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Colloquium: Heavy-electron quantum criticality and single-particle spectroscopy

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Heavy-electron quantum criticality and single-particle spectroscopy

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Angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy (STM) have become indispensable tools in the study of correlated quantum materials. Both probe complementary aspects of the single-particle excitation spectrum. Taken together, ARPES and STM have the potential to explore properties of the electronic Green function, a central object of many-body theory. In this article, we explicate this potential with a focus on heavy-electron quantum criticality, especially the role of Kondo destruction. We discuss how to probe the Kondo destruction effect across the quantum critical point using ARPES and STM measurements. We place particular emphasis on the question of how to distinguish between the signatures of the initial onset of hybridization-gap formation, which is the “high-energy” physics to be expected in all heavy-electron systems, and those of Kondo destruction, which characterizes the low-energy physics and, hence, the nature of quantum criticality. We survey

recent progress and possible challenges in the experimental investigations, compare the STM and ARPES spectra for several quantum critical heavy-electron compounds, and outline the prospects for further advances.

CONTENTS

I. Introduction	2
II. Quantum criticality	3
A. High-energy excitations, temperature evolution and mass enhancement	5
B. Isothermal evolution at low temperatures	6
C. Further considerations	6
D. Summary of Section II	6
III. ARPES, STM, and the single-particle Green function	6
A. The one-particle Green function	7
B. ARPES and STM	7
C. Probing quantum criticality in the Kondo lattice	8
D. Summary of Section III	9
IV. Quantum criticality in YbRh_2Si_2	9
Summary of Section IV	11
V. The Cerium-based 115 family: photoemission vs. tunneling spectroscopy	11
A. CeIrIn_5	11
B. CeCoIn_5	12
C. CeRhIn_5	14
D. Further considerations	15
E. Summary of Section V	16
VI. Progress, challenges and prospects	16
A. High energy Kondo features	16
B. Isothermal evolution at low temperatures	17
C. Outlook	17
VII. Conclusion	19
Acknowledgements	19
References	20

I. INTRODUCTION

A major objective of quantum materials research is to link observable properties to the nature of quantum mechanical many-body ground states properties and to the characteristics of the excitation spectrum above the ground state. In particular, it aims at understanding and predicting the emergence of novel phases in terms of a minimal set of variables, most notably symmetries and broken symmetries of the ground state and the ensuing classification of the excitation spectrum. The typical energy window commonly involved in the materials of interest can cover a wide range, from a few percent of a meV to several eV. A well-known example is the high-temperature superconductors, which have stimulated research since their discovery more than 30 years ago.

The quest for a unified understanding of different classes of quantum materials has led to the notion of quantum critical points (QCPs) as an economic and powerful way of organizing their phase diagrams (Coleman and Schofield, 2005; Kirchner *et al.*, 2013; Sachdev, 1999; Si and Steglich, 2010). Such continuous zero-temperature phase transitions not only separate different ground states but also give rise to a characteristic behavior; this is the quantum-critical fan, which can extend to comparatively large energies and temperatures, *cf.* Fig. 1. Within this fan, universal scaling behavior is expected up to some material-specific high-energy cutoff. Among the materials classes that are currently attracting particular interest are the cuprates, iron pnictides, pyrochlore iridates, transition metal dichalcogenides, and heavy-electron compounds. An underlying theme of most if not all these materials classes is the tendency of their charge carriers to localize in response to the large effective Coulomb repulsion experienced by the itinerant degrees of freedom. The tendency towards localization gives rise to the bad-metal behavior of these materials.

In heavy-electron compounds, which most commonly are based on Ce, Yb, and U, the primary degree of freedom is the f electron. In the lanthanide-based materials, the $4f$ electron is localized close to the ionic core as a result of atomic physics and thus has a characteristic energy of order eV. For the same reason, the wavefunction overlap between the $4f$ orbitals and the band (or c) electrons, *i.e.*, the hybridization, is typically small. As a result, the $4f$ electron appears localized at high temperatures or energies in the entire range of phase space as long as the valency of the lanthanide ion remains near its localized limit. In this regime each $4f$ electron contributes a finite amount $\sim \ln N_f$ to the entropy, where N_f is the angular momentum degeneracy. N_f is affected by spin-orbit coupling and the crystal electric fields but as long as $N_f > 1$, the spin entropy remains macroscopically large.

Similar arguments in principle apply to actinide-based heavy-electron compounds (Fisk *et al.*, 1985). In contrast to their $4f$ counterparts, $5f$ orbitals are substantially less localized. As a result, the associated heavy-electron bands are more dispersive, $f - c$ hybridization is stronger and crystal electric fields are less-well defined. Collectively, these properties frequently lead to more complex behaviors compared to Ce- or Yb-based intermetallics (Lawrence *et al.*, 2011), and so we will use the lanthanide-based heavy-electron materials as exemplary of the essential physics. As temperature is lowered and the ground state is approached, the spin entropy associated with the localized $4f$ ($5f$) electron needs to

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be quenched. Evidently, the system possesses several options for releasing this entropy, which lead to different ground states. At zero temperature, the system can transition from one ground state to another upon changing coupling constants in the Hamiltonian. At values of these coupling constants where the ground state energy is non-analytic, the system undergoes a quantum phase transition. Experiments, however, are performed at non-zero temperatures. The challenge, then, is how to distinguish the approach to different ground states with only a limited, intermediate temperature window accessible to experiment. This task is made even more difficult given that high-energy properties are largely insensitive to the changes in the coupling constants that take a system through different ground states.

The primary tools for exposing the underlying physics that accompanies the entropy release as the temperature or energy is lowered include spectroscopic methods that can trace excitations over some energy range of interest. For example, spin excitations can be probed with the help of inelastic neutron scattering. Among the various spectroscopic techniques, angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy/spectroscopy (STM) stand out as these allow to most directly trace properties of the one-particle Green function, the basic building block in almost every many-body theory.

We survey and compare recent ARPES and STM experiments performed on quantum critical heavy-electron compounds that are located close to ground state instabilities at the border of magnetism. In particular, we focus on how critical Kondo destruction (Coleman *et al.*, 2001; Si *et al.*, 2001), *i.e.* the breakdown of Kondo entanglement at zero temperature right at the onset of magnetism, is reflected in ARPES and STM data at elevated temperatures.

The Colloquium is organized as follows. After a brief introduction of quantum criticality in heavy-electron systems, we recapitulate the relation between APRES and STM measurements and their link with the single-particle Green function. We then discuss recent STM measurements on YbRh_2Si_2 , a heavy-electron antiferromagnet that features a Kondo-destruction QCP as a function of applied magnetic field, before turning to high-resolution ARPES measurements on the Cerium-115 family that consists of CeMIn_5 ($M=\text{Co,Rh,Ir}$). We close with an outlook on current challenges and future directions. To facilitate the reading, each of Sections II-V ends with a brief summary of the salient points discussed in the section.

