Spontaneous rotational symmetry breaking in KTaO$_3$ interface superconductors

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We report the observation of spontaneous rotational symmetry breaking of the superconductivity at the interface of YAlO$_3$/KTaO$_3$ with a superconducting transition temperature of 1.86 K. Both the magnetoresistance and upper critical field in an in-plane field manifest striking asymmetric twofold oscillations deep inside the superconducting state, whereas the anisotropy vanishes in the normal state, demonstrating that this is an intrinsic property of the superconducting phase. We attribute this behavior to the mixed-parity superconducting state, which is an admixture of $s$-wave and $p$-wave pairing components induced by strong spin-orbit coupling. Our work demonstrates the unconventional nature of the pairing interaction in the KTaO$_3$ interface superconductor, and provides a new platform to clarify a delicate interplay of electron correlation and spin-orbit coupling.

The intriguing properties of interface superconductivity has been a central theme in condensed matter physics in recent years [1]. Indeed, the presence of inversion symmetry breaking and the interfacial coupling between two constitute materials are expected to promote the strong interplay between electron correlations, providing an ideal platform for unveiling the inherent pairing mechanism of unconventional superconductivity and promising avenues for the development of superconductor-based devices [2–4]. The prototype interface superconductivity has been experimentally observed at the interface of LaAlO$_3$/SrTiO$_3$ with a superconducting transition temperature ($T_c$) of 250 mK [5]. Subsequently, the superconductivity at the interface of La$_{1.55}$Sr$_{0.45}$CuO$_4$/La$_2$CuO$_4$ has been also observed [6]. Interestingly, monolayer FeSe films grown on SrTiO$_3$ substrates show superconductivity above 100 K by compared to their bulk with a $T_c$ below 8 K [7,8]. This suggests that the electron correlations and the coupling at the interface cooperatively contribute to the remarkable enhancement of superconductivity [10].

Very recently, unexpected crystalline-orientation dependent superconductivity has been experimentally observed at the interface between EuO (or LaAlO$_3$) and KTaO$_3$ ($T_c \sim 2$ K) [11,13], which shows near two orders of magnitude enhancement in $T_c$ compared to its three-dimensional counterpart [14]. This findings suggest that superconductivity at KTaO$_3$ interfaces is an intrinsic interface property. In particular, we conjecture that the dimensional reduction induces strong electron correlations, which play a crucial role in the spontaneous symmetry breaking of the ground state of (111)-oriented KTaO$_3$ interfaces. Interestingly, an indication of broken symmetry phase with a strong in-plane anisotropy of the electrical resistance has been reported at the interface of ferromagnetic EuO and KTaO$_3$ [11]. Theoretically, this symmetry breaking phase in EuO/KTaO$_3$ may come from the joint effect of the ferromagnetic order in EuO and the strong electron correlation at the interface [15], leading the intrinsic nature of rotational symmetry breaking in KTaO$_3$ interface superconductors to be elusive.

In this Letter, we carry out an experimental study on nonmagnetic YAlO$_3$ thin films with a wide-band gap of 7.9 eV grown on the polar KTaO$_3$(111) substrates. This gap is significantly larger than that of LaAlO$_3$ (5.6 eV) [16], enabling strong confinement potential to restrict the interfacial conducting electrons to a thinner interfacial layer, thus prompting an intriguing quantum behaviors at their interface [17]. Electrical transport measurements on the as-grown films reveal two-dimensional superconductivity with a $T_c$ of 1.86 K, and a superconducting layer thickness of 4.5 nm. By tuning the in-plane azimuthal angle $\varphi$-dependent magnetic field, both the magnetoresistance and upper critical field display pronounced asymmetric twofold oscillations deep inside the superconducting state, while they vanish in the normal state. These results unambiguously demonstrate that the anisotropy with in-plane rotational symmetry breaking is an intrinsic property of the superconducting phase in YAlO$_3$/KTaO$_3$. We thus classify the inversion symmetry breaking KTaO$_3$ interface superconductor as a mixed-parity unconventional superconductivity with an admixture of $s$-wave and $p$-wave pairing components.

The YAlO$_3$/KTaO$_3$ heterostructures are prepared by depositing YAlO$_3$ films on a (111)-oriented KTaO$_3$ single crystal substrate using pulsed laser deposition (see Supplementary Materials [18]). Atomic force microscopy characterizations show that the surface of KTaO$_3$ substrates and YAlO$_3$ films are atomically flat (see Fig. S1 [18]). X-ray diffraction confirms the absence of epitaxial peaks of YAlO$_3$ (see Fig. S2 [18]), thus suggesting that the YAlO$_3$ film is not in a well-defined crystalline phase. The microstructure of the interface is further examined by aberration-corrected scanning transmission electron
magneto-resistance for fields (b) parallel and (c) perpendicular to the plane surface of Sample #1. (d) Temperature dependence of the upper critical field $\mu_0 H_{c2}$.

