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Tuning the pairing interaction in a *d*-wave superconductor by paramagnons injected through interfaces

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Unconventional superconductivity and magnetism are intertwined on a microscopic level in a wide class of materials. A new approach to this most fundamental and hotly debated issue focuses on the role of interactions between superconducting electrons and bosonic fluctuations at the interface between adjacent layers in heterostructures. Here we fabricate hybrid superlattices consisting of alternating atomic layers of heavy-fermion superconductor CeCoIn₅ and antiferromagnetic (AFM) metal CeRhIn₅, in which the AFM order can be suppressed by applying pressure. We find that the superconducting and AFM states coexist in spatially separated layers, but their mutual coupling via the interface significantly modifies the superconducting properties. An analysis of upper critical fields reveals that, upon suppressing the AFM order by applied pressure, the force binding superconducting pairing can be tuned non-trivially by magnetic fluctuations (paramagnons) injected through the interface.

In diverse families of strongly correlated electron systems, including cuprates, iron-pnictides, and heavy fermion compounds, superconductivity is often found near a quantum critical point (QCP) where a magnetic phase vanishes in the limit of zero temperature, pointing to a magnetic glue as the source of electron pairing [1-3]. In these materials, microscopic coexistence of superconducting and magnetically ordered phases both involving the same charge carriers is a striking example of unusual emergent electronic phases. Moreover, superconductivity is frequently strongest near the QCP, suggesting that the proliferation of critical magnetic excitations emanating from the QCP plays an important role in Cooper pairing. Despite tremendous research, however, the entangled relationship between superconductivity and magnetism has remained largely elusive.

Recently, realization that interactions between superconducting electrons and bosonic excitations through an atomic interface can have a profound influence on Cooper-pair formation has raised the exciting possibility of a new route to controlling superconductivity. For instance, when a monolayer of FeSe is grown on a SrTiO₃ substrate, the interaction between FeSe electrons and $SrTiO_3$ phonons via the interface enhances the pairing interaction, giving rise to the highest transition temperature T_c among iron-based superconductors [4–7]. This discovery raises the possibility of a magnetic analogue in which the pairing interaction is influenced by magnetic fluctuations through an interface between an unconventional superconductor and a magnetic metal. This concept is illustrated schematically in Figs. 1(a) and 1(b). Besides allowing a new approach to revealing the entangled relationship between magnetism and unconventional

superconductivity, this concept has the advantage that magnetic excitations are tunable and can be enhanced as a magnetic transition is driven toward zero temperature, unlike phonon excitations in SrTiO₃. The state-of-theart molecular beam epitaxy (MBE) technique enables realization of this idea through fabrication of artificial Kondo superlattices with alternating layers of Ce-based heavy fermion superconductors and magnets that are

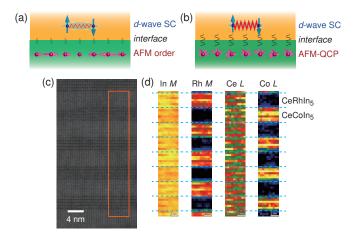


FIG. 1: (a) Schematic figure of the interaction between *d*-wave superconductivity (SC) and static antiferromagnetic (AFM) order via the interface. (b) Interaction between two competing orders under pressure near a quantum critical point (QCP), where AFM order disappears. (c), (d) High resolution cross-sectional (c) TEM and (d) EELS images for CeCoIn₅(5)/CeRhIn₅(5) superlattice. The EELS images were measured in the boxed area in the TEM image, for In M, Rh M, Ce L, and Co L edges.

atomic layer thick [8–10]. These artificially engineered materials are particularly suitable systems to elucidate the mutual interaction through the interface, providing a new platform to study the interplay of competing orders.

