ($>10,000$ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$) and mobility ($\mu$) of the resistivity. The carrier density $n$ (in (b)) of SrTiO$_3$ single crystal reduced in $\sim$$10^{-7}$ Torr vacuum at 950 $^\circ\text{C}$ for 1 h. Inset of (a): linear fitting of $T^2$ dependence of the resistivity. The carrier density $n$ in (b) was averaged over the entire crystal thickness.

II. ELECTRICAL PROPERTIES

The temperature dependence of resistivity ($\rho$-$T$), carrier density ($n$-$T$) ($n$ is an average carrier density value over the entire crystal thickness) and mobility ($\mu$-$T$) of samples reduced for 1 h is depicted in Fig.1, showing a metallic behaviour over the whole temperature range from 300 to 2 K (Fig. 1(a)). The origin of this metallic behavior can be understood in terms of the Mott criterion [13]. The critical carrier density for a metal-insulator transition is given by the Mott critical carrier density $n_c$ $\approx (0.25/\alpha^*)^3$, where $\alpha^* = 4\pi\varepsilon_r\varepsilon_0\hbar^2/m_D\epsilon^2$ is the effective

![FIG. 1:](attachment:image.png)

(a) $\rho (\Omega \cdot \text{cm})$ vs. $T$ ($\text{K}$)

(b) $\mu (\text{cm}^2\text{V}^{-1}\text{s}^{-1})$ vs. $T$ ($\text{K}$)

(c) $n (10^{18} \text{ cm}^{-3})$ vs. $T$ ($\text{K}$)
Bohr radius and \( \varepsilon_0 \) the vacuum permittivity. The measured carrier density at 300 K is \( \sim 1.5 \times 10^{18} \text{ cm}^{-3} \), which is more than three times \( n_e \sim 4.9 \times 10^{17} \text{ cm}^{-3} \), considering the room temperature \( \varepsilon_r \approx 300 \) and \( m_D \approx 5m_0 \) for STO. Interestingly, the resistivity from 2 up to \( \sim 80 \) K exhibits an obvious behavior of a strongly correlated Fermi liquid as fitted in the inset of Fig. 1(a), reminiscent of the normal state of electron-doped cuprate superconductors [14] and the \( p \)-wave superconductor \( \text{Sr}_2\text{RuO}_4 \) [15], noting that semiconducting STO is also superconducting at very low temperatures [12]. Moreover, the common Fermi liquid origin of the \( T^2 \) resistivity and superconductivity of \( n \)-type \( \text{SrTiO}_3 \) has been elaborately discussed by Marel et al. [16].

The detailed \( n-T \) curve determined by Hall measurements is shown in Fig. 1(b). At high temperatures, the carrier density slightly fluctuates; however, it increases with decreasing temperature at low temperatures especially between 100 and 10 K. This unexpected behavior is unphysical from the viewpoint of thermal activation. In fact, this behavior was also observed previously in semiconducting STO single crystals [7,8] and \( \text{LaAlO}_3/\text{STO} \) heterostructures grown at low oxygen pressures [3,17]. Therefore there should be another intrinsic mechanism affecting the carrier density.

One unique property of STO is that its \( \varepsilon_r \) increases with lowering temperature (especially from \( \sim 100 \) K) and saturates at 4 K because of the quantum-mechanical stabilization of the paraelectric phase [5]. It seems plausible to assume that in \( \text{SrTiO}_3-x \) part of carriers are trapped by the Coulomb potentials of the majority of positively charged defects due to the strongly ionic nature of the lattice. As the \( \varepsilon_r \) increases, the Coulomb potentials will be suppressed due to dielectric screening since the screened Coulomb potential [18] is inversely proportional to \( \varepsilon_r \). Hence the increase of \( \varepsilon_r \) could serve as a kind of detrapping mechanism and consequently account for the increase of carrier density in \( \text{SrTiO}_3-x \) at low temperatures.

