

Interplay between Polar Distortions and Superconductivity in SrTiO₃

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SrTiO₃ is the first oxide superconductor discovered, but the nature of superconductivity is still heavily debated in this material. SrTiO₃ shows a superconducting state in extremely dilute regimes, in which Debye temperature is significantly larger than Fermi temperature and superconductivity is at odds with Bardeen-Cooper-Schrieffer description. Superconductivity frequently emerges near an electronic order in unconventional superconductors. Superconductivity in SrTiO₃ occurs near a polar instability and recent observations suggest a direct link between the enhancement of and ferroelectric Curie temperature [1], [2]. Tuned SrTiO₃ shows polar superconductivity in which spin-orbit coupling can result in an unconventional superconducting state[3]–[5]. Sensitivity to disorder is a probe to reveal the nature of superconductivity in quantum materials. Conventional superconductors are protected against non-magnetic disorders. Atomic resolution high-angle annular dark-field imaging in scanning transmission electron microscopy (HAADF-STEM) is a powerful technique that is capable of detecting subtle structural disorders.

In this study, we use HAADF-STEM to measure Ti column displacement vectors in the Sm-doped compressively strained SrTiO₃ thin films. These films undergo successive ferroelectric and superconducting transitions. SrTiO₃ thin films were grown using hybrid molecular beam epitaxy (MBE) on (001) LSAT single crystals. Films were doped with Sm with carrier densities of n_{3D} of $6 \times 10^{19} \text{ cm}^{-3}$, $1.4 \times 10^{20} \text{ cm}^{-3}$, and $2.8 \times 10^{20} \text{ cm}^{-3}$. HAADF-STEM images obtained at room temperature and polarization vector assigned to each Ti column. The polarization vector is defined as the difference between the center of mass of the four neighboring Sr columns and the Ti column position obtained by 2D Gaussian fitting.

Figure 1(a) and (b) show the direction of polarization vector for two Sm-doped samples with n_{3D} of $6 \times 10^{19} \text{ cm}^{-3}$, and $2.8 \times 10^{20} \text{ cm}^{-3}$, respectively. The color wheel shows the polarization direction in 30-degree intervals. The large clusters of red vectors indicate the formation of [001] nano-domains on the low doped sample which undergoes ferroelectric transition at 92 K and becomes superconducting at ~600 mK. The displacements of Ti-columns become completely random with increasing the carrier density to $2.8 \times 10^{20} \text{ cm}^{-3}$. The polar phase does not set in for this sample and becomes superconducting at ~200 mK. Sm dopant atoms introduce lattice distortions, destabilizing the cooperative Ti displacements and, as a result, suppress the ferroelectric transition and superconductivity. Figure 2 shows the distribution of the magnitude of the displacements measured for doped and undoped strained SrTiO₃ obtained from 7000 Ti columns.

The undoped strained SrTiO₃ films have the largest polar distortions associated with large polar domains and high Curie temperature. Increasing the dopant concentration suppresses the average magnitude of the polar distortions until the strain field of dopants overlap and distortion increases. Here, we observe a systematic suppression of the correlated polar distortions and ferroelectricity with the introduction of the Sm dopants. We also observe an inverse relationship between the critical temperature of superconductivity and the magnitude of polar distortions. The peak of the superconductivity dome corresponds to the smallest polar distortions, highlighting the sensitivity of superconductivity to non-magnetic disorder in SrTiO₃. The interaction of magnetic disorders and superconductivity in SrTiO₃ provides insight into the nature of the superconducting state [6].

