Band-selective gap opening by a \( C_4 \)-symmetric order in a proximity-coupled heterostructure \( \text{Sr}_2\text{VO}_3\text{FeAs} \)

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Complex electronic phases in strongly correlated electron systems are manifested by broken symmetries in the low-energy electronic states. Some mysterious phases, however, exhibit intriguing energy gap opening without an apparent signature of symmetry breaking (e.g., high-\( T_c \) cuprates and heavy fermion superconductors). Here, we report an unconventional gap opening in a heterostructured, iron-based superconductor \( \text{Sr}_2\text{VO}_3\text{FeAs} \) across a phase transition at \( T_0 \approx 150 \text{ K} \). Using angle-resolved photoemission spectroscopy, we identify that a fully isotropic gap opens selectively on one of the Fermi surfaces with finite warping along the interlayer direction. This band selectivity is incompatible with conventional gap opening mechanisms associated with symmetry breaking. These findings, together with the unusual field-dependent magnetoresistance, suggest that the Kondo-type proximity coupling of itinerant Fe electrons to localized V spin plays a role in stabilizing the exotic phase, which may serve as a distinct precursor state for unconventional superconductivity.

Strongly correlated electron systems often exhibit a variety of self-organized forms with broken symmetry. Particularly, in correlated itinerant electron systems, exemplified by doped copper- and iron-based superconductors (FeSCs) (1, 2), multiple many-body instabilities promote rich and complex landscape of electronic orders, including different types of spin, charge, or orbital orders. The resulting broken symmetry renders the footprint in the electronic structure such as band folding, band splitting, and the gap opening. Investigating those footprints thus in turn provides characteristics of phase—what type of phase transition occurs, what kind of interaction is involved, and how the system lowers the energy across the phase transition. For instance, an anisotropic gap opening only at a segment of the Fermi surface (FS) indicates the breaking of translational symmetry and thus suggests a density wave and spin ordering as the corresponding phase (3–5). An isotropic gap opening, on the other hand, can be attributed to a phase without translational symmetry breaking, such as Mott phase. In addition, the loss of coherent spectral weight near the Fermi level could allow us to determine the underlying interaction, such as strong electron correlation (6–9) and coupling with local magnetic moment (10, 11).

Recently, in a correlated heterostructure system \( \text{Sr}_2\text{VO}_3\), FeAs, a mysterious phase transition is found to occur at \( T_0 \approx 150 \text{ K} \). The order parameter and thus symmetry of the phase are still elusive, despite the clear thermodynamic evidences for its existence. The system is composed of iron arsenide (SrFeAs) and Mott-insulating transition metal oxide (SrVO\(_3\)) layers (Fig. 1A and B) (12–14). The two constituent layers are known to favor distinct \( C_2 \) and \( C_4 \) symmetric magnetic phases, respectively. Through a proximity coupling, however, localized spins in the SrVO\(_3\) layers are expected to couple with Fe 3d electrons in the FeAs layers and obtain strong magnetic frustration, which may lead to a transition to a distinct phase. Indeed, recent experimental investigations revealed that the known magnetic phases such as collinear antiferromagnetic (AF) or nematic order of SrFeAs layer and Néel-type AF order in SrVO\(_3\) layer are not stabilized in this system (15). Furthermore, no evidence of time-reversal or lattice symmetry breaking has been observed across this mysterious phase transition (15–17), suggesting an intriguing intraunit cell order with \( C_4 \) symmetry.

