Artificial heavy fermions in a van der Waals heterostructure

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Heavy-fermion systems represent one of the paradigmatic strongly correlated states of matter1–3. They have been used as a platform for investigating exotic behaviour ranging from quantum criticality and non-Fermi liquid behaviour to unconventional topological superconductivity4–12. The heavy-fermion phenomenon arises from the exchange interaction between localized magnetic moments and conduction electrons leading to Kondo lattice physics, and represents one of the long-standing open problems in quantum materials3. In a Kondo lattice, the exchange interaction gives rise to a band with heavy effective mass. This intriguing phenomenology has so far been realized only in compounds containing rare-earth elements with 4f or 5f electrons1,4,13,14. Here we realize a designer van der Waals heterostructure where artificial heavy fermions emerge from the Kondo coupling between a lattice of localized magnetic moments and itinerant electrons in a 1T/1H-TaS2 heterostructure. We study the heterostructure using scanning tunnelling microscopy and spectroscopy and show that depending on the stacking order of the monolayers, we can reveal either the localized magnetic moments and the associated Kondo effect, or the conduction electrons with a heavy-fermion hybridization gap. Our experiments realize an ultimately tunable platform for future experiments probing enhanced many-body correlations, dimensional tuning of quantum criticality and unconventional superconductivity in two-dimensional artificial heavy-fermion systems15–17.

In recent years, van der Waals (vdW) heterostructures have become the de facto platform to engineer artificial electronic phenomena18–22. They have only weak interactions between the different layers, which allows each layer to retain its intrinsic properties. The interfaces between the layers are essentially free of contamination and defects and well defined down to the atomic level. These factors make it possible to combine materials with seemingly competing electronic orders.

For the realization of artificial heavy fermions, we need to couple a material hosting local moments with an itinerant electron bath13,14. In particular, some monolayer transition metal dichalcogenides show metallic behaviour23,24, while others are known to realize a correlated, charge density wave (CDW)-driven state hosting local magnetic moments and potentially a quantum spin liquid state25–29.

In this Article, we use molecular beam epitaxy (MBE) to grow bilayer 1T-TaS2/1H-TaS2 heterostructures (Fig. 1a) on highly oriented pyrolytic graphite (HOPG) and characterize them using low-temperature scanning tunnelling microscopy (STM) and spectroscopy (STS). We demonstrate that the localized moments in 1T-TaS2, Kondo couple with the itinerant electrons of the Ta 4d-band in 1H-TaS2 and give rise to a heavy-fermion band structure. The use of vdW materials in general allows unprecedented external control—using, for example, light and electrostatic gating—that cannot be reached in the traditional heavy-fermion systems. These results mark the latest advance in realizing strongly correlated states in vdW heterostructures and open up a pathway towards even more exotic phases such as artificial heavy-fermion superconductivity in the future.

Heavy fermions in a vdW heterostructure

1T-TaS2 exhibits a CDW state that results in a large unit cell hosting a single localized moment25–27,30. This local moment can be exchange-coupled with the Ta 4d-band of the underlying 1H-TaS2 layer forming a Kondo ground state. When these Kondo impurities form a lattice (defined by the CDW unit cell), the strongly localized half-filled limit, the Kondo sites create a nearly flat pseudo-fermion band at the chemical potential (Fig. 1b, details of the theory can be found in Supplementary Section 1)31. A finite coupling between Kondo sites would give rise to a small dispersion of the resonant pseudo-fermion Kondo modes. In our case, coupling in the Kondo lattice is sufficiently weak and does not modify the picture qualitatively (Supplementary Section 1). The hybridization between the resonant modes (Fig. 1b) and the conduction electrons (Fig. 1c) leads to an avoided crossing in the hybridized electronic structure (Fig. 1d), in an exact analogue of a natural heavy-fermion compound. Interestingly, the physics of this heterostructure is potentially simpler than those of typical heavy-fermion compounds with hidden order.

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The previous treatment is valid in the Kondo insulating regime of a heavy-fermion system, the regime observed experimentally, and would not emerge for a two-dimensional Kondo lattice on a surface of a three-dimensional metal. In the present case, we would expect to observe the Kondo resonances when probing the local density of states (LDOS) of the IT-TaS₂ layer. On the other hand, the LDOS on the IH-TaS₂ side should exhibit a signature of a heavy-fermion hybridization gap at the Fermi level (Fig. 1e). We will show in the following that we can realize precisely this response in our experiments.