The layered heavy fermion compounds $CeMIn_5$ (M = Co, Rh) are ideal model systems in which the interplay between magnetism and superconductivity can be explored, because of their high purity and small energy scales [11–13]. They have similar Fermi surface structures and similar pressure-temperature (p-T) phase diagrams. At ambient pressure, $CeCoIn_5$ is a superconductor $(T_c=2.3 \text{ K})$ with $d_{x^2-y^2}$ -wave symmetry [14–16]. The normal state displays non-Fermi-liquid properties in zero field, including T-linear resistivity, indicative of a nearby underlying QCP [17, 18]. In contrast, CeRhIn₅ orders antiferromagnetically at atmospheric pressure $(T_{\rm N}=3.8\,{\rm K})$ [19]. Its magnetic transition is suppressed by applying pressure and the ground state becomes purely superconducting state at $p > p^* \approx 1.7 \,\text{GPa}$, indicating the presence of a pressure induced QCP [20-23]. As disorder may greatly influence physical properties especially near a QCP, there is a great benefit in examining quantum critical systems which are stoichiometric, and hence, relatively disorder free; both compounds are examples of a small number of such systems. Both host a wide range of fascinating superconducting properties including an upper critical field H_{c2} that is limited by extremely strong Pauli pair-breaking [14, 21].

To realize hybrid heterostructures shown in Figs. 1(a) and 1(b), we fabricate superlattice films with alternating block layers (BLs) of n unit-cell-thick (UCT) CeCoIn₅ and m-UCT CeRhIn₅, CeCoIn₅(n)/CeRhIn₅(m). We demonstrate that the pairing interaction in a d-wave superconductor is tuned by injecting magnetic fluctuations through the atomic interface.

The hybrid superlattices $CeCoIn_5(n)/CeRhIn_5(m)$ with c axis oriented structure are grown on a MgF₂ substrate by the MBE technique [8–10] and are terminated by a CeCoIn₅ layer at the top surface. Figures 1(c)and 1(d) display high-resolution cross-sectional transmission electron microscope (TEM) image and electron energy loss spectroscopy (EELS) images, respectively, of a $CeCoIn_5(5)/CeRhIn_5(5)$ superlattice. The EELS images clearly resolve the 5-UCT CeCoIn₅ and CeRhIn₅ BLs, demonstrating sharp interfaces. The EELS images, along with analysis of the TEM intensity and lateral satellite peaks in an X-ray diffraction pattern [24], indicate no discernible atomic inter-diffusion between the neighboring Co and Rh layers. High-pressure resistivity measurements have been performed under hydrostatic pressure up to 2.4 GPa using a piston cylinder cell with Daphne oil 7373 as pressure transmitting medium.

Figures 2(a) and 2(b) depict the resistively determined p-T phase diagrams of separate, MBE-grown epitaxial thin films of CeCoIn₅ and CeRhIn₅, whose resistivities (ρ) are shown in Figs. 2(c) and 2(d), respectively. The

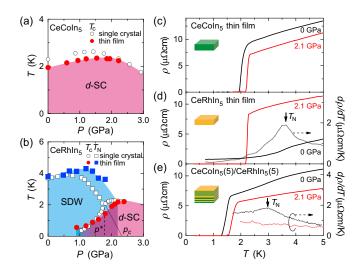


FIG. 2: (a), (b) p-T phase diagrams of thin films and single crystals of (a) CeCoIn₅ and (b) CeRhIn₅. (c) Temperature dependence of the resistivity of CeCoIn₅ thin film at ambient pressure and at p = 2.1 GPa. (d) and (e) show temperature dependence of the resistivity (solid lines, left axes) and its temperature derivative $d\rho(T)/dT$ (dotted lines, right axes) for CeRhIn₅ thin film and CeCoIn₅(5)/CeRhIn₅(5) superlattice at ambient pressure and at p = 2.1 GPa, respectively. The peak of $d\rho(T)/dT$ corresponds to AFM transition.

p-T phase diagrams of both films are essentially those of single crystals. T_c (=2.0 K) in the CeCoIn₅ thin film, however, is slightly reduced from the bulk value, whereas $T_{\rm N}$ (=3.7 K) of CeRhIn₅ thin film is almost the same as that in a single crystal. With pressure, T_c of the CeCoIn₅ thin film increases and shows a broad peak near $p \sim 1.7$ GPa. Similar to CeRhIn₅ single crystals [17, 20, 22], superconductivity in the thin films develops at $p \gtrsim 1$ GPa where it coexists with magnetic order. In analogy to CeRhIn₅ single crystals, there appears to be a purely superconducting state at $p \gtrsim 2.1$ GPa (Fig. S3 in [24]), which is a slightly higher pressure than that required to remove evidence for AFM order in single crystals.

