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Article

Experimental signature of topological superconductivity and Majorana zero modes on β-Bi_2Pd thin films

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ABSTRACT

The search for Majorana fermions in topological superconductors is one of paramount research targets in physics today. Using a cryogenic scanning tunneling microscopy, we here report the signature of topologically nontrivial superconductivity on a single material of β-Bi_2Pd films grown by molecular beam epitaxy. The superconducting gap associated with spinless odd-parity pairing opens on the surface and appears much larger than the bulk one due to the Dirac-fermion enhanced parity mixing of surface pair potential. Zero bias conductance peaks, probably from Majorana zero modes supported by such superconducting states, are identified at magnetic vortices. The superconductivity exhibits resistance to nonmagnetic defects, characteristic of time-reversal-invariant topological superconductors. Our study reveals β-Bi_2Pd as a prime platform to generate, manipulate and braid Majorana zero modes for quantum computation.

1. Introduction

Topological superconductors (TSCs) are a novel quantum phase of matter characterized by a fully gapped bulk state and gapless boundary states hosting exotic Majorana fermions that are their own anti-particles [1]. The Majorana fermions obey non-Abelian braiding statistics and could be useful for fault-tolerant quantum computers [2,3]. Following theoretical proposals [4–8], several experiments have disclosed their signatures in semiconductor nanowires [9,10], iron atomic chains [11] and topological insulators [12,13] by proximity to classical superconductors, all sharing complex hybrid heterostructures. Alternatively, the newly discovered single-component superconductors, such as Cu/Sr/Nb-doped Bi_2Se_3 [14–16], Weyl semi-metal TaAs single crystal [17], In-doped SnTe [18] and $PbTaSe_2$ [19], have been suggested as potential TSC candidates, but far from a final conclusion [20].

Tetragonal Bi_2Pd (hereafter, β-Bi_2Pd) crystallizes into a simple $CuZr_2$-type (I4/mmm) structure (Fig. 1a), and exhibits classical *s*-wave bulk superconductivity with a transition temperature (T_c) close to 5.4 K [21]. Intriguingly, it was recently demonstrated from angle-resolved photoemission spectroscopy (ARPES) that β-Bi_2Pd holds several topologically protected surface bands cross the Fermi level (E_F) [22]. The nontrivial surface states of β-Bi_2Pd are subject to a classical *s*-wave bulk pairing, which naturally satisfies the key ingredients of proximity-induced two-dimensional (2D) topological superconductivity near the surface [4]. Here the proximity induced electron pairing on the spin-momentum-locked topological surface has a nontrivial topology and is obliged to be effectively spinless *p*-wave so as to guarantee the pair wave function antisymmetric [4,23,24]. Such superconducting states are anticipated to carry Majorana zero modes (MZMs) at the end of magnetic vortex lines, and thus reignite numerous research interests in β-Bi_2Pd. However, the subsequent studies consistently reveal a conventional *s*-wave superconductivity [25–27] and no MZM at vortices of β-Bi_2Pd single crystals [28]. In this work, we used a state-of-the-art molecular beam epitaxy (MBE) in ultrahigh vacuum (UHV) to prepare β-Bi_2Pd thin films on $SrTiO_3$(001) substrate and characterized their superconductivity with *in situ* scanning tunneling microscope (STM). We found the experimental evidence for nontrivial and impurity-resistant superconducting gap opening on the surface, as well as possible MZMs at vortices.

2. Experimental

Our experiments were conducted in a cryogenic STM apparatus, connected with a MBE chamber for sample preparation. The base

