Enhancing superconductivity in SrTiO$_3$ films with strain

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The nature of superconductivity in SrTiO$_3$, the first oxide superconductor to be discovered, remains a subject of intense debate several decades after its discovery. SrTiO$_3$ is also an incipient ferroelectric, and several recent theoretical studies have suggested that the two properties may be linked. To investigate whether such a connection exists, we grew strained, epitaxial SrTiO$_3$ films, which are known to undergo a ferroelectric transition. We show that, for a range of carrier densities, the superconducting transition temperature is enhanced by up to a factor of two compared to unstrained films grown under the same conditions. Moreover, for these films, superconductivity emerges from a resistive state. We discuss the localization behavior in the context of proximity to ferroelectricity. The results point to new opportunities to enhance superconducting transition temperatures in oxide materials.

INTRODUCTION

Although SrTiO$_3$ was the first oxide superconductor to be discovered (1), the nature of its superconducting state has been a longstanding subject of debate in the literature (2–10), reflecting in many ways the elusiveness of other families of superconductors, such as the cuprates. A striking feature is that superconductivity already appears at very low carrier densities (2, 11), when the Fermi temperature is lower than the Debye temperature, which is at odds with the Bardeen-Cooper-Schrieffer (BCS) theory. Bulk, undoped SrTiO$_3$ is an incipient ferroelectric for which quantum fluctuations suppress a low-temperature transition to a ferroelectric ground state at the lowest temperatures (12, 13). Several recent theoretical proposals have suggested that a connection between the ferroelectric and superconducting properties exists (14–17), providing strong motivation for developing experimental approaches that search for such a link.

The complex relationship between metallicity and ferroelectricity is one of the main challenges in experimental tests of the role of (incipient) ferroelectricity in the superconducting pairing mechanism of SrTiO$_3$. In particular, free carriers, needed for superconductivity, and ferroelectricity do not easily coexist. For example, sufficiently large concentrations of mobile carriers can screen the splitting of the transverse and longitudinal optical phonon modes (18, 19), which is essential for ferroelectricity in materials such as SrTiO$_3$ (20). Furthermore, conducting samples cannot sustain sufficiently large electric fields that are needed to switch a ferroelectric polarization. Thus, although there are no fundamental reasons why the two properties cannot coexist (21), practically, they are difficult to obtain within a homogeneous material.

Despite these challenges, several recent experiments have reported on the superconductivity of SrTiO$_3$ crystals that were tuned toward ferroelectricity by using approaches that were previously known (22–24) to stabilize ferroelectricity in insulating SrTiO$_3$. In particular, oxygen isotope doping (25) and alloying with Ca (26) were found to result in modest changes in $T_c$ (superconducting transition temperatures) of doped single crystals.

Epitaxial coherency strains are well known to stabilize ferroelectricity in SrTiO$_3$ films (27–30). Films under different epitaxial strains thus offer an attractive platform to compare the superconducting properties of films poised to undergo a ferroelectric transition with those that remain paraelectric. Here, we show that doped, compressively...
strained SrTiO$_3$ films exhibit $T_C$ values that are enhanced by a factor of two compared to unstrained films. Moreover, films with enhanced $T_C$ exhibit a pronounced upturn in the normal state resistivity with decreasing temperature, which is highly unusual. In contrast, films with higher carrier densities remain metallic and do not exhibit enhanced $T_C$, although they are strained by the same amount. We discuss the implications of the results with regard to the connection between superconductivity and (incipient) ferroelectricity in SrTiO$_3$.

**RESULTS**

Electrical measurements were carried out on epitaxial SrTiO$_3$ films, which were grown on (001) LSAT [(LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$] and SrTiO$_3$ substrates using a hybrid molecular beam epitaxy (MBE) technique (31, 32). The films were doped with different amounts of Sm$^{3+}$ to obtain a range of carrier densities.

Figure 1A shows the resistivities ($\rho$) of strained Sm:SrTiO$_3$ films grown on LSAT with different carrier densities, measured between 1 K and 10 mK without an applied magnetic field. The legend indicates the Hall carrier densities measured at 300 K. Lines are a guide for the eye.

**Fig. 2.** Superconducting transitions for strained Sm-doped SrTiO$_3$ films with different carrier densities. The resistivity as a function of temperature between 1 K and 10 mK without an applied magnetic field is shown. The legend indicates the Hall carrier densities measured at 300 K. Lines are a guide for the eye.

