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Yu-Shiba-Rusinov bands in ferromagnetic superconducting diamond

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The combination of different exotic properties in materials paves the way for the emergence of their new potential applications. An example is the recently found coexistence of the mutually antagonistic ferromagnetism and superconductivity in hydrogenated boron-doped diamond, which promises to be an attractive system with which to explore unconventional physics. Here, we show the emergence of Yu-Shiba–Rusinov (YSR) bands with a spatial extent of tens of nanometers in ferromagnetic superconducting diamond using scanning tunneling spectroscopy. We demonstrate theoretically how a two-dimensional (2D) spin lattice at the surface of a three-dimensional (3D) superconductor gives rise to the YSR bands and how their density-of-states profile correlates with the spin lattice structure. The established strategy to realize new forms of the coexistence of ferromagnetism and superconductivity opens a way to engineer the unusual electronic states and also to design better-performing superconducting devices.

INTRODUCTION

The interplay between different coexisting physical properties induces emergent phenomena, opening a way to design new electronic devices. The investigated mutual interactions between electrical conductivity/superconductivity and magnetism give rise to, for example, colossal magneto-resistance in manganese oxides (1) and superconducting pairing by spin fluctuation in iron pnictides (2,3). In this context, a new intriguing case where superconductivity and ferromagnetism coexist was recently found in hydrogenated boron-doped diamond films (4).

To assess the effect of magnetic perturbation on superconductivity, one powerful approach is the analysis of the influence of individual magnetic impurities or groups of impurities on the superconducting state. Their effect has been extensively revealed in various superconductors (7), where it was found that individual local magnetic moments interacting with superconducting electrons give rise to in-gap localized states, called Yu-Shiba–Rusinov (YSR) states (8–10). For the investigation of such localized YSR states, scanning tunneling microscopy/spectroscopy (STM/S) is a methodology of choice, because it directly provides the in-gap local density of states (DOS) with high spatial and energy resolution (7, 11–16).

RESULTS

Hydrogenated boron-doped diamond

Heavily boron-doped polycrystalline chemical vapor deposited diamond films with a hydrogen-terminated surface were used for this research. Structural and compositional analyses indicate that the polycrystalline diamond films are metal free (6) and have a thickness of ~900 nm, a mean grain size of ~800 nm, and a boron concentration of ~1.5 × 1021 cm−3. Raman spectroscopy confirms that the sample surface is hydrogen-terminated (6).

Coexistent superconductivity and ferromagnetism

The onset of the superconducting state (ρ = 0) takes place at 2.7 K in the hydrogenated boron-doped diamond. To gain insight into the dimensionality of the system, we measured the resistive superconducting transition ρ(T) in different magnetic fields, μ0H, which were applied perpendicular and parallel to the sample (see Fig. 1, A and B). In contrast to the linear dependences of the offset temperature of the resistive superconducting transition on the applied magnetic fields, the μ0H2–T phase boundaries, as determined by the onset temperature of the resistive superconducting transition, are in good agreement with quadratic fits (see Fig. 1C). Because of the quantum
Fig. 1. Coexistent superconductivity and ferromagnetism in hydrogenated boron-doped diamond. Resistive superconducting transitions in (A) perpendicular (\(\perp\)) and (B) parallel (//) applied magnetic fields, respectively. (C) The temperature dependences of the critical field, \(\mu_0H_{C2}\), measured in magnetic fields perpendicular and parallel to the sample. To build up the \(\mu_0H-T\) phase boundaries, a criterion was set at 95% of the normal-state resistivity to determine the onset critical temperature, and the resistive superconducting transition was linearly extrapolated down to \(\rho = 0\) for the determination of the offset critical temperature in different magnetic fields. The \(\mu_0H-T\) phase boundaries are extrapolated to zero temperature by quadratic fits (solid curves) and linear fits (dashed lines), respectively. (D) Magnetization hysteresis loops measured below the superconducting transition temperature. The map was constructed by normalizing the tunneling spectra to the \(dI/dV\) value at 5 mV, far outside the superconducting gap, and measuring the zero-bias conductance. The arrows and dashed lines indicate the same locations in (A), respectively. (C) Magnifications (39 nm × 39 nm) of the “puddles” where the characteristic YSR bands in Figs. 3 and 4 are observed.