Figure 2(e) compares the *T*-dependence of $\rho(T)$ and its temperature derivative $d\rho(T)/dT$ for a CeCoIn₅(5)/CeRhIn₅(5) superlattice at ambient pressure and at p = 2.1 GPa. At ambient pressure, a distinct peak in $d\rho(T)/dT$ associated with an AFM transition can be seen at 3K in addition to a superconducting transition at ~ 1.4 K [21]. T_c and T_N of the hybrid superlattice are lower than that of the CeCoIn₅ and CeRhIn₅ thin films, respectively, suggesting that they are influenced by dimensionality and electronic structures. However, they are still larger than that of respective CeCoIn₅/YbCoIn₅ and CeRhIn₅/YbRhIn₅ superlattices (Fig. S4 in [24]) [9, 25], indicating the importance of mutual interaction between the CeCoIn₅ and CeRhIn₅ BLs. On the other hand, at p = 2.1 GPa, there is no signature

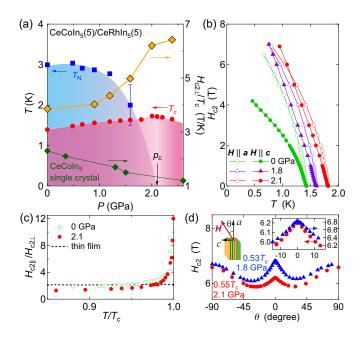


FIG. 3: (a) p-T phase diagram of CeCoIn₅(5)/CeRhIn₅(5) superlattice. Out-of-plane upper critical field $H_{c2\perp}$ normalized by T_c , $H_{c2\perp}/T_c$, measures the coupling strength of the superconductivity. (b) Temperature dependence of in-plane and out-of-plane upper critical fields at ambient pressure and at p = 1.8 and 2.1 GPa. (c) Anisotropy of upper critical field, $H_{c2\parallel}/H_{c2\perp}$, near T_c of superlattices at ambient pressure and at 2.1 GPa, along with the data of CeCoIn₅ thin film. (d) Angular dependence of upper critical field of superlattice at p = 1.8 and 2.1 GPa. The inset is an expanded view of the low angle region.

for magnetic order, while the superconducting state remains with slightly higher T_c than at ambient pressure. In Fig. 3(a), we plot the *p*-dependence of T_c and T_N determined by the peak in $d\rho(T)/dT$. At $p \sim 2$ GPa, T_c is at a maximum, forming a dome-shaped *p*-dependence. With pressure, T_N is suppressed gradually at low *p*, followed by a rapid suppression at $p \gtrsim 1$ GPa (Fig. S4 in [24]). At $p \gtrsim 1.6$ GPa, evidence for magnetic order is hidden beneath the superconducting dome. Although there is a large ambiguity in a determining a critical pressure p_c , a simple extrapolation of $T_N(p)$ gives $p_c \sim 2$ GPa where T_c is maximized. Further, this critical value is very close to that of p_c in CeRhIn₅ single crystals.