II. QUANTUM CRITICALITY

Quantum phase transitions occur at zero temperature and like their finite temperature counterparts, they can

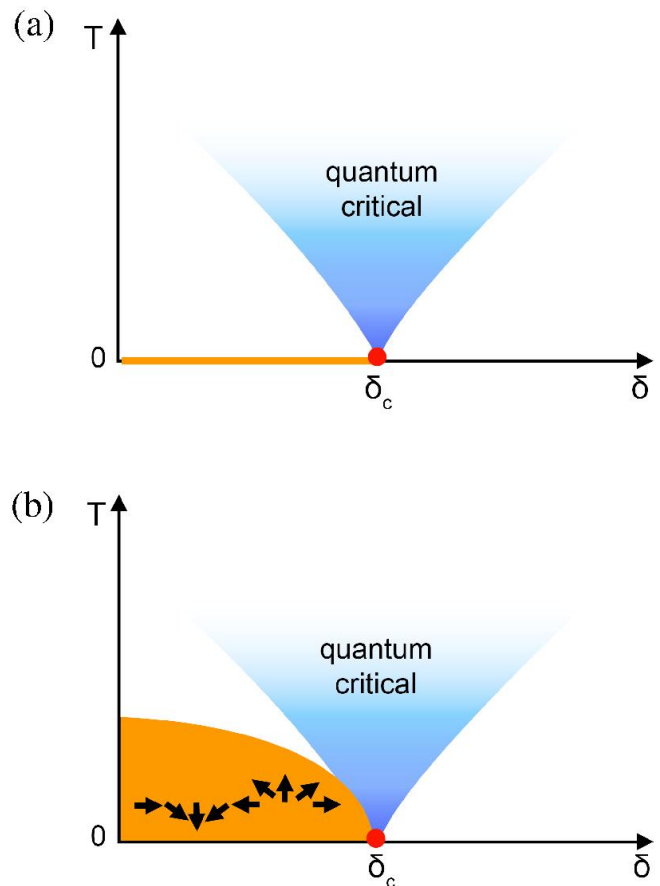


FIG. 1 The quantum critical fan: (Color online) (a) A quantum phase transition occurs at zero temperature for a critical value (δ_c) of a non-thermal tuning parameter δ . It separates the distinct behaviors of the ground state wavefunction, *i.e.*, being ordered for $\delta < \delta_c$ (indicated by the orange line) and disordered for $\delta > \delta_c$. At non-zero temperatures vestiges of the quantum phase transition lead to distinctive scaling behavior in a quantum critical fan that spreads out of the quantum critical point and extends up to a problem specific cutoff temperature. Unlike the scaling behavior, the existence of a fan of quantum critical behavior is generic, independent of the nature of the criticality. (b) Frequently, quantum phase transitions occur as order is suppressed and the transition temperature $T_c(\delta)$ of a classical phase transition vanishes, *i.e.*, $T_c(\delta \rightarrow \delta_c) \rightarrow 0$. In heavy-electron materials, the most common types of quantum criticality separate anti-ferromagnetic and paramagnetic phases. Shown here is a type of antiferromagnetic order, indicated by the variation of the magnetic moment density over one wavelength. This Colloquium explores the potential of single-electron spectroscopies to distinguish different types of quantum criticality.

be either first order or continuous (Gegenwart *et al.*, 2008; v. Löhneysen *et al.*, 2007; Sachdev, 1999; Si and Steglich, 2010). In contrast to the finite temperature case where thermal fluctuations drive the transition, quantum fluctuations, encoded already at the Hamiltonian level, are responsible for the occurrence of a quantum phase transition. A classical transition can be accessed by vary-

ing the temperature through a critical value T_c , while the zero-temperature transition is approached by tuning a non-thermal control parameter, denoted δ in Fig. 1, to its critical value (δ_c). If the transition is continuous, characteristic, critical scaling ensues in its vicinity which reflects the singular behavior of the ground state wavefunction at δ_c . At non-zero temperatures, this singular behavior leads to the quantum critical fan in which characteristic behavior is observed in various quantities below a system-specific cutoff energy, see Fig. 1.

In stoichiometric heavy-electron compounds containing, *e.g.*, Ce or Yb elements, $4f$ electrons in a partially-filled $4f$ shell are strongly correlated, provided the Ce or Yb ions possess a valence close to +III. The spin-orbit interaction and the crystal electric field generated by the ligands surrounding the Ce or Yb ion in the crystalline environment reduce the degeneracy of the $4f$ shell. Most commonly, the lowest-lying atomic $4f$ -levels correspond to a Kramers doublet. As a result, the $4f$ electrons behave as a lattice of effective spin-1/2 local moments. This leads to an effective description in terms of the Kondo lattice Hamiltonian:

$$H_{\text{KL}} = H_0 + \sum_{ij} I_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_i J_K \mathbf{S}_i \cdot \mathbf{s}_i^c, \quad (1)$$

where $H_0 = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}$ describes the conduction electrons. The RKKY interaction I_{ij} and the Kondo coupling J_K typically are antiferromagnetic, *i.e.*, $I_{ij} > 0$, $J_K > 0$. The competition between these two types of interactions lies at the heart of the microscopic physics for heavy-electron systems (Doniach, 1977).

In the heavy-electron compounds described by the Kondo lattice Hamiltonian, Eq. (1), a QCP may arise from tuning the ratio of RKKY to Kondo interactions, which is parameterized by the non-thermal control parameter $\delta \equiv T_K^0/I$. Here, the Kondo scale (for the $N_f = 2$ case) is $T_K^0 \approx \rho_0^{-1} \exp(-1/\rho_0 J_K)$, with ρ_0 being the density of states of the conduction electrons at the Fermi energy, whereas I parameterizes the RKKY interaction. This RKKY exchange interaction between the localized moments is mediated by the conduction electron spin density. It is perturbatively generated from the Kondo coupling term $\sim J_K$, resulting in $I_{ij}(J_K)$. In Eq. (1), we have added I_{ij} as an independent exchange interaction to facilitates the discussion of the phase diagram, because tuning the ratio between the explicit I_{ij} and J_K is more convenient. It accesses the quantum phase transition that otherwise would have been induced in the tuning of δ through the variation of the ratio of J_K to the conduction electron bandwidth $2D \sim 1/\rho_0$. Formally, one may think of Eq.(1) as arising from a more complete starting Hamiltonian through the process of integrating out additional conduction electron degrees of freedom; this procedure results in the explicit I_{ij} term in the effective Hamiltonian of Eq.(1). One needs to be sure that there is no double counting of the explicit and generated