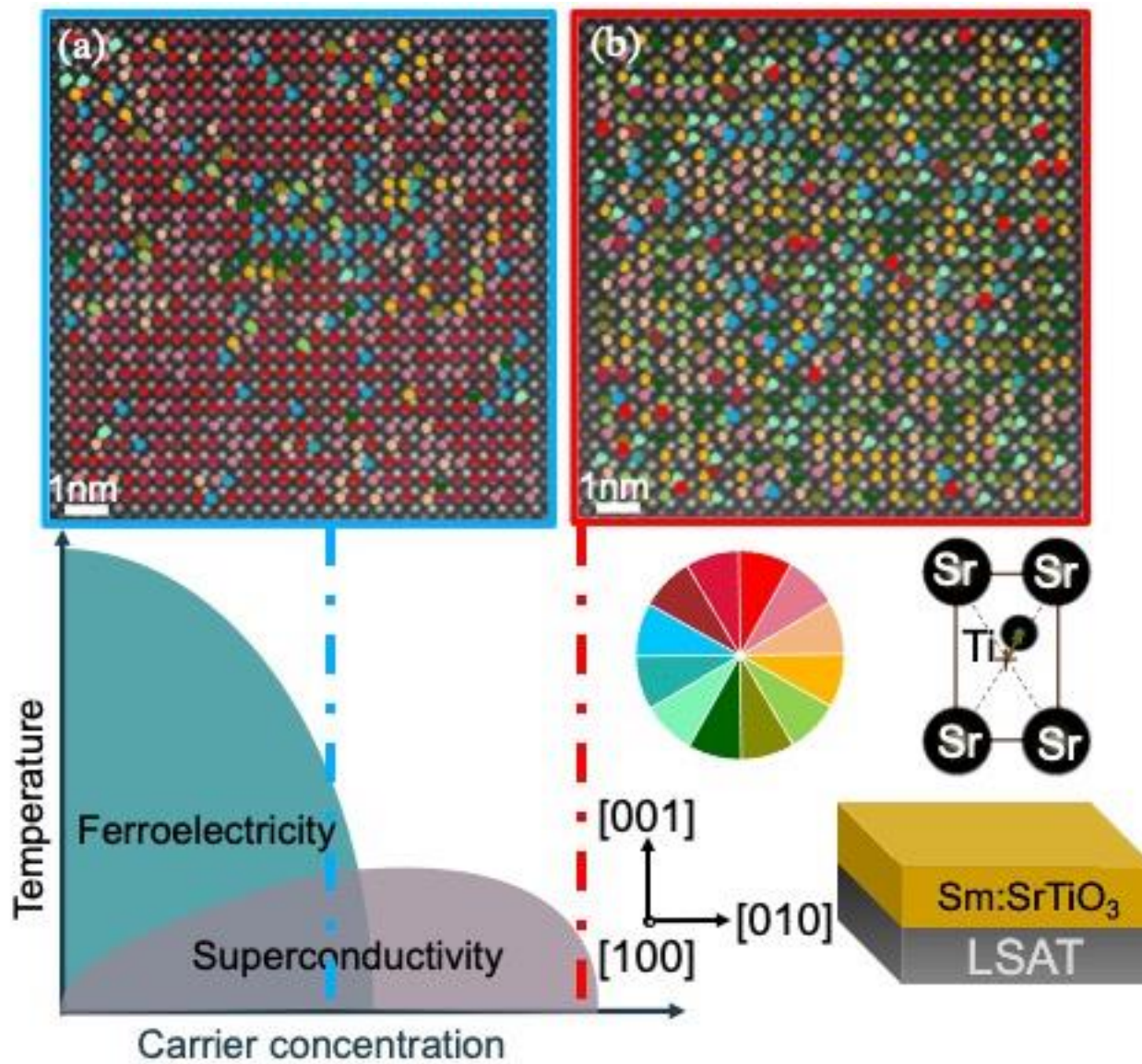


Figure 1. Figure 1- HAADF-STEM images of polarization maps of epitaxially strained Sm doped SrTiO₃ films with n_{3D} of (a) $6 \cdot 10^{19} \text{ cm}^{-3}$ and (b) $2.8 \cdot 10^{20} \text{ cm}^{-3}$ Increasing the doping, polar domains shrink and superconductivity and ferroelectricity get suppressed.