Fig. 2. Topography and spectroscopic mapping of zero-bias conductance in hydrogenated boron-doped diamond. (A) Topography of the diamond surface. Arrows (S1 to S6) indicate the location of the characteristic YSR bands as shown in Figs. 3A and 4A, and dashes (L1 to L4) indicate the direction of the spectroscopic mapping in Fig. 3A. (B) Zero-bias conductance map acquired at 0.5 K, well below the superconducting transition temperature. The map was constructed by normalizing the tunneling spectra to the \(dI/dV\) value at 5 mV, far outside the superconducting gap, and measuring the zero-bias conductance. The arrows and dashed lines indicate the same locations in (A), respectively. (C) Magnifications (39 nm × 39 nm) of the “puddles” where the characteristic YSR bands in Figs. 3 and 4 are observed.

Confinement of the superconducting order parameter in the presence of out-of-plane grain boundaries and twin boundaries, the \(\mu_0H_{C2}-T\) phase boundary measured in perpendicular fields overshoots the in-plane phase boundary at finite temperatures, revealing a crystalline geometry-induced anomalous superconducting anisotropy in the diamond films. When extrapolating the \(\mu_0H_{C2}-T\) phase boundaries down to zero temperature, the quadratic fits converge at \(\mu_0H_{C2}(0 \text{ K}) \approx 4 \text{ T}\) (see Fig. 1C). According to \(\xi_{GL} = [\Phi_0/2\pi \mu_0H_{C2}(0 \text{ K})]^{0.5}\) with \(\Phi_0\) being the flux quantum, the Ginzburg–Landau coherence length \(\xi_{GL}\) is determined to be 9.1 nm. Despite the superconducting anisotropy at finite temperatures, the substantial distinction between \(\xi_{GL}\) and film thickness indicates the 3D nature of the superconductivity.

The magnetization hysteresis loops measured below the superconducting transition temperature show characteristics of both superconductivity and ferromagnetism (see Fig. 1D). The former is featured with the central peak and the low-field diamagnetism in the virgin curve, while the latter is reflected in the overall shape of the loops. The temperature-induced evolution of such anomalous hysteresis loops has been reported in (6). The presence of ferromagnetism in various forms of metal-free carbon has remained an intriguing question for decades. Although both \(sp^3/sp^2\) defects and hydrogenation have been commonly used to explain the ferromagnetism observed in different forms of carbon (6), polycrystalline diamond rich in \(sp^3/sp^2\) defects but without hydrogenation did not show any ferromagnetic behavior (17). Furthermore, theoretical calculations demonstrated that unhydrogenated carbon sites and hydrogen impurities can give rise to local magnetic moments in graphene and diamond, respectively (18–20).

The substantial distinction between the in-plane and out-of-plane magnetization loops indicates the in-plane easy axis of magnetization (see Fig. 1D). Taking into account the robust 3D nature of the superconductivity, the diamond films cannot be simply treated as conventional ferromagnets in the form of thin film, where similar magnetic anisotropy is mainly caused by shape anisotropy (21). The anomalous superconducting anisotropy mentioned above cannot be a cause of the magnetic anisotropy, either, because at 1.8 K and in the magnetic field range of 0 to 0.4 T, the diamond films demonstrate no superconducting anisotropy (see Fig. 1C). The observed magnetic anisotropy is highly likely due to the anisotropic distribution of magnetic impurities.

**YSR bands observed in direct local measurements**

As revealed by STM/S measurements performed at 0.5 K on the surface of the hydrogenated boron-doped diamond (see Fig. 2A), the zero-bias tunneling conductance map demonstrates strong modulations of the local DOS on the nanoscale, which appear as “puddles” (see Fig. 2B and C). The tunneling conductance spectra acquired over the puddles show symmetric maxima and minima around the zero energy inside the superconducting gap 2\(\Delta\) (see Fig. 3A), which are fingerprints of the YSR effect. In contrast to the localized YSR states that are restricted to a few atomic distances in 3D systems (14), these collective excitations extend up to tens of nanometers, revealing the delocalization of the YSR states and the emergence of YSR bands. The extraordinarily large spatial extension of our observed YSR bands can also be seen in the spatial dependence of the integrated in-gap DOS (see Fig. 3B).