The \( \mu-T \) curve is plotted in Fig. 1(c) on a logarithmic scale. The high mobility, up to \( \sim 11,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1} \) at 2 K, decreases with temperature rapidly and varies in accordance with certain power laws above 30 K, where the scattering of electrons by polar optical phonons dominates and results in \( \mu \propto T^{-\beta} \) [19]. The effect of the structural phase transition in STO at \( \sim 105 \) K on the mobility can be apparently seen from the linear fittings owing to the variation in the activation of phonon modes. Below 30 K, the scattering of electrons is dominated by ionized defect scattering and electron-electron Umklapp scattering because of the Fermi liquid behavior [15]. However, the electron-electron scattering should be enhanced with increasing carrier density, so the ionized defect scattering is the more pronounced mechanism for the continuously increasing mobility similarly due to the dielectric screening of ionized scattering potentials [7]. Finally, the trend to saturation appears from 5 K possibly corresponding to the quantum paraelectric phase in STO.

![Fig. 2. (Color online) (a) \( \rho-T \) curves under different magnetic fields. (b) Extracted resistivity minimum temperature from (a) versus magnetic field. (c) Large field Hall effect at 2 K from -5 to 5 T.](image)

III. MAGNETOTRANSPORT PROPERTIES

The \( \rho-T \) curves under different magnetic fields perpendicular to the surface are shown in Fig. 2(a). The transverse MR effect (\( \Delta \rho/\rho(0) = \rho(B)/\rho(0) \)) is notable only below \( -50 \) K and always positive. Intriguingly, the \( \rho-T \) curves under sufficiently strong magnetic fields (\( \geq 2.5 \) T) exhibit a resistivity minimum at a temperature \( T_{min} \), which increases with \( B \) monotonically as plotted in Fig. 2(b). The resistivity minimum cannot pertain to a Kondo effect or weak localization since they are inherently antagonistic towards magnetic fields. The antilocalization effect is also ruled out since it is typically very small (of the order of few percent). One possible origin for this behavior could be magnetic field induced carrier freeze-out due to the considerable shrinking of electron wave functions if the magnetic field strength is much larger than the Coulomb forces [20]. The magnetic freeze-out would cause an increase of Hall coefficient. To examine this, Hall measurements was performed up to 5 T at 2 K. However, the observed linear Hall effect indicates that large magnetic fields are not affecting the carrier density at all as seen in Fig. 2(c).

To explore the origin of the magnetic-field induced resistivity minimum, the out-of-plane transverse MR was measured up to 9 T. As shown in Fig. 3(a), both the 2 and 10 K MR curves exhibit an obvious quadratic shape. For simplicity, we analyze our data using the single band picture, where the quadratic MR can be described by the classical orbital scattering. The Fermi energy at 2 K is \( E_F \approx 1.08 \text{ meV} \) and more than \( \sim 6kT \) (2 K) taking \( m_0 \approx 5m_0 \) and \( n \) as the average carrier density over the entire crystal thickness at 2 K as shown in Fig. 1(b), corresponding to a degenerate gas state (\( E_F \geq kT \)). Indeed, the carrier density of the surface region should be larger than the average value due to the inhomogeneity of oxygen vacancies (this will be discussed later), which
would thus lead to an even larger $E_F$. However, no signature of quantum oscillations in MR was seen in spite of the high mobility. This could be because both the $m_0$ and $m_e$ have been largely enhanced from strong electron correlations of the Fermi liquid and eventually the initially assumed degenerate gas actually exists in a non-degenerate ($E_F \leq kT$)“liquid” state. In addition, the Shubnikov-de Haas oscillation was observed typically at lower temperatures below 2 K [10].

The MR of 9 T at 2 K is extremely large, more than 2,900% and decreases to $\sim 1,200\%$ at 10 K. The classical transverse orbital scattering can be described by $\Delta \rho/\rho(0) = \alpha^2 \mu^2 B^2$, where $\alpha$ is a material dependent constant. By fitting the MR curves of 2 and 10 K, the average value of $\alpha$ is obtained to be $\sim 0.32$, which is comparable to the $\alpha \sim 0.38$ in $n$-type InSb [21]. The mobility of 2 K is larger than that of 10 K and therefore the MR of 2 K is far larger as shown in Fig. 3(a). Under a sufficiently large $B$, the MR difference between 2 and 10 K is so large that the overall resistivity $\rho(B, T) = \rho(0, T) + \alpha \mu^2 B^2 \rho(0, T)$ at 2 K can become larger than that of 10 K although $\rho(0, 2 \text{K})$ is smaller than $\rho(0, 10 \text{K})$ in the normal metallic state. In this way, the intriguing resistivity minimum under a large magnetic field can be understood.