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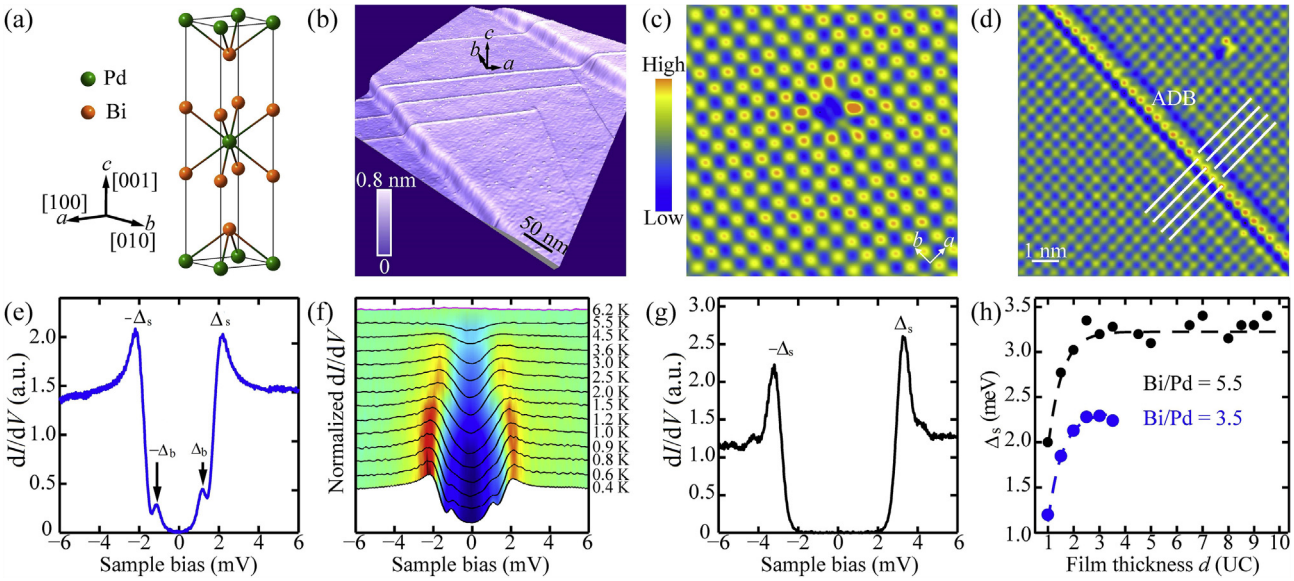

Fig. 1. (Color online) STM characterization of topologically superconducting β-Bi_2Pd films on $SrTiO_3$. (a) Crystal structure of β-Bi_2Pd. Each Pd atom sits at the center of the square prism of eight Bi atoms and the resulting Bi_2Pd motifs are stacked alternately by weak van der Waals forces, forming a body-centered layered structure. The *a*, *b* and *c* axes are taken along the crystal orientations. Along the *c*(001) direction, each unit cell consists of two Bi-Pd-Bi triple layers. (b) STM topography (V = 4.0 V, I = 30 pA, 400 nm × 400 nm) of ~20 UC thick β-Bi_2Pd films, grown with a Bi/Pd flux ratio of 3.1. The bright spots and linear protrusions correspond to Bi adatoms and ADB defects, respectively. The ADBs are orientated along either of the two in-plane crystallographic axes. Unless otherwise specified, our measurements are performed on this films. (c) Zoom-in image of β-Bi_2Pd surface (V = 2 mV, I = 280 pA, 4.5 nm × 4.5 nm), displaying a single Bi vacancy. The bright spheres indicate Bi atoms in the top layer. (d) A magnified ADB (V = −1 mV, I = 300 pA, 8 nm × 8 nm). The eight white lines show a lattice shift in the *b*–*c* atomic planes across the ADB by half of the lattice parameter along the *a* axis (1.7 Å). (e) Differential conductance dI/dV spectrum on the thick β-Bi_2Pd films grown with a Bi/Pd flux ratio of 5.5 (b) at 0.4 K, revealing two distinct superconducting gaps, denoted as Δ_s and Δ_b, respectively. Spectra are acquired with a sample bias voltage of V = 10 mV and tunneling current of I = 300 pA throughout. (f) Temperature dependence of dI/dV spectra in β-Bi_2Pd, displaying completely vanishing gap at 6.2 K (magenta curve). (g) dI/dV spectrum on β-Bi_2Pd films prepared with a larger Bi/Pd flux ratio of 5.5. (h) Pairing gap Δ_s versus both the film thickness d and Bi/Pd flux ratio. Dashed lines are guide to the eye.