To understand this mysterious phase transition, it is desired to investigate the electronic structure evolution across the transition. Here, via systematic temperature-dependent angle-resolved photoemission spectroscopy (ARPES) studies, we show that the transition accompanies an isotropic gap opening on only one of the FSs with strong interlayer hopping. Our measurements reveal that the lattice \( C_4 \) and time-reversal symmetries are retained across the phase transition, which is incompatible with known gap opening mechanisms such as band-selective Mott (6–9) or spin/charge density-wave transitions (3–5). The band selectiveness, which should be from the

**Significance**

Heterostructures of correlated electronic systems offer versatile platforms for various types of quantum phases and their transitions. A common wisdom states that the proximity coupling between constituent layers plays a secondary role, because it is much weaker than the intralayer interactions. In this work, we present a counterexample of the belief. Namely, the proximity coupling between localized spins and itinerant electrons stabilizes an exotic electronic state with band-selective gap opening whose observation is done in a correlated heterostructure \( \text{Sr}_2\text{VO}_3\text{FeAs} \). Our finding highlights that the proximity coupling can be an effective knob for exotic phases in correlated heterostructures.
interlayer hopping strength, together with an unusual magnetoresistance (MR) below \( T_0 \) emphasizes the importance of proximity coupling with localized V spins to induce the exotic \( C_4 \) symmetric ordered phase. This phase may serve as a distinct parent state for superconductivity in \( \text{Sr}_2\text{VO}_3\text{FeAs} \), in which the usual pairing channel is suppressed by the gap opening.

**Results**

An intriguing phase transition at \( T_0 \approx 150 \) K is clearly indicated by the temperature-dependent resistivity \( \rho(T) \) (Fig. 1C) and specific heat \( C_p(T) \) (Fig. 1D) of our single crystalline samples. Neither thermal hysteresis nor magnetic field dependence in \( T_0 \) is observed in \( \rho(T) \) up to 14 T, whereas significant MR develops below \( T_0 \) as we discuss below. The anomaly in \( C_p(T) \) is sharper than expected in the mean-field behavior of the conventional second order transition, indicating the weakly first-order character. As compared to typical phase transitions found in FeSCs, the observed specific-heat anomaly is sharper than at the nematic transition but smoother than at the magnetic transition in other FeSCs (18, 19), showing significant coupling with lattice degree of freedom (20). In \( \text{Sr}_2\text{VO}_3\text{FeAs} \), a sudden drop of the \( c \)-axis lattice parameter at \( T_0 \) indicates a sizable coupling of the \( c \)-axis lattice contraction to the phase transition (15). No evidence for broken tetragonal \( C_4 \) or time-reversal symmetry across \( T_0 \) was found in a recent nuclear magnetic resonance (NMR) study using both As and V nuclei (15). Also, no additional Bragg peak observed in X-ray and neutron diffraction patterns indicates intact translational symmetry across \( T_0 \) (15–17). Therefore, despite the clear thermodynamic signature of the phase transition as shown in Fig. 1, the phase transition cannot be described by the conventional magnetic, nematic, or density-wave ordering reported in other FeSCs (21, 22).

To understand the electronic response across the intriguing transition, we study the low-energy electronic structure using ARPES. Since the Mott-insulating \( \text{Sr}_2\text{VO}_3 \) layers do not contribute to low-energy states near the Fermi energy (\( E_F \)) (23, 24), the overall electronic structure of \( \text{Sr}_2\text{VO}_3\text{FeAs} \) shown in Fig. 2A and B follows the general band topology of other FeSCs (21, 22, 25). There are three bands centered at the \( \Gamma \) point, one electron band (\( \alpha \)) and two hole bands (\( \beta \) and \( \gamma \)), while two electron bands (\( \delta \) and \( \epsilon \)) are located at the \( M \) point of the Brillouin zone (BZ). For each band, the contributions from Fe orbitals (mostly \( d_x^2, d_y^2, \) and \( d_z^2 \)) differ, as summarized by the color-coded lines in Fig. 2C, which is determined by the systematic, polarization-dependent ARPES study (24). The orbital characters of these bands are similar to those of other FeSCs except for the \( \gamma \) hole band. For other FeSCs, the \( \gamma \) hole band near \( E_F \) is derived almost entirely from the in-plane \( d_{xy} \) orbital and shows a strong two-dimensional character. However, the \( \gamma \) hole band of \( \text{Sr}_2\text{VO}_3\text{FeAs} \) was found to be of admixture of atoms \( \Gamma \) and \( \beta \) orbitals, which leads to the finite \( k \) dispersion.