**STM and STS results**

We have tuned our MBE growth to produce both IT and IH monolayers as well as bilayer islands within all possible combinations: IH and IT bilayers, and IH/IT and IT/IH heterobilayers. Details of the sample growth and STM experiments are given in the Methods. Figure 2 shows results of STM and STS experiments for the IT- and IH-TaS₂ monolayers and for the IT/IT-TaS₂ vertical heterostructure. The tunnelling spectrum of the monolayers matches with the expected electronic structure of the IT- and IH-TaS₂ (Fig. 2b)14-17. IT-TaS₂ (green curve) is a Mott insulator with the upper Hubbard band at V = 0.2 V, the lower Hubbard band at V = -0.3 V and a charge gap of ~0.3 V, while IH-TaS₂ (yellow curve) is a metal with finite LDOS at the Fermi level. The LDOS of the IT-TaS₂ on top of IH-TaS₂ (red curve) has a strong peak at the Fermi level, which appears periodically on every site of the star of David CDW (Extended Data Figs. 1 and 2). This is consistent with the Kondo lattice effect (Supplementary Section 1) and with the previous experiments on an analogous IT/IH-TaSe₂ heterostructure20. The exact value of the Kondo exchange coupling depends on fine chemical details, and therefore can be different between the IT/IT-TaS₂ and IT/IH-TaSe₂ heterostructures. For IT-TaS₂ directly on top of HOPG, the Kondo temperature would be much smaller owing to the reduced density of states (DOS) of HOPG.

To confirm the Kondo nature of the peak in the tunnelling spectrum at zero bias and to rule out a possible explanation of a narrow band reported in a related bulk compound 4Hb-TaS₂ (ref. 32), we have measured its evolution with the temperature and applied magnetic field33. The peak at zero bias is getting gradually wider and lower in intensity with temperature (Fig. 2c), which cannot be accounted for by the effect of thermal broadening. The tunnelling spectra at different temperatures were also fitted to a Fano lineshape (Methods and Extended Data Figs. 3 and 4), resulting in a temperature dependence of the Kondo resonance width Γ (Fig. 2d, black circles). This width follows a well-known temperature dependence and a fit (red line) yields a Kondo temperature of T_K = 18 K. Applying a magnetic field perpendicular to the sample surface broadens the zero-bias peak at lower magnetic fields and eventually splits it at high magnetic fields (Fig. 2e), a behaviour expected for the Zeeman splitting of a Kondo resonance33. There are also additional spectroscopic features around the Fermi level. Our theoretical model (Supplementary Section 1) does not account for such features, which could be a sign of richer physics.

**STS of this heterostructure**

STS of this heterostructure probes the LDOS of the top layer. As shown above, the IT-TaS₂ on top of IT-TaS₂ develops a Kondo resonance at the Fermi level. When the layers in the heterostructure are inverted (that is, with IH-TaS₂ on top of IT-TaS₂), we observe a ~7 mV gap around the Fermi level (Fig. 3). Figure 3b shows a large-bias tunnelling spectrum of the IH/IT-TaS₂ with a dip at zero bias. The tunnelling conductance at zero bias is finite because of the larger lock-in bias modulation used for large-bias spectroscopy. However, by performing higher-resolution tunnelling spectroscopy, we find that the tunnelling conductance at zero bias is zero at the lowest temperature (Fig. 3c). This is not the case for IH-TaS₂ on HOPG or for bilayer IH-TaS₂, even though both of these develop a dip around the Fermi level (Extended Data Fig. 5). This dip is a common feature of monolayer metallic transition metal dichalcogenides32,34, and it could be explained as a CDW gap or as a dip in the DOS of two-dimensional metals caused by a Fermi liquid renormalization35. More importantly, it is not expected to be a heavy-fermion hybridization gap, as no magnetic moments are present in IH-TaS₂ on HOPG.

As can be seen in Fig. 3c, the gap is continuously filling up with increasing temperature, which is consistent with the findings of previous experiments measuring the temperature dependence of a heavy-fermion hybridization gap36,37. Once again, this filling of the gap cannot be accounted for only by the thermal broadening of the tunnelling spectrum. This can be seen in Fig. 3d, where we take the lowest-temperature tunnelling spectrum (blue curve) and simulate
the same spectrum with a thermal broadening corresponding to $T = 25$ K (purple curve). This thermally broadened spectrum is notably different from the measured one at 25 K (green curve), which is due to the combined effect of thermal broadening and the filling of the ~7 mV gap. The difference between simulated thermal broadening of the lowest-temperature spectrum and the actually measured temperature dependence can also be seen in the temperature dependence of a zero-bias tunnelling conductance. This is shown in Fig. 3e, where we look at the temperature dependence of the depth of the zero-bias dip. In the experimental data, the features wash out with increasing temperature much more rapidly than in the case of simulated thermal broadening.