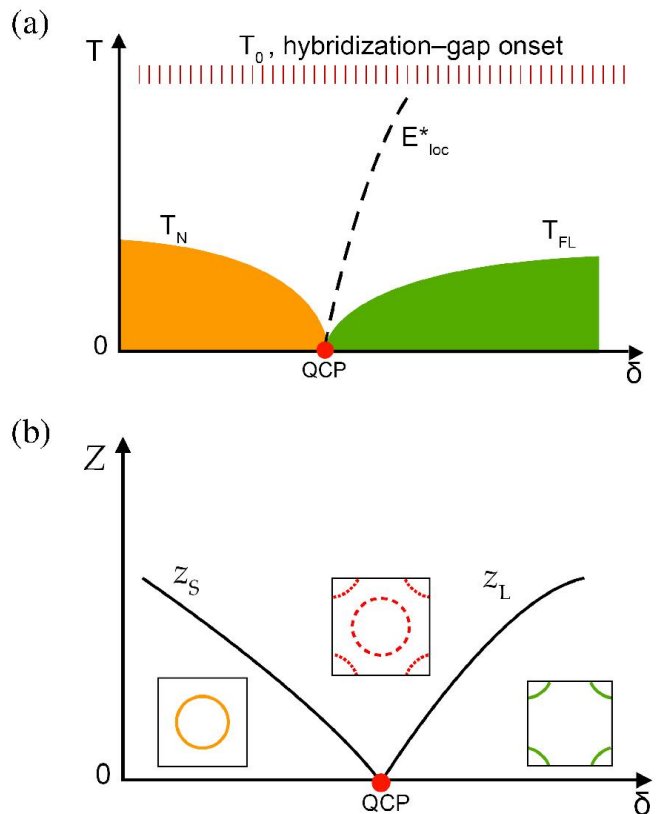


FIG. 2 Illustrations of basic concepts of Kondo-destruction quantum criticality: (Color online) (a) Local quantum criticality with Kondo destruction, under the variation of the control parameter δ . Here, T_0 is a high-energy scale that describes the initial onset of dynamical Kondo correlations and that smoothly evolves across the QCP δ_c . This high-energy scale is reflected in the onset of hybridization-gap formation. The low-energy physics is described in terms of T_N and T_{FL} , which are respectively the temperatures for the Néel transition and the crossover into the paramagnetic Fermi liquid state. This phase diagram also involves the Kondo-destruction energy scale E_{loc}^* , which characterizes the Kondo destruction. The E_{loc}^* line divides the phase diagram in terms of the flow of the system towards either the Kondo-screened or the Kondo-destruction ground state. In the conventional model of spin-density wave quantum criticality, the line E_{loc}^* extrapolates to zero temperature in the ordered phase so that the Fermi surface already is large before reaching the QCP with increasing δ and evolves smoothly across the QCP (Coleman *et al.*, 2001; Si *et al.*, 2001). (Adapted from Ref. (Stockert *et al.*, 2012)). (b): The small (left) and large (right) Fermi surfaces, and the associated quasiparticle weights z_S and z_L that are discussed in Section III. The fluctuating Fermi surfaces (middle) are associated with the QCP. (Sketch adapted from (Pfau *et al.*, 2012))

contributions to I_{ij} , and this can be consistently done in practice. For a technical discussion of this point we refer the reader to Ref. (Si *et al.*, 2005).

On the paramagnetic side, the ground state is characterized by the amplitude of the *static* Kondo singlets that

are formed between the local moments and conduction electron spins (Hewson, 1993). For a Kondo-destruction QCP, this static Kondo-singlet amplitude is continuously suppressed when the system approaches the QCP from the paramagnetic side (Si *et al.*, 2014, 2001; Zhu *et al.*, 2003).

As illustrated in Fig. 2(a), the Kondo-destruction energy scale E_{loc}^* goes to zero as the control parameter δ approaches the QCP at δ_c from the paramagnetic side, and the antiferromagnetic order sets in when δ goes across δ_c . The Kondo destruction goes beyond the Landau framework of quantum criticality. The latter is based on order-parameter fluctuations, which in the present context of antiferromagnetic heavy-electron systems is referred to as a spin-density-wave (SDW) QCP (Hertz, 1976; Millis, 1993; Moriya, 1985). It arises when E_{loc}^* stays non-zero when decreasing δ to δ_c , and approaches zero only inside the ordered regime at $\delta < \delta_c$. In this case, the asymptotic quantum critical behavior at energies below $E_{loc}^*(\delta_c)$ is the same as in the type of phase diagram shown in Fig. 1(b), where E_{loc}^* is not part of the critical physics.

The Kondo destruction gives rise to a dynamical spin susceptibility which displays unusual scaling at the QCP (Si *et al.*, 2014, 2001). This includes a fractional exponent (Glossop and Ingersent, 2007; Grepel and Si, 2003; Zhu *et al.*, 2003, 2007) in the singular dependence on frequency (ω) and temperature (T), and ω/T scaling. These features have in fact been observed by inelastic neutron scattering measurements on the $5f$ electron system $UCu_{5-x}Pd_x$ (Aronson *et al.*, 1995) and the $4f$ electron-based metal $CeCu_{6-x}Au_x$ (Schröder *et al.*, 2000).

For $CeCu_{6-x}Au_x$ at its critical doping $x_c \approx 0.1$, the exponent in the ω/T scaling analysis (Schröder *et al.*, 2000) was found to be $\alpha = 0.75(5)$, which compares well with the value $\alpha = 0.72-0.78$ calculated at the Kondo-destruction QCP (Glossop and Ingersent, 2007; Grepel and Si, 2003; Zhu *et al.*, 2003, 2007). In the case of a standard SDW QCP, no such ω/T scaling is expected as this QCP is described by a Ginzburg-Landau functional above its upper critical dimension (Hertz, 1976; Millis, 1993; Moriya, 1985).

