Inducing and tuning Kondo screening in a narrow-electronic-band system

Shiwei Shen1, Chenhaoping Wen1, Pengfei Kong1, Jingjing Gao2,3, Jianguo Si2, Xuan Luo2, Wenjian Lu2, Yuping Sun2,4,5, Gang Chen6 & Shichao Yan1,7

Although the single-impurity Kondo physics has already been well understood, the understanding of the Kondo lattice where a dense array of local moments couples to the conduction electrons is still far from complete. The ability of creating and tuning the Kondo lattice in non-f-electron systems will be great helpful for further understanding the Kondo lattice behavior. Here we show that the Pb intercalation in the charge-density-wave-driven narrow-electronic-band system 1T-TaS2 induces a transition from the insulating gap to a sharp Kondo resonance in the scanning tunneling microscopy measurements. It results from the Kondo screening of the localized moments in the 13-site Star-of-David clusters of 1T-TaS2. As increasing the Pb concentration, the narrow electronic band derived from the localized electrons shifts away from the Fermi level and the Kondo resonance peak is gradually suppressed. Our results pave the way for creating and tuning many-body electronic states in layered narrow-electronic-band materials.

1 School of Physical Science and Technology, ShanghaiTech University, Shanghai 201210, China. 2 Key Laboratory of Materials Physics, Institute of Solid State Physics, HFIPS, Chinese Academy of Sciences, Hefei 230031, China. 3 University of Science and Technology of China, Hefei 230026, China. 4 High Magnetic Field Laboratory, HFIPS, Chinese Academy of Sciences, Hefei 230031, China. 5 Collaborative Innovation Centre of Advanced Microstructures, Nanjing University, Nanjing 210093, China. 6 Department of Physics and HKU-UCAS Joint Institute for Theoretical and Computational Physics at Hong Kong, The University of Hong Kong, Hong Kong, China. 7 ShanghaiTech Laboratory for Topological Physics, ShanghaiTech University, Shanghai 201210, China.

✉email: yanshch@shanghaitech.edu.cn
In the narrow-electronic-band or even the flat-band-like systems, the electron kinetic energy is strongly suppressed, and the electron correlation is then enhanced. When the electron correlation energy becomes comparable to the kinetic energy, the system is strongly correlated. This makes the narrow-electronic-band system a versatile platform for exploring the exotic correlated electronic phases. Well-known examples include the narrow band of f-like electrons with Kondo lattice and heavy fermions, and recently, the magic-angle twisted bilayer graphene superlattices with Mott-insulating gap and topological electronic phases.

In layered narrow-electronic-band materials, the Coulomb interaction and interlayer coupling often complex their electronic states. One of the notable examples is the charge density wave (CDW) driven narrow-electronic-band material, 1T-TaS$_2$. 1T-TaS$_2$ is a layered transition metal dichalcogenide (TMD) and undergoes a commensurate CDW transition around 180–190 K. In this commensurate CDW state of 1T-TaS$_2$, the in-plane lattice distortion leads to the formation of Star-of-David (SD) clusters which consist of 13 Ta atoms, and there is one unpaired electron in each SD cluster. Further band structure calculations indicate the unpaired electrons in the SD clusters form a narrow electronic band that may be susceptible to the Mott-Hubbard transition by the in-plane Coulomb interaction. The resulting Mott insulating state is quoted as a cluster Mott insulator where the unpaired electron is localized on the SD cluster rather than on the original Ta lattice site. In the cluster Mott insulating state, this unpaired electron forms an effective spin-1/2 local magnetic moment. No conventional magnetic ordering has been detected in 1T-TaS$_2$ down to millikelvin temperature, and 1T-TaS$_2$ has been proposed to be a spin-1/2 triangular lattice quantum spin liquid candidate. This local moment formation and spin liquid scenario have been supported by the recent muon spin relaxation measurement in 1T-TaS$_2$ and the scanning tunneling microscopy (STM) measurement in the epitaxially-grown isostuctural material (monolayer 1T-TaSe$_2$).