We have verified that the presented phenomenon occurs on all of our 1H/1T-TaS$_2$ heterostructures (see Extended Data Fig. 6 for examples). In addition, as our system has low chemical disorder, the heavy-fermion response is expected to be more homogeneous than in natural heavy-fermion systems. We have verified this by spatially dependent spectroscopy experiments (Extended Data Figs. 7 and 8).

**Origin of the spectral gap**

Besides the heavy-fermion hybridization gap, there are also other possible explanations for the gap in the tunnelling spectrum of a 1H/1T-TaS$_2$ heterostructure that we need to consider. The first possibility is a Coulomb gap, where the repulsive Coulomb interactions of quasiparticles confined in an island would lead to a gap around the Fermi level. The repulsive Coulomb interactions should increase with decreasing island size; however, we do not observe a correlation between the gap width and island size in our experiments (Extended Data Fig. 6). Another possibility would be a CDW gap. Even though the 1H-TaS$_2$ exhibits a 3 x 3 CDW, in the case of 1H-TaS$_2$ on top of 1T-TaS$_2$, we observe no CDW modulation in STM images with atomic resolution (Extended Data Fig. 9). Finally, the observed gap could also in principle be of a superconducting origin. 1H-TaS$_2$ is already a superconductor, and by creating a heterostructure one could increase the electron–phonon interaction, or an unconventional magnon-mediated superconductivity could arise in this heterostructure. In addition, bulk 4Hb-TaS$_2$ that combines alternating layers of 1T- and 1H-TaS$_2$ has been shown to be a chiral superconductor with out-of-plane critical field below 1 T (ref. 42). In general, two-dimensional superconductivity should be strongly influenced by applying out-of-plane magnetic field, and we see no notable changes to the observed gap up to 10 T (Extended Data Fig. 10). This is also consistent with the reported magnetic field dependence of a hybridization gap of similar size, where no major changes in the overall shape of the spectra were observed (see also discussion in Supplementary Section 2). Considering all of these findings, we conclude that there are strong arguments against other possible origins of the spectral gap and that the gap is probably a heavy-fermion hybridization gap (see also discussion in Supplementary Section 3).
offset for clarity. d, Tunnelling spectra of the heavy-fermion gap measured at 300 mK (blue) and 25 K (green). The purple curve corresponds to a tunnelling spectrum measured at 300 mK with a thermal broadening of 25 K.

e, Temperature evolution of the zero-bias dip depth (black circles) with a guide for the eye (black line). The red line is a simulated temperature evolution acquired by applying thermal broadening to the 300 mK spectrum.

Conclusions

We have realized an artificial vdW heterostructure hosting heavy-fermion physics. Our STM and STS experiments demonstrate the presence of the fundamental ingredients for this, the Kondo screening of local moments and the development of a heavy-fermion gap that emerges in our heterostructures. These results open a pathway towards a whole new family of two-dimensional materials showing the emergence of a strongly correlated state of matter that previously required rare-earth elements. The use of vdW materials allows for an unprecedented level of control over the system parameters that is not available in rare-earth compounds, such as changing the twist angle or tuning the chemical potential by the gating of a sample. These samples are also easy to synthesize using MBE growth or exfoliation, making them more accessible compared to the rare-earth compounds. Growth of such high-quality heterostructures with larger-scale uniformity could lead to further insights into the band structure of this artificial heavy-fermion system using quasiparticle interference. Ultimately, these systems will enable the study of heavy-fermion superconductivity, quantum criticality and non-Fermi liquid phases tunable by gating and twist engineering.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-021-04021-0.
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Methods

Sample preparation
TaS₂ was grown by MBE on HOPG under ultrahigh-vacuum conditions (base pressure $-1 \times 10^{-10}$ mbar). HOPG crystal was cleaved and subsequently out-gassed at $-800\,\text{°C}$. High-purity Ta and S were evaporated from an electron-beam evaporator and a Knudsen cell, respectively. Before growth, the flux of Ta was calibrated on an Au(111) at $-1$ monolayer per hour. The ratio of 1T- to 1H-TaS₂ can be controlled via the substrate temperature and the overall coverage. Before growth, the HOPG substrate temperature was stabilized at $-680\,\text{°C}$. The sample was grown in a S pressure of $-5 \times 10^{-8}$ mbar and the growth duration was 25 min.