In the single-particle excitations, the collapse of E_{loc}^* implies a sudden reconstruction of the Fermi surface across the QCP. To contrast this picture with the more traditional scenario of an SDW transition (Hertz, 1976; Millis, 1993; Moriya, 1985), where critical fluctuations are tied to nesting properties of the Fermi surface, we will refer to quantum criticality exhibiting critical Kondo destruction as *local quantum criticality* (Coleman *et al.*, 2001; Pépin, 2007; Senthil *et al.*, 2004; Si *et al.*, 2001). At zero temperature,

- for $\delta > \delta_c$, the Fermi surface is large and is given by the combination of the $4f$ and conduction electrons. A non-zero amplitude of the static Kondo

singlet will be referred to as defining a *Kondo-screened* ground state. It produces a Kondo resonance, which reflects electronic excitations produced by entanglement of the $4f$ -moments with the conduction electrons. The Kondo effect is responsible for the large mass enhancement and a small quasiparticle weight z_L [Fig. 2(b)]. There is a small gap for the single-particle excitations at the small Fermi surface.

- for $\delta < \delta_c$, the Fermi surface is small as determined by the conduction electrons alone. This is because, when the amplitude of the static Kondo singlet vanishes, there is no longer a well-defined Kondo resonance. We refer to this state as a *Kondo-destruction* ground state.
- at the QCP, single-particle excitations are gapless and have a non-Fermi-liquid form, both at small and large Fermi surfaces.

A. High-energy excitations, temperature evolution and mass enhancement

Figure 2(a) also contains a high-energy scale T_0 which describes the *initial onset* of dynamical Kondo correlations. This scale is generally affected by the presence of higher crystal electric field doublets (or quartets) that together form the $4f$ multiplet (Chen *et al.*, 2017; Cornut and Coqblin, 1972; Kroha *et al.*, 2003; Pal *et al.*, 2019). It is important to note that this scale smoothly evolves across the QCP at δ_c . The development of the hybridization gap is associated with the initial onset of dynamical Kondo correlations, as illustrated in Fig. 3, and will appear on both sides of δ_c .

For $\delta > \delta_c$, the temperature evolution of the physical properties reflects the flow of the system towards the Kondo-screened ground state. For instance, the initial onset of dynamical Kondo correlations results in the Kondo-screened ground state; the single-particle excitations develop into fully coherent heavy-quasiparticles at the large Fermi surface as the temperature is lowered below T_{FL} , the crossover temperature into the paramagnetic Fermi-liquid state.

For $\delta < \delta_c$, the initial onset of dynamical Kondo correlations still takes place, even though it does, in the end, *not* lead to a well-defined Kondo resonance and the Kondo-singlet amplitude vanishes in the ground state. Still, as the temperature is further lowered, vestiges of the Kondo effect will be observed at any non-zero temperature. In particular, the effective mass is a dynamical quantity, measuring the dispersion of the Landau quasiparticles, and is enhanced through the dynamical Kondo effect; further discussions of this point can be found in Refs. (Cai *et al.*, 2019; Si *et al.*, 2014).

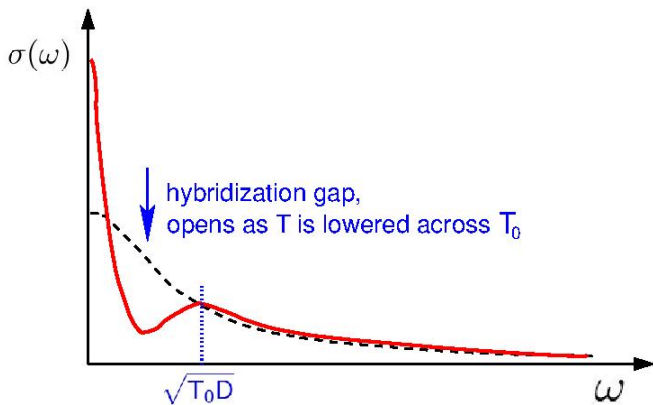


FIG. 3 Sketch of the optical conductivity $\sigma(\omega)$ for temperatures well above (black dashed line) and well below (continuous red line) the crossover temperature T_0 : (Color online) Here, the lowering of temperature through T_0 is accompanied by the onset of the hybridization gap. The characteristic frequency scale for the hybridization gap is $\sqrt{T_0 D}$ (marked by the blue vertical dotted line), where D is an energy scale of the order of the conduction electron bandwidth. At low energies, *i.e.*, for $\omega \ll T_0$ and $T \ll T_0$, and sufficiently far away from quantum criticality (*i.e.* $\delta < \delta_c$ or $\delta > \delta_c$), a pronounced Drude peak reflects the mass enhancement in the Fermi liquid regimes that surround the QCP in the phase diagram [Fig. 2(a)]. The behavior of $\sigma(\omega)$ at high energies, including the hybridization gap, is a generic feature of heavy-electron systems and is seen throughout the high-energy part of the phase diagram of Fig. 2(a).

B. Isothermal evolution at low temperatures

The distinction between the two sides of δ_c can be sharply made at low temperatures, where well-defined quasiparticles reside at the small Fermi surface for $\delta < \delta_c$ and at the large Fermi surface for $\delta > \delta_c$. At zero temperature, the distinction appears as a sudden reconstruction of the Fermi surface as δ passes through δ_c . At non-zero but low temperatures, this becomes a crossover. The crossover width increases with increasing temperature. When the crossover width becomes large, the difference between the two sides becomes ambiguous. We will illustrate this point below, especially through the experiments carried out on YbRh_2Si_2 .

C. Further considerations

In the Kondo-destruction description, the *static* Kondo effect is suppressed in the antiferromagnetic phase at $\delta < \delta_c$. However, dynamical Kondo-singlet correlations remain at non-zero frequencies in this regime. They lead to the development of $4f$ -electron spectral weight near the Fermi energy, which we will refer to as Kondo-resonance-like features. The dynamical Kondo effect (see Ref. (Cai *et al.*, 2019), as well as earlier discussions in Refs. (Si *et al.*, 2014; Zhu *et al.*, 2003)) still produces a

large mass enhancement and a small quasiparticle weight z_S [Fig. 2(b)]. There is a small gap for the single-particle excitations at the large Fermi surface.

The inelastic neutron scattering result on $\text{CeCu}_{6-x}\text{Au}_x$ ($x_c \approx 0.1$) (Schröder *et al.*, 2000) has been confirmed by the recent inelastic neutron scattering measurements (Poudel *et al.*, 2019) in $\text{CeCu}_{6-x}\text{Ag}_x$ ($x_c \approx 0.2$). When analyzing the data in terms of the one-component form, as arising in the Kondo-destruction description, Ref. Poudel *et al.*, 2019 found a similar form of ω/T scaling with a similar value for the critical exponent, $\alpha = 0.73(1)$. Ref. Poudel *et al.*, 2019 also analyzed the data in terms of a multicomponent spin fluctuation spectrum, with one of the (weaker) components conforming to the expectation of an SDW QCP. However, thermodynamic singularities have provided evidence for the one-component description (Grube *et al.*, 2017).

