Switching Magnetism and Superconductivity with Spin-Polarized Current in Iron-Based Superconductor

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We explore a new mechanism for switching magnetism and superconductivity in a magnetically frustrated iron-based superconductor using spin-polarized scanning tunneling microscopy (SPSTM). Our SPSTM study on single-crystal Sr$_2$VO$_3$FeAs shows that a spin-polarized tunneling current can switch the Fe-layer magnetism into a nontrivial $C_4$ symmetric antiferromagnetic order, which cannot be achieved by thermal excitation with an unpolarized current. Our tunneling spectroscopy study shows that the induced $C_4$ (2 × 2) order has characteristics of plaquette antiferromagnetic order in the Fe layer and strongly suppresses superconductivity. Also, thermal agitation beyond the bulk Fe spin ordering temperature erases the $C_4$ state. These results suggest a new possibility of switching local superconductivity by changing the symmetry of magnetic order with spin-polarized and unpolarized tunneling currents in iron-based superconductors.

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Iron-based superconductors (FeSCs) have shown intriguing phenomena related to the coexistence of magnetism and superconductivity below the superconducting transition temperature ($T_c$) [1–3]. Although an understanding of their detailed interplay is still under debate, certain magnetic orders seem to be very crucial in realizing coexistent superconductivity [3–15]. Recent studies have shown new reentrant $C_4$ symmetric antiferromagnetic phases ($C_4$ magnetism from now on) coexisting with superconductivity and have reported that the superconducting $T_c$ is suppressed by $C_4$ magnetic order [16–19]. Direct atomic-scale control of the Fe layer’s magnetic symmetry and the determination of its correlation with superconductivity may be useful for an in-depth understanding of the interplay between superconductivity and magnetism. To our knowledge, there has been no report of a direct real-space observation of such a control by local probes and atomic-scale demonstration of the correlation of magnetism and superconductivity.

In this regard, the parent compound tetragonal iron-based superconductor Sr$_2$VO$_3$FeAs with $T_c ≈ 33$ K [20] is an ideal candidate where the interplay between magnetism and superconductivity can be directly demonstrated due to its nearly degenerate magnetic ground states. Sr$_2$VO$_3$FeAs has two types of square magnetic ion lattices: a square Fe lattice in the FeAs layer and a square V lattice in the two neighboring VO$_2$ layers. At optimal doping, the FeAs layer usually prefers $C_2$ magnetism harboring superconductivity, while the VO$_2$ layer prefers $C_4$ magnetism [1–3,21]. Previous experimental studies of Sr$_2$VO$_3$FeAs [22–28], however, have reported inconsistent results about magnetic order; recent nuclear magnetic resonance (NMR) measurements on single crystals [29] and neutron diffraction [30] experiments show that there is no long-range magnetic order in the V lattice at any temperature, while in the Fe lattice a magnetic order with a small moment of $\sim 0.05\mu_B$, possibly due to frustration, is developed below 50 K. Indeed, a theoretical generalized gradient approximation (GGA) calculation has suggested that there can be a number of competing metastable magnetic states composed of different symmetries in V and Fe layers [21]. This is a reasonable theoretical prediction considering the coupling...
and frustration between V and Fe layers (Supplemental Material Sec. II [31]). Therefore, it has been quite a challenging and interesting experimental task to determine the possible magnetic ground states of the heterostructure superconductor Sr$_2$VO$_3$FeAs and the possible methods to adjust their balances.

One possible way to explore the potentially frustrated magnetic states and their relation to superconductivity is using a spin-polarized scanning tunneling microscope (SPSTM) to locally modify the magnetic environment with a spin-polarized tunneling current. Our density functional theory (GGA) calculation (Supplemental Material Sec. I [31]) showed a possibility that a nonzero net spin density by the injection of a spin-polarized tunneling current can induce a $C_4$ magnetic order from a pristined $C_2$ magnetic order due to the Hund interaction, as illustrated schematically in Figs. 1(a)–1(d). The spin transfer torque and Joule heating effects will then provide the energies to overcome the characteristic potential barriers between the different magnetic states [33].

In this Letter, using a SPSTM we demonstrate that a spin-polarized tunneling current can induce a nontrivial metastable $C_4$ magnetic order in the Fe layer not usually achievable through thermal excitation. We also show that a thermal annealing beyond the bulk Fe magnetic ordering temperature erases the induced $C_4$ magnetic order. From the tunneling spectroscopy analysis measured inside and outside of the region of the induced $C_4$ magnetic order, we also find a signature of suppressed superconductivity in the $C_4$ order region, which is shown to be consistent with the nesting and spin fluctuation scenario of iron-based superconductivity.

We grew single crystals of Sr$_2$VO$_3$FeAs with a self-flux method [29], which are then cleaved in situ at a temperature $\sim$15 K just before mounting on the STM head. Because of the weakly van der Waals–coupled SrO-SrO layers, the cleaved surface is almost always terminated with a symmetrically cleaved SrO layer. For real-space magnetic imaging and injection of a spin-polarized current, we have developed a technique of SPSTM with an antiferromagnetic Cr-cluster tip (Cr tip, from now on), which is created in situ on a Cr(001) surface (Supplemental Material Sec. III [31]). Each Cr tip is confirmed on Cr(001) steps for spin contrasts [Fig. S2(c)] and no gap in the $dI/dV$ spectrum on Cr(001) [Fig. S2(e)].

