

Magnetism in f -electron superlattices

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We analyze antiferromagnetism in f -electron superlattices. We show that the competition between the Kondo effect and the RKKY interaction in f -electron materials is modified by the superlattice structure. Thus, the quantum critical point which separates the magnetic phase and the Fermi-liquid phase depends on the structure of the f -electron superlattice. The competition between the Kondo effect and the RKKY interaction is also reflected in the magnetic interlayer coupling between different f -electron layers. We demonstrate that in the case of a weak Kondo effect the magnetic interlayer coupling behaves similarly to other magnetic heterostructures without the Kondo effect. However, close to the quantum phase transition, the dependence of the interlayer coupling on the distance between the f -electron layers is modified by the Kondo effect. Another remarkable effect, which is characteristic for an f -electron superlattice, is that the magnetic interlayer coupling does vanish stepwise depending on the distance between different f -electron layers. As a consequence, the quantum critical point depends also stepwise on this distance.

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I. INTRODUCTION

Recent experimental progress has made it possible to create thin layers of f -electron materials and artificial superlattices, which consist of a periodic structure of f -electron materials such as CeCoIn₅ and CeIn₃ and metals without f electrons [1–6]. Thus, it has become feasible to change the electronic structure and tune the properties of f -electron materials. This is particularly important when one thinks of the interesting phenomena which can be observed in these materials, such as magnetism, unconventional superconductivity, and quantum criticality. Usually, a quantum critical point in f -electron materials (if it exists) occurs at a certain pressure or magnetic field. These parameters are fixed by the electronic structure. In an artificially created f -electron superlattice, on the other hand, it is now possible to change the electronic structure of the material and thus tune the quantum critical point. Furthermore, by combining layers of different f -electron materials the competition/interplay between quantum critical layers, superconducting layers, magnetic layers, and metallic heavy-fermion layers can be studied, which presents invaluable opportunities to study novel phenomena.

An example of an f -electron superlattice which has recently been created in the laboratory is CeIn₃(n)/LaIn₃(4). It has been experimentally shown that the Néel temperature in CeIn₃(n)/LaIn₃(4) superlattices decreases to 0 when the Ce-layer thickness n is reduced to $n = 2$, which is accompanied by a linear temperature dependence of the resistivity [1]. This demonstrates the influence of the superlattice structure on the magnetic state in f -electron material and the ability to tune the quantum critical point. In other experiments, using the heavy fermion CeCoIn₅ and the conventional metal YbCoIn₅, superconductivity has been observed in thin CeCoIn₅ layers [2].

The properties of f -electron materials are determined by the competition of the RKKY interaction and the Kondo effect. While the RKKY interaction favors a magnetically ordered state, the Kondo effect screens the magnetic moments of strongly interacting f electrons, which results in a paramagnetic Fermi-liquid state. This competition is described and visualized in the Doniach phase diagram [7], which contrasts the energy scales of both effects; the RKKY interaction depends quadratically on the coupling between conduction electrons and f electrons and the Kondo effect exponentially. Therefore, for weak coupling the RKKY interaction is stronger than the Kondo effect, while for strong coupling the Kondo effect prevails. When both effects are equal in strength, quantum criticality accompanied by non-Fermi-liquid behavior can frequently be observed [8–10].

While the dependence of the RKKY interaction and the Kondo effect on the coupling between conduction electrons and f electrons is well understood [7], it is unclear how the competition of both depends on the superlattice structure. In this paper we analyze the influence of the superlattice structure on the RKKY interaction and the competition with the Kondo effect and, furthermore, study the dependence of the magnetic interlayer coupling on the distance between different f -electron layers and how it is modified by the Kondo effect. We find that especially close to the quantum critical point, there are large modifications in the dependence on the distance between the f -electron layers compared to non- f -electron superlattices. The magnetic interlayer coupling vanishes stepwise when the distance between different f -electron layers is increased. Thus, the quantum critical point changes stepwise when the number of spacer layers changes.