D. Summary of Section II

For heavy-electron metals, the Landau form of quantum criticality corresponds to an SDW QCP. A new type of quantum criticality has been advanced, in the form of a Kondo-destruction (local) QCP. It goes beyond the Landau framework in that, the critical destruction of Kondo entanglement characterizes the physics beyond the slow fluctuations of the magnetic order parameter. The Kondo destruction is characterized by a new energy scale, E_{loc}^* , vanishing at the QCP, as illustrated in Fig. 2(a); a sudden reconstruction from large to small Fermi surface as the system is tuned from the paramagnetic side through the QCP, along with a vanishing quasiparticle weight on approach of the QCP from both sides, as illustrated in Fig. 2(b).

III. ARPES, STM, AND THE SINGLE-PARTICLE GREEN FUNCTION

The unusual ω/T scaling of the dynamical spin susceptibility sets apart the QCP featuring critical Kondo destruction from the more traditional QCP based on the Landau framework of order-parameter fluctuations. It means that the dynamical susceptibility $\chi(\omega, T)$, in the regime where scaling operates, can be scaled to depend on ω or T only through the combination ω/T . Such scaling has been observed in $\text{CeCu}_{6-x}\text{Au}_x$ at its antiferromagnetic QCP (Schröder *et al.*, 1998) and indicated for YbRh_2Si_2 (Friedemann *et al.*, 2010). Recent measurements of the optical conductivity in thin films of YbRh_2Si_2 have demonstrated a singular response in the charge sector with an ω/T scaling (Prochaska *et al.*, 2020). A scaling form of this kind for both the optical conductivity and dynamical spin susceptibility is strongly suggestive of the presence of ω/T scaling in the single-

particle excitations encoded in the one-particle Green function.

A. The one-particle Green function

This Green function can quite generally be cast into the form

$$G(\omega, \mathbf{k}, T) = \frac{1}{\omega - \varepsilon_{\mathbf{k}} - \Sigma(\omega, \mathbf{k}, T)}, \quad (2)$$

where $\varepsilon_{\mathbf{k}}$ is the bare electron dispersion and the proper selfenergy $\Sigma(\omega, \mathbf{k}, T)$ encodes the effects of electron-electron interaction. In a Fermi liquid, this function can be decomposed into two parts,

$$G(\omega, \mathbf{k}, T) = G_{\text{coh}}(\omega, \mathbf{k}, T) + G_{\text{incoh}}(\omega, \mathbf{k}, T), \quad (3)$$

where the incoherent part is non-singular close to the Fermi surface while the coherent part G_{coh} near E_F describes the quasiparticle contribution and assumes the form

$$G_{\text{coh}}(\omega, \mathbf{k}, T) = \frac{z}{\omega - v_F(k - k_F) + i\Gamma(\omega, T)}, \quad (4)$$

where z is the quasiparticle weight and v_F is the Fermi velocity. The lifetime of a quasiparticle is given by the inverse of the decay rate Γ . The amplitude of the static Kondo screening discussed above is related to a pole in the selfenergy, which in the Fermi liquid regime can at sufficiently low ω and T be written

$$\Sigma(\omega, \mathbf{k}, T) = \frac{a}{\omega - b} + \delta\Sigma(\omega, \mathbf{k}, T), \quad (5)$$

where a and b are parameters that capture the strength of Kondo screening and energy of the Kondo resonance, respectively. The pole in Σ shifts the Fermi momentum from its initial, 'small' value to a new, 'large' value as long as $a \neq 0$. In contrast to the SDW QCP case, where $a \neq 0$ on either side of the critical point, Kondo screening is critically destroyed at the local QCP and $a = 0$ in the antiferromagnetic side with a Fermi surface that is determined by the conduction electrons alone.

The Hall effect turns out to be a particularly useful quantity in this context as it is a measure of the carrier density on either side of the QCP. This is a consequence of the Fermi liquid nature of the two phases separated by the QCP, in which the Hall coefficient is completely determined by the renormalized dispersion of the single-electron excitations, to the leading order of elastic scattering (quenched disorder) when it is nearly isotropic, and is independent of the quasiparticle weight z or any Landau parameters, regardless of the strength of electron-electron (and electron-phonon) interactions. This can be seen through the kinetic equations of a Fermi liquid, or using the Kubo formalism (Betbeder-Matibet and Nozières, 1966; Kohno and Yamada, 1988)

and related Feynman-diagrammatic means (Khodas and Finkel'stein, 2003).

The dynamical spin susceptibility $\chi(\omega, \mathbf{k}, T)$ and also the optical conductivity $\sigma(\omega, T)$ can be written as convolutions of the Green function with itself and specific vertex functions. On the other hand, ARPES and STM measurements depend directly on $G(\omega, \mathbf{k}, T)$. Single-electron spectroscopies are thus, at least in principle, particularly useful in distinguishing between the two types of quantum criticality.

B. ARPES and STM

ARPES and STM measurements probe the single-particle spectrum and thus give access to the spectral function $2\pi A(E, \mathbf{k}) = -\text{Im}G(\omega = E + i\delta, \mathbf{k}, T)$. Although the single-particle Green function appears in the theoretical description of both ARPES and STM, both spectroscopic techniques are complementary. While ARPES directly probes the single-particle excitations as a function of energy and momentum, STM measures a conductance that is local in real space. Both methods are surface sensitive, albeit to different degrees. Furthermore, through variation of the photon energy, the bulk sensitivity of ARPES can be enhanced. By construction, ARPES only probes the occupied part of the single-particle excitation spectrum, which, especially at low temperatures, leads to a sharp cutoff at the Fermi energy (Hüfner, 2003). ARPES therefore measures only part of the full spectral function, *i.e.* the imaginary part of the retarded Green function below the Fermi energy. A sketch of the spectral function of a Fermi liquid is shown in Fig. 4 (a). It consists of contributions from the quasiparticle pole and an incoherent background. The quasiparticle pole contributes a factor z to the total area beneath the spectral function, while the incoherent background contributes $(1 - z)$ times the total area. z is commonly called the wave-function renormalization factor and it is inversely proportional to the quasiparticle mass in a Fermi liquid. The evolution of z with tuning parameter is plotted schematically in Fig. 2(b) where we see that z vanishes at a local QCP. The position of the quasiparticle pole as a function of momentum defines the dispersion. *Per se*, ARPES is not able to distinguish between the quasiparticle peak and the incoherent part of the spectral function. Provided the energy and momentum resolution is not a limiting factor, however, the characteristic broadening $\delta\epsilon$ of the quasiparticle peak in energy ($\sim |\epsilon_{\mathbf{k}} - E_F|^2$) and with temperature ($\sim T^2$) should be discernible in the momentum distribution curves provided by ARPES. The weight of the quasiparticle peak, in principle, could also be extracted based on the total incoherent part. However, since ARPES only probes occupied states, the complete spectral function is inaccessible. Although, inverse photoemission is in principle able to probe states above

the Fermi energy, it is haunted by rather poor energy resolution.

