Extremely strong-coupling superconductivity in artificial two-dimensional Kondo lattices

Y. Mizukami¹, H. Shishido^{1†}, T. Shibauchi¹, M. Shimozawa¹, S. Yasumoto¹, D. Watanabe¹, M. Yamashita¹, H. Ikeda¹, T. Terashima², H. Kontani³ and Y. Matsuda^{1*}

When interacting electrons are confined to low dimensions, the electron–electron correlation effect is enhanced dramatically, which often drives the system into exhibiting behaviours that are otherwise highly improbable. Superconductivity with the strongest electron correlations is achieved in heavy-fermion compounds, which contain a dense lattice of localized magnetic moments interacting with a sea of conduction electrons to form a three-dimensional Kondo lattice¹. It had remained an unanswered question whether superconductivity would persist on effectively reducing the dimensionality of these materials from three to two. Here we report on the observation of superconductivity in such an ultimately strongly correlated system of heavy electrons confined within a two-dimensional square lattice of Ce atoms (two-dimensional Kondo lattice), which was realized by fabricating epitaxial superlattices^{2,3} built of alternating layers of heavy-fermion CeCoIn₅ (ref. 4) and conventional metal YbCoIn₅. The field-temperature phase diagram of the superlattices exhibits highly unusual behaviours, including a striking enhancement of the upper critical field relative to the transition temperature. This implies that the force holding together the superconducting electron pairs takes on an extremely strong-coupled nature as a result of two-dimensionalization.

The layered heavy-fermion compound CeCoIn₅ has the highest superconducting transition temperature ($T_c = 2.3$ K) among rare-earth-based heavy-fermion materials⁴. Its electronic properties are characterized by an anomalously large value of the linear contribution to the specific heat (Sommerfeld coefficient $\gamma \sim 1$ J mol⁻¹ K⁻²), indicating heavy effective masses of the 4f electrons, which contribute greatly to the Fermi surface. The tetragonal CeCoIn₅ crystal structure is built from alternating layers of CeIn₃ and CoIn₂ stacked along the [001] direction. This compound possesses several key features for understanding the unconventional superconductivity in strongly correlated systems^{5–7}. The superconductivity with $d_{x^2-y^2}$ pairing symmetry^{8–11} which occurs in the proximity of a magnetic instability is a manifestation of magnetic-fluctuation-mediated superconductivity^{5–7,12}. A very strong coupling superconductivity, where electron pairs are bound together by strong forces, is revealed by a large specific-heat jump⁴ at T_c , representing a steep drop of the entropy below T_c , and a large superconducting energy gap Δ needed to break the electron pair⁹. Despite its layered structure, the largely corrugated Fermi surface¹³, three-dimensional-like antiferromagnetic fluctuations in the normal state¹⁴ and small anisotropy of upper critical field¹⁵ all indicate that the electronic, magnetic and superconducting properties are essentially three dimensional rather than two dimensional. Therefore, it is still

unclear to what extent the three-dimensional nature is essential for the superconductivity of CeCoIn₅.

Recently, a state-of-the-art technique has been developed to reduce the dimensionality of the heavy electrons in a controllable fashion by layer-by-layer epitaxial growth of Ce-based materials. Previously, a series of antiferromagnetic superlattices CeIn₃/LaIn₃ were successfully grown², but it remains open whether heavy electrons in a single Ce layer forming a two-dimensional Kondo lattice can be superconducting. Here we fabricate multilayers of CeCoIn₅ sandwiched by non-magnetic and non-superconducting metal YbCoIn₅ (the Yb ion is divalent in closed-shell 4f(14) configuration) forming an ($n:m$) c -axis oriented superlattice structure, where n and m are the number of layers of CeCoIn₅ and YbCoIn₅ in a unit cell, respectively. Small lattice mismatch between CeCoIn₅ and YbCoIn₅ offers a possibility of providing an ideal heterostructure. The high-resolution cross-sectional transmission electron microscope (TEM) results (Fig. 1a–c), and distinct lateral satellite peaks in the X-ray diffraction pattern (Fig. 1d), demonstrate the continuous and evenly spaced CeCoIn₅ layers, with no discernible interdiffusion even for $n = 1$ cases (see Supplementary Fig. S1 for quantitative analysis of interdiffusion by X-ray). The epitaxial growth of each layer with atomic flatness is shown by the streak patterns of the reflection high-energy electron diffraction (Fig. 1e) and atomic force microscopy images (Fig. 1f).