The remainder of this paper is organized as follows: In the next section we describe the model and methods which we use to analyze the f -electron superlattice. This is followed by Sec. III, where we analyze the magnetization dependence in f -electron superlattices. Thereafter, we analyze the interlayer coupling in Sec. IV and the spectral functions in Sec. V. Conclusions finish the paper (Sec. VI).

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II. MODEL AND METHOD

Similarly to our previous paper, in which we analyze the Kondo effect in f -electron superlattices [11], we study a system consisting of a periodic structure made of Kondo lattice layers [7,12,13], described by H_{KLL} , and normal metallic layers, described by H_{NL} . We use the notation of a (N, M) superlattice, where N Kondo lattice layers are separated by M normal metal layers, but focus only on $(1, M)$ superlattices. We model each layer as a square lattice. The model Hamiltonian, which is visualized in Fig. 1, thus reads

$$\begin{aligned} H &= H_{\text{NL}} + H_{\text{KLL}} + H_{\text{inter}}, \\ H_{\text{NL}} &= t_{\text{NL}} \sum_{(i,j)\sigma} c_{i z \sigma}^\dagger c_{j z \sigma}, \\ H_{\text{KLL}} &= t_{\text{KLL}} \sum_{(i,j)\sigma} c_{i z \sigma}^\dagger c_{j z \sigma} + J \sum_i \vec{S}_{i z} \cdot \vec{s}_{i z}, \\ H_{\text{inter}} &= t_z \sum_i \sum_{\langle z_1, z_2 \rangle \sigma} c_{i z_1, \sigma}^\dagger c_{i z_2, \sigma}. \end{aligned}$$

The operator $c_{i z \sigma}^\dagger$ creates an electron at lattice site (i, z) with spin direction $\sigma = (\uparrow, \downarrow)$, where i is an index describing the x and y direction. t_{NL} and t_{KLL} are hopping constants within the normal metallic layers and the Kondo lattice layers, respectively. t_z describes the hopping between layers. In our calculation we study only isotropic electron hopping, $t = t_{\text{NL}} = t_{\text{KLL}} = t_z$. J describes the spin-spin interaction between the magnetic moments and the conduction electrons in the Kondo lattice layers. Throughout this paper we assume an antiferromagnetic coupling, $J > 0$. Furthermore, we take the hopping t as the unit of energy. All calculations in this study are performed for half-filled lattices, $\langle n_\uparrow \rangle + \langle n_\downarrow \rangle = 1$. The doped case is left for future studies.

We use the real-space dynamical mean-field theory (RDMFT) [14] to calculate the magnetic properties of this system. The RDMFT approximates the self-energy for each lattice site as local; the self-energy vanishes between different atoms, thus nonlocal fluctuations are not included in this framework. This approximation becomes exact in infinite dimensions. Because the self-energy can depend on the lattice site and spin direction, this approach is suitable for analyzing the competition of the Kondo effect and magnetism in f -electron superlattices. In order to calculate the self-energy, each lattice site is mapped onto its own impurity model. This is

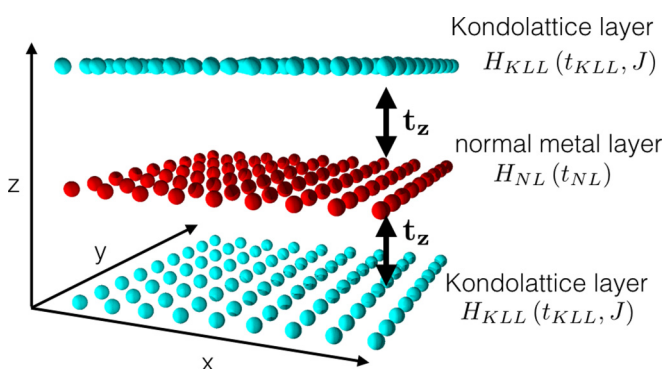


FIG. 1. Visualization of the Hamiltonian.

done by calculating the local Green's function for each lattice site, which can be written as

$$G_{i z \sigma}(\omega + i\eta) = \frac{1}{\omega + i\eta - \Delta_{i z \sigma}(\omega + i\eta) - \Sigma_{i z \sigma}(\omega + i\eta)},$$

where $\Sigma_{i z \sigma}(\omega + i\eta)$ is the self-energy for this lattice site. The hybridization function $\Delta_{i z \sigma}(\omega + i\eta)$ describes the coupling between an impurity and conduction electrons. The resulting impurity model for each lattice site is then solved using the numerical renormalization group (NRG) [15–18].