We demonstrate that two-dimensional (2D) superconductivity is realized in CeCoIn₅ BLs in the whole pressure regime. Figures 3(b) and 3(c) depict the *T*-dependence of the upper critical field determined by the mid point of the resistive transition in a magnetic field *H* applied parallel $(H_{c2\parallel})$ and perpendicular $(H_{c2\perp})$ to the *ab* plane and the *T*-dependence of the anisotropy of upper critical fields, $H_{c2\parallel}/H_{c2\perp}$, respectively. The anisotropy diverges on approaching T_c , in sharp contrast to the CeCoIn₅ thin film whose anisotropy shows little *T*-dependence up to T_c . This diverging anisotropy in the superlattice is a characteristic feature of 2D superconductivity, in which $H_{c2\parallel}$ increases as $\sqrt{T_c - T}$ due to the Pauli paramagnetic limiting, but $H_{c2\perp}$ increases as $T_c - T$ due to orbital limiting near T_c [9]. This result, along with the fact that the thickness of the CeCoIn₅-BL is comparable to the perpendicular superconducting coherence length $\xi_{\perp} \sim 3-4$ nm, indicates that each 5-UCT CeCoIn₅ BL effectively acts as a 2D superconductor [9]. The 2D superconductivity is reinforced by the angular variation of $H_{c2}(\theta)$. Figure 3(d) and its inset show $H_{c2}(\theta)$ below and above p^* . For both pressures, at $T \ll T_c$, $H_{c2}(\theta)$ in the regime $|\theta| \leq 30^\circ$ is enhanced with decreasing $|\theta|$ and exhibits a sharp cusp at $\theta = 0$. This cusp behavior is typical for a Josephson coupled layered superconductor [26].

We note that in stark contrast to CeRhIn₅ single crystal and our thin film, each CeRhIn₅ BL in $CeCoIn_5(5)/CeRhIn_5(5)$ superlattice is not fully superconducting even when the AFM order is suppressed under pressure, which leads to the realization of 2D superconductivity in a wide range of pressure. In fact, as shown in Fig. 3(d), overall angle dependence of $H_{c2}(\theta)$ including the cusp structure near $\theta = 0$ is observed at p = 1.8 GPa, where the bulk superconductivity is not observed in CeRhIn₅ thin film (Fig. 2(b) and Fig. S3 in [24]). Essentially a very similar angle dependence of $H_{c2}(\theta)$ is observed at $p = 2.1 \,\text{GPa}$. These results imply that 2D superconductivity occurs in CeCoIn₅ BLs even above p_c . Moreover, in a CeRhIn₅(5)/YbRhIn₅(7) superlattice zero resistivity is not attained under pressure (Fig. S6 in [24]), in contrast to $CeCoIn_5(n)/YbCoIn_5(5)$, where zero resistivity is attained even for n=3 [9]. These results indicate that, with the reduction of BL thickness, the superconductivity of CeRhIn₅ is strongly suppressed. This may be related to the incommensurate magnetic structure of CeRhIn₅ with ordering vector q = (0.5, 0.5, 0.297) [19], in which the long-wave-length AFM fluctuations perpendicular to the layers are suppressed in CeRhIn₅ BLs with atomic layer thickness. In CeCoIn₅, on the other hand, AFM fluctuations with different q = (0.45, 0.45, 0.5) are dominant [27]. This commensurability along the c axis would be better compatible with the superlattice structure, and as a result, the superconductivity is robust against the reduction of BL thickness [28].

Recent site-selective nuclear magnetic resonance measurements on a CeCoIn₅(5)/CeRhIn₅(5) superlattice at ambient pressure report that the AFM order is not induced in CeCoIn₅ BLs [29]. As pressure suppresses the magnetic ordering in CeRhIn₅ and tunes CeCoIn₅ towards a Fermi liquid state, it is unlikely that AFM order is induced in the CeCoIn₅ BLs under pressure.

We here comment on the low temperature anisotropy of H_{c2} of the CeCoIn₅(5)/CeRhIn₅(5) superlattice (Fig. 3(b)). At p = 2.1 GPa, $H_{c2\perp}$ exceeds $H_{c2\parallel}$ at low temperatures. Such a reversed anisotropy of H_{c2} has been reported in CeRhIn₅ single crystal above the pres-