The 4.6 K STM topographic image of the as-cleaved SrO top layer of Sr$_2$VO$_3$FeAs taken with an unpolarized W tip in Fig. 2(c) shows small randomly oriented domains of quasi-$C_2$-symmetric atomic corrugations. These show no preference for any particular fourfold lattice direction over large scales, consistent with their identity as surface reconstructions (SRs) in the absence of bulk orthorhombicity [30].

In contrast, our SPSTM images with a spin-polarized Cr tip show (above a small bias threshold $\sim$30 meV, $\sim$25 pA), a $C_4$ symmetric ($2 \times 2$) order with intra-unit-cell topographic modulations [Figs. 2(d) and 2(e)] without any signature of SR seen in unpolarized tip images [Fig. 2(c)]. This observation implies that the spin-polarized current induces randomly fluctuating SRs with a flat time average (see Fig. S7). At the same time, any magnetic signal of a Fe layer observed on the top layer oxygen should be the average of the four neighboring Fe spins connected to the As ions in each vertical O-V-As tunneling path as shown in Figs. 2(a) and 2(b). Hence, the most natural explanation for the observed ($2 \times 2$) pattern with three groups of apparent height levels is the plaquette order in the Fe lattice with flat time-averaged SRs. The Fourier-transformed $q$-space image [the inset in Fig. 2(d)] also shows the double wave vectors $Q = (\pi/2, \pi/2)_F$ and $Q^* = (\pi/2, -\pi/2)_F$ expected from the plaquette order in Ref. [8].

To understand the nature of magnetic metastability in this system, we performed a comparative study of bias-dependent topographic measurements using unpolarized (W) and spin-polarized (Cr) tips at 4.6 K. Using the unpolarized tip, shown in Figs. 3(a)–3(c), we found that the surface starts to change at biases beyond $V_B^0 \approx$ 300 meV and the fluctuation becomes so rapid above 400 meV that the surface starts to appear essentially flat as a result of time averaging of the fluctuations. Returning to the low bias condition, as shown in Fig. 3(d), we observe that the square area which experienced the high bias scanning has completely changed with sharply defined boundaries, as shown in Fig. 4(a).

![Image](image_url)

**FIG. 1.** (a)–(d) Schematic illustrations of FeAs-layer configuration potential landscapes for Sr$_2$VO$_3$FeAs in various situations. (a) The imaginary case of FeAs and Sr$_2$VO$_3$ layers being separated sufficiently while apart the electron doping from the Sr layer retained near optimal. The $C_2$ magnetism in the Fe layer with strong superconductivity is preferred. (b) The natural separation found in Sr$_2$VO$_3$FeAs results in interlayer coupling and near degeneracy among the magnetic states with different symmetries, with the $C_2$ magnetism with strong superconductivity still being the ground state. (c) If a sufficiently strong spin-polarized current is injected, the balances among these states may change, possibly resulting in $C_4$ magnetic states with weak superconductivity in the FeAs layer. (d) When the sample is thermally annealed globally or locally with a high bias tunneling current injection, it may return to the ground states with $C_2$ magnetism and strong superconductivity.
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Returning to the very low bias condition shown in Fig.3(h), an unpolarized tip as will be discussed further with Fig.4. The significantly lower bias threshold voltage for a spin-polarized tip is indicative of the final state and the transition mechanism qualitatively different from those achieved by polarized tip case at low bias near 10 meV, but the surface starts to conducting pairing (Refs. [21,29], and Supplemental Material Sec. I [31]). Neither of these two kinds of spin-polarized tip [Fig.3(e)] can be understood from the fact that the pristine state will probably have either C4 single-stripe correlations or (in the presence of disorder) short-range C4 single-stripe orders, both supporting superconducting pairing (Refs. [21,29], and Supplemental Material Sec. I [31]). Neither of these two kinds of C4 magnetism is detectable by SPSTM due to the particular tunneling geometry of this material [Fig. S3(d)].

In order to explore the possibility of erasing of the C4 order by thermal excitation, we performed a variable temperature Cr-tip SPSTM measurement [Figs. 3(i)–3(l) and Fig. S10]. We found that the C4 order can be erased near 60 K, right above the Fe magnetic ordering temperature found in NMR measurement [29]. On the other hand, the application of a magnetic field up to 7 T does not induce any qualitative change in the C4 (2 × 2) pattern in the Cr tip SPSTM topograph [Fig. S11]. These show that the induced C4 order is an antiferromagnetic order in the Fe layer and the switching of the Fe magnetism is reversible by thermal agitation beyond the bulk Fe magnetic ordering temperature.

