Divalent EuRh$_2$Si$_2$ as a reference for the Luttinger theorem and antiferromagnetism in trivalent heavy-fermion YbRh$_2$Si$_2$

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Application of the Luttinger theorem to the Kondo lattice YbRh$_2$Si$_2$ suggests that its large 4f-derived Fermi surface (FS) in the paramagnetic (PM) regime should be similar in shape and volume to that of the divalent local-moment antiferromagnet (AFM) EuRh$_2$Si$_2$ in its PM regime. Here we show by angle-resolved photoemission spectroscopy that paramagnetic EuRh$_2$Si$_2$ has a large FS essentially similar to the one seen in YbRh$_2$Si$_2$ down to 1 K. In EuRh$_2$Si$_2$ the onset of AFM order below 24.5 K induces an extensive fragmentation of the FS due to Brillouin zone folding, intersection and resulting hybridization of the Fermi-surface sheets. Our results on EuRh$_2$Si$_2$ indicate that the formation of the AFM state in YbRh$_2$Si$_2$ is very likely also connected with similar changes in the FS, which have to be taken into account in the controversial analysis and discussion of anomalies observed at the quantum critical point in this system.
he determination of Fermi surfaces (FSs) plays a central role in understanding the many-body physics of strongly correlated metals. This is because most of the exciting phenomena in these systems are governed by thermally excited quasiparticles confined to a narrow window around the Fermi energy ($E_F$)\(^1\)\(^-\)\(^5\). Furthermore, FSs are often the key to discriminating between rivaling theoretical models describing particular correlated metals\(^1\)\(^-\)\(^5\). For periodic Kondo lattice (KL) materials, where local orbital electrons both carry a magnetic moment and are coupled to conduction bands, the central issue is the extent to which the moment bearing electrons are included in the FS and the way in which this might be accomplished. A fundamental guiding theoretical principle was established by the seminal realization of R. M. Martin\(^6\)\(^,\)\(^7\) that the Luttinger FS sum rule\(^7\) can be applied even to such strongly correlated systems.

The sum rule states that the FS volume is conserved in the presence of interactions so long as the interactions do not produce a phase transition. In this context, the essence of the sum rule is that if the local orbital magnetic moments are quenched, for example by the Kondo effect, then the underlying local orbital electrons must be counted in the FS. If the moments are not quenched, for example in a magnetically ordered state, the underlying electrons are excluded from the FS. For the KL, the sum rule is satisfied by the formation of heavy quasi-particle states at the Fermi energy, the analog of the Kondo resonance for a local orbital impurity site. The behavior along the line of the Fermi energy labeled $E_F^{\text{Yb}}$ in Fig. 1a shows, for the hole situation that will be relevant for this paper, how hybridization of the local orbital quasiparticle into the conduction bands enlarges the hole-like Fermi surface size from $k_F$ to $k'_F$ to count the local orbital hole. These two situations are now known as the small and large Fermi surfaces, respectively\(^6\)\(^,\)\(^7\).

Even though the initial theoretical realization is now many years in the past, clear experimental demonstrations of the both Luttinger theorem and its workings for KL materials are still very few. Measurements with the de Haas van Alphen (dHvA) technique have provided some observations of both the small and large FS\(^10\). But for angle resolved photoemission spectroscopy (ARPES) the large FS has been observed thus far only in YbRh\(_2\)Si\(_2\)\(^1\)\(^-\)\(^3\),\(^9\), while for another paradigm system CeCoIn\(_5\) the situation remains problematic and controversial\(^11\)\(^,\)\(^12\). This paucity is particularly unfortunate because ARPES is the FS technique most evocative for this concept, in that it allows to see directly both the Fermi surface and the underlying dispersing quasiparticles that define it.

Here, we present an ARPES study which reveals both the workings of the Luttinger sum rule and also another very general FS principle pointed out by Müller–Hartmann\(^13\), that if the single particle self energy for a dispersing quasi-particle is independent of the crystal momentum $k$, as is highly plausible for the 4f electrons of a rare-earth material, not only the FS size but also its shape is conserved for symmetry preserving interactions. These two concepts together are the reason why the FSs found in density functional theory (DFT) can often be in good agreement with experiment for rare-earth materials.