In terms of energy, the band disperses within the relatively small range of about 10 meV, as illustrated in Fig. 2C with thickness. Since the \( \gamma \) hole band top is located close to \( E_F \), it can provoke the stronger significant \( k \) warping of the \( \gamma \) hole pocket (i.e., even stronger than that of \( \delta \) and \( \epsilon \) electron pockets) (SI Appendix, Fig. S1). This unusual orbital mixing and the resulting \( k \) dispersion of the \( \gamma \) hole band are critical to its response across \( T_0 \). Such behavior is distinct from that of other FSs, as discussed below.

As well as identify the overall band structure of \( \text{Sr}_2\text{VO}_3\text{FeAs} \), we also investigate the detailed low-energy electronic structure near the \( E_F \). We first compare gap opening in the electron (\( \delta \) and \( \epsilon \)) and hole (\( \gamma \)) bands (Fig. 2D and E). The symmetrized energy distribution curves (EDCs) taken at \( T \gtrsim 15 \) K (i.e., well below the \( T_C \) of \( 
\sim 30 \) K) exhibit a clear gap for both the \( \delta \) electron and \( \gamma \) hole bands. The gap for the \( \delta \) electron bands, estimated with the Dynes formula, differ significantly from each other, as shown in Fig. 2F (\( \Delta_\delta \sim 10 \) meV and \( \Delta_\gamma \sim 35 \) meV for electron and hole bands, respectively). A gap is also observed for the small \( \alpha \) electron band, and its size follows the superconducting gap function \( \Delta = \Delta_0 \cos k_x \cos k_y \) (Fig. S3), together with that of the \( \delta \) band (SI Appendix, Fig. S3). The gap in the \( \delta \) electron band obtained from the temperature-dependent spectra (Fig. 2D) follows the Bardeen-Cooper-Schrieffer (BCS)-like temperature evolution and closes at \( T_C \) (Fig. 2F). \( 2 \Delta_0/k_BT_C \) is found to be \( \approx 8 \), which is a typical value for strongly coupled superconductors.

In contrast to the gap behavior of electron bands, surprisingly, the size of the larger gap in the \( \gamma \) hole band remains similar across \( T_C \) (Fig. 2E and F). To investigate the behavior of the anomalous gap in the \( \gamma \) hole band, we obtained data at higher temperatures, in the range between 70 and 170 K, as shown in Fig. 2G. We take the polynomial fit of the spectrum at 170 K as the normal-state spectrum and overlay them as dashed lines in Fig. 2G. First, compared to the normal-state spectrum, it should be noted from the 70 K data are that the spectral weight is transferred within the narrow energy range from \( E_F \) to a peak at a higher binding energy of \( \sim 40 \) meV, forming a gap with a full suppression of spectral weight at \( E_F \) (SI Appendix, Figs. S2 and S4). The overlaid spectra in Fig. 2H clearly show that the gap is gradually filled, and the peak is suppressed as the temperature increases past \( T_C \). Both the gap and peak finally disappear near \( T_0 \approx 150 \) K (Fig. 2I and SI Appendix, Fig. S5). A clear “peak-and-gap” line shape is the key feature distinct from the
so-called pseudogap behavior of other FeSCs (26, 27) and high-$T_C$ cuprates (28, 29)—a partial gap feature only due to spectral weight transfer to a much-wider energy window. We can see that the gap size has little temperature dependence, which is in contrast to the gap evolution expected in the mean-field theory but is consistent with the sharp anomaly in the specific-heat data (Fig. 1D). These observations unambiguously reveal that across $T_0$, unusual electronic gap opening occurs in the hole Fermi pocket with a gap ratio $2\Delta/T_0 \sim 6$, while the electron pockets remain gapless above $T_C$.