**Fig. 3.** Comparison of $T_C$ of strained and unstrained SrTiO$_3$. $T_C$ values of strained and unstrained La- and Sm-doped SrTiO$_3$ films with different carrier densities grown on LSAT and SrTiO$_3$ substrates, respectively (filled circles), are shown. $T_C$ values of SrTiO$_3$ crystals taken from the literature are also shown, some of which were tuned toward a ferroelectric transition (triangles). Literature data are from Lin et al. (11), Stucky et al. (25), Rischau et al. (26), Koonce et al. (33), and Suzuki et al. (34). Lines are a guide for the eye.
300 and 2 K. Here, the resistivities were determined from the sheet resistance and film thickness (200 nm) measured by cross-sectional transmission electron microscopy. The highest doped film ($n_{3D} = 2.8 \times 10^{20} \text{ cm}^{-3}$ at 300 K, where $n_{3D}$ is the carrier density determined from Hall measurements) shows metallic behavior, $d\rho/dT > 0$, down to 1.8 K. In contrast, lower doped films ($n_{3D} = 2 \times 10^{19}$, $6 \times 10^{19}$, and $1.4 \times 10^{20} \text{ cm}^{-3}$) exhibit a crossover to $d\rho/dT < 0$ upon lowering the temperature. The transition temperatures, defined as $d\rho/dT = 0$, for films with $n_{3D} = 6 \times 10^{19}$ and $1.4 \times 10^{20} \text{ cm}^{-3}$ are ~110 and ~60 K, respectively (see arrows). The lowest doped film with $n_{3D} = 2 \times 10^{19} \text{ cm}^{-3}$ not only demonstrates an upturn in resistivity at low temperatures but also shows a more complicated temperature dependence. All films exhibit an abrupt decrease in the Hall carrier density upon lowering the temperature (Fig. 1B). Roughly the same mobile carrier density (4 $\times$ $10^{19}$ to 7 $\times$ $10^{19} \text{ cm}^{-3}$) is lost in all films.

Figure 2 shows $\rho$ between 1 K and 10 mK. All films become superconducting at low temperatures. Taking $T_C$ as the temperature for which $\rho$ corresponds to $\rho_e$, where $\rho_e$ is the normal state value and $e$ is Euler’s number, the values for $T_C$ are 0.37, 0.60, 0.67, and 0.16 K for films with $n_{3D} = 2 \times 10^{19}$, $6 \times 10^{19}$, $1.4 \times 10^{20}$, and $2.8 \times 10^{20} \text{ cm}^{-3}$, respectively.

Figure 3 compares the $T_C$ values for the films shown in Fig. 2 with those of other SrTiO$_3$ samples with similar carrier densities reported in the literature, including SrTiO$_3$ crystals that were tuned toward ferroelectricity using the approaches mentioned in the Introduction (11, 25, 26, 33, 34). The $T_C$ of the Sm: SrTiO$_3$ film with the highest doping density, which remains metallic, is comparable to previous reports. In contrast, films with lower doping densities have a substantially increased $T_C$, which reaches almost a factor of two near the peak of the superconducting dome (“optimal doping”). Thus, the enhancement of $T_C$ is seen for underdoped and optimally doped films but not on the overdoped side of the superconducting dome of SrTiO$_3$. For direct comparison with unstrained films grown by the same MBE method and $T_C$ defined in the same way, Fig. 3 also shows $T_C$ of unstrained Sm- and La-doped SrTiO$_3$ thin films grown on SrTiO$_3$ substrates, which show $T_C$ similar to La-doped SrTiO$_3$ crystals reported in the literature (34). All samples shown in Fig. 3, except for the three lower doped films on LSAT, which have enhanced $T_C$, exhibit metallic behavior.