III. MAGNETIZATION

As stated above, the properties of f -electron materials are strongly influenced by the competition between the RKKY interaction and the Kondo effect. A similar competition will also occur in an f -electron superlattice. However, while in an ordinary f -electron material the strength of the RKKY interaction and the energy scale of the Kondo effect are fixed, in an f -electron superlattice they depend on the width of the layers used in the superlattice.

Thus, we start our analysis by showing the magnetization of the conduction electrons within the f -electron layers for different superlattice structures at $T = 0$ (see Fig. 2). In the strong-coupling region, $J/t > 2.6$, all magnetization curves vanish independently of the superlattice. In this region all layers of the superlattice become paramagnetic, i.e., the localized spins are screened by the Kondo effect. In this phase the density of states vanishes exactly at the Fermi energy in all f -electron layers due to the formation of a Kondo insulating state, while the spacer layers remain metallic.

The critical interaction strength, at which the magnetization vanishes, depends therefore on the superlattice structure. The three-dimensional (3D) Kondo lattice without any spacer layers (red curve in Fig. 2) has the highest critical interaction strength, $J_C^{3\text{D}}/t \approx 2.6$. We observe that the magnetization of the (1,1) superlattice vanishes at nearly the same critical interaction strength as the 3D lattice. Increasing the number of spacer layers further, the maximum value of the magnetization as well as the critical interaction strength decreases.

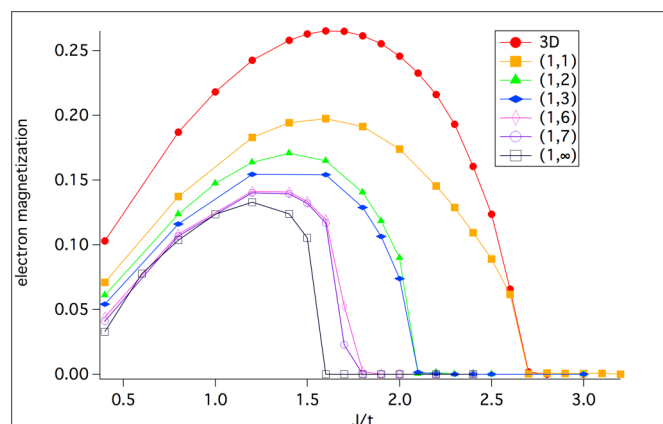


FIG. 2. Magnetization of the conduction electrons plotted against the coupling strength, J , for different superlattices, $(1, M)$. The magnetization curves interpolate between a single layer embedded in a 3D metallic host, $(1, \infty)$, and the 3D Kondo lattice model, $(1, 0)$.

Remarkably, we find that the (1,2) and (1,3)-superlattices and the (1,4) and (1,5) superlattices exhibit similar magnetization curves. The same behavior can also be observed for larger distances between f -electron layers; (1,2*n*) and (1,2*n* + 1) superlattices exhibit similar magnetization curves. Thus, the quantum critical point in the f -electron superlattice changes stepwise. We elucidate this point below after presenting more data.

We find that even a single f -electron layer, (1,∞) in Fig. 2, which is embedded in a 3D metallic host, shows magnetism for $J/t < 1.6$. This system corresponds to a superlattice with infinitely many spacer layers.