The observed YSR bands demonstrate a variety of DOS profiles (see Figs. 3A and 4, A and B). Apart from the pair of peaks at finite energies (\(E_{\text{YSR}} \approx \pm 0.39\Delta\)), the tunneling spectra S1 and S4 both show a third peak at zero energy inside the superconducting gap 2\(\Delta \approx 3 \text{ meV}\). Along with the peaks at \(E_{\text{YSR}} \approx \pm 0.22\Delta\) in spectrum S2, a pair of kinks (\(E_{\text{YSR}} \approx \pm 0.39\Delta\)) emerge at the shoulders of the peaks. Spectrum S3 exhibits two peaks at \(E_{\text{YSR}} \approx \pm 0.35\Delta\) and two kinks at \(E_{\text{YSR}} \approx \pm 0.21\Delta\), while spectrum S5 reveals the emergence of two pairs of peaks at \(E_{\text{YSR}} \approx \pm 0.61\Delta\) and \(E_{\text{YSR}} \approx \pm 0.2\Delta\), respectively. Note that among these YSR bands, even S5 shows a spatial extent of ~9 nm (see L4 in Fig. 3A). In contrast to these YSR bands, the tunneling spectra outside the puddles exhibit Bardeen-Cooper-Schrieffer-like gap structures (see spectrum S6 in Fig. 4, A and B).
Theoretical modeling of YSR bands with a tight-binding BdG approach

To give insights into the origin of the various DOS profiles (see Fig. 4, A and B), we developed a microscopic theory. The ferromagnetism can arise from both bulk hydrogen impurities (20, 22, 23) and hydrogen islands at the surface, and the mechanism is still largely unknown. Because we observed extended but finite (several tens of nanometers) regions with nonnegligible in-gap DOS, we speculate that its origin is the presence of islands of magnetic impurities located at or close to the surface, which interact with the bulk superconducting state. Moreover, although the present STM/S measurements do not have atomic resolution and the character of the distribution of the impurities cannot be determined (see Materials and Methods), it should have strong correlation with the type of the surface. The surface of the polycrystalline nanodiamond films is far from flat, and there are (111) and (100) surfaces (6), giving rise to triangular and rectangular spin lattices on them, respectively (see Fig. 5). Given the strong exchange interaction between the magnetic centers (with Curie temperature $>400$ K), the spins on the surface can be treated as classical ones.

The electronic bands in the presence of a spin lattice were calculated with a tight-binding BdG approach with the localized YSR states on spin sites as the basis (4, 5). The BdG equation in the presence of the classical spin $S$ at $R_j$ is given by

$$\begin{bmatrix} \xi_k \Delta \\ \Delta & -\xi_k \end{bmatrix} \mathbf{w}_k - \sum_j S \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} e^{-i\mathbf{k} \cdot \mathbf{R}_j} \mathbf{w}(R_j) = E \mathbf{w}_k$$

where $\mathbf{w}_k$ stands for the quasi-particle states characterized by wave vector $\mathbf{k}$, $\mathbf{w}(R_j)$ indicates the localized YSR state at site $j$, $E$ is energy, $\xi_k$ is the free electron energy with respect to chemical potential, $J$ is the exchange coupling parameter, $S = 1/2$ is the spin, and $\Delta$ is the superconducting order parameter. We transformed this equation into the tight-binding form on the basis of localized YSR states and then calculated the quasi-particle bands (see Materials and Methods). To reproduce the experimental DOS, we treated the dimensionless exchange interaction $\alpha = \pi v_0 S$ between the spins and conduction electrons ($v_0$ is the DOS of the electronic band at the Fermi energy) and the distance between the spin sites as variables.

Among the experimentally obtained DOS, we focused on S1, S2, and S3, which clearly show the band nature (Fig. 3A). Figure 4C shows the theoretical DOS calculated using rectangular lattices for S1 and S2 and a triangular lattice for S3 with $\alpha = 0.7$ (see Materials and Methods for other parameters). The dimensions of the rectangles are $9 \times 10$ and $9 \times 13$ for S1 and S2, respectively, and the length of the side of the equilateral triangle is 12 for S3, all in units of diamond lattice constant, $a = 3.57$ Å. The numbers and the positions of the maxima of the DOS within the superconducting gap are in good agreement with the experimental data. Qualitatively different DOS profiles were obtained for S1 and S2 using the same spin lattice structure and $\alpha$, because the interaction between the YSR states oscillates with respect to the distance. The good agreement between the theoretical modeling and the experimental data confirms that the observed in-gap excitations are YSR bands. Moreover, the diameter of the YSR localized state (14) and the distance between the nearest lattice points are comparable to each other, which is consistent with the absence of the spatial variation of the DOS in our experimental spectra S1 to S3 obtained with the present spatial resolution of STM/S (see Fig. 3A). Despite the fact that the magnetic impurities tend to destroy the superconductivity, the superconducting temperature of the ferromagnetic diamond is almost unchanged, compared to other doped diamond superconductors (24, 25). This is explained by the low concentration of the hydrogen-related magnetic impurities on surface as readily understood from the dimensions of the lattices used for the calculations. The theoretical analysis also suggests that the present system is close to the regime of gapless superconductivity due to the classical spins (9).