We investigate the transport properties for the ($n:5$) superlattices by varying n . The resistivity $\rho(T)$ of the CeCoIn₅ thin film (Fig. 2a) reproduces well that of bulk single crystals⁴. Below ~ 100 K, $\rho(T)$ increases on cooling owing to the Kondo scattering, decreases after showing a peak at around the coherence temperature $T_{\text{coh}} \sim 30$ K and drops to zero at the superconducting transition. The hump structure of $\rho(T)$ at $\sim T_{\text{coh}}$ is also observed in the superlattices but becomes less pronounced with decreasing n . The superconducting transition to zero resistance is observed in the superlattices for $n \geq 3$ (Fig. 2b). For $n = 2$ and 1, $\rho(T)$ decreases below ~ 1 K, but it does not reach zero. However, when the magnetic field is applied perpendicular to the layers for $n = 1$, $\rho(T)$ increases and recovers to the value extrapolated above 1 K at 5 T, whereas the reduction of $\rho(T)$ below 1 K remains in the parallel field of 6 T (Fig. 2c). The observed large and anisotropic field response of $\rho(T)$ is typical for layered superconductors, demonstrating superconductivity even in the $n = 1$ superlattice with a two-dimensional square lattice of Ce atoms. The critical temperature T_c determined by the resistive transition gradually decreases with decreasing n (Fig. 2d). The residual resistivity ρ_0 of the superlattices is of the same order as ρ_0 of single-crystalline film (Fig. 2d) and is much lower than ρ_0 of Yb-substituted CeCoIn₅ single crystals¹⁶.

¹Department of Physics, Kyoto University, Kyoto 606-8502, Japan, ²Research Center for Low Temperature and Materials Sciences, Kyoto University, Kyoto 606-8501, Japan, ³Department of Physics, Nagoya University, Furo-cho, Nagoya 464-8602, Japan. [†]Present address: Department of Physics and Electronics, Osaka Prefecture University, Sakai, Osaka 599-8531, Japan. *e-mail: matsuda@scphys.kyoto-u.ac.jp.

Figure 1 | Epitaxial superlattices ($n:5$) of $\text{CeCoIn}_5(n)/\text{YbCoIn}_5(5)$. **a**, High-resolution cross-sectional TEM image of $n=1$ superlattice. The bright dot arrays are identified as the Ce layers and the less bright dots are Yb atoms, which is consistent with the designed superlattice structure in the left panel. **b**, The intensity integrated over the horizontal width of the image plotted against vertical position indicates a clear difference between the Ce and Yb layers, showing no discernible atomic interdiffusion between the neighbouring Ce and Yb layers. **a** (upper right inset), **c**, The fast Fourier transform (FFT) of the TEM image, which shows clear superspots along the [001] direction (**c**). **d**, Cu K α 1 X-ray diffraction patterns for $n=1, 2, 3, 4, 5$ and 7 superlattices with a typical total thickness of 300 nm show first (red arrows) and second (blue arrow) satellite peaks. The positions of the satellite peaks and their asymmetric heights can be reproduced by the step-model simulations (green lines) ignoring interface and layer-thickness fluctuations²⁹ (see also Supplementary Information for the detailed analysis of the satellite peak intensity). **e**, Streak patterns of the reflection high-energy electron diffraction image during the growth. **f**, Typical atomic force microscopy image for $n=1$ superlattice. The typical surface roughness is within 0.8 nm , comparable to one unit-cell thickness (uct) along the c axis of CeCoIn_5 .