As mentioned before, DMFT does not include nonlocal fluctuations. It can be expected that the inclusion of nonlocal fluctuations will further reduce this critical value. However, a finite critical coupling strength, $J/t > 0$, even for a single f -electron layer embedded in a 3D metallic host is consistent with studies of (isolated) two-dimensional Kondo lattices including nonlocal fluctuations [19,20], which indeed show a finite critical coupling strength.

Different superlattices interpolate between the 3D Kondo lattice model and a single f -electron layer within a 3D metallic host. We note that a two-dimensional Kondo lattice, without any coupled metallic layers, exhibits a different magnetization curve with a larger maximal magnetization than the (1,∞) superlattice and cannot be directly compared to a superlattice.

The coupling between the f -electron layer and the metallic spacer layers induces a magnetization into the spacer layers, which we show in Fig. 3. Due to a Fermi-surface nesting of (π, π) , the sign of the magnetization oscillates from layer to layer. Thus, depending on the distance between f -electron layers, these layers are coupled either ferromagnetically or antiferromagnetically, which has also been found in a weak-coupling study of the periodic Anderson model for superlattices [21].

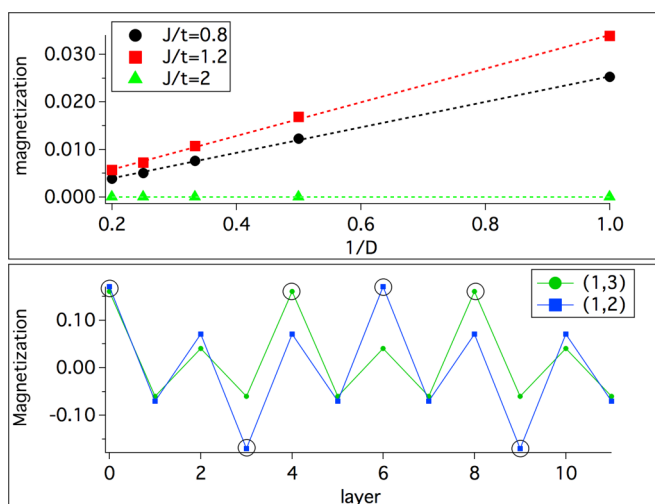


FIG. 3. Top: Local magnetization of the conduction electrons of an f -electron layer embedded in a 3D metal, (1,∞) superlattice, for different distances from the f -electron layer. Bottom: Magnetization of the conduction electrons plotted against the layer for (1,3) and (1,2) superlattices and $J/t = 1.6$. The f -electron layers of both superlattices are shown by circles.

Naturally, the magnetization in a spacer layer decreases with increasing distance to the f -electron layer. The magnetism in the superlattice is purely due to the interaction of the localized spins and conduction electrons in the f -electron layer. We show the absolute value of the magnetization of the conduction electrons as a function of the distance of an f -electron layer in the top panel in Fig. 3. We see that the induced magnetization behaves as $|n_{\uparrow} - n_{\downarrow}| \sim 1/D$, where D is the distance from the conduction electron to the f -electron layer. We note that these data could also be fitted by different power laws, because the DMFT data include only short distances. However, an analysis of the electron susceptibility and a test calculation using 100 layers predict behavior as $1/D$. The prefactor of the $1/D$ law depends therefore on the coupling between the conduction electrons and the magnetic moments; for weak coupling the prefactor first increases, while for strong coupling the prefactor decreases and finally vanishes when the whole superlattice becomes nonmagnetic. In the bottom panel in Fig. 3, we show the magnetization of the conduction electrons in different layers in the (1,2) and the (1,3) superlattices. We have marked the f -electron layers in both superlattices with circles. We observe that in the (1,2) superlattice f -electron layers are coupled antiferromagnetically, while in the (1,3) superlattice they are coupled ferromagnetically. As mentioned above the magnetization curves of the (1,2) and (1,3) superlattices lie very close together. In Fig. 3, we see that the magnetization is similar not only in the f -electron layer, but also in the next layer (see, e.g., layer 1).