**DISCUSSION**

In summary, we unveiled the presence of long-range coherent YSR bands in a 3D ferromagnetic superconductor, heavily boron-doped diamond with hydrogen-terminated surface. In contrast to localized YSR states, the observed YSR bands show a spatial extent up to tens of nanometers. Limited by the resolution of STM/S measurements on polycrystalline diamond, the spatial distribution of magnetic impurities cannot be extracted from our experiments. By developing a microscopic theory for a 3D superconductor with a 2D spin lattice at its surface, we reproduced the experimental DOS profiles of the observed YSR bands in ferromagnetic superconducting diamond reasonably well. Apart from the Fulde-Ferrell-Larkin-Ovchinnikov state (26, 27) and the so-called domain wall superconductivity (28, 29), introduction of the sparse magnetic impurity lattice is another form...
of the coexistence of the two mutually antagonistic orderings, ferromagnetism and superconductivity. The present results suggest the possibility to construct hybrid systems consisting of, e.g., sparse 2D ferromagnetic lattice and superconductor with spin-orbit coupling (5) to realize unconventional topological superconducting phases.

MATERIALS AND METHODS

Sample preparation

The heavily boron-doped and hydrogenated polycrystalline diamond films were grown using hot filament chemical vapor deposition (CVD). The substrates were undoped single-crystal Si (100) with a 30-nm-thick layer of SiO2 on the surface. The wafers were seeded with a colloidal suspension of diamond nanoparticles (diameter ~20 nm) using an electrospray process (seeding density ~3 × 10^10 cm^-2) (30).

The seeded substrate was then placed 3 mm below a 2200°C tantalum filament in the CVD reactor. A gas mixture of 0.6% CH4 in H2 (total flow, 200 standard cubic centimeters per minute) was thermally dissociated using the hot filament for diamond growth onto the substrate, which was maintained at 800°C. Boron doping was realized by adding diborane (B2H6) to the gas mixture with a B2H6/CH4 ratio of 5%. To ensure that the diamond surface was hydrogen-terminated, after deposition was complete, the CH4 and B2H6 flows were switched off, while the sample remained under the filaments for 1 min in pure hydrogen gas. The filaments were then switched off, allowing the sample to cool back to room temperature under flowing H2 in ~30 min (6).

Electrical transport and magnetization measurements

The electrical transport properties and magnetization of the samples were measured using a Heliox ³He cryostat and an MPMS3 system, respectively.

Scanning tunneling microscopy/spectroscopy

The differential tunneling conductance spectra were acquired by means of a homemade STM (31) cooled to 0.5 K by a commercial Janis SSV cryomagnetic system with a ³He refrigerator. The atomically sharp gold STM tip was obtained in situ by systematically impaling the tip into a clean gold surface. To measure the tunneling current spectra, the feedback loop was turned off, and the tunneling current at different bias voltages applied to the tip was recorded. The applied bias voltage was swept from -10 to +10 mV with an interval of ~20 µV, which is smaller than the width of the in-gap peaks by one order of magnitude. The initial tunneling resistance was 5 megohms. Because the gold tip features a constant DOS, the differential conductance derived numerically from the current spectra represents the local DOS of the diamond surface. Despite the atomic sharpness of the STM tip, the spatial resolution of our STMS measurements was limited to ~3 nm because of the surface roughness of the lab-grown polycrystalline diamond films. As a result of the newly formed nucleation sites, the surface of the diamond crystallites is corrugated on the nanoscale and far from flat. The nanoscale corrugations give rise to additional noise, which limits the spatial resolution of the STMS measurements.

Theoretical modeling

The BdG equation was transformed into the tight-binding form on the basis of localized YSR states (4).

\[
\mathbf{w}(\mathbf{R}) = \sum_j \langle j | S | \mathbf{R} \rangle \left( \begin{array}{c} I_0(\mathbf{R}_j) + I_1(\mathbf{R}_j) & \Delta I_0(\mathbf{R}_j) \\ \Delta I_0(\mathbf{R}_j) & E I_0(\mathbf{R}_j) - I_1(\mathbf{R}_j) \end{array} \right) \mathbf{w}(\mathbf{R}_j) \tag{1}
\]

where \( I_0(\mathbf{R}) = | \mathbf{R}_i - \mathbf{R}_j | \), \( I_0(\mathbf{R}) = (2\pi)^{-3} \int d^3 \mathbf{k} \frac{e^{i \mathbf{k} \cdot \mathbf{R}}}{(E - \xi_{\mathbf{k} \mathbf{l}^2} - \Delta^2)} \), and \( I_1(\mathbf{R}) = (2\pi)^{-3} \int d^3 \mathbf{k} \xi_{\mathbf{k}} e^{i \mathbf{k} \cdot \mathbf{R}} \left( E - \frac{\xi_{\mathbf{k}^2}}{\Delta^2} \right) \). These integrals were evaluated under the condition of \( (\hbar k_F)^2/2m_e \gg \hbar \nu_F >> \Delta \gg E \) as