Having established that the unusual band-selective gap opening occurs only in the hole pocket below $T_0$, we now examine the in-plane momentum dependence of the gap to determine whether there is any signature of $C_4$ symmetry breaking in the gap structure related to the phase transition. Fig. 3A depicts symmetrized EDCs taken along the hole FS (Fig. 3C) at $T = 60$ K ($T_C < T < T_0$) for $k_z = 0$. Note that the peak-and-gap feature is present in all of the spectra, that is, in all directions. The gap sizes of the spectra are again estimated with the Dynes fit, as indicated by the overlaid curves in Fig. 3A, and the results are plotted in Fig. 3C. The anomalous gap at $T_C < T < T_0$ is almost independent of the in-plane momentum. Assuming a possible azimuthal angle ($\phi$) dependence in the $k_z$-$k_y$ plane, expressed by $\Delta(\phi) = \Delta_0 + \Delta_n \cos n\phi$, the $n$-fold gap anisotropy $\Delta_n$ should be less than $\sim 5\%$ of isotropic gap $\Delta_0$.

It is still conceivable that the gap may not be isotropic at other $k_z$ values, as there is fairly large $k_z$ dispersion for the $\gamma$ hole pocket (24); the ARPES intensity map at the binding energy of 10 meV in the $k_z$-$k_y$ plane near the BZ center in Fig. 3D indeed clearly shows that the $\gamma$ hole pocket has $k_z$ dispersion (SI Appendix, Fig. S1). We therefore took the same set of $\gamma$ hole band gap data for $k_z = \pi$ and plotted these in Fig. 3B. The
peak-and-gap feature is developed below $T_0$ (SI Appendix, Fig. S6), as is the case for the symmetrized EDCs taken at $k_x = 0$ (Fig. 3A). The procedure used to extract the gap from the $k_x = 0$ data yields an isotropic gap for $k_x = \pi$, as plotted in Fig. 3C. It should be noted that the gap sizes for $k_x = 0$ and $\pi$ are almost the same, as shown in Fig. 3C. The EDCs taken with various photon energies between $h\nu = 80$ and 86 eV (corresponding $k_z$ values are indicated by the black dashed lines in Fig. 3D) are also found to have essentially the same gap size (Fig. 3E). The observed anomalous gap above $T_C$ is therefore independent of both the in-plane and out-of-plane momentum, indicating a fully isotropic gap in the $\gamma$ pocket. The fully isotropic gap confirms that the rotational and translational symmetries of the underlying crystal lattice are retained across $T_0$, consistent with the results of previous studies (15).

The observed, anomalous, full gap opening cannot easily be explained by conventional mechanisms of gap opening. In particular, the band-selective and fully isotropic gap opening set strong constraints on its origin. The isotropic gap in the $\gamma$ pocket, centered at the $\Gamma$ point, together with the lack of splitting of degenerate $d_{xz}$ and $d_{yz}$ states of $\delta$ and $\epsilon$ bands (Fig. 2B and SI Appendix, Fig. S7), reflects the $C_4$ nematic order, which is one of the most common orders in FeSCs (21, 22). A band-selective Mott transition, which is often found in FeSCs (6–9), is also unlikely. In a band-selective Mott transition, a particular heavy band loses its coherent spectral weight and becomes localized due to the strong electron correlation effect, while the other bands remain itinerant. In this case, as found for the $d_{xy}$ bands in $K_xFe_2Se_2$ (6–8), the spectral weight is completely suppressed for the entire band in the whole $BZ$ and is transferred to the Hubbard state located far away from $E_F$ by $\sim U$, which is much larger than the bandwidth. In $Sr_2VO_3FeAs$, however, spectral weight transfer occurs only near $E_F$ within an energy window much smaller than the bandwidth; the rest of the $\gamma$ band remains almost intact.