IV. INTERLAYER MAGNETIC COUPLING

The induced magnetization leads to a magnetic coupling between different f -electron layers, which corresponds to the RKKY interaction between different magnetic layers. Such a magnetic coupling between different layers is well known from magnetic heterostructures. While in the general case the magnetic interlayer coupling behaves as $1/D^2$, it has been shown that for a nested Fermi surface, as in our model, it behaves as $1/D$ [22]. In an f -electron superlattice, however, this magnetic interlayer coupling is modified by the Kondo effect. Such a competition is absent in ordinary magnetic heterostructures without f electrons. We, therefore, analyze the influence of the Kondo effect on this interlayer magnetic coupling.

To determine the interlayer coupling we prepare the following setup:

- (1) We start with a fully converged magnetic solution of an f -electron superlattice.
- (2) We select a single f -electron layer and set the self-energies of this layer to 0.
- (3) For all other layers, we use the self-energies of the converged solution.
- (4) We apply a staggered magnetic field to the selected f -electron layer, which points opposite to the original magnetization of the layer.

We perform DMFT calculations for this setup and study the magnetization of the selected layer as a function of the magnetic field. The self-energy of the selected layer is thereby updated during the DMFT iterations and calculated

self-consistently. In these calculations, there are two competing effects. On the one hand, due to the magnetic interlayer coupling, the f -electron layers of the superlattice without an applied magnetic field try to restore the original solution. On the other hand, the magnetic field has a tendency to stabilize a solution where the magnetization of the selected layer is flipped. Thus, without an applied magnetic field the solution for this calculation will be identical to the input. Furthermore, if there is only the f -electron layer with an applied magnetic field, an infinitesimally small magnetic field will be sufficient to stabilize a solution with flipped magnetization. The strength of the magnetic field at which the magnetization flips corresponds to the magnetic coupling of the selected layer to all other f -electron layers in the superlattice.

We show the magnetic interlayer coupling, determined by the above procedure, in Fig. 4. The top panel shows the magnetic-field dependence of the magnetization for an (1,2) superlattice with coupling strength $J/t = 1.6$. For a weak magnetic field, we observe that the magnetization of the probed layer points in the same direction as in the initial solution. When the strength of the magnetic field is increased, the magnetization is reduced and flips at a critical strength of the magnetic field. This flipping of the magnetization is clearly visible as a jump. We take the value of this critical magnetic field strength as the magnetic interlayer coupling for this superlattice and interaction strength.

In the middle and bottom panels in Fig. 4, we show this interlayer coupling plotted against $1/D$, where D is the distance between the f -electron layers. Thus, the distance in a (1, M) superlattice is $D = M + 1$. We find that for weak coupling between the magnetic moments and the conduction electrons, $J/t = 1.2$, the magnetic interlayer coupling can be well described as $1/D$. This agrees with the behavior of the magnetization shown in Fig. 3. Thus, there is a long-range interlayer coupling between different f -electron

layers. Only for $D \rightarrow \infty$ does the interlayer coupling vanish completely. For a low coupling strength, J , the Kondo effect is exponentially weak and does not play an important role. All the calculations have been performed for a half-filled system so that the noninteracting Fermi surface is nested. Our results thus agree for weak coupling with the magnetic interlayer coupling of usual magnetic heterostructures.

For strong coupling (see bottom panel in Fig. 4) the determined interlayer coupling cannot be fitted as $1/D$. The magnetic interlayer coupling clearly deviates from $1/D$ behavior and vanishes already for $1/D > 0$, where the superlattice becomes nonmagnetic. This results from the competition between the RKKY interaction and the Kondo effect and is, thus, a characteristic of f -electron superlattices. The Kondo effect screens the magnetic moments arising from the interacting f electrons and, thus, affects the magnetic interlayer interaction. A remarkable effect can be observed for strong coupling; the magnetic interlayer couplings for $1/D = 0.2$ ($M = 3$) and $1/D = 0.25$ ($M = 2$) are nearly identical. The interlayer coupling vanishes pairwise for an increasing number of spacer layer. We have thus included separate least-squares fits (dashed lines) for an even number and an odd number of spacer layers in Fig. 4. Even for weak coupling we see that superlattices with even and odd numbers of spacer layers have slightly different least-squares fits. The difference between these two lines corresponds qualitatively to the strength of the Kondo effect in the superlattice.