As explained further below, we compare two isostructural and isostructural rare-earth materials, YbRh\(_2\)Si\(_2\) (YRS) and EuRh\(_2\)Si\(_2\) (ERS), both in the paramagnetic (PM) state, the former where the FS encloses one strongly correlated 4f hole and $N$ weakly correlated conduction band holes, and the latter where the FS encloses $N + 1$ weakly correlated holes, i.e. the same total hole count for

**Fig. 1** Formation of the large Fermi surface in a Kondo lattice. a Schematic hybridization model for Yb case: the localized and renormalized level of the 4f hole (dashed red line) hybridizes with a light hole-like conduction band (blue) and forms a heavy-quasiparticle band (purple) right below the Fermi level, $E_F$. The Fermi vector shifts from $k_F$ to $k'_F$. The red sun symbol indicates the region of $f-d$ hybridization. b ARPES band map of YbRh\(_2\)Si\(_2\) (compare ref. 9). The Brillouin zone cut corresponds to the black line shown in d. The crystal-field-split Yb 4f bands (highlighted in red) hybridize with a bunch of projected bulk bands (shaded in blue) forming a heavy hole-like quasi-particle band at low temperature and shifting the Fermi vector outwards. c, d Schematic representation of a small and large Fermi surface: due to the formation of the Kondo lattice and the Luttinger theorem, the small hole pocket in c transforms into a large hole pocket in d, which incorporates now the additional 4f hole.
Results

Strongly correlated YbRh$_2$Si$_2$ and weakly correlated EuRh$_2$Si$_2$.

Heavily studied YRS\textsuperscript{14–17} is a canonical heavy-fermion KL material, with antiferromagnetic (AFM) order below a Néel temperature $T_N$ of 70 mK and superconductivity below 2 mK\textsuperscript{17}. The AFM transition lies very close to a quantum-critical point (QCP) induced by a tiny magnetic field of 60 mT applied along the basal plane, that suppresses the magnetic order. The Yb valence $v$ is near 3$^+$ which gives one hole in the 4$f$ shell. If magnetic moments are Kondo quenched in the low temperature paramagnetic phase just above $T_N$, we would expect this hole to be included in the FS. As will be seen in more detail below, DFT shows that the generic RERh$_2$Si$_2$ (RE = rare earth) FS consists of two sheets called the Doughnut $(D)$ and the Jungle-gym $(J)$\textsuperscript{8–10,18}. The Jungle-gym forms a largely interconnected Fermi surface sheet across the whole $k$-space, whereas the Doughnut consists of a square-like, large hole pocket around the Z-point of 3D Brillouin zone (BZ).

The topology of the Doughnut can be used to distinguish the large (itinerant 4$f$ hole) and the small (localized 4$f$ hole) FSs\textsuperscript{8–10}, via the size of its necks which, respectively, do or do not extend into neighboring BZs, as shown schematically in Fig. 1c, d. Our past ARPES work for YRS clearly shows the large FS of Fig. 1d down to 800 mK\textsuperscript{8}. The YRS ARPES spectrum of Fig. 1b shows the heavy 4$f$ quasi-particle and the itinerant band states dispersing across $E_F$ marked with colors corresponding to the schematic of Fig. 1a. As a calibration, the small FS of Fig. 1c has been included conclusively verified by ARPES and DFT for the stable trivalent, local moment AFM, weakly correlated, isoelectronic reference system YbCo$_2$Si$_2$\textsuperscript{8,19}, see also Supplementary Note 1.

ERS, the material studied in this paper, is also a weakly correlated, local moment AFM isoelectronic reference system\textsuperscript{20–22}, but with a valency of 2$^+$ it is divalent. Thus, in accordance with the Luttinger sum rule, ERS in its paramagnetic phase should possess the large FS, and in the first part of the paper we will show that this is indeed the case. Further, not only can the electronic structure and Fermi surface for the AFM phase of ERS be calculated easily with DFT, but also the transition at $T_N = 24.5$ K can be explored conveniently with ARPES. In the second part of the paper we present the respective DFT and ARPES results for the AFM phase and, finally, we use these data to discuss possible changes in the electronic structure of YRS in its AFM phase below 70 mK and the possible implications for its QCP behavior.

Paramagnetic phase of EuRh$_2$Si$_2$ from DFT and UV-ARPES.