To further confirm the origin of this resistivity minimum, the magnetic field dependence of resistivity ($\rho-B$) at 2 and 10 K are compared in Fig. 3(b). There is an evident crossover between the two $\rho-B$ curves at $\sim 2.5 \text{ T}$, above which the resistivity at 2 K exceeds that at 10 K. The crossover field is consistent with the critical $B$ in Fig. 2(a). The low temperature resistivity $\rho(B, T) = \rho(0, T) + \alpha \mu^2 B^2 \rho(0, T)$ can be readily simulated up to 80 K by considering the $T^2$ dependence of $\rho(0, T)$ and mathematically representing the mobility below 30 K with an exponential fitting. The simulated results (not shown) were consistent with the curves in Fig. 2(a), which suggests that the above resistivity comparison is not only true between 2 and 10 K but also valid for other temperatures. Finally we conclude that the magnetic field induced resistivity minimum originates from the extremely large MR and its pronounced increase with decreasing temperature at low temperatures. The large MR is achieved by the fairly high $\mu$, sufficiently large $B$ and the possible stabilization of the classical limit by strong electron correlations of the Fermi liquid.

Similar behavior was also observed in SrTiO$_{3-x}$ single crystals reduced for 2 h, which possess a larger room temperature carrier density and also a high mobility at 2 K. The Fermi liquid behavior, i.e. the $T^2$ dependence of the resistivity, also exists but up to $\sim 65$ K. Nevertheless, as the reducing time was prolonged to 8 h for a STO single crystal, the room temperature carrier density reaches $\sim 6 \times 10^{18} \text{ cm}^{-3}$ and consequently the mobility at 2 K is only $\sim 2,500 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. As a result there is no observable resistivity minimum even under a 9 T magnetic field.

The condition for the strong-field region $\hbar \omega_c \gg kT$, in which most of the carriers are in the lowest Landau magnetic quantum level, was coined as the “quantum limit” [22]. The theoretical analyses [23-25] indicate that for both degenerate and nondegenerate statistics the transverse MR has a quadratic field dependence in the classical low-field case with $\hbar \omega_c \ll kT$ but a linear dependence in the quantum limit. Moreover, the other criterion $n \ll (eB/\hbar)^{3/2}$ [25] should also be fulfilled for the usual quantum linear MR as observed in high mobility InSb [21,26], Ge [27], graphite [28] and also recently in the topological insulator Bi$_2$Se$_3$ [29]. As shown in Fig. 4(a), the MR curves exhibit highly linear field dependence while the $B$ is applied in plane and transverse to the current. The magnitude of the in-plane MR is on average $\sim 92\%$ at 2 K and $\sim 66\%$ at 10 K under 9 T, which are far smaller than the out-of-plane values. Accordingly neither the crossover in the $\rho-B$ curves (inset of Fig. 4(a)) nor the magnetic field induced resistivity minimum was observed. In our case, the linear MR starts from a very small field $< 0.5 \text{ T}$ as seen from Fig. 4(a), which in turn corresponds to a critical carrier density of $\sim 1.36 \times 10^{17} \text{ cm}^{-3}$ for the usual quantum linear MR. The average carrier density at 2 K in our case is already $\sim 1.85 \times 10^{18} \text{ cm}^{-3}$ and more than one order of magnitude larger than the critical carrier density. In this case, the linear MR may be out of the usual quantum linear MR picture [30].

On the other hand, the linear MR starting from small fields was previously also observed in nonstoichiometric silver chalcogenides Ag$_{2+x}$Se and Ag$_{2+x}$Te by Xu et al. [31]. In that case, the criteria for the usual quantum linear MR can also not be fulfilled. However, Abrikosov [25,32] proposed another model of quantum linear MR for the Ag$_{2+x}$Se and Ag$_{2+x}$Te case with two assump-
FIG. 4: (Color online) (a) In-plane transverse MR at 2 K and 10 K up to 9 T. The upper and lower insets are the corresponding ρ-B curves of the two temperatures and the schematic of measurement geometry respectively. (b) The parameter \( \frac{\rho(B)/\rho(0) kT}{\mu_B B} \) plotted as a function of magnetic field.