The reason for this even-odd effect, i.e., pairwise vanishing of the interlayer coupling, is that the Kondo temperature and the RKKY interaction change, but the two effects cancel each other. Not only the RKKY interaction, but also the Kondo effect and the Kondo temperature depend on the superlattice [11]. The Kondo temperature shows even-odd oscillations in f -electron superlattices depending on whether the number of spacer layers is even or odd. This effect becomes important when analyzing the competition between the RKKY interaction and the Kondo effect. The Kondo effect becomes weaker when the number of spacer layers is changed from even to odd. Thus, the magnetic moments are less screened, which effectively increases the magnetic interlayer coupling. On the other hand, increasing the distance between the f -electron layers leads to a decrease in the interlayer coupling. As a result, the observed magnetic interlayer coupling, which takes into account both RKKY and Kondo effect, does not change. This explains the stepwise decrease in the interlayer coupling and the existence of two different superlattices with similar magnetization curves in Fig. 2.

Taking the results of the magnetic interlayer coupling, we can now understand the competition between the RKKY interaction and the Kondo effect in f -electron superlattices. We can distinguish different situations, which we qualitatively show in Fig. 5. The interlayer RKKY interaction behaves as $1/D$ in the superlattice. Furthermore, there is a magnetic intralayer coupling between the localized moments within the same layer, which does not depend on the structure of the superlattice. For weak coupling (see bottom panel in Fig. 5) the Kondo temperature is exponentially low and even the magnetic intralayer coupling is stronger than the Kondo effect. In this situation, we find a magnetic ground state independent of the number of spacer layers. Furthermore,

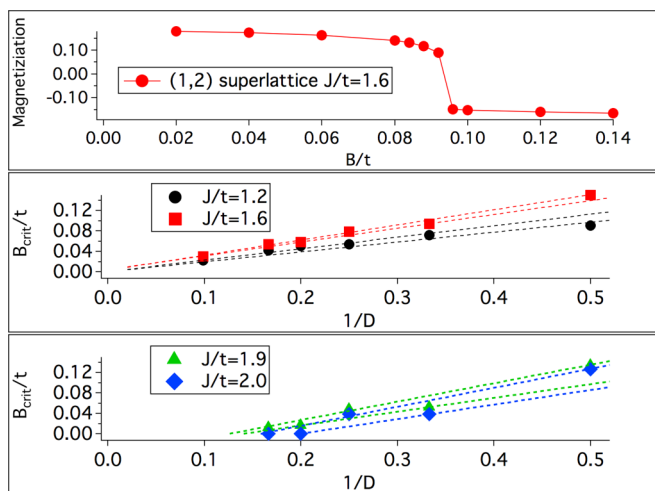


FIG. 4. Interlayer magnetic coupling calculated by applying a small magnetic field. Top: Magnetization of the chosen layer as a function of the magnetic field for a (1,2) superlattice with $J/t = 1.6$. Middle and bottom: Interlayer coupling strength plotted against $1/D$ for weak coupling and strong coupling, respectively.