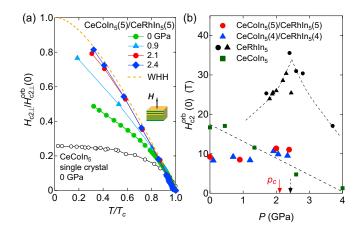


FIG. 4: (a) Out-of-plane upper critical field $H_{c2\perp}$ normalized by the orbital-limited upper critical field at T = 0 K, $H_{c2\perp}/H_{c2\perp}^{c2\pm}(0)$, for CeCoIn₅(5)/CeRhIn₅(5) superlattice is plotted as a function of the normalized temperature T/T_c . Two extreme cases, i.e. the result of the bulk CeCoIn₅ dominated by Pauli paramagnetic effect and the WHH curve with no Pauli effect, are also shown. (b) Pressure dependence of $H_{c2}^{crb}(0)$ of CeCoIn₅(n)/CeRhIn₅(n) superlattices with n = 4and 5 for $H \parallel c$. For comparison, $H_{c2}^{crb}(0)$ of CeRhIn₅ single crystals for $H \parallel a$ and that of CeCoIn₅ single crystal for $H \parallel c$ are shown. Solid and dashed arrows represent p_c for CeCoIn₅(n)/CeRhIn₅(n) superlattices and CeRhIn₅ single crystal, respectively.

sure where the AFM order disappears [11, 22]. However, similar reversed anisotropy $(H_{c2\perp} > H_{c2\parallel})$ is preserved at p = 1.8 GPa, where $H_{c2\parallel}$ exceeds $H_{c2\perp}$ in CeRhIn₅ single crystal and thin film. This indicates that anisotropy reversal of H_{c2} occurs under pressure in 5-UCT CeCoIn₅ BLs. Based on these results, we conclude that 2D superconducting CeCoIn₅ BLs in CeCoIn₅(5)/CeRhIn₅(5) are coupled by the Josephson effect in the whole pressure regime.

Application of pressure leads to a drastic change in the nature of superconductivity in the hybrid superlattices. Figure 4(a) depicts the *T*-dependence of $H_{c2\perp}$, normalized by the orbital-limited upper critical field at T = 0 K, $H_{c2\perp}^{orb}(0)$, which is obtained from the Werthamer-Helfand-Hohenberg (WHH) formula, $H_{c2\perp}^{orb}(0) = -0.69T_c(dH_{c2\perp}/dT)_{T_c}$. We also include two extreme cases: $H_{c2\perp}/H_{c2\perp}^{orb}(0)$ for bulk CeCoIn₅ [30], in which H_{c2} is dominated by Pauli paramagnetism, and the WHH curve with no Pauli effect. Pressure dramatically enhances $H_{c2\perp}/H_{c2\perp}^{orb}$. What is remarkable is that, in the high pressure regime near p_c , $H_{c2\perp}/H_{c2\perp}^{orb}$ nearly coincides with the WHH curve, indicating that $H_{c2\perp}$ is limited solely by orbital pair-breaking.

The fact that $H_{c2\perp}$ approaches the orbital limit provides important insight on superconductivity of the hybrid superlattice. In CeCoIn₅/YbCoIn₅, where YbCoIn₅ is a conventional metal, Pauli pair-breaking effect is