\[
\begin{align*}
I_0(\mathbf{R}) &\sim -\frac{\pi \nu_0}{\sqrt{\Delta^2 - E^2}} \frac{\sin(k_F R)}{k_F R} \exp \left( -\frac{\sqrt{\Delta^2 - E^2}}{\hbar \nu_F} \right), \\
I_1(\mathbf{R}) &\sim \frac{\pi \nu_0}{k_F R} \left[ \exp \left( -\frac{\sqrt{\Delta^2 - E^2}}{\hbar \nu_F} \right) - 1 \right] \cos(k_F R) + \frac{\hbar \nu_F}{k_F R} - \cos(k_F R) \frac{\sin \left( \frac{\omega_D \nu_F}{\nu} \right)}{\sin(k_F R) \frac{\sin \left( \frac{\omega_D \nu_F}{\nu} \right)}{k_F R}} \tag{2}
\end{align*}
\]

where \( h \) is the reduced Planck constant; \( k_F \) and \( \nu_F \) are the wave vector and velocity at the Fermi energy, respectively; \( m_e \) is the mass of electron; \( \omega_D \) is the Debye frequency; and \( \sin(x) \) is the sine integral. The electron attraction and \( \Delta \) are assumed to be finite when \( |\xi_{\mathbf{k}}| < \omega_D \). The expression of \( I_1 \) was improved so that small distances can be accurately treated in comparison with the previous work based on asymptotic treatment (4).

When there is only one impurity, the eigenvalues \( E \) of Eq. 1 reduce to (8–10)

\[
E = \pm \sqrt{\Delta^2 - E^2} \tag{3}
\]

where \( \alpha = \pi \nu_0 / S \). The energy levels appear within the superconducting gap.

Assuming the periodicity of the impurity centers, Eq. 1 was Fourier-transformed with the period of impurity centers

\[
\lambda_q w'_q = \begin{pmatrix} \sqrt{1 - \lambda_q^2} & -1 \\ -1 & \sqrt{1 - \lambda_q^2} \end{pmatrix} w_q' \begin{pmatrix} I_{0,q} + I_{1,q} & I_{0,q} \\ I_{0,q} & \lambda_q I_{0,q} - I_{1,q} \end{pmatrix} w_q' \tag{4}
\]
Here, $w_q = \sum_j w(R_j) \exp(-i q \cdot R_j)$ is the Fourier transformation of $w(R_j), \lambda_q = E/\Delta_{n,q}(n=0,1)$ is defined by

$$\hat{I}_{n,q} = \sum_{j(0)} e^{-i q \cdot R_j} \hat{I}_n(R_j)$$

or $q = 0$, $I_q(R) = I_{q}(R)/\Delta_{0}(R)$, and $I_{q}(R) = I_{q}(R)/[\Delta_{0}(R)]$. Last, by solving the eigenvalue problem (Eq. 3), we obtained a nonlinear equation for each wave vector $q$

$$\lambda_q = \sqrt{1 - \frac{a^2}{\alpha}} - \lambda_q I_{0,q} + \sqrt{(1 + I_{0,q})^2 + I_{1,1}^2}$$

Neither spin-orbit coupling nor edge of the lattice is considered in our theory. However, the scenario of the emergence of exotic edge states discussed in (5) does not apply.

By numerically solving this equation, we obtained the quasi-particle energy bands. For the simulations, $k_F = 1 \times 10^3$ to $3 \times 10^3$ m$^{-1}$ and $\omega_F \approx 7$ to 8 eV Å were taken from (32), and $a = 3.57$ Å and $\omega_D \approx 160$ meV for diamond were taken from (33). With these parameters, $k_F a = 1$, $\hbar \nu_F a / 2 \approx 2$ eV, $\nu_F / (\nu_D a) \approx 0.08$, and $\Delta / (\hbar \nu_F a) \approx 0.0005$. The shape of the impurity lattice is either a rectangle or regular triangle. The DOS was convoluted with Gaussian function with standard deviation $\sigma/\Delta = 0.14, 0.10,$ and $0.09$ for $S_1$, $S_2$, and $S_3$, respectively. The height of the DOS, which depends on the distance between the STM tip and the surface (14), was rescaled to match the experimental DOS.

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