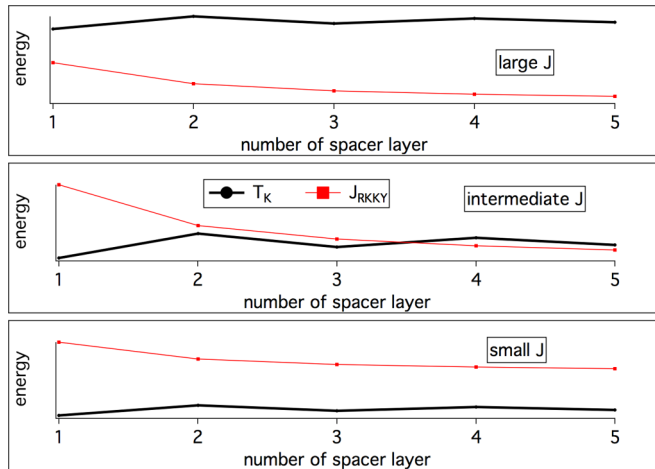


FIG. 5. Visualization of the competition between the Kondo effect and the RKKY interaction. We model the strength of the Kondo effect as an oscillating function in accordance with our previous study. The strength of the RKKY interaction decreases as $1/D$ with increasing distance. Top: For large J , the Kondo effect is stronger than the RKKY for any superlattice. Middle: For intermediate J , the RKKY interaction is stronger than the Kondo effect for small M but becomes weaker for large M . Bottom: For the weak-coupling J , the RKKY interaction is stronger than the Kondo effect for any superlattice.

because the Kondo effect is negligible, we observe the $1/D$ dependence of the interlayer coupling as for usual magnetic heterostructures. For strong coupling between magnetic moments and conduction electrons, as shown in the top panel in Fig. 5, the Kondo effect is stronger than the RKKY interaction in any superlattice. In this situation we can only find paramagnetic ground states. There is no magnetic interlayer coupling, because all magnetic moments are completely screened by the Kondo effect. The most interesting case occurs when the intralayer RKKY coupling is smaller than the Kondo temperature, but the sum of intralayer and interlayer RKKY interactions is larger (see middle panel in Fig. 5). Increasing the number of metallic spacer layers then leads to a reduction in the RKKY interaction, which finally becomes weaker than the Kondo effect and therefore leads to a vanishing of the magnetic order. For this intermediate coupling strength, the Kondo effect and the RKKY interaction strongly influence each other. The Kondo effect becomes slightly weaker when the number of spacer layers increases from even to odd. However, because at the same time also the strength of the RKKY interaction decreases, the two effects may cancel each other out.

V. SPECTRAL FUNCTIONS

Finally, we discuss the spectral functions in the ordered phase, shown in Fig. 6. In the top panel, we show the spectral functions of the conduction electrons in the f -electron layer for $J/t = 1.6$ and different superlattices. At this interaction strength, the magnetic order persists for the superlattices shown and the spectral functions of the conduction electrons form a gap at the Fermi energy, $\omega = 0$. This gap is partly due to the magnetic order and partly due to the Kondo effect which appears in this layer. Due to the absence of f electrons in the