...tions, i.e., (1) the substance is inhomogenous, consisting of clusters of excess silver atoms with a high electron density, surrounded by a medium with a much lower electron density; (2) in this medium the electron energy spectrum is close to a gapless semiconductor with a linear dependence of energy on momentum, and thereby explained all the data in [31] satisfactorily. Our scenario seems quite close to the above case since the inhomogeneity of oxygen vacancies in SrTiO\(_3\)-x single crystals has been a long-standing issue [33, 34]. Moreover, the oxygen vacancy clustering in SrTiO\(_3\)-x has been well studied both theoretically by Cuong et al. [35] and experimentally by Muller et al. [36]. Similar to clusters of excess silver atoms in nonstoichiometric silver chalcogenides, oxygen vacancy clusters have a high electron density. Thus, the linear MR in our case could be another example for the unusual quantum linear MR induced by inhomogeneities and conductive clusters.

Considering the further spin-orbital splitting of the Landau magnetic levels under a magnetic field \( B \), the energy scale \( \mu_B B \) is involved, where \( \mu_B \) is the Bohr magneton. The dimensionless parameter \( \frac{\rho(B)/\rho(0) kT}{\mu_B B} \) is theoretically predicted to be relatively independent of temperature for the quantum limit as illustrated in [26]. It is plotted in Fig. 4(b) as a function of field for two different temperatures and the parameter is approaching a constant value. This further supports the idea that the linear in-plane transverse MR is intrinsically a kind of quantum MR.

In early studies on the transport properties of semiconducting STO, STO single crystals were thermally reduced in vacuum and high temperature, which are similar to what we used, for quite a long time (typically up to 10 days [6]) to achieve uniform oxygen vacancy distribution. However, the oxygen vacancy distribution obtained by that long time reduction was found [34] to be still not ideally uniform over the entire thickness of single crystals. Furthermore, the O\(_{vac}\) in short-time (like 1 h in our case) reduced STO single crystals are therefore far from uniform leading to a large gradient in O\(_{vac}\) concentration [34] and thus also in carrier density [37] from the surface to the interior. Typically the oxygen vacancy doping can increase carrier density but on the other hand can decrease mobility in SrTiO\(_3\)-x due to electron-impurity scattering, enhanced electron-electron scattering and suppressed screening effect. However, the lowering of mobility by oxygen vacancies is not as significant as the increase of carrier density by oxygen vacancy doping as can be seen from the electronic transport property studies of SrTiO\(_3\)-x single crystals by Frederikse et al. [6] and SrTiO\(_3\)-x films by Ohtomo and Hwang [38]. Thus, the higher oxygen vacancy concentration results in smaller resistivity and accordingly the conducting channel.

Consequently the measured transport data should only well represent the properties of the surface layers for this kind of samples. The highly anisotropic transport properties of the Fermi liquid can therefore be understood by the strong surface interlayer scattering due to the large inhomogeneity of O\(_{vac}\) and accordingly the density-of-states along the out-of-plane direction. Even though the free carriers in our STO samples are not only confined to the very top surface, the sharp gradient of the carriers with the largest concentration at the top surface can result in the highly anisotropic transport properties.

IV. CONCLUSIONS

In conclusion, we studied the electrical and magneto-transport properties of SrTiO\(_3\)-x single crystals. It was found that the Fermi liquid exists at low temperatures and the dielectric constant of STO plays an important role in carrier density and mobility. A magnetic-field induced resistivity minimum was explored to originate from the high mobility and the possible strengthening of the classical limit by mass enhancement of strong electron correlations. The linear in-plane transverse MR, potential for linear MR sensors, was observed and attributed to the unusual quantum linear MR due to the inhomogeneity of oxygen vacancies and also oxygen vacancy clustering. The large anisotropy in transverse MRs reveals the strong surface interlayer scattering due to the inhomogeneity of O\(_{vac}\) at the surface of SrTiO\(_3\)-x. By this work, we demonstrate a potential route to quantum linear MR in virtue of inhomogeneities.
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