Apart from the novel Mott localization of the SD cluster in 1T-TaS$_2$, the previous theoretical and experimental studies indicate that the interlayer stacking effect plays a crucial role in the electronic properties of 1T-TaS$_2$. Due to the interlayer coupling in 1T-TaS$_2$, the interlayer stacking pattern of the SD clusters yields two distinct surface terminations. The Type-I surface terminates at the end of the dimerization, while the Type-II surface terminates in the middle of the dimerization and thus cuts the dimerization (see Fig. 1a). In the differential conductance (dI/dV) spectra measured with STM, there are insulating gaps for both surface terminations, and the Type-I surface has a slightly larger insulating gap than that on the Type-II surface. It has recently been demonstrated that the insulating gap on the Type-I surface is an interlayer-dimerization-induced insulating gap and the smaller insulating gap on the Type-II surface is likely a Mott-insulating gap.

More surprisingly, the very recent study demonstrates that when the monolayer 1T-TaS$_2$ is grown on the monolayer metallic 1H-TaS$_2$ by molecular beam epitaxy, the Kondo resonance peak appears in the 1T-TaS$_2$ layer, which confirms the local moment formation and possible cluster Mott physics in the monolayer 1T-TaS$_2$. If the insulating gap of the Type-II surface in the bulk 1T-TaS$_2$ is truly a Mott-insulating gap, there should be local magnetic moment formation that is similar as the monolayer 1T-TaS$_2$. The nature of a band-insulating gap instead of a Mott-insulating gap would indicate the absence of such a magnetic moment formation. To address this issue, the Kondo resonance could be an ideal experimental diagnosis of the local moment formation on the Type-II 1T-TaS$_2$ surface. Here we report a low-temperature and high-magnetic-field STM study about 1T-TaS$_2$. We observe the insulating gap to Kondo resonance transition on the Type-II 1T-TaS$_2$ surface by the Pb intercalation. We further establish the relationship between the narrow electronic band and the Kondo resonance peak. These observations are helpful for understanding the origin of the insulating gap on the Type-II 1T-TaS$_2$ surface and the Kondo resonance behavior in the narrow-electronic-band systems.

Results and discussion
After cleaving the 1T-TaS$_2$ sample in ultrahigh vacuum, we evaporate the Pb atoms onto the cleaved 1T-TaS$_2$ sample. We find both the Type-I and Type-II surface terminations exist. In Fig. 1d, g, we depict the STM topographies taken on the Type-I and Type-II surface terminations of 1T-TaS$_2$. The Pb atoms form regular islands on these two surfaces (the detailed structure of the Pb islands are shown in Supplementary Figs. S1, S2). However, the bare 1T-TaS$_2$ regions of the two surfaces appear quite different. The bare 1T-TaS$_2$ of the Type-I surface is the same as the pristine 1T-TaS$_2$ surface with uniform SD clusters and a few intrinsic atomic defects (see Fig. 1d). However, in the bare 1T-TaS$_2$ region of the Type-II surface, except the intrinsic atomic defects, some of the SD clusters appear as dark spots (see Fig. 1g). These dark SD clusters are almost indistinguishable in STM topographies taken with positive bias voltages (Supplementary Fig. S3), and the position of the dark SD cluster can be changed but rarely during the STM measurements (Supplementary Fig. S4). The boundary between these two types of the surfaces can also be seen on the same 1T-TaS$_2$ sample (Supplementary Fig. S5). In comparison with the SD clusters on the bare Type-I surface, we conclude that for the Type-II surface there are intercalated Pb atoms beneath the dark SD clusters and no intercalated Pb atoms under the bright SD clusters (Supplementary Fig. S5). Our STM data indicate that Pb atoms are prone to intercalate into the van der Waals gap below the Type-II surface. This is likely because the van der Waals gap below the Type-II surface is slightly larger than that below the Type-I surface.