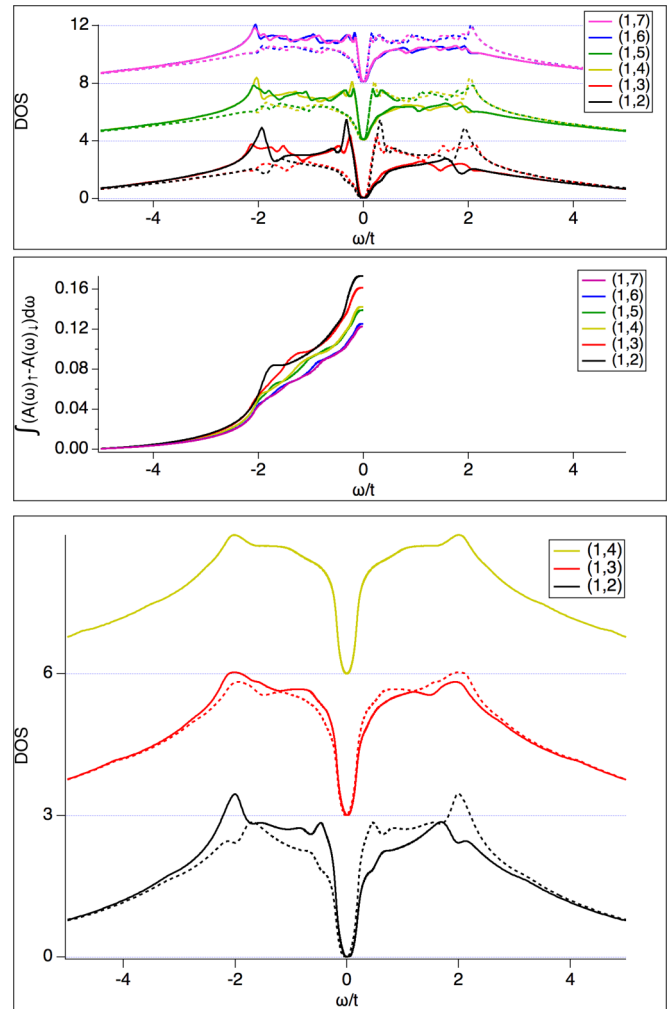


FIG. 6. Spectral functions of the conduction electrons in the f -electron layer in the ordered phase. Solid lines and dashed lines correspond to the majority and minority spin direction at the analyzed lattice site, respectively. Top: Spectral functions for six different superlattices and $J/t = 1.6$. (We have shifted the origin of the spectral functions of the upper curves for clarity.) We always show the spectral functions of two superlattices with similar magnetization together. Middle: Integrated spectral weight $\int (A_{\uparrow} - A_{\downarrow})$ for the spectral functions in the top panel. Bottom: Spectral functions for different superlattices and $J/t = 2$. For $M > 3$ the magnetization of the superlattice vanishes.

spacer layer, the spectral functions in the spacer layer remain metallic, although they are magnetically ordered. Besides the gap, the most prominent features in the spectral functions are a van Hove singularity close to the gap and an excitation at $\omega = 2t$. The strength of the van Hove singularity decreases with an increasing number of spacer layers M , which might be related to the decrease in the magnetization. In the middle panel we show $\int (A_{\uparrow}(\omega) - A_{\downarrow}(\omega))d\omega$, which corresponds to the magnetization, for the superlattices shown in the top panel. We observe again the stepwise change in the magnetization when the number of spacer layers is increased, i.e., for $M > 3$ these magnetizations are nearly equal for superlattices (1,4) and (1,5) as well as for (1,6) and (1,7). In the top panel we demonstrate that not only is the magnetization similar in these

superlattices, but also the spectral function. Excitations lie approximately at the same energies and have the same strength. In the bottom panel in Fig. 6, we show the spectral functions for $J/t = 2$. The magnetization vanishes when the number of spacer layers M is increased, although the coupling between localized moments and conduction electrons is constant.

VI. CONCLUSIONS

We have analyzed the magnetic order in f -electron superlattices and have demonstrated that the quantum critical point of the f -electron material can be tuned by changing the superlattice structure similarly to experimental results on $\text{CeIn}_3(n)/\text{LaIn}_3(4)$ superlattices [1]. We have focused in this study on the influence of the superlattice on the competition between the RKKY interaction and the Kondo effect. Besides the RKKY interaction and the Kondo effect also the reduced dimensionality of the f -electron material will become important, especially when the number of f -electron layers becomes small as in the experiments on $\text{CeIn}_3(n)/\text{LaIn}_3(4)$ superlattices. Increased magnetic fluctuations due to the reduced dimensionality can further reduce the critical coupling of the magnetic order. An analysis of the interplay of RKKY interaction, Kondo effect, and nonlocal fluctuations is left

for a future study using cluster extensions of the dynamical mean-field theory.

Furthermore, we have studied the competition of the Kondo effect and the RKKY interaction in superlattices. We have demonstrated that while the magnetic interlayer coupling between different f -electron layers behaves as $1/D$ for a weak Kondo effect, as in non- f -electron superlattices, it differs strongly from this behavior for a strong Kondo effect. In this case the magnetic interlayer coupling vanishes already for a finite number of spacer layers. Furthermore, due to the interplay between the Kondo effect and the RKKY interaction, the interlayer coupling vanishes stepwise when the number of spacer layers is increased. As a consequence, two superlattices with different numbers of spacer layers show similar magnetization curves.

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