pressure of both systems is better than 1.0×10^{-10} Torr (1 Torr ≈ 133.322 Pa). Nb-doped (0.05 wt%) $SrTiO_3$(001) substrates were outgassed in UHV and then annealed at 1200 °C to obtain clean surface. The β-Bi_2Pd films were prepared by co-evaporating high-purity Pd (99.99%) and Bi (99.999%) sources from standard Knudsen cells under Bi-rich condition, with the substrate held at 300–350 °C. More details are discussed in the Electronic Supplementary Material (online). Polycrystalline PtIr tip was cleaned in UHV and calibrated on MBE-grown Ag films prior to all STM measurements at 0.4 K, unless otherwise specified. The differential conductance dI/dV spectra and maps were collected using a standard lock-in technique with a small bias modulation of 0.1 mV at 913 Hz.

3. Results and discussion

MBE growth of β-Bi_2Pd films on $SrTiO_3$ substrate proceeds in Volmer-Weber mode. Epitaxial islands down to a single unit-cell (UC, two Bi-Pd-Bi triple layers) thick with lateral size of several hundreds of nanometers could be prepared, as seen from Fig. S1 (online). We have also established the growth dynamics of high-quality β-Bi_2Pd crystalline films by MBE, as detailed in the Electronic Supplementary Material (online). Fig. 1b shows a constant-current STM topographic image on the atomically flat Bi-terminated (001) β-Bi_2Pd films, grown by using a Bi/Pd flux ratio of 3.1. The films are found to straddle continuously over the neighboring terraces, indicative of a "carpetlike" growth of β-Bi_2Pd across the underlying $SrTiO_3$ steps. The adjacent Bi atoms are spaced ~3.4 Å apart (Fig. 1c), while the out-of-plane lattice constant is approximate to 1.3 nm (Fig. S1 online). They match excellently with those in β-Bi_2Pd crystals [21] and β-Bi_2Pd thin films prepared on Bi/Si(111) substrate [29], justifying the chemical identity of epitaxial films studied here as β-Bi_2Pd. Because other Pd-Bi intermediate compounds all exhibit sharply different crystal structures and lattice parameters from our experimental observations [30]. Furthermore, the similar electron band structure between the MBE-grown thin films and β-Bi_2Pd crystals, revealed by ARPES [21,29], unambiguously backups this claim. Finally, we found no other spurious phase in all samples investigated and the lattice constants alter little with film thickness d. This indicates a negligibly small strain involved, as expected for quasi van der Waals epitaxy of layered β-Bi_2Pd on $SrTiO_3$. Two distinct kinds of surface defects, namely Bi vacancy (Fig. 1c) and Bi adatom (Fig. 1b), as well as anti-phase domain boundary (ADB) (Fig. 1d) are identified.

Scanning tunneling spectroscopy (STS) probes the local density of states (DOS) and can measure the superconducting gap at E_F. In order to minimize the possible strain effects, we measured the tunneling conductance dI/dV spectrum on a thick (~26 nm) β-Bi_2Pd films. Fig. 1e depicts a typical dI/dV spectrum at the base temperature of 0.4 K. In sharp contrast to single-gap superconductivity reported for bulk counterpart [25–28], two pairs of conductance peaks at two different energy scales are noticed, indicating double superconducting gaps in the β-Bi_2Pd films. The smaller one with a low spectral weight is estimated to be (1.0 ± 0.1) meV, close to the reported values of 0.76–0.92 meV in bulk β-Bi_2Pd [25–28]. This hints at its possible origin from the bulk states and we thus dub it as Δ_b for simplicity. On the other hand, the newly discovered gap, which we reveal below stems from the topological surface states of β-Bi_2Pd and is dubbed as Δ_s, is more prominent and significantly enhanced in magnitude.

The temperature-dependent dI/dV spectra have been performed to decipher the origin of Δ_s in the epitaxial β-Bi_2Pd films, as plotted in Fig. 1f. At elevated temperatures, the Δ_s is gradually smeared out and vanishes below 6.2 K. This result is surprising: the larger Δ_s of 2.15 meV would have led to a much higher T_c (13 K or higher assuming the reduced gap ratio $2\Delta/k_BT_c$ = 3.7–4.1 for β-Bi_2Pd [25–28]) if it stems from another bulk band in β-Bi_2Pd [21]. The