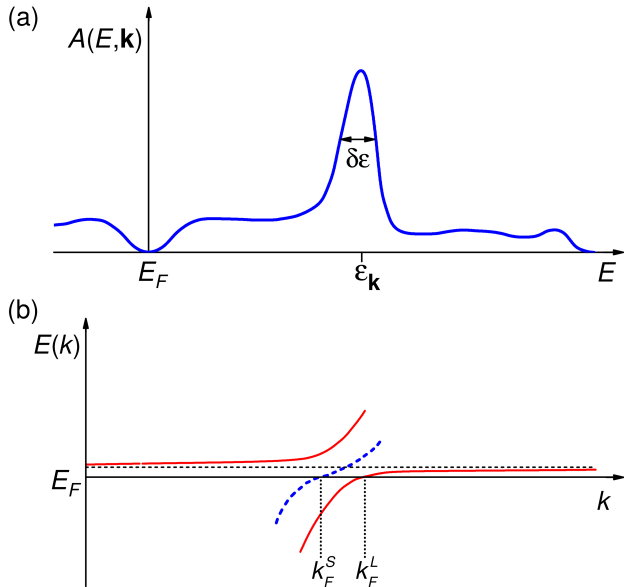


FIG. 4 Electronic characteristics of the Fermi liquid state of a Kondo lattice: (Color online) (a) Spectral density of a Fermi liquid: The quasiparticle pole at $\epsilon_{\mathbf{k}}$ has a characteristic width $\delta\epsilon$ that increases with the distance from the Fermi energy E_F as $\delta\epsilon \sim |\epsilon_{\mathbf{k}} - E_F|^2$. A similar broadening occurs due to finite-temperature effects. The incoherent part of $A(E, K)$ vanishes at E_F . (b) Quasiparticle dispersion in the Fermi liquid to either side of the QCP: k_F^L and k_F^S refer to large and small Fermi surfaces, respectively. Across a Kondo-destruction QCP, the one-electron spectrum is gapless at k_F^L and develops a small gap at k_F^S for $\delta > \delta_c$, and the converse is valid for $\delta < \delta_c$. The flattening of the dispersion near k_F^S for $\delta < \delta_c$ (dashed blue curve) reflects the effective mass enhancement due to the dynamical Kondo effect.

In general, when interpreting ARPES spectra, one needs to keep in mind that in order to relate the photoemission intensity to the spectral function, the one-electron dipole matrix element enters, which generally is unknown. In addition, k_z broadening can be important, where k_z is the component of the electron momentum perpendicular to the surface, and depends on the photon energy (Strocov, 2003; Wadati *et al.*, 2006).

STM, on the other hand, measures a local-in-real-space conductance. In the linear-response regime, the current-voltage characteristics is related to the local density of states (DOS) of the material under investigation (Bardeen, 1961; Tersoff and Hamann, 1985). Therefore, at low bias voltage and temperature, the spatially resolved spectral density can be obtained. As the applied bias voltage shifts the chemical potential at which the local density of states is probed, STM is, unlike ARPES, not confined to only occupied states. It is, however, important to realize that the assumption that the spectral function is independent of the bias voltage has to break

down at some sample-dependent value of the bias voltage beyond which the tunneling current can no longer be related to the local density of states. Moreover, the properties of the STM tip, *e.g.* its DOS, may affect the results.

C. Probing quantum criticality in the Kondo lattice

One of the strongest diagnostic tools to distinguish the local QCP from the SDW QCP are Hall conductivity measurements, as the local QCP manifests itself by a jump of the Hall coefficient across the QCP. This is a consequence of the Hall coefficient being inversely proportional to the carrier density (in the isotropic case or, in general, the curvatures of the quasiparticle dispersion on the Fermi surface) while being independent of the quasiparticle weight z in a Fermi liquid. The continuous nature of the local QCP is ensured by the vanishing of the quasiparticle weight from either side of the transition. For an SDW QCP, on the other hand, z will remain non-zero (except at isolated points on the Fermi surface) as δ is tuned through δ_c .

In a Kondo lattice at sufficiently high temperatures, where in first approximation the effect of the RKKY interaction can be ignored, the single-impurity Anderson model is expected to capture the overall physical behavior. This model is given by

$$H_{\text{AND}} = \sum_{\sigma} \epsilon f_{\sigma}^{\dagger} f_{\sigma} + \sum_{\mathbf{k}, \sigma} \epsilon_{\mathbf{k}} c_{\mathbf{k}, \sigma}^{\dagger} c_{\mathbf{k}, \sigma} + \frac{U}{2} \sum_{\sigma \neq \sigma'} f_{\sigma}^{\dagger} f_{\sigma'}^{\dagger} f_{\sigma'} f_{\sigma} + \sum_{\mathbf{k}, \sigma} \left(V_{\mathbf{k}} f_{\sigma}^{\dagger} c_{\mathbf{k}, \sigma} + \text{h.c.} \right) \quad (6)$$

where f_{σ}^{\dagger} (f_{σ}) is the set of local $4f$ electron creation (destruction) operators of spin projection σ . The conduction electron operators are c_{σ}^{\dagger} and c_{σ} . The band structure of the conduction electrons is encoded in $\epsilon_{\mathbf{k}}$, and the matrix element $V_{\mathbf{k}}$ that mixes $4f$ and c electrons is referred to as the hybridization. (For the case of the periodic Anderson model in the local moment limit, with the $4f$ electron occupancy being close to unity, it reduces to the Kondo-lattice model given in Eq. (1) when the charge degrees of freedom of the $4f$ electrons are projected out (Schrieffer and Wolff, 1966; Zamani *et al.*, 2016a).)