To study the connection between superconductivity and the C4 magnetic order, we performed a comparative spectroscopic study. We first acquired a large-area topograph using a unpolarized tip with a bias condition below threshold $V_{th}^{N}$. We then scanned over a smaller square area near the center [black dotted square in Fig. 4(a)] with a bias condition exceeding the threshold $V_{th}^{N}$, simulating thermal annealing in this area. Figure 4(a) shows the topograph taken immediately afterwards with a bias condition below $V_{th}^{N}$. It shows the changed surface topographic pattern,
which corresponds to another instance of the nearly degenerate ground states achievable by tunneling current-induced nonuniform thermal excitation. Then we measured the $\frac{dI}{dV}$ spectra inside [annealed, Fig. 4(c), blue solid curve] and outside [as-cleaved, Fig. 4(c), green solid curve] of the central high-bias-scanned region. The tunneling spectra measured in both regions identically show various features: a pair of superconducting coherence peaks near $-6$ and $+6$ meV and SDW gap-edge-like features near $-18$ and $+14$ meV. These spectral features are virtually independent of the changes in SRs as demonstrated in Supplemental Material of Ref. [31]. This implies that the difference in both regions is only the modification of SR due to thermal agitation by the tunneling current and that most of the spectral features, including the superconducting gap, are the physics in the FeAs layer beneath the topmost Sr$_2$VO$_3$ layer [35].

In the case of a spin-polarized (Cr) tip, the results are qualitatively different. Figure 4(b) shows a large-area topograph taken with a bias condition below $V_{\text{SP}}$ after scanning over the smaller square region (black dotted square) with biases over $V_{\text{th}}^\text{SP}$ [Figs. 3(e)–3(g)]. The central square region shows the well-defined $C_4$ domains (and various domain walls) induced by the spin-polarized current causing a sustained spin polarization lowering the $C_4$ order energy under the tip. The $dI/dV$ spectra measured in the region with $C_4$ order [red and purple curves in Fig. 4(d)] show that the superconducting coherence peaks and the SDW-gap-edge-like features are both significantly suppressed in the presence of $C_4$ magnetic order. One plausible explanation for suppressed superconductivity in this particular $C_4$ (plaquette) order is related to the mutual relationship of the spin-wave dispersion in Fig. 2(f) (derived from Ref. [8]) and the overlaid Fermi surfaces observed in angle-resolved photoemission spectroscopy (ARPES) measurement [34]. For the $C_4$ plaquette order, the low-energy spin fluctuations with wave vectors $Q$ and $Q'$ do not satisfy the nesting condition between any pair of the Fermi surfaces $\Gamma$ and $M$ ($X$) and thus are unable to effectively mediate pairing in the spin-fluctuation-based theory of iron-based superconductivity. According to this scenario, the suppression of the nesting condition by the induced $C_4$ plaquette order will have a more drastic effect on superconductivity compared with switching between $C_2$.
FIG. 4. (a) [(b)] W-tip (Cr-tip) topograph with bias conditions below threshold \( V_{\text{th}}^{N} (V_{\text{th}}^{SP}) \) taken after the higher bias scans shown in Figs. 3(a)–3(c) [Figs. 3(e)–3(g)] performed only in the area inside the dotted square. The inset image in the solid square in (b) is a Cr-tip topograph with a higher bias above \( V_{\text{th}}^{SP} \) showing the domains and the domain walls more clearly (see Fig. S8). (c) [(d)] shows the tunneling spectra measured at marked positions with corresponding marker colors in (a) with a W tip [(b) with a Cr tip]. The W-tip spectra were measured at bias [40 meV, 120 pA] inside the \( C_{4} \) region and at the set point of [30 meV, 5 pA] in the pristine region with a larger averaging time to avoid inducing a \( C_{4} \) state during the \( dI/dV \) measurement. All the data are taken at 4.6 K.

and \( C_{4} \) orders that maintain the nesting conditions, as shown in the recent studies on \( \text{Ba}_{1-x}\text{K}_{x}\text{Fe}_{2}\text{As}_{2} \) [17] and \( \text{Ba}_{1-x}\text{Na}_{x}\text{Fe}_{2}\text{As}_{2} \) [18], where a more subtle \( T_{c} \) reduction was observed. Among multiple theories of iron-based superconductivity based on spin fluctuations [36,37] and orbital fluctuations [38,39], our experimental results on this material seem to favor the former.

In summary, we carried out a real-space study of correlation between superconductivity and \( C_{4} \) magnetism in an iron-based superconductor by changing the magnetic symmetry using spin-polarized STM. In this magnetically frustrated material, a spin-polarized tunneling current induced a nontrivial metastable \( C_{4} \) order not usually accessible through thermal excitation, while thermal agitation beyond the bulk Fe spin ordering temperature erased the \( C_{4} \) state. We also observed suppressed superconductivity in the \( C_{4} \) order region induced by a spin-polarized current consistent with the spin-fluctuation-based theories. These are a unique and clear demonstration of switching the Fe-layer magnetism and superconductivity by spin-polarized current injection and thermal agitation.

As suggested in Fig. S12, our findings may be extended toward future studies for heterostructure superconductor devices manipulating magnetism and superconductivity using spin-polarized and unpolarized currents.

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