An important question is whether the superconducting electrons in the superlattices are heavy and if so what their dimensionality is. As shown in Figs 2c, 3a and 4a, the parallel and perpendicular (to the layers) upper critical fields, $H_{c2\parallel}$ and $H_{c2\perp}$, of the superlattices at low temperature are significantly larger than those in conventional superconductors with similar T_c . The magnetic field destroys the superconductivity in two distinct ways, the orbital pair-breaking effect (vortex formation) and the Pauli paramagnetic effect, a breaking up of pairs by spin polarization. The zero-temperature value of the orbital upper critical field in perpendicular field $H_{c2\perp}^{\text{orb}}(0)$ reflects the effective electron mass in the plane m_{ab}^* , $H_{c2\perp}^{\text{orb}}(0) \propto m_{ab}^{*2}$, and is estimated to be 6, 11 and 12 T for $n=3, 5$ and 7 superlattices from the initial slope of $H_{c2\perp}$ at T_c by the relation $H_{c2\perp}^{\text{orb}}(0) = 0.69T_c(-dH_{c2\perp}/dT)_{T_c}$. These magnitudes are comparable with or of the same order as $H_{c2\perp}^{\text{orb}}(0)$ ($=14\text{ T}$) in bulk single crystal, providing strong evidence for the superconducting ‘heavy’ electrons in the superlattices. We stress that even a slight deviation of the f -electron number from unity leads to a serious reduction of the heavy-electron mass¹⁷. Moreover, the band-structure calculation for the $n=1$ superlattice shows that the number of f -electrons in each CeCoIn_5 layer is very close to

unity (Supplementary Information). These facts indicate that the f -electron wavefunctions are essentially confined to Ce layers. The magnetic two-dimensionality is shown by estimating the strength of the Ruderman–Kittel–Kasuya–Yosida interaction, an intersite magnetic exchange interaction between the localized f moments, which decays with the distance as $1/r^3$. This interaction between the Ce ions in different layers of ($n:5$) superlattices reduces to $(4.568/44.716)^3 \sim 1 \times 10^{-3}$ of that between neighbouring Ce ions within the same layer (see Supplementary Table S1).

The superconducting order parameters in the CeCoIn_5 layers of the superlattices are expected to be coupled weakly by the proximity effect through the normal-metal YbCoIn_5 layers. The proximity-induced superconductivity in YbCoIn_5 layers is expected to be very fragile and destroyed when a weak field is applied¹⁸. If the thickness of the CeCoIn_5 layer is comparable to the perpendicular coherence length $\xi_{\perp}$ ($\sim 2.1\text{ nm}$ for CeCoIn_5), and the separation of superconducting layers ($\sim 3.7\text{ nm}$ for ($n:5$) superlattices) exceeds $\xi_{\perp}$, each CeCoIn_5 layer acts as a two-dimensional superconductor¹⁹. This two-dimensional feature is revealed by the diverging $H_{c2\parallel}/H_{c2\perp}$ of $n=3, 5$ and 7 superlattices on approaching T_c (Fig. 3b), in sharp contrast to bulk CeCoIn_5 (ref. 20), and by a cusplike angular

Figure 2 | Superconductivity in superlattices ($n:5$) of $\text{CeCoIn}_5(n)/\text{YbCoIn}_5(5)$. **a**, Temperature dependence of electrical resistivity $\rho(T)$ for $n = 1, 2, 3, 5, 7$ and 9 , compared with those of 300-nm-thick CeCoIn_5 and YbCoIn_5 single-crystalline thin films. **b**, Low-temperature part of the same data as in **a**. **c**, $\rho(T)$ for $n = 1$ at low temperatures in magnetic field parallel (dotted line) and perpendicular (solid lines) to the ab plane. **d**, Superconducting transition temperature as a function of n (left axis). The circles are the mid-points of the resistive transition and the bars indicate the onset and zero-resistivity temperatures. The residual resistivity ρ_0 as a function of n is also shown (right axis).

dependence $H_{c2}(\theta)$ near parallel field for the $n = 3$ superlattice (Fig. 3c), which is qualitatively different from that expected in the three-dimensional anisotropic-mass model but is well fitted by the model in the two-dimensional limit¹⁹. On the basis of the above two-dimensional features observed in all electronic, magnetic and superconducting properties, we conclude that the observed heavy-electron superconductivity is mediated most probably by two-dimensional electron-correlation effects.