In Fig. 1e, h, we show the zoom-in STM topographies taken on the two different surface terminations. Figure 1f, i are the differential conductance (dI/dV) spectra taken on the Pb island and the bare 1T-TaS$_2$ region for the two surface terminations, respectively. As shown in Fig. 1f, i, the Pb islands on the two surfaces are both metallic (see the green spectra). The dI/dV spectrum taken on the bare 1T-TaS$_2$ region of the Type-I surface is the same as that taken on the pristine Type-I surface, where the ~150 mV insulating gap can be seen (the purple spectrum shown in Fig. 1f). In contrast, instead of showing a Mott-insulating gap as on the pristine Type-II surface, a pronounced zero bias peak (ZBP) appears in the dI/dV spectrum taken on the bare Type-II surface (the purple spectrum shown in Fig. 1i). Although the dark SD clusters appear in most of the bare Type-II regions, we can still find a small region on the bare 1T-TaS$_2$ surface where there are no dark SD clusters (as shown by the dashed black lines in Fig. 2a). In this small region, the ~50 mV insulating gap as shown on the pristine Type-II surface can be seen (Fig. 2b and Supplementary Fig. S6). This demonstrates that the strong ZBP shown in Fig. 1i is indeed induced by the intercalation of Pb atoms below the Type-II surface. As shown by the yellow circles in Fig. 2a, there is no CDW domain wall in the Pb intercalated region, and the SD clusters in the Pb intercalated region and the ~50 mV gap region are also well aligned. This indicates that the intercalated Pb atoms do not create CDW domain walls (see Supplementary Fig. S7 for more details), which is different from the Se substituted 1T-TaS$_2$ and copper intercalated 1T-TiSe$_2$ where the substituted or intercalated atoms create CDW domain walls. We note that on the Type-II surface, we also observe a few
**Fig. 1** Emergence of the zero bias peak in the Pb intercalated 1T-TaS2. a Schematic showing the alternating interlayer stacking of the SD clusters in 1T-TaS2 which results in two inequivalent cleavage planes. b Schematic for the structure of the Type-I surface after evaporating Pb atoms. c Schematic for the structure of the Type-II surface after evaporating Pb atoms. The red, blue and yellow balls in b, c represent the Pb, Ta and S atoms, respectively. d Constant-current STM topography taken on the Type-I surface ($V_s = -500 \text{ mV}$, $I = 20 \text{ pA}$). e Zoom-in view of the bare Type-I surface and Pb islands ($V_s = -500 \text{ mV}$, $I = 20 \text{ pA}$). f Differential conductance ($dI/dV$) spectra taken on the spots indicated by the purple and green arrows shown in e. g Constant-current STM topography taken on the Type-II surface ($V_s = -500 \text{ mV}$, $I = 30 \text{ pA}$). h Zoom-in view of the bare Type-II surface and Pb islands ($V_s = -500 \text{ mV}$, $I = 20 \text{ pA}$). i $dI/dV$ spectra taken on the spots indicated by the purple and green arrows in h. The data shown in this figure are taken at $T = 4.3 \text{ K}$. Source data are provided as a Source Data file.

**Fig. 2** Spatial distribution of the ZBP. a Constant-current STM topography taken on the Type-II 1T-TaS2 surface ($V_s = -500 \text{ mV}$, $I = 20 \text{ pA}$). The yellow circles indicate the centers of the SD clusters. The dashed black lines show the region without the dark SD clusters. b $dI/dV$ spectrum taken on the spot shown by the red arrow in a. c Constant-current STM topography taken on the Type-II surface with dark SD clusters ($V_s = -150 \text{ mV}$, $I = 30 \text{ pA}$). d $dI/dV$ spectra taken on the bright and dark SD clusters shown by the purple and green arrows in c. The black arrow indicates the electronic state located slightly above the Fermi level in the dark SD cluster. e Spatially distributed $dI/dV$ spectra along the red arrow in c. f Constant-current STM topography taken on the Type-II surface with dark SD clusters ($V_s = -200 \text{ mV}$, $I = 30 \text{ pA}$). g, h $dI/dV$ maps taken on the same region as shown in f with 0 mV (g) and +40 mV (h) bias voltages, respectively. The green arrows in f–h indicate the positions of the dark SD clusters. The data shown in this figure are taken at $T = 4.3 \text{ K}$. Source data are provided as a Source Data file.
individual Pb adatoms, and there is no ZBP in the $dI/dV$ spectra taken on the Pb adatoms (Supplementary Fig. S8).