The DFT-derived FS of ERS in the PM phase is shown in Fig. 2a, b as a side and top view, respectively. It very closely resembles the one established for YRS in the Kondo PM regime and both the Doughnut and the Jungle-gym can clearly be seen. The center and inset panels of Fig. 2c compare, respectively, DFT and UV-ARPES data for PM ERS, taken with 45 eV photon energy. For this photon energy and the resulting photoelectron kinetic energy, the photoelectron has a very small relaxation time and a short mean free path. In consequence\textsuperscript{23} our UV-ARPES data is both surface sensitive and $k_z$ broadened so that the bulk electronic structure is mostly $k$-summed along $k_z$. Therefore the DFT-derived electronic structure is shown projected along $k_z$ onto the 2D surface BZ, which is depicted as a dashed-red line in Fig. 2b. High symmetry points of the surface BZ are marked in both panels with overlined symbols. The large Doughnut sheet can be well distinguished and is seen as a homogenous gray area. The projected Jungle-gym is weakly visible in the first BZ, but due to photoemission cross-section effects it is much more pronounced in the second BZ. Those parts that build the border of the projected gap around the M-point and are not masked by the Doughnut, are indicated by arrows. In addition to these features predicted by the bulk band-structure calculations, sharp spectral patterns around the Γ-point as well as the diamond-shaped...
feature around the M-point result from surface resonances and Shockley surface states. The Shockley state has been discussed in detail in earlier works\textsuperscript{24} and green (hν = 730 eV), respectively. c Fermi surface sections derived from soft X-ray ARPES performed at 40 K (for the PM phase) and at 10 K (for the AFM phase).

**Soft X-ray ARPES on EuRh$_2$Si$_2$.** At such high photon energies, the probing depth is quite high compared to that of UV-ARPES, the probability to excite surface-related states is significantly reduced, and we can study the bulk $k_z$ dependence (a complete $k_z$ - $k_x$ map obtained by soft X-ray ARPES is presented in Supplementary Figure 2 and discussed in Supplementary Note 3). The energy resolution is reduced but nonetheless adequate for the purpose at hand. We used photon energies of 670 eV and 730 eV, which probe the electronic structure along the measurement arcs shown in Fig. 3b. These provide planes of approximately constant $k_z$ near the $\Gamma$- and Z-point of the paramagnetic BZ. In Fig. 3c, we compare ARPES-derived Fermi surface maps obtained in the PM phase at $T = 40$ K and the AFM phase at $T = 10$ K, see also Supplementary Figure 3 and Supplementary Note 4. As expected, in the PM phase we see distinctly different spectra in the regions of the inequivalent \( \Gamma \) - and Z-points. In particular, a hole-like pocket can be seen around Z, whereas no such pocket exists around \( \Gamma \). Upon cooling the system down to 10 K below the Néel temperature, a similar hole-like band as that around Z now appears around the $\Gamma$-point, where the intensity was previously depleted. At the same time, the hole pocket around Z seems to be unaffected by the AFM transition. The appearance of spectral
weight around \( \Gamma \) in the AFM phase, which in the PM phase could be seen exclusively around \( Z \), is fully consistent with the change of the BZ and the resulting back-folding of electronic bands.

**Antiferromagnetic phase of EuRh\(_2\)Si\(_2\) from DFT and UV-ARPES.** Having experimentally established the back folding of electron bands along the \( \Gamma - Z \) direction across the AFM transition, we turn now to the results of our band structure calculations to analyze in more detail the changes of the electronic structure upon entering the AFM phase. The DFT-derived FS is shown in Fig. 4a, b as a side and top view, respectively. The Doughnut and parts of the Jungle-gym are folded into the new BZ. The Doughnut is folded onto the \( \Gamma \)-point along with the previous Z-point of the PM phase. The new Z-plane, which lies halfway between the \( \Gamma \)- and Z-points of the PM structure, cuts the folded Doughnut slightly, resulting in small oval hole pockets labeled (3) in Fig. 4b and marked in green (see Supplementary Figure 4). These pockets are oriented towards the corners of the simple-tetragonal BZ and are a further important indicator for the folded FS in the AFM phase. The large Jungle-gym then hybridizes with the Doughnut, splitting up into two distinct sheets. The resulting splitting of the FS is labeled (2) in Fig. 4b and marked in cyan. The outermost sheet enclosing the BZ corners is labeled (1) in Fig. 4b and marked in red. This banana-shaped feature results from the folding of the Jungle-gym onto itself, splitting off due to the exchange interaction to form long tubes with almost parallel surfaces along \( k_z \). Similarly, the outer of the two sheets of feature (2) also forms long, but warped, tubes along \( k_z \). As a general conclusion, the bulk DFT calculation predicts a strong fragmentation of the PM FS upon the AFM transition.