STM measurements
After preparation of the sample, it was inserted into the low-temperature STM (Unisoku USM-1300) housed in the same ultrahigh-vacuum system, and all subsequent experiments were performed at $T = 300$ mK. STM images were obtained in the constant-current mode. $dI/dV$ spectra were recorded by standard lock-in detection while sweeping the sample bias in an open feedback loop configuration, with a peak-to-peak bias modulation of 0.4–1 mV at a frequency of 731 Hz; a modulation of 5 mV was used for $dI/dV$ spectra at larger biases.

Kondo resonance analysis
To determine the intrinsic Kondo resonance width, the tunnelling spectra shown in Extended Data Fig. 3 (black lines) were fitted to a thermally convolved Fano lineshape (red lines):

$$\frac{df}{dV}(eV) \propto \int_{-\infty}^{\infty} (\xi + q)^2 \left(1 + \xi^2\right)(-\gamma (E - eV, T))dE, \quad \xi = \frac{2(E - E_0)}{\Gamma}.$$

Here, $q$ is the Fano parameter, $\Gamma$ is the intrinsic Kondo resonance width, $E_0$ is the energy of a resonance and $f(E, T) = 1/(e^{E/kT} + 1)$ is the Fermi distribution function. We point out that the extraction of the intrinsic resonance width in our case is phenomenological, which is also reflected in a larger uncertainty at higher temperatures. The fitting procedure results in obtaining the temperature dependence of the intrinsic Kondo resonance width shown in Fig. 2d (black circles), which was consequently fitted to the formula

$$\Gamma(T) = \sqrt{(\pi k_B T)^2 + 2(k_B T_K)^2}.$$

The fit (red line) yields a Kondo temperature $T_K = 18$ K. This functional form of $\Gamma(T)$ is in principle not formally valid in the whole temperature regime of a heavy-fermion system, and it is expected to break down in the temperature regime separating the Fermi liquid regime and the non-Fermi liquid regime. A potential explanation is that the Fermi liquid temperature of our system resides above the temperatures measured and shown in the fit.

To show that this temperature dependence of the peak at zero bias is not just an effect of thermal broadening, we have performed a thermal broadening simulation:

$$\frac{df}{dV}(eV) \propto \int_{-\infty}^{\infty} \rho(E)(-\gamma (E - eV, T))dE,$$

where $\rho(E)$ is the DOS. As the thermal broadening at the temperature of 300 mK is negligible, we took the 300 mK tunnelling spectrum as the DOS and used the Fermi distribution function at 18 K. The result can be seen in Extended Data Fig. 4, where the measured and simulated spectrum at 18 K clearly do not match, which means that this is not just an effect of thermal broadening. The same procedure was applied in Fig. 3d.

Data availability
All of the data supporting the findings are available from the corresponding authors upon request.

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Acknowledgements
This research made use of the Aalto Nanomicroscopy Center (Aalto NMC) facilities and was supported by the European Research Council (ERC-2017-AdG no. 788185 “Artificial Designer Materials”), the Academy of Finland (Academy professor funding nos. 318995 and 320555, Academy postdoctoral researcher no. 309975, Academy research fellow nos. 331342 and 336243) and the Jane and Aatos Erkko Foundation. We acknowledge the computational resources provided by the Aalto Science-IT project.

Author contributions
V.V., J.L.L., S.K. and P.L. conceived the experiment, V.V., M.A., S.C.G. and S.K. carried out the sample growth and the low-temperature STM experiments. V.V. analysed the STM data. G.C. and J.L.L. developed the theoretical model. V.V., J.L.L. and P.L. wrote the manuscript with input from all co-authors.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41586-021-04021-0.

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Peer review information Nature thanks Milan Allan, Stefan Kirchner and the other, anonymous, reviewer(s) for their contribution to the peer review of this work. Peer reviewer reports are available.

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Extended Data Fig. 1 | Grid spectroscopy measurement of a 1T-TaS$_2$/1H-TaS$_2$ heterostructure. a, STM image of a 1T/1H-TaS$_2$ heterostructure ($V = 200$ mV and $I = 10$ pA). b, Tunneling spectra across the red line shown in (a), starting from top left. We observe a Kondo peak located on each of the CDW centers. c, $dI/dV$ map at $V = 0$ mV measured on the area shown in (a) showing the Kondo lattice. Due to a Kondo peak, there is a higher $dI/dV$ intensity on each of the CDW centers. Middle and bottom rows: $dI/dV$ maps at given bias voltages, all measured on the area shown in (a) and using the same colour scale as in (c). Each of the $dI/dV$ curves was normalized by dividing it by its mean value. Single tunneling spectra on different CDW centers and on a different area are shown in Extended Data Fig. 2.
Extended Data Fig. 2 | Single tunneling spectra in the centre of different CDW unit cells. Left: STM image of a 1T-TaS$_2$/1H-TaS$_2$ heterostructure ($V = 200$ mV and $I = 20$ pA). Right: tunneling spectra measured at locations highlighted by the same-coloured dots on the STM image, spectra are vertically offset for clarity.
Extended Data Fig. 3 | Fitting of the Kondo resonances. Left: Temperature dependence of a Kondo resonance in tunneling spectroscopy (black lines), and their respective fits to the Fano lineshape (red lines). The spectra are vertically offset for clarity. Right: Table of the fit parameters.