In order to understand the nature of the ZBP, we perform the spatially-resolved $dI/dV$ measurements on the dark SD clusters and the bright SD clusters on the Type-II surface. As shown in Fig. 2d, the strong ZBP appears on the bright SD cluster (shown with the purple arrow in Fig. 2c). On the dark SD cluster (shown with the green arrow in Fig. 2c), the intensity of the ZBP is strongly suppressed and a new electronic peak located slightly above the Fermi level emerges (shown with the black arrow in Fig. 2d). The $dI/dV$ linecut along the red arrow in Fig. 2c indicates the ZBP persist in every bright SD cluster (Fig. 2e and Supplementary Fig. S9). Figure 2g, h are the $dI/dV$ maps taken on the same region as shown in Fig. 2f, which clearly show the spatial distributions of the electronic states on the Pb intercalated Type-II surface. As we can see, the ZBP peak has higher intensity at the center of each bright SD cluster (Fig. 2g). The +40 mV electronic state locates at the dark SD clusters (shown by the green arrows in Fig. 2h). Since the Type-II surface is an undimerized 1T-TaS$_2$ surface where there is an unpaired electron localized in each SD cluster, the ZBP is likely due to the Kondo screening of the unpaired electrons in the SD clusters.

To further confirm the Kondo nature of the ZBP, we perform the temperature and magnetic field dependent $dI/dV$ measurements. In order to reduce the broadening effect induced by the modulation voltage, we reduce the modulation voltage to be 0.2 mV. As shown in Fig. 3a, the ZBP at each temperature is fitted reasonably well with the thermally convolved Fano line shape with intrinsic width.$^{11,23}$ Fitting the half width at half maximum of the temperature-dependent ZBP with the well-known Kondo expression yields a Kondo temperature $T_K \sim 37$ K (Fig. 3b)$^{23,24}$. In addition, we perform the magnetic field dependent $dI/dV$ measurements at 0.6 K. As shown in Fig. 3c, no clear splitting of the Kondo resonance peak is observed with external magnetic field up to 8.5 T. However, the Kondo resonance peak in the single layer 1T-TaS$_2$ is clearly split with 10 T magnetic field.$^{20}$ The difference is that the width of Kondo resonance peak in Fig. 3c is about four times the width of the Kondo resonance peak in the single layer 1T-TaS$_2$ (Supplementary Fig. S10). This makes the Kondo resonance peak in Fig. 3c less sensitive to the splitting induced by the magnetic field, and larger magnetic field is needed to induce the splitting of the Kondo resonance on the Pb intercalated Type-II 1T-TaS$_2$ surface.$^{25,26}$ Similar effect has also been observed in the heavy fermion metal where the Kondo lattice peak has no clear splitting with magnetic field up to 11 T.$^{27}$

Having identified the Kondo nature of the ZBP, we next discuss the origin of the electronic state located slightly above the Fermi level in the dark SD cluster marked by the green arrow in Fig. 2c. In the CDW state of 1T-TaS$_2$, the unpaired electrons in each SD cluster form a narrow electronic band near the Fermi level.$^{18}$ The new electronic band at the dark SD cluster is the narrow electronic states derived from the unpaired electrons in the SD clusters. This also indicates that the intercalated Pb atom depletes the localized electron in the dark SD cluster, which makes the narrow electronic band almost unoccupied and locate slightly above the Fermi level.