Furthermore, the almost isotropic gap over the whole $\gamma$ FS (Fig. 3C) is also inconsistent with the conventional density-wave
transitions. In density wave–type transitions with a nonzero \( \mathbf{Q} \) modulation, for example, strong gap opening occurs at sections of FS that are connected with the modulation vector \( \mathbf{Q} \), leading to significant gap anisotropy in momentum space (3–5). As shown in our simulations based on mean-field theory for a density-wave order with \( \mathbf{Q} = (\pi,0) \) or \((0,\pi), \) the relative gap anisotropy with respect to the isotropic gap \( \Delta_D/\Delta_0 \) is larger than \( -1 \) (SI Appendix, Fig. S13), clearly distinct from our experimental results. The remaining candidates are associated with an intraunit cell order with a zero \( \mathbf{Q} \), which are often called vestigial phases of a \( C_4 \) symmetric double-\( \mathbf{Q} \) spin order (30, 31). These phases are described by composite spin-orbital order patterns, retaining the \( C_4 \) symmetry. However, even in these phases, characteristic band shift or splitting rather than gap opening is expected to occur, in stark contrast to experimental results as summarized in Fig. 3F. Thus, the selective gap opening only at the \( \gamma \) hole pocket is highly nontrivial and inconsistent with theoretical models describing the various phases due to the broken symmetry known or proposed in FeSCs so far.

While the relevant symmetry breaking of the gap opening is unclear, its band selectiveness may further provide a clue regarding the nature of the \( C_4 \) symmetric order. In other FeSCs, the \( \gamma \) hole pocket mainly arises from the \( d_{xy} \) orbital, which is believed to have a stronger correlation effect than \( d_{x^2-y^2} \)-related bands (22, 32). In \( \text{Sr}_2\text{VO}_3\text{FeAs}, \) however, there are significant contributions from \( d_{xy} \) orbitals to the \( \gamma \) hole pocket, which thus shows finite dispersion along the \( k_z \) direction. This contrasts with the \( \delta \) and \( \epsilon \) electron pockets at the \( M \) point, which have negligible \( k_z \) warping with strong two-dimensionality. The \( \epsilon \) electron pocket at the \( \Gamma \) point appears to have a weak \( k_z \) dispersion compared to the \( \gamma \) hole pocket. These two-dimensional FSs—\( \alpha, \delta, \) and \( \epsilon \) pockets—remain gapless below \( T_0 \) until superconductivity develops below \( T_c; \) that is, the full gap below \( T_0 \) selectively develops at the \( \gamma \) hole pocket that has a distinctive \( k_z \) dispersion and thus strong interlayer hopping. This suggests that proximity coupling to the neighboring Mott-insulating layers plays an important role in triggering the \( C_4 \) symmetric state.

The importance of the proximity coupling is also reflected in the unusual transport properties seen under magnetic fields. Fig. 4 shows the in-plane MR, \( \rho_{ab}(H) \), under magnetic fields of up to 60 T for \( H \parallel ab \) and \( H \parallel c \) in the transverse configuration. At high temperatures above \( T_0 \) positive MR is seen for both \( H \parallel c \) and \( H \parallel ab \), with that for \( H \parallel c \) being slightly larger. This can be understood in terms of the conventional orbital effect typically found in quasi–two-dimensional systems (SI Appendix, Fig. S8). However, an additional contribution of the negative MR starts to emerge for \( H \parallel c \) below \( T_0 \) and dominates the field dependence until it is saturated at higher magnetic fields. Taking the MR for \( H \parallel ab \) as the reference, we estimate the negative contribution of the MR for \( H \parallel c \) by \( \Delta \rho_{ab} = \rho_{ab}(H \parallel c) - \rho_{ab}(H \parallel ab) \) at \( H = 60 \) T, which shows a clear onset at \( T_0 \) (Fig. 4H). The similar behavior is also observed in the longitudinal MR along the \( c \)-axis, as shown in \( \Delta \rho_c (H) = \rho_c (H) - \rho_c (0) \) for \( H \parallel c \) (SI Appendix, Fig. S9). The negative contribution in MR is clearly developed below \( T_0 \), further confirming that observed unusual MR is not due to the orbital effect. Such negative MR has not been observed in any FeSCs, except \( \text{EuFe}_2\text{As}_2 \) (33), in which the spin scattering due to localized Eu spins above \( T_N \) is important. The observed complex, field-dependent MR in \( \text{Sr}_2\text{VO}_3\text{FeAs} \) below \( T_0 \) can only be understood by taking into account the significant scattering of itinerant electrons with fluctuating localized spins, which is suppressed under high magnetic fields. This shows that the phase transition across \( T_0 \) in \( \text{Sr}_2\text{VO}_3\text{FeAs} \) is intimately tied to the hybridization between the itinerant Fe electron fluid and localized V spins caused by proximity coupling.