A fascinating issue is how the two-dimensionalization changes the pairing nature. The fact that $H_{c2\perp}^{\text{orb}}(0)$ estimated from the initial slope of $H_{c2\perp}(T)$ at T_c well exceeds the actual value at low temperatures indicates the predominant Pauli paramagnetic pair-breaking effect even in perpendicular field. Therefore, $H_{c2}(\theta)$ at low temperatures is dominated by the Pauli effect in any field direction. This is reinforced by the result that the cusplike behaviour of $H_{c2}(\theta)$ becomes less pronounced well below T_c (Fig. 3c), which is the opposite trend to the $H_{c2}(\theta)$ behaviour of conventional multilayer systems²¹. In fact, the Pauli-limited upper critical field H_{c2}^{Pauli} given by

$$H_{c2}^{\text{Pauli}} = \sqrt{2}\Delta/g\mu_B \quad (1)$$

where g is the gyromagnetic ratio determined by the Ce crystalline electric field levels, varies smoothly with field direction, consistent with $H_{c2}(\theta)$ of the present superlattices at low temperatures. Figure 4a shows the H - T phase diagram of the superlattices. What is remarkable is that, with decreasing n , T_c decreases rapidly from the bulk value, whereas H_{c2} does not exhibit such a reduction, for both field directions. In fact, at low temperatures, $H_{c2\parallel}$ of $n = 5$ and 7 is even larger than that of the bulk. This robustness of H_{c2} (and hence of Δ) against n reduction indicates that the superconducting pairing interaction is hardly affected by two-dimensionalization. This provides strong evidence that the superconductivity in bulk CeCoIn_5 is mainly mediated by two-dimensional spin-fluctuations as in the superlattices with two-dimensional magnetic structure, although the neutron spin resonance mode in bulk CeCoIn_5 is observed in the three-dimensional (π, π, π) position below T_c (ref. 10).

In sharp contrast to H_{c2} , the thickness reduction dramatically enhances H_{c2}/T_c from the bulk value (Fig. 4b). A comparison to the pressure-dependence results²², which represent the increased three-dimensionality, reveals a T_c dome and a general trend of enhanced H_{c2}/T_c with reduced dimensionality. Through the relation of equation (1), this trend immediately implies a

Figure 3 | Superconducting anisotropy in superlattices (n:5) of CeCoIn₅(n)/YbCoIn₅(5). **a**, Magnetic-field dependence of resistivity for $n = 3$ superlattice at several field angles from $\theta = 0$ ($H \perp ab$) to 90° ($H \parallel a$) (10° step) at $T = 0.15$ K. **b**, Anisotropy of H_{c2} , $H_{c2||}/H_{c2\perp}$, as a function of reduced temperature T/T_c for $n = 3, 5$ and 7 superlattices and for the bulk CeCoIn₅. **c**, Upper critical field $H_{c2}(\theta)$ at several temperatures as a function of field angle θ . H_{c2} is determined by the mid-point of the transition except for 1.0 K, where an 80% resistivity criterion has been used. The solid blue and red lines are the fits to the three-dimensional anisotropic mass model represented as $H_{c2}(\theta) = H_{c2||}/(\sin^2\theta + \gamma^2 \cos^2\theta)^{1/2}$ with $\gamma = H_{c2||}/H_{c2\perp}$ and Tinkham's formula $|H_{c2}(\theta)\cos\theta/H_{c2\perp}| + |H_{c2}(\theta)\sin\theta/H_{c2||}|^2 = 1$ for a two-dimensional superconductor, respectively¹⁹.