To further characterize the superconducting behaviors in YAlO$_3$/KTaO$_3$, we measure the magnetoresistance $R_x(\mu_0 H)$ (here, $\mu_0$ is the vacuum permeability) at various temperatures between 0.5 K and 5 K with fields parallel ($\mu_0 H_\parallel$) and perpendicular ($\mu_0 H_\perp$) to the magnetic field. The fundamental superconducting behavior is clearly resolved. Indeed, the magnetoresistance $R_x(\mu_0 H)$ varies differently with $\mu_0 H_\parallel$ and $\mu_0 H_\perp$, and both the superconducting critical fields $\mu_0 H_{c1}$ and $\mu_0 H_{c2}$ parallelly shift to a lower value with the increase of the field, where $\mu_0 H_{c2}$ are evaluated at the midpoints of the normal-state resistance at 5 K. These results provide an indication of a two-dimensional superconducting feature in YAlO$_3$/KTaO$_3$. The temperature-dependent upper critical fields $\mu_0 H_{c2}$ are shown in Fig. 2(d) and are well fitted by the phenomenological two-dimensional Ginzburg-

FIG. 1. (a) HAADF-STEM image of YAlO$_3$/KTaO$_3$ viewed along the [112] zone axis. The inset shows the enlarged HR-STEM image of KTaO$_3$ overlapped with atomic configuration (colored). (b) HR-STEM image and the corresponding EDS elemental mapping of interface. (c) Distribution of Ta$^{5+}$ ions along the [111] crystal axis of KTaO$_3$(111) surface. Ta$^{5+}$ ions are shown with progressively smaller sizes in the three adjacent (111) planes, which are labeled as Ta-I, Ta-II, and Ta-III, respectively. (d) Hall bar structure on YAlO$_3$/KTaO$_3$(111) heterostructure. Here, $\varphi$ is defined as the in-plane azimuthal angle between the applied magnetic field B and the [110]-axis of the lattice, shown in the inset of (d).
Landau (G-L) model [19]: \( \mu_0 H_{c2}^\perp (T) = \frac{\Phi_0}{2\xi_{GL} d_{SC}} \sqrt{1 - \frac{T}{T_c}} \) and \( \mu_0 H_{c2}^\parallel (T) = \frac{\Phi_0}{2\xi_{GL} d_{SC}} \sqrt{1 - \frac{T}{T_c}} \), where \( \Phi_0 \), \( \xi_{GL} \), and \( d_{SC} \) denote a flux quantum, the in-plane superconducting coherence length at \( T = 0 \) K, and the effective thickness of superconductivity, respectively. Using the extrapolated \( \mu_0 H_{c2}^\perp (0) = 0.98 \) T and \( \mu_0 H_{c2}^\parallel (0) = 13.81 \) T, we find \( \xi_{GL} = 18.4 \) nm and \( d_{SC} = 4.5 \) nm, where \( \xi_{GL} \) is significantly larger than \( d_{SC} \), suggesting a two-dimensional nature of superconductivity. Additionally, the in-plane \( \mu_0 H_{c2}^\parallel (0) \) is substantially larger than the Pauli-paramagnetic pair-breaking field \( B_P \approx 3.46 \) T based on the BCS theory in the weak-coupling limit [20, 21]. High values of \( \mu_0 H_{c2}^\parallel (0) \) exceeding \( B_P \) can be realized in the presence of strong spin-orbit coupling owing to the elastic scattering, which results in the suppression of spin paramagnetism effects. The violation of this paramagnetic limit is a common phenomenon in interface superconductors [11, 13, 22–25], especially when the superconducting layer thickness is in the range \( d_{SC} < 20 \) nm. However, the mechanism for realizing \( \mu_0 H_{c2}^\parallel (0) \) value in excess of \( B_P \) remains an open question [11]. Furthermore, the thickness of the superconducting layer in YAI03/KTaO3(111) is \( d_{SC} = 4.5 \) nm, which is the thinnest value recorded in the heterointerface superconductors so far [11, 13, 22, 20]. This result is intuitively expected, since the strong confinement potential induced by YAI03 significantly restricts the superconducting electrons to a thinner superconducting layer [17]. On the other hand, the out-of-plane polar angle \( \theta \)-dependent critical field \( H_{c2}^\theta \) at 1.5 K quantitatively verifies the behavior expected from a two-dimensional structure in YAI03/KTaO3, as shown in Fig. S3 [18]. The \( \theta \)-dependent \( \mu_0 H_{c2}^\theta \) are well fitted by the two-dimensional Tinkham formula and the three-dimensional anisotropic G-L model, given by \( \mu_0 H_{c2}^\theta = \frac{H_{c2}^\theta \cos \theta}{H_{c2}} + \left( \frac{H_{c2}^\theta \sin \theta}{H_{c2}} \right)^2 = 1 \) and \( \left( \frac{H_{c2}^\theta \cos \theta}{H_{c2}} \right)^2 + \left( \frac{H_{c2}^\theta \sin \theta}{H_{c2}} \right)^2 = 1 \), respectively [27, 28]. A cusp-like peak is clearly observed at \( \theta = 90^\circ \) (see Fig. S3 [18]), which is well described by the two-dimensional Tinkham model, as frequently observed in interface superconductivity [11, 20] and layered transition metal dichalcogenides [28, 30].