In \( \text{Sr}_2\text{VO}_3\text{FeAs}, \) the localized V spins are known to remain fluctuating at least down to 5 K, even with a large Curie–Weiss temperature (\( \Theta \sim 100 \) K) (15, 16, 34). The absence of magnetic order for V spins in the square lattice by itself evidences additional Kondo-like coupling \( J_K \) with Fe electrons across the interface, which is frustrated with the intralayer superexchange interaction \( J_0 \) of localized V spins (15). This is consistent with the enhancement of the interlayer \( J_K \) interaction across \( T_0 \) observed in the MR behavior as shown in Fig. 4. A significant spin interaction between the SrFeAs and the SrVO3 layers is also captured in recent measurements of the spin relaxation time, which showed a spin-gap–like behavior with unusual suppression of the stripe-type AF fluctuation (15). Furthermore, we found that \( T_0 \) systematically increases with external pressure (Fig. 4f and SI Appendix, Fig. S10). External pressure mostly reduces the interlayer distance and thus enhances proximity coupling in layered compounds like \( \text{Sr}_2\text{VO}_3\text{FeAs}. \) This pressure dependence is consistent with the notion that the hybridization between itinerant electron in the FeAs layers and the localized spin in the SrVO3 layers is responsible for stabilizing the \( C_4 \) symmetric state.

**Discussion**

Based on these results, we show that the observed band-selective gap opening is independent of symmetry breaking. Employing the highest two-dimensional group on a square lattice, we consider all possible subgroups and investigate the effect of their breaking on band structures (SI Appendix, Table S3). The resulting energy gap opening either occurs in both electron and hole bands or becomes highly anisotropic in the momentum space, which is in sharp contrast to our experimental observations. Thus, we conclude that any symmetry-breaking channels cannot explain the band-selective, fully isotropic gap opening in \( \text{Sr}_2\text{VO}_3\text{FeAs}. \)

Although the precise mechanism of the observed exotic gap opening that preserves the symmetry of normal phase is yet to be identified, we stress that the very existence of the exotic gap opening indicates importance of the Kondo-like proximity coupling between itinerant Fe 3d electron and localized V spins. Since itinerant electrons in FeAs layers are spatially separated from the localized spins in SrVO3 layer and coupled through As orbitals (Fig. 1B), the coupling \( J_K \) is essentially nonlocal. It can thus introduce strong momentum-space anisotropy in hybridization (35, 36). In addition, itinerant Fe electrons have an internal orbital degree of freedom, particularly in the \( \gamma \) hole band with highly mixed \( 3d_{x^2-y^2} \) and \( 3d_{xy} \) orbitals. These nonlocal and multiorbital features may play a role in the abrupt change in \( J_K \) across \( T_0 \) which could lead to the band-selective gap opening (Fig. 2), the onset of negative MR (Fig. 4), and a reduction in the \( c \)-axis lattice parameter (15).