Figure 4 | Superconducting phase diagrams of superlattices (n:5) of CeCoIn₅(n)/YbCoIn₅(5). **a**, Magnetic-field versus temperature phase diagram of $n = 3, 5$ and 7 superlattices in magnetic field parallel (open symbols) and perpendicular (closed symbols) to the ab plane, compared to the bulk CeCoIn₅ data. The mid-point of the transition in $\rho(T)$ (circles) and $\rho(H)$ (squares) has been used to evaluate $H_{c2}(T)$. **b**, Superconducting transition temperature, T_c (open triangles), and reduced critical fields H_{c2}/T_c in parallel (filled blue circles) and perpendicular (filled red squares) fields as a function of dimensionality parameter $1/n$ (right panel). The pressure dependence of these quantities²⁸ is also shown for comparison (left panel). Note the different scales for parallel (blue) and perpendicular (red) fields.

remarkable enhancement of Δ/T_c by two-dimensionalization. (As the anisotropy of H_{c2} at low temperatures does not depend on n , as shown in Fig. 3b, we assume that the g -value of the superlattices deviates little from the bulk value.) We note that the enhanced impurity scatterings cannot be primary origins of the T_c reduction, as these effects do not significantly enhance the Δ/T_c ratio in d -wave superconductors²³. This is supported by no discernible interdiffusion by TEM results and a ρ_0 of superlattices of the same order as ρ_0 of the bulk CeCoIn₅. The reduction of T_c may be caused by the reduction of density of states in the superlattices, but this scenario is also unlikely because the density-of-states reduction usually reduces the pairing interaction, which results in the reduction of Δ .

Using the reported value of $2\Delta/k_B T_c = 6$ in the bulk single crystal⁹, $2\Delta/k_B T_c$ for the $n = 5$ superlattice is estimated to exceed 10, which is significantly enhanced from the weak-coupling

Bardeen–Cooper–Schrieffer value of $2\Delta/k_B T_c = 3.54$. It has been suggested theoretically that d -wave pairing mediated by antiferromagnetic fluctuations in two dimensions can be much stronger than that in three dimensions^{24–26}. The striking enhancement of $2\Delta/k_B T_c$ associated with the reduction of T_c , a situation resembling that of underdoped high- T_c cuprates, implies that there seem to be further mechanisms, such as two-dimensional phase fluctuations²⁷ and a strong pair-breaking effect due to inelastic scattering²⁸. Further investigation, particularly probing electronic and magnetic excitations in the normal and superconducting states, is likely to bridge the physics of highly unusual correlated electrons in the two-dimensional Kondo lattice and in the two-dimensional CuO₂ planes of cuprates. The fabrication in a wide variety of nanometric superlattices also opens up a possibility of nanomanipulation of heavy electrons, providing a unique opportunity to produce a new superconducting system and its interface.

Methods summary

CeCoIn₅/YbCoIn₅ superlattices are grown by the molecular beam epitaxy technique. The pressure of the molecular beam epitaxy chamber was kept at 10^{-7} Pa during the deposition. The (001) surface of MgF₂ with rutile structure ($a = 0.462$ nm, $c = 0.305$ nm) was used as a substrate. The substrate temperature was kept at 550 °C during the deposition. Atomic layer-by-layer molecular beam epitaxy provides for digital control of layer thickness, which we measure by counting the number of unit cells². Each metal element was evaporated from individually controlled Knudsen cells. 15-unit-cell-thick (uct) YbCoIn₅ was grown after CeIn₃ (28 nm) was grown on the (001) surface of substrate MgF₂ as a buffer layer. Then n -uct CeCoIn₅ layers and m -uct YbCoIn₅ (typically $m = 5$) were grown alternately, typically repeated 30–60 times. The deposition rate was monitored by a quartz oscillating monitor and the typical deposition rate was $0.01\text{--}0.02$ nm s⁻¹.

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Author contributions

The first two authors contributed equally to this work. Y. Matsuda and T.S. planned the experiments, analysed the data and wrote the paper. Y. Mizukami, H.S. and T.T. fabricated the superlattices. M.S., S.Y., D.W. and M.Y. contributed to the measurements. H.I. carried out the band-structure calculation and H.K. analysed the data. All authors critically reviewed the paper.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturephysics. Reprints and permissions information is available online at <http://www.nature.com/reprints>. Correspondence and requests for materials should be addressed to Y. Matsuda