Figure 4 shows the superconducting upper critical magnetic field, $H_{c2}$, as a function of temperature. Data follow the relation $H_{c2}(0) = \frac{t^2}{1 + t^2}$, where $t = T/T_C$, which is shown as dashed lines, similar to the behavior observed in La-doped films (35). The values of $H_{c2}$ (i.e., 1.75 T at 50 mK for the film with $n_{3D} = 6 \times 10^{19} \text{ cm}^{-3}$) are comparable to those reported for La-doped SrTiO$_3$ films (35). Table 1 lists quantities derived from the data, including the values for the BCS superconducting gap, $\Delta = 1.75 n_B T_C$, where $n_B$ is the magnetic quantum flux. Here, films with $n_{3D} = 2 \times 10^{19}$ and $1.4 \times 10^{20} \text{ cm}^{-3}$, despite having different critical temperatures, show relatively similar $H_{c2}$ at low temperatures. Thus, $T_C$ peaks near $n_{3D} = 1.4 \times 10^{20} \text{ cm}^{-3}$, while $H_{c2}$ peaks near $n_{3D} = 6 \times 10^{19} \text{ cm}^{-3}$. The behavior of $H_{c2}$ might be related to the multiband nature of this superconductor (36) and requires further investigations beyond the scope of the present study.

### DISCUSSION

To briefly summarize the results, our main findings are as follows: (i) There is an enhancement in $T_C$ of compressively strained SrTiO$_3$ films up to a factor of two, and (ii) the increase in $T_C$ depends on the carrier density and (iii) it is connected to the presence of a crossover in the resistivity in the normal state to $d\rho/dT < 0$ at low temperatures.

The first important conclusion from these results is that the increase in $T_C$ is caused not simply by the epitaxial strain, such as strain-induced modification of phonon modes. If this were the case, then all films, independent of their carrier density, should exhibit enhanced $T_C$, because they are all under the same epitaxial film strain. Instead, the carrier density dependence of the enhancement in $T_C$ and the resistivity upturn point to a direct connection between a (proximal) ferroelectric state and the modified superconducting properties.

As mentioned above, SrTiO$_3$ films grown on LSAT substrates are known to transition to a ferroelectric state as a result of the epitaxial coherence strain (27–30). Even SrTiO$_3$ films containing substantial amounts of carriers (on the order of $10^{19} \text{ cm}^{-3}$) were found to undergo a ferroelectric transition around 140 K (29). Thus, the films in the present study are, at minimum, in proximity to a ferroelectric transition.

Beyond this, the observed crossover in the temperature dependence of the resistivity also hints at the emerging ferroelectric or polar nature of the films. The behavior is in marked contrast to unstrained SrTiO$_3$ films grown on SrTiO$_3$ (Fig. 3; see also (37) for more data) or bulk crystals of SrTiO$_3$ (38), which remain metallic even with orders of magnitude lower carrier densities. The reason is the high dielectric constant of SrTiO$_3$. The Mott criterion (39) for metallic behavior, $n^{1/3} \rho_B \approx 0.25$, where $\rho_B$ is the Bohr radius, is thus easily exceeded, even if we assume a substantially reduced dielectric constant (~1000), which is more typical for ferroelectric SrTiO$_3$ (29). While carrier localization can be caused by

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**Table 1. Values for $T_C$, $H_{c2}$, $\Delta$, and $\xi$ for Sm-doped SrTiO$_3$ films on LSAT with different carrier densities.**

| $n_{3D}$ at 300 K (cm$^{-3}$) | $T_C$ (K) | $H_{c2}$ at 50 mK (T) | $\Delta$ (\mu eV) | $\xi$ (nm) |
|-----------------------------|-----------|----------------------|------------------|--------|
| $2 \times 10^{19}$          | 0.37      | 0.83                 | 56               | 18     |
| $6 \times 10^{19}$          | 0.6       | 1.75                 | 90               | 14     |
| $1.4 \times 10^{20}$        | 0.67      | 0.91                 | 100              | 19     |
| $2.8 \times 10^{20}$        | 0.16      | 0.07                 | 20               | 67     |

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disorder or traps, unstrained films with similar dopant densities and disorder remain metallic. Screening of a polar charge is a reasonable explanation for the observed localization of a fixed amount of the mobile charge density (Fig. 1B). Remotely doped ferroelectric BaTiO$_3$ films undergo a transition that is very similar to the one observed here (40). In contrast, $T_C$ is not increased at higher carrier densities on the overdoped side of the superconducting dome, which remains metallic. At high carrier densities, we expect the long-range interactions needed for ferroelectricity to be screened (20). While there is currently no agreement in the literature as to the pair-breaking mechanism that causes $T_C$ to decrease on the overdoped side, it is expected to play a role here as well.