| Temperature (K) | Γ (meV) | q  | E₀ (meV) |
|-----------------|---------|----|----------|
| 0.3             | 1.58    | >100 | 0.19    |
| 2               | 1.64    | >100 | 0.00    |
| 4               | 1.94    | >100 | 0.00    |
| 6               | 2.09    | >100 | 0.00    |
| 8               | 2.79    | >100 | −0.49   |
| 10              | 3.04    | >100 | −0.50   |
| 12              | 3.40    | >100 | −1.00   |
| 14              | 4.01    | >100 | −1.00   |
| 16              | 4.50    | >100 | −1.30   |
| 18              | 5.00    | >100 | −1.99   |
Extended Data Fig. 4 | Thermal broadening of the Kondo resonance.
Tunneling spectroscopy of a Kondo resonance at 300 mK (blue), 18 K (green), and a simulated tunneling spectrum at 18 K (red), where the 300 mK spectrum was taken as the density of states.
Extended Data Fig. 5 | Tunneling spectra of 1H-TaS2 on different substrates. Tunneling spectroscopy of 1H-TaS2 on HOPG (purple line), bilayer 1H-TaS2 (blue line) and 1H-TaS2 on monolayer 1T-TaS2 (green line). The spectra are measured on positions highlighted by the same-coloured dots in the STM image on the left. The spectra exhibit a dip around the Fermi level, but while 1H-TaS2 on HOPG (purple line) and bilayer 1H-TaS2 (blue line) have finite zero-bias conductance, only the spectroscopy of 1H/1T-TaS2 (green line) exhibits a heavy-fermion gap with zero conductance at zero bias.
Extended Data Fig. 6 | Examples of different 1H/1T-TaS₂ heterostructures. STM images (left) and corresponding tunneling spectra measured on top of a heterostructure (right). Purple dots highlight the positions, where the tunneling spectra were taken. All the heterostructures exhibit a heavy-fermion gap with zero conductance at zero bias and approximately the same gap width, regardless of the island size.
Extended Data Fig. 7 | Tunneling spectroscopy across middle of the 1H/1T-TaS$_2$ island. a, STM image of a 1H/1T-TaS$_2$ vertical heterostructure ($V = 900$ mV and $I = 20$ pA). b, Tunneling spectra across the red line shown in (a). c, Average tunneling spectrum from the spectra across line in (b).
Extended Data Fig. 8 | Spatial dependence of the heavy-fermion gap. a, STM image of a 1H-TaS2/1T-TaS2 heterostructure (V = 50 mV and I = 500 pA). Larger scale topographic image is shown in Extended Data Fig. 9e. b, Tunneling spectra across the red line shown in (a). c, dI/dV map at V = 0 mV measured on the area shown in (a). Middle and bottom rows: dI/dV maps at given bias voltages, all measured on the area shown in (a) and using the same colour scale as in (c).
Extended Data Fig. 9 | High-resolution STM images of different heterostructures. a-c, e-g, Large area STM images. b, STM image of a 1H-TaS$_2$ on HOPG ($V$ = 50 mV and $I$ = 500 pA). Inset shows fast Fourier transform of the image. d, STM image of a monolayer 1T-TaS$_2$ on HOPG ($V$ = 1 V and $I$ = 20 pA). f, STM image of a 1H/1T-TaS$_2$ vertical heterostructure ($V$ = 50 V and $I$ = 500 pA). g, STM image of a 1T/1H-TaS$_2$ vertical heterostructure ($V$ = 0.3 V and $I$ = 50 pA). 1H-TaS$_2$ on HOPG exhibits a strong 3×3 CDW, while 1H-TaS$_2$ on 1T-TaS$_2$ shows no signs of CDW. Both 1T-TaS$_2$ on HOPG and 1T-TaS$_2$ on 1H-TaS$_2$ exhibit a strong $\sqrt{13} \times \sqrt{13}$ CDW.
Extended Data Fig. 10 | Magnetic field dependence of a heavy-fermion gap.
Tunneling spectroscopy of a heavy-fermion gap measured on a 1H/1T-TaS₃ vertical heterostructure at different applied magnetic fields, the spectra are vertically offset for clarity.