The next question is why the intercalated Pb atoms can induce the transition from the ~50 mV insulating gap to the sharp...
Kondo resonance peak on the Type-II 1T-TaS₂ surface. Recently, motivated by the Kondo resonance in single layer 1T-TaSe₂ grown on single layer 1H-TaSe₂, Chen et al. have developed a model for the correlated electrons coupled by tunneling to a layer of itinerant metallic electrons to explain the transition to the Kondo resonance. We think the insulating gap to Kondo resonance is related with the energy position of the narrow electronic band formed by the unpaired electrons in the SD clusters. As shown in Fig. 4a–d (see Supplementary Fig. S12 for more details), when the intercalant concentration of the Pb atoms increases, the narrow electronic band is gradually shifted to be below the Fermi level (Fig. 4e–h). At the same time, the Kondo peak is gradually reduced (Fig. 4c, f), and it can be fully suppressed (Fig. 4g, h). The evolution of dI/dV spectra in the dark SD clusters are shown in Supplementary Fig. S13. This means that as increasing the Pb concentration, the narrow electronic band is gradually electron doped at the positions of the bright SD clusters, and the Kondo resonance peak is related with its filling factor.

Furthermore, we find that as increasing the intercalant concentration of Pb, the dark and bright SD clusters on the Type-II surface form a new $\sqrt{3} \times \sqrt{3}$ superstructure with respect to the SD lattice (Fig. 5a). The green and purple dots in Fig. 5a indicate the dark and bright SD clusters. Figure 5b is the STM topography related with the energy position of the narrow electronic band. As shown in the dI/dV spectra taken on the bright and dark SD clusters (Fig. 5c), the narrow electronic band in the dark SD cluster is fully unoccupied (the green spectrum), and it is fully occupied in the bright SD cluster (the purple spectrum). Figure 5d–f show the spatial distributions of the electronic states at the energies indicated by the vertical dashed lines in Fig. 5c. More dI/dV maps are shown in the Supplementary Fig. S14. As we can see, the $-80$ mV and the $+40$ mV states show contrast inversion, and they have higher intensity in the bright and dark SD clusters, respectively. Our data indicate the intercalated Pb atoms make the localized electrons in the SD clusters of the top 1T-TaS₂ layer to be spatially redistributed and the new $\sqrt{3} \times \sqrt{3}$ superstructure results from the spatial charge modulation of the localized electrons. In this case, the narrow electronic band in the bright SD clusters is locally electron doped and it corresponds to local hole doping in the dark SD clusters (insets in Fig. 5c).
Our work demonstrates that the intercalated Pb atoms below the undimerized 1T-TaS$_2$ layer induce the insulating gap to Kondo resonance transition. The strength of the Kondo resonance peak can be deliberately tuned by the intercalant concentration of Pb. Our data not only demonstrate the interplay between the narrow electronic band and the Kondo resonance peak, but also shed new light on the origin of the insulating gap in the undimerized 1T-TaS$_2$ layer. Our results should be helpful for understanding the Kondo lattice behavior in heavy fermion materials and the many-body resonance in kagome metals. We believe this method should be able to be extended to other foreign atoms or molecules to induce many-body electronic states in layered narrow-electronic-band materials.

**Methods**

**Sample preparation.** 1T-TaS$_2$ samples were cleaved at room temperature and in an ultrahigh-vacuum chamber. After cleaving, Pb atoms were deposited onto the 1T-TaS$_2$ sample from a Knudsen cell. During dosing Pb, the 1T-TaS$_2$ sample was kept at room temperature, and the temperature of the Pb source was 470 °C. After dosing Pb, the 1T-TaS$_2$ sample was kept at room temperature for 5 to 10 min and then inserted into the STM head for measurements. The intercalation concentration of the Pb atoms below the Type-II surface was related with the room-temperature annealing time after dosing Pb (see Supplementary Fig. S12).

**STM measurements.** The STM experiments at 4.3 K were conducted with a standard lock-in detection technique.

**Data availability**

The data that support the findings presented here are available from the corresponding author upon reasonable request. Source data are provided with this paper.

**Code availability**

The codes for fitting the data are available from the corresponding author upon reasonable request.

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Author contributions
S.Y. conceived the experiment. S.S., C.W. and P.K. carried out the experiments and performed the analysis. J.G., L.X. and Y.S. were responsible for the sample growth. J.S. and W.L. performed the first-principles calculations. S.S., G.C. and S.Y. wrote the manuscript with the input from all authors.

Competing interests
The authors declare no competing interests.

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Correspondence and requests for materials should be addressed to Shichao Yan.

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