Since the superconductivity in YAI03/KTaO3 is two-dimensional, the Berezinskii-Kosterlitz-Thouless (BKT) transition describes superconducting phase coherence [31, 32]. Here, the BKT transition temperature defines the vortex unbinding transition, and can be determined using current-voltage (I-V) measurements as a function of temperature \( T \), as shown in Fig. 3(a). Below \( T_c \), we find a critical current \( I_c \), whose value decreases with increase in temperature. The maximal value of \( I_c \) is 330 A at 0.5 K, which is substantially larger than that observed in EuO/KTaO3 [11] and LaAlO3/KTaO3 [13]. Such a high critical current value originates from the high charge carrier concentration (about 1.45×10^14 cm^-2) confined in a thinner superconducting layer of YAI03/KTaO3, promising for large-scale applications in superconductor-based devices. In Fig. 3(b), we also plot the characteristics I-V on a log-log scale, and observe that the slope of the I-V curve smoothly evolves from the normal ohmic state, \( V \sim I \), to a steeper power law resulting from the current exciting vortex-anti-void vortex motion, \( V \sim I^\alpha \), with \( \alpha = 3 \) as shown in Fig. 4(b). In this case, the anisotropic magnetoresistance \( R_\alpha \) is two-dimensional, and becomes minimum when the field is directed along the [110]-axis [19], which is consistent with \( T_c \) as defined in Fig. 2(a). In addition, close to \( T_{BKT} \), an \( R_\alpha \)-vs.-\( T \) dependence, where \( R_0 \) and \( b \) are material parameters, is expected [33]. As shown in Fig. 3(d), the measured \( R_\alpha(T) \) is also consistent with this behavior and yields \( T_{BKT} = 1.85 \) K, in good agreement with the analysis of the \( \alpha \) exponent.

Next, we discuss the in-plane anisotropy of superconductivity in YAI03/KTaO3 using an in-plane azimuthal angle \( \varphi \)-dependent magnetoresistance, where \( \varphi \) is defined as the azimuthal angle between the magnetic field and the [110]-axis of the lattice, as indicated in Fig. 1(d). In the normal state \( T = 5 \) K in Fig. 4(a)], the magnetoresistance \( R_\alpha \) is found to be essentially independent of \( \varphi \), displaying isotropic behavior. While in the superconducting state \( T = 1.5 \) K in Fig. 4(a)], we observe a pronounced asymmetric twofold oscillations of the magnetoresistance \( R_\alpha \) [see Fig. 4(b)]. In this case, the anisotropic magnetoresistance \( R_\alpha \) attains the maximum value when the magnetic field is directed along the [112]-axis (\( \varphi = 90^\circ \)) and becomes minimum when the field is directed along the [110]-axis (\( \varphi = 0^\circ \)) (also see Fig. S4 [18]). Considering that the existence of striking asymmetric twofold oscillations in magnetoresistance manifests deep inside the superconducting region and vanishes in the normal state, we can straightforwardly rule out the possibilities of ex-
trinsic contributions, such as the magnetic field induced Lorentz force effects and the electronic band structure inherent to the KTaO$_3$ with respect to the underlying threefold lattice symmetry [35] [also see Fig. 1(c)], and thus demonstrate that this anisotropy is an intrinsic property of the superconducting phase in YAlO$_3$/KTaO$_3$.