In summary, our experimental results, especially the carrier density dependence of the observed $T_C$ enhancement, should be of interest for testing the different theoretical models that have been proposed in the literature that relate superconductivity in SrTiO$_3$ to a (proximal) ferroelectric state (14–17). Independent of the precise mechanism, the results point to opportunities to enhance $T_C$ by searching for superconducting oxides that are in proximity to ferroelectricity. It would also be interesting to explore whether similar enhancement could be obtained in superconducting/ferroelectric composite structures.

Last, we would like to note that, during the preparation of this article, we became aware of a recent report of increased $T_C$ in SrTiO$_3$ crystals under uniaxial tensile strain (41). The increase in $T_C$ was attributed to a strain-induced modification of phonon modes, suggesting that other promising approaches to enhance $T_C$ exist for SrTiO$_3$.

**MATERIALS AND METHODS**

Epitaxial SrTiO$_3$ films, which were doped with Sm$^{3+}$, were grown by a hybrid MBE technique described elsewhere (31, 32). The LSAT substrate temperature was 900°C (thermocouple reading), and the growth rate was ~130 nm/hour. The carrier density scaled with the Sm flux during growth, consistent with all Sm dopants acting as donors (+3 formal charge density (Fig. 1B). Remotely doped ferroelectric BaTiO$_3$ films were grown by a hybrid MBE technique described elsewhere (42). A combination of high-resolution x-ray diffraction (see fig. S1) and reflection high-energy electron diffraction oscillations was used to calibrate the film thickness. Laue thickness fringes were visible in x-ray diffraction and confirmed the film thickness. Reciprocal space mapping (see fig. S2) was carried out around the 113 reflection of SrTiO$_3$ on LSAT (42) while being sufficiently thick to avoid substantial carrier depletion from the well-known surface depletion of SrTiO$_3$ (43). A combination of high-resolution x-ray diffraction (see fig. S1) and reflection high-energy electron diffraction oscillations was used to calibrate the film thickness. Reciprocal space mapping (see fig. S2) was carried out around the 113 reflection of SrTiO$_3$ and used to determine the in- and out-of-plane lattice parameters of the film and to confirm that the films remained coherently strained to the LSAT. Cross-sectional high-angle annular dark-field (HAADF) imaging in scanning transmission electron microscopy (STEM) was also used to determine the film thickness and structural quality (fig. S3). We also investigated unstrained Sm-doped SrTiO$_3$ and La-doped SrTiO$_3$ films on (001) SrTiO$_3$ substrates grown under the same conditions.

Temperature-dependent measurements of the longitudinal and Hall resistances were carried out using a Quantum Design Physical Property Measurement System. The Hall carrier densities extracted from the Hall measurements, $n_{3D} = 1/(4eR_H)$, where $t$ is the thickness of the Sm$_x$Sr$_{1-x}$TiO$_3$ thin film, $e$ is the electron charge, and $R_H$ is the Hall coefficient $R_{11} = dR_{xy}/dB$, were extracted from linear fits to the transverse resistance $R_{xy}(B)$ with the magnetic field ($B$). Magnetotransport measurements below 1 K were carried out in a dilution refrigerator (Triton, Oxford Instruments Group). Transport measurements were carried out in van der Pauw geometry with square-shaped samples (5 mm × 5 mm). Ohmic contacts (40-nm Ti/400-nm Au) were deposited on the sample corners (<0.5 mm × 0.5 mm) through a shadow mask using an electron beam evaporator. The superconductivity measurements were carried out using a lock-in amplifier (SR830, Stanford Research Systems) in AC mode with an excitation current of 1 μA and a frequency of 33.33 Hz. The critical field was obtained from measuring the longitudinal resistance while sweeping an out-of-plane magnetic field at different temperatures (see fig. S4).

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/4/eaaw0120/DC1

**Fig. S1.** 2D scan of a SrTiO$_3$ film on LSAT near the 001 reflections.

**Fig. S2.** RSM of a SrTiO$_3$/LSAT heterostructure around the 113 reflection.

**Fig. S3.** Cross-sectional HAADF-STEM images.

**Fig. S4.** Magnetic field dependence of the superconducting transition.

**Fig. S5.** Superconducting transitions for Sm- and La-doped SrTiO$_3$ films grown on SrTiO$_3$ substrates.

**Fig. S6.** Resistivity as a function of temperature for Sm-doped SrTiO$_3$ films grown on SrTiO$_3$ substrates.

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