STM spectra of single-site Kondo problems possess the structure of Fano resonances (Fano, 1961) and depend on the ratio of tunneling into the Kondo impurity vs tunneling into the embedding host. This ratio is encoded in the so-called Fano parameter. Rigorous derivations of the tunneling current and the form of the Fano parameter are given in (Plihal and Gradzuk, 2001; Schiller and Hershfield, 2000; Újsághy *et al.*, 2000). If tunneling occurs predominantly into the conduction band the measured local DOS features the suppression of conduction electron states near the Fermi energy as the Kondo

effect develops. The first scanning tunneling studies of dense Kondo systems appeared about a decade ago (Aynajian *et al.*, 2010; Ernst *et al.*, 2010; Schmidt *et al.*, 2010). The pronounced variation of STM spectra with the type of surface for Kondo lattice compounds is largely due to variations in the Fano parameter, see *e.g.* Fig. 10 for tunneling into differently terminated surfaces. This has been explicitly demonstrated based on mean field and dynamical mean field theory approximations for the Kondo lattice (Benlagra *et al.*, 2011; Figgins and Morr, 2010; Maltseva *et al.*, 2009; Wölfle *et al.*, 2010). At sufficiently high temperatures, however, STM spectra in the vicinity of each Ce moment are expected to be similar to those for the single-ion Kondo case. Kondo screening is a predominantly local phenomenon and thus its onset and evolution are easily probed in real space, *i.e.*, via STM. For a study of the single-particle Green function in the paramagnetic Fermi liquid regime of the Kondo lattice far away from any QCP, in the context of photoemission, see Refs. (Costi and Manini, 2002; Reinert *et al.*, 2001). ARPES measurements at similar temperatures, around and above the energy scale T_0 , provide the band structure $\varepsilon_{\mathbf{k}}$ of the occupied conduction electron states. A flat band near the $4f$ electron atomic level ε (see Eq. (6)), which is far from the Fermi energy, and the formation of a flat band near the Fermi energy induced by the Kondo effect at each Ce moment, reflect the $4f$ electron spectral weight. This can be enhanced using resonant ARPES, see (Chen *et al.*, 2017).

At (sufficiently) low temperatures, in the Fermi liquid regime to either side of the QCP at δ_c [Fig. 2(a)], the band structure is shown in Fig. 4(b). For $\delta < \delta_c$, the small Fermi surface prevails and the band structure is that of the blue dashed line crossing the Fermi energy E_F at k_F^S . Still, incoherent spectral weight, a vestige of incomplete Kondo screening, develops but is ultimately gapped near k_F^L . For $\delta > \delta_c$, the Fermi surface incorporates the $4f$ moments and the Fermi wavevector changes from k_F^S at high temperatures (without the $4f$ moments) to k_F^L at low temperatures. On this side of the QCP, any spectral weight near k_F^S is due to incoherent single-particle excitations and is ultimately gapped. In other words, for $\delta > \delta_c$ in the Fermi liquid regime the spectral weight near the blue dashed line of Fig. 4(b) has developed a small gap at k_F^S . This should in principle be directly detectable via ARPES, provided the energy and momentum resolution is sufficiently high, and low enough temperatures can be reached.

On the other hand, the change k_F^S to k_F^L has only indirect vestiges in real space as the Fermi liquid is a momentum-space concept. The ensuing difficulties when tracing single-electron excitations in real space can already be read off from Fig. 4(b): The Fermi liquid is described by a low-energy effective theory and is valid only in the vicinity of k_F . (The spectral function is a more general concept but it only assumes a form similar to that

shown in Fig. 4(a) in the Fermi liquid regime.) Fourier transforming the momentum-resolved spectral function to real space necessarily will sum up spectral weight outside of the Fermi liquid regime, where the characteristic broadening that identifies the quasiparticle peak is no longer valid.

One possible way forward is to perform quasiparticle interference (QPI) experiments to map out the band structure near the Fermi energy (Derry *et al.*, 2015; Yazdani *et al.*, 2016). We will return to this possibility in Section VI. Another is to perform isothermal STM measurements at low temperatures through the phase diagram connecting $\delta < \delta_c$ with $\delta > \delta_c$. While this by itself does not provide any direct information on the size of the Fermi surface, it has been recently demonstrated that such a measurement is able to pick up the critical slowing down at the Kondo-destruction energy scale (Seiro *et al.*, 2018), as discussed in the next Section.

D. Summary of Section III

The nature of quantum criticality in heavy-electron metals is manifested in the evolution of the single-particle excitations across the QCP. It is natural to probe this behavior using the ARPES and STM spectroscopies, given that they are established means of studying single-particle excitations in metals. However, there is challenge to this task, mostly because heavy-electron systems have the distinction that the required energy scale is very low.

For ARPES, this requirement poses a challenge to access the quantum critical behavior, as is the limitation that even the state-of-the-art setups cannot yet reach temperatures below about 1 K. Still, ARPES should be informative in elucidating i) the onset of hybridization gap, which represents the high-energy physics for the quantum criticality of heavy-electron metals [see Fig. 2(a)]; and ii) the evolution of the dynamical Kondo effect as temperature is lowered towards either the antiferromagnetic/paramagnetic ground state or the quantum-critical regime.

STM spectroscopy has superb energy resolution and can reach low temperatures, but more demanding setups (such as those suited for QPI) are needed to access the information in the momentum space. Still, STM probes the single-particle physics in a way that is complementary to ARPES. In addition, it provides a promising means to probe the isothermal evolution of single-particle excitations at low temperatures, across the Kondo-destruction energy scale.

IV. QUANTUM CRITICALITY IN YbRh_2Si_2

YbRh_2Si_2 is a prototype system for local quantum criticality, as illustrated by its temperature (T)-magnetic

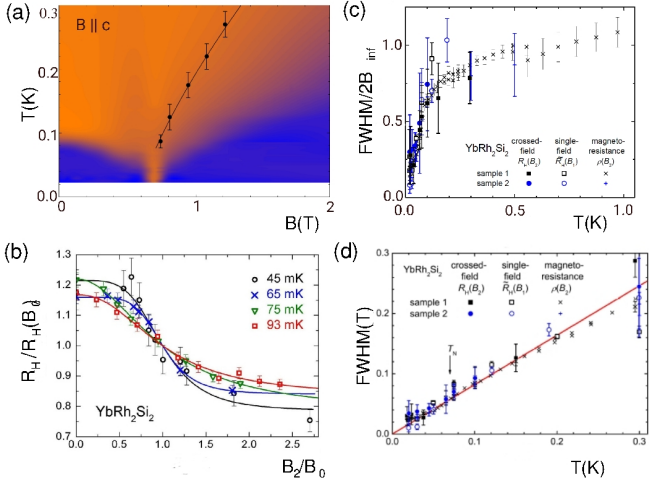


FIG. 5 Quantum criticality in YbRh_2Si_2 (Color online): (a) The temperature vs. field phase diagram of YbRh_2Si_2 from (Custers *et al.*, 2003). The blue regions marks Fermi liquid behavior, *i.e.*, $\rho(T) - \rho(0) \sim T^2$, while orange indicates the quantum critical area of the phase diagram where $\rho(T) - \rho(0) \sim T^x$, with $x \approx 1$. The continuous line in the quantum critical region is the E^* -line as derived from thermodynamic and transport properties. (b) Normalized Hall coefficient across the critical field for different temperatures. The inverse of R_H is a measure of the carrier density. The lower T , the sharper is the crossover. At $T = 0$ and $B = B_c$, a jump of R_H corresponds to the sudden localization of $4f$ electrons as B is taken trough B_c from above. From (Paschen *et al.*, 2004). (c) Comparison between the isothermal magnetotransport crossover width and the crossover field as specified by the ratio of the $\text{FWHM}/2$ to the crossover inflection field, B_{inf} . FWHM denotes the full width at half maximum. From (Paschen *et al.*, 2016). (d) The “sharpness” of the crossover: The FWHM vanishes in a linear-in- T fashion indicating a jump of R_H at B_c in the zero-temperature limit.