weakened in the superlattice compared with the bulk due to local inversion symmetry breaking at the interfaces, which splits the Fermi surfaces with spin texture and thus effectively suppresses the Zeeman effect [31, 32]. This leads to the Rashba-induced anisotropic suppression of the Zeeman effect [10], which may be partly responsible for the observed reversed anisotropy $H_{c2\parallel}/H_{c2\perp} < 1$ at low temperatures (Fig. 3(d)). However, this effect is less important in $CeCoIn_5(n)/CeRhIn_5(n)$ superlattices compared with $CeCoIn_5/YbCoIn_5$, which is evidenced by the fact that $H_{c2\perp}/H_{c2\perp}^{\rm orb}(0)$ does not strongly depend on n (Fig. S5 in [24]). Moreover, such an effect is not expected to have significant pressure dependence. Therefore, there must be a different mechanism that significantly enhances the Pauli-limiting field $H_{c2}^{\text{Pauli}} =$ $\sqrt{2}\Delta/g\mu_B$, where g is the g-factor of electrons and μ_B is the Bohr magneton. An enhancement of H_{c2}^{Pauli} is not due to a dramatic suppression of g. As g is enhanced by pressure in both CeCoIn₅ and CeRhIn₅ [21], g is expected to be enhanced with pressure in the superlattice. Therefore the enhancement of $H_{c2\perp}^{\text{Pauli}}$ is attributed to a strong increase in the superconducting gap Δ . This is supported by the observed enhancement of $H_{c2\perp}/T_c$ upon approaching p_c shown in Fig. 3(a). We note that $H_{c2\perp}$ below p^* exceeds that of CeRhIn₅ single crystals, implying that the enhancement of $H_{c2\perp}/T_c$ should be attributed to the CeCoIn₅ layers. Because $H_{c2\perp} \approx H_{c2\perp}^{\text{Pauli}} \ll H_{c2\perp}^{\text{orb}}(0)$ in the low *p* regime and $H_{c2\perp} \approx H_{c2\perp}^{\text{orb}}(0) \ll H_{c2\perp}^{\text{Pauli}}$ near $p \sim p_c$, the enhancement of $H_{c2\perp}/T_c$ directly indicates an enhancement of $H_{c2\perp}^{\text{Pauli}}/T_c$ and hence Δ/k_BT_c . This behavior contrasts with observations on $CeCoIn_5$ single crystals, in which H_{c2}/T_c decreases with pressure. The enhancement of Δ/k_BT_c is a consequence of an enhanced pairing interaction. In a spin fluctuation mediated scenario, the pairing interaction is mainly provided by high energy spin fluctuations whose energy scale is well above Δ and low energy fluctuations cause pair-breaking. Since the high energy fluctuations enhance T_c while low energy ones reduce T_c , enhancement of the pairing interaction can give rise to an increase of Δ/k_BT_c without accompanying a large enhancement of T_c , which is consistent with the observed behavior. Thus, the present results demonstrate that the pairing interaction in CeCoIn₅ BLs is strikingly enhanced as a result of the quantum critical magnetic fluctuations that develop in CeRhIn₅ BLs, which are injected into CeCoIn₅ BLs through the interface.

It is well established that quantum fluctuations strongly influence normal and superconducting properties in many classes of unconventional superconductors. One of the most striking is a diverging effective quasiparticle mass m^* upon approaching the QCP, as reported in cuprate, pnictide and heavy-fermion systems [3, 23, 33]. Such a mass enhancement gives rise to a corresponding enhancement of H_{c2}^{orb} , which is proportional to $(m^*\Delta)^2$. Here we stress that there is a fundamental difference in the present hybrid superlattices. Figure 4(b) depicts the *p*-dependence of $H_{c2\perp}^{orb}$ of the CeCoIn₅(*n*)/CeRhIn₅(*n*) superlattices with n = 4 and 5, along with results for CeCoIn₅ and CeRhIn₅ single crystals [13, 22]. In contrast to a CeRhIn₅ single crystal which shows a sharp peak at the critical pressure, $H_{c2\perp}^{orb}$ of the superlattices depends weakly on pressure with no significant anomaly at p_c . Compared to the monotonic decrease observed in single crystal CeCoIn₅, this weak dependence is consistent with an enlarged gap Δ , but the results suggest the absence of mass enhancement in the CeCoIn₅ BL. Such a behavior is in contrast to usual expectations for quantum criticality, details of which deserve further studies.

In summary, we have designed and fabricated the hybrid superlattice CeCoIn₅/CeRhIn₅ formed by alternating atomically thick layers of a *d*-wave heavy fermion superconductor CeCoIn₅ and an AFM metal CeRhIn₅. The present results demonstrate the importance of the interface between which unconventional superconducting and nonsuperconducting magnetic layers can interact with each other. In particular, the strength of the pairing interaction can be tuned by magnetic fluctuations, or paramagnons, injected through the interface, highlighting that the pairing interaction can be maximized by the critical fluctuations emanating from the magnetic QCP without an accompanying mass enhancement. The fabrication of a wide variety of hybrid superlattices paves a new way to study the entangled relationship between unconventional superconductivity and magnetism, offering a route to exploring the emergence of novel superconducting systems and the roles of their interface.

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