Our results suggest the important role of the magnetic proximity coupling, while it leaves several interesting open questions. Recently, the Jacarino–Peters effect has been observed in the upper critical fields of \( \text{Sr}_2\text{VO}_3\text{FeAs}, \) revealing that the interlayer coupling \( J_K \sim 2 \) meV is AF (37). The relatively small \( J_K \) as compared to the observed gap energy scale \( \Delta \sim 30 \) meV, implies that interactions with larger energy scales are involved in the phase transition. Magnetic exchange interactions in the FeAs and VO3 layers are generally expected to be \( \sim 50 \) meV (38) and \( \sim 10 \) meV (39, 40), respectively. The energy scale of orbital ordering in FeSCs is \( \sim 50 \) meV (41), while that in \( \text{RO}_3 \) (\( R \) = rare earth ion or \( Y \)) is in the range of 50 to 100 meV (40). We stress that the symmetry broken phases associated with those larger energy scales are not stabilized in the present heterostructure, which could lead to a nonlinear dependence of the gap on \( J_K \) as found in correlated systems such as quantum criticality (42, 43) and Kondo effect (44).
Furthermore, the band-selective gap opening is expected to affect the superconducting properties in the FeAs layers significantly. Below $T_C$, the $\gamma$ hole FS disappears, and the typical superconducting pairing channel connecting the $\gamma$ hole and the $\delta$ and $\epsilon$ electron FSs is suppressed. This can invoke other pairing interactions [e.g., with the incipient $\beta$ hole bands (45, 46) or via other $C_4$ symmetric AF fluctuations (30)]. The possibility of more exotic scenarios associated with deconfined phases, such as fractionalized Fermi liquids and orthogonal metals (47–50) or more exotic scenarios associated with deconfined phases, such as fractionalized Fermi liquids and orthogonal metals (47–50) or via other $C_4$ symmetric AF fluctuations (30)], the coexistence of a superconducting order, remains to be explored. Our findings regarding the $C_4$ symmetric order of Sr$_2$VO$_3$FeAs highlight that interfacing transition pnictides and oxides could host diverse exotic phases with entangled spin, orbital, and charge degrees of freedom in tunable manner through proximity coupling.

Materials and Methods

Single Crystal Growth. Sr$_2$VO$_3$FeAs single crystals were grown using self-flux techniques as follows. The mixture of SrO, VO$_3$, Fe, SrAs, and FeAs powders with a stoichiometry of Sr$_2$VO$_3$FeAs:FeAs = 1:2 was pressed into a pellet and sealed in an evacuated quartz tube under Ar atmosphere. The sample was heated to 1,180 °C, held at this temperature for 80 h, cooled slowly first to 950 °C at a rate of 2 °C/h, and then furnace-cooled. The plate-shaped single crystals were mechanically extracted from the flux. High crystallinity and stoichiometry are confirmed by the X-ray diffraction and energy-dispersive spectroscopy. The typical size of the single crystals is $200 \times 200 \times 10 \, \mu m^3$.

Transport Properties and Specific Heat. Magnetotransport properties were measured using conventional four-probe configuration on a single crystal in a 14-T Physical Property Measurement System (Quantum Design), a 33-T Bitter magnet at the National High Magnetic Field Laboratory, Tallahassee, and a 60-T pulse magnet at Dresden High Magnetic Field Laboratory, Dresden. Specific-heat measurements were done on several pieces of Sr$_2$VO$_3$FeAs single crystals ($\sim 1$ mg) using the relaxation method in a 14-T Physical Property Measurement System (Quantum Design).

ARPES. ARPES measurements were performed at beam lines 10.0.1 and 4.0.3 of the Advanced Light Source, Lawrence Berkeley National Laboratory. Samples were cleaved at 10 K in an ultra-high vacuum better than $3 \times 10^{-11}$ Torr. Spectra were acquired with Scienta R4000 analyzer at beam line 10.0.1 and Scienta R8000 analyzer at beam line 4.0.3. Several photon energies, particularly between 40 and 70 eV at beam line 10.0.1 and between 68 and 100 eV at beamline 4.0.3, were used for the ARPES measurements including photon energy dependence. The overall energy resolution was 18 meV or better.

Data Availability. All study data are included in the article and/or SI Appendix.

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