We now compare the band structure calculation to the FS experimentally obtained by UV-ARPES at a temperature of 10 K, sufficiently below the Néel temperature. As with the PM phase we must account for the \( k_z \) broadening of bulk features and the sensitivity to surface features. We consider the bulk features first and show in the upper left part of Fig. 4c the calculated bulk FS projected along \( k_z \). As labeled, it still shows nicely the same three key features already called out in Fig. 4b. The gap labeled (2) lies within the projected Doughnut close to the borders. The ARPES-derived FS in the lower right half of Fig. 4c shows these same three bulk features, as indicated by dashed lines with the same color coding. Although the ARPES intensity of the PM phase Jungle-gym is quite low in the first BZ, the borders of the outer FS sheet, which coincide with the banana-shaped feature (1), are clearly visible. The ARPES-derived FS also shows the gap labeled (2) and additionally there are four oval gap-like features which appear exactly where the oval hole pockets (3) are expected.

Considering next the surface, by comparing the ARPES data to slab DFT calculations we find that all remaining features can be understood as a superposition of spectra from both Si and Eu terminations present together on our measured surfaces. In our slab DFT calculations, a supercell consisting of a thick slab of four conventional unit cells (32 atomic layers) surrounded by empty space mimics the two surfaces of the material with several bulk layers in between and enables us to study the surface modifications of the band structure. In Fig. 4d, we overlaid the projected bulk bands (shaded in gray) with the surface states and surface resonances of the FS obtained from our slab calculation (colored). To identify the surface character, only states with a predominant contribution in the topmost four atomic layers are extracted from the overall slab band structure. The two segments at the top colored cyan and red/blue are calculated for the Eu-terminated half of the slab, the two segments at the bottom colored green and red/blue are obtained from the Si-terminated part. The cyan and green states indicate surface states and resonances for the respective terminations. The red and blue coloring on the right side additionally points out the spin polarization of these states.

We have already identified in the PM phase UV-ARPES spectra a pronounced well known Shockley state around the M-point. We see that this strictly two-dimensional Shockley state splits off from the border of the projected bulk band gap, occurs only for Si-termination, and is strongly spin split due to exchange interactions with the ferromagnetically polarized Eu \( 4f \) moments, both in the calculated AFM FS and in the AFM ARPES data. This characteristic fingerprint thus clearly indicates the presence of Si-terminations in UV-ARPES.

For Eu-termination there are a bunch of surface resonances that arise from bulk states, are pinned to the topmost Eu surface layer, and are further spin split. These account nicely for what is perhaps the most striking and instructive feature in the UV-ARPES derived FS, a dense bunch of bright and rather sharp states, which appear within the Doughnut following roughly its shape. Multiply repeated experiments convincingly indicate reproducibility of this spectral pattern. These states obviously cannot be observed in the PM phase (compare Fig. 2c) and are not present in the calculated bulk FS of the AFM phase.

**Discussion**

What are the relevance and the implications of our observations on the AFM state of EuRh\(_2\)Si\(_2\) for the system YbRh\(_2\)Si\(_2\)? In the debate on the nature of the field induced quantum critical point in YbRh\(_2\)Si\(_2\), the evolution of the Fermi surface from the magnetically non-ordered state to the AFM state is a pivotal issue\(^{15,16,27}\). Most of the discussion has focused on the possibility of a change in the nature of the quasiparticle, while the effect of the magnetic ordering on its own has not been considered.
However, the present observations on EuRh$_2$Si$_2$ show that magnetic ordering results in large changes in the Fermi surface through band folding and band splitting. These results cannot be taken over directly to YbRh$_2$Si$_2$ because the AFM structure of YbRh$_2$Si$_2$ is yet unknown. However, results on both static magnetic properties (e.g. magnetization) as well as dynamic magnetic properties (e.g. relaxation behavior) (see Supplementary Note 5) indicate that magnetic correlations in YbRh$_2$Si$_2$ are very similar to those in EuRh$_2$Si$_2$. In particular both display a strong competition between FM and AFM order. With the Fermi surfaces being so similar in the PM states of EuRh$_2$Si$_2$ and YbRh$_2$Si$_2$, and also a strong similarity in their magnetic correlations, EuRh$_2$Si$_2$ becomes a relevant model system for gauging the impact of AFM order on the Fermi surface of YbRh$_2$Si$_2$.