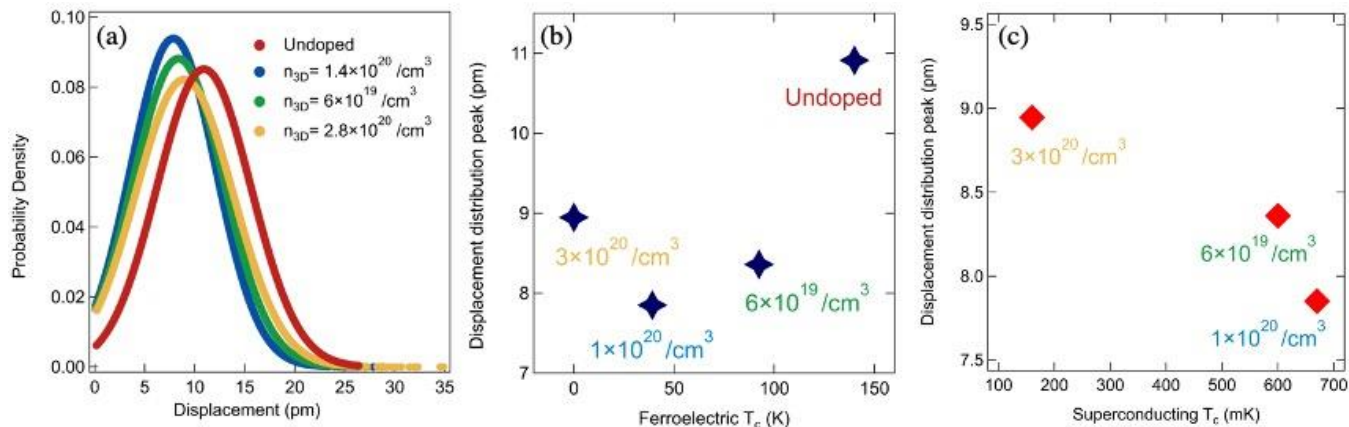


Figure 2. Figure 2 (a) Distribution of the magnitude of the displacements at room temperature (b) change in Curie temperature with the average magnitude of the displacements (c) Superconducting critical temperature as a function of the average magnitude of the displacement

References

- [1] K. Ahadi, L. Galletti, Y. Li, S. Salmani-Rezaie, W. Wu, and S. Stemmer, “Enhancing superconductivity in SrTiO₃ films with strain,” *Sci. Adv.*, vol. 5, no. 4, 2019, doi: 10.1126/sciadv.aaw0120.
- [2] R. Russell, N. Ratcliff, K. Ahadi, L. Dong, S. Stemmer, and J. W. Harter, “Ferroelectric enhancement of superconductivity in compressively strained SrTiO₃ films,” *Phys. Rev. Mater.*, vol. 3, no. 9, p. 091401, Sep. 2019, doi: 10.1103/PhysRevMaterials.3.091401.
- [3] S. Salmani-Rezaie, K. Ahadi, and S. Stemmer, “Polar Nanodomains in a Ferroelectric Superconductor,” *Nano Lett.*, vol. 20, no. 9, pp. 6542–6547, Sep. 2020, doi: 10.1021/acs.nanolett.0c02285.
- [4] S. Salmani-Rezaie, K. Ahadi, W. M. Strickland, and S. Stemmer, “Order-Disorder Ferroelectric Transition of Strained SrTiO₃,” *Phys. Rev. Lett.*, vol. 125, no. 8, p. 087601, Aug. 2020, doi: 10.1103/PhysRevLett.125.087601.
- [5] T. Schumann *et al.*, “Possible signatures of mixed-parity superconductivity in doped polar SrTiO₃ films,” *Phys. Rev. B*, vol. 101, no. 10, p. 100503, Mar. 2020, doi: 10.1103/PhysRevB.101.100503.
- [6] The authors acknowledge support by the U.S. Department of Energy (Grant No. DEFG02-2ER45994).