To further reveal the twofold asymmetric superconductivity in YAlO$_3$/KTaO$_3$ in terms of the superconducting gap structure, we extract the upper critical field $\mu_0 H_{c2}$ from the $\varphi$-dependent magnetoresistance $R_\varphi$ in the superconducting region. Interestingly, the in-plane $\varphi$-dependent $\mu_0 H_{c2}$ also displays asymmetric twofold oscillations, providing additional strong evidence for the twofold rotational asymmetry of the superconductivity in YAlO$_3$/KTaO$_3$. In addition, the oscillation of $\mu_0 H_{c2}$ has a $\pi$ phase shift compared with that of the magnetoresistance $R_\varphi$ [see Fig. 1(a)] such that for the $\varphi$ value where superconductivity is hardest to suppress, $\mu_0 H_{c2}$ is the largest and the magnetoresistance $R_\varphi$ is the lowest [Figs. 1(a) and 1(c)], as expected from our intuitions [34, 35]. Since $\mu_0 H_{c2}$ achieves its maximum for the field applied parallel to the [110]-axis, and minimum for the directions $90^\circ$ from the [110]-axis, the superconducting gap leads to a maximum along the [110]-axis and a minimum along the [112]-axis, whose topological contour is similar to that of $sp$-hybridization in molecule [37] [see the inset figure of Fig. 1(d)], signaling that the pairing symmetry of YAlO$_3$/KTaO$_3$ most likely belongs to the $s+p$-wave.

Having experimentally established the asymmetric twofold anisotropy of the superconducting state of YAlO$_3$/KTaO$_3$, we now proceed to elaborate about its origin using the underlying symmetries of the crystal structure without requiring the details of the pairing mechanisms based on the group theoretical formulation of the Ginzburg-Landau theory [38]. This allows us to obtain fundamental information about the superconducting ground state in YAlO$_3$/KTaO$_3$ superconductor. From the viewpoint of group symmetry, if a superconductor possesses an inversion symmetry, the Pauli principle requires a totally antisymmetric Cooper pair wavefunction, which imposes the condition that the superconducting states should be either spin-singlet or spin-triplet, whereas mixed-parity states are forbidden [38]. In the YAlO$_3$/KTaO$_3$ the lack of inversion symmetry, however, tends to mix spin-singlet and spin-triplet driven by strong spin-orbit coupling [39]. Indeed, the conducting electrons with strong spin-orbit coupling originating from the heavy Ta 5$d$ orbitals has been revealed at KTaO$_3$ interfaces [35, 40]. Due to the absence of a mirror plane parallel to the interface of YAlO$_3$/KTaO$_3$, the point group of YAlO$_3$/KTaO$_3$ is $C_{3v}$, which does not contain the symmetry element of an inversion. This situation is analogue to non-centrosymmetric superconductors [39]. Upon inspecting the character table of $C_{3v}$ point group listed in Table S1 [18], we notice that the mixed-parity superconducting state only belongs to the $A_1+E$-representation with the possible basis function of $s+p$. Interestingly, the two-dimensional irreducible representation of $E$ can spontaneously break the threefold rotational symmetry of the crystal [see Fig. 1(c)], leading to a subsidiary uniaxial anisotropy [41], such as a uniaxial $p_x$-wave or $p_y$-wave pairing. Since the upper critical field is proportional to the superconducting gap amplitude $\mu_0 H_{c2} \propto |\Delta_{gap}(\varphi)|$, only the $s+p_x$-wave pairing can give rise to an overall asymmetric twofold anisotropic gap shown in Fig. 1(c). In Fig. 1(d), we theoretically evaluate the mixed-parity superconducting gap $|\Delta_{gap}|$ as a function of the ratio of $s$-wave $\Delta_x$ and $p$-wave $\Delta_p$ components, $\gamma = \Delta_x/\Delta_p$. In the two limiting cases of $s$-wave and $p$-wave pairing, the $|\Delta_{gap}|$ exhibits isotropy and perfect twofold modulations, respectively. For increasing $\gamma$, the asymmetric anisotropy becomes more and more significant. These theoretical results are in qualitative agreement with the experimental observations shown in Fig. 1(c) (also see Fig. S5 [18]). Therefore, we have evidence that the KTaO$_3$ interface with inversion symmetry breaking represents an anisotropic mixed-parity superconductor with an admixture of $s$-wave and $p$-wave pairings driven by strong spin-orbit coupling. This is a relevant and appealing example obtained from heterostructure engineering of non-centrosymmetric bulk superconductors, and itself opens a new avenue to investigate unconventional superconductivity. In particular, it represents a novel platform to assess and clarify the emergence of unconventional superconductivity in systems showing a delicate interplay...
of strong electron correlation and spin-orbit coupling, inherently linked to inversion symmetry breaking in the heterostructures.

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