One concern with this approach might be that EuRh$_2$Si$_2$ is an RKKY mediated large local moment system whereas the extremely small moment of YbRh$_2$Si$_2$ strongly suggests that it should be regarded as a spin-density-wave (SDW) system (here SDW refers to its original sense as the ordering of delocalized electrons). However, for the relevance and resulting change of the Fermi surface, both cases are actually very similar. Because in RKKY exchange the transfer of information on the polarization from one 4f site to the next 4f site is exclusively due to conduction electrons, the generalized susceptibility $\chi(q)$ of the conduction electrons, and therefore e.g. nesting properties, is as much relevant for local moment ordering as for the SDW scenario. Accordingly, modifications of the band structure, at least on a qualitative level, should be very similar in both kinds of systems. Both our experimental ARPES results and our calculations show that for this Fermi surface, AFM order leads to significant changes, because of both the large amount of band folding and the large band splittings. Such large changes cannot be ignored in discussing the anomalies seen in a number of transport properties only within the AFM state, but not the critical behavior of the Kondo system YbRh$_2$Si$_2$ (with nearly trivalent Yb). The Si terminated surface reveals mainly itinerant magnetism confined to the surface due to the presence of Shockley surface states, which couple via exchange interaction with the Eu 4f moments in the fourth layer below the Si surface. However, the Eu-terminated surface exhibits mainly bulk electron states, which show strong resonant behavior at the Eu surface. Due to the exchange coupling with the topmost polarized Eu 4f moments these states become themselves spin-polarized in the vicinity of the surface. These results clearly show that a magnetically ordered hetero system may reveal curious and profoundly different magnetism at its surface depending on the constituting sorts of surface atoms which is challenging to predict and can be interesting for technological applications.

### Methods

**Experiment.** UV-ARPES and Soft X-ray ARPES experiments were carried out at the Swiss Light Source (SIS X09LA instrument) and SPring-8 synchrotron radiation facility (Beamline BL23SU), respectively. The UV-ARPES experiments were discussed in details in24, while the Soft X-ray ARPES experiments were comprehensively presented in ref. 25. The single crystals of EuRh$_2$Si$_2$ were grown in iodium flux as described in20,21.

**Theory.** The full-relativistic band structure calculations have been performed with the FPLO code26, a density functional theory code based on the full-potential local orbital method, within the generalized gradient approximation (GGA)27. The Eu 4f basis states have been fixed to the experimentally found occupation ($n_{4f} = 7.0^{+2}_{-1}$, frozen core approximation) using either an unpolarized configuration in the paramagnetic phase or a local-moment scenario of 7 $\mu_B$ per Eu atom with ferromagnetic coupling within the Eu a-b-planes and antiferromagnetic interaction along the c-axis in the AFM phase. Surface effects were simulated by an asymmetric slab with 32 atomic layers terminated on one side by Si and on the other side by Eu atoms. Further details on the computations can be found in28,29.

### Data availability

All relevant data are available from the corresponding author upon request.

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Author contributions
C.G. and D.V.V. designed research; S.S., C.K., and C.G. grew and characterized the samples; M.G., A.G., A.C., S.D., and D.V.V. performed the UV-ARPES experiments; S.I.F. performed Soft X-ray ARPES experiment; Theoretical studies were performed by M.G., Yu.M.K. and E.V.C.; Operation of the UV-ARPES facility was carried out by M.R., M.S., and N.C.P. The obtained results were discussed together with K.K., C.L., and J.W.A.; The manuscript was written by M.G., C.G., J.W.A. and D.V.V.

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