field (B) phase diagram [Fig. 5(a)]. Here, the Fermi surface jump and the Kondo-destruction energy scale have been extensively studied through magnetotransport and thermodynamic measurements. At a given temperature, the isothermal Hall coefficient [Fig. 5(b)] and other transport and thermodynamic quantities display a rapid crossover (Friedemann *et al.*, 2010; Gegenwart *et al.*, 2007; Paschen *et al.*, 2004).

From these measurements, a $E^*(B)$ line is thus specified in the phase diagram. This line relates to each T a B^* -scale: $B^*(T) \geq B_c$ with $B^*(T = 0) = B_c$. The full-width at half-maximum (FWHM) of the crossover [Fig. 5(b)] extrapolates to zero in the zero-temperature limit [Fig. 5(d)], which implicates a jump of the Fermi surface across the QCP. It follows that, in the low-temperature limit, at $B < B_c$, the Fermi surface is small.

On the non-magnetic side, $B > B_c$, the mass enhancement diverges as B approaches B_c from above. This has been established by measurements of both the T -linear specific-heat coefficient γ , which is proportional to the

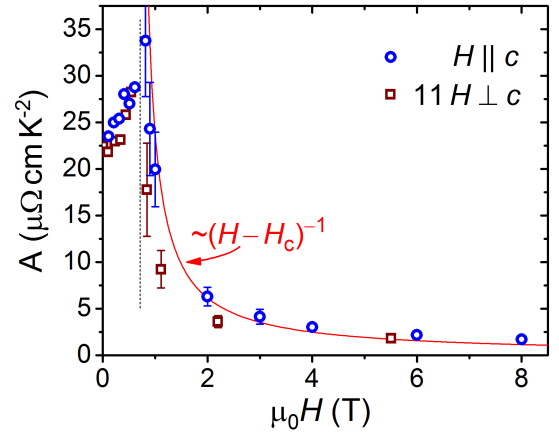


FIG. 6 Divergence of the A coefficient at the QCP in YbRh_2Si_2 (Color online): The effective mass diverges on approach to the critical field $B_c = \mu_0 H_c$ from either side of the QCP. Data for $H \perp c$ have been scaled by a factor of 11. Adapted from Ref. (Gegenwart *et al.*, 2002).

effective mass m^* , and the T^2 coefficient A of the resistivity, which was found to obey the Kadowaki-Woods relation (Tsuji *et al.*, 2005). The divergence of A is shown in Fig. 6.

For $B < B_c$, the mass enhancement is also large. This is compatible with the large C/T measured in the antiferromagnetic state, although to reliably extract γ is a challenge because of the interference of the large specific-heat feature at the magnetic transition temperature T_N . The mass enhancement can be more reliably extracted from the A -coefficient, because the effect of the magnetic transition at T_N on the resistivity is relatively minor. The evolution of the A -coefficient with B is consistent with the destruction of the Kondo effect as the QCP is approached from the non-magnetic side as well as the dynamical Kondo effect inside the antiferromagnetic phase.

The effect of increasing temperature on the Hall crossover can be quantified in terms of the ratio of the crossover width to the crossover magnetic field. For $T \gtrsim 0.5$ K, the ratio quickly increases towards unity, as shown in Fig. 5(c). This implies that, for such temperatures, YbRh_2Si_2 falls in the quantum critical fluctuation regime already for zero magnetic field. Thus, the single-electron spectral weight will be significant at both the small and large Fermi surfaces. In this temperature range, significant spectral weight is thus to be expected at the large Fermi surface. The ARPES measurements in YbRh_2Si_2 , which have been reported for $T > 1$ K (Kummer *et al.*, 2015), are consistent with this prediction (Paschen *et al.*, 2016).

The temperature evolution of the single-particle excitations in YbRh_2Si_2 has been studied by STM measurements, which were first carried out down to 4.6 K in Ref. (Ernst *et al.*, 2011) and were recently extended down to 0.3K (Seiro *et al.*, 2018). The lattice Kondo

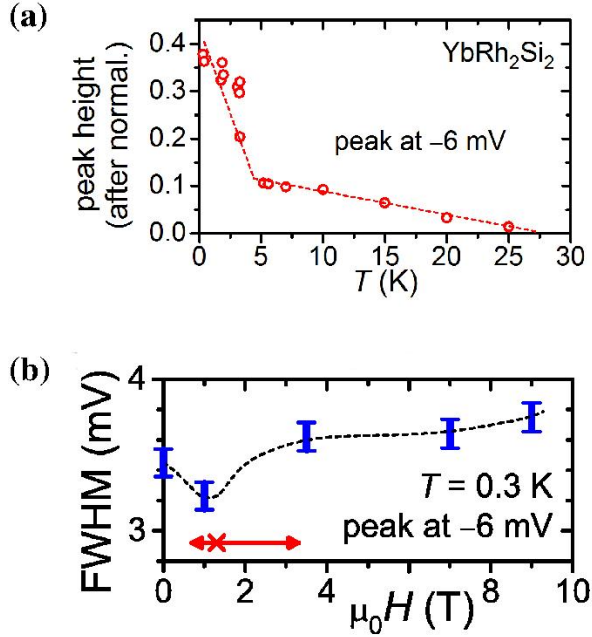


FIG. 7 STM spectroscopy of the lattice Kondo feature at -6meV in YbRh_2Si_2 (Color online): (a) The temperature evolution of the peak height of the -6meV peak. A strong increase in the peak height is observed below 5K . (b) The full width at half maximum (FWHM) of the -6meV peak across the critical field at the base temperature $T = 0.3\text{K}$. Note that at this temperature all field values place the system within the quantum critical fan. The decrease of the peak width near $B^* = \mu_0 H^*(T = 0.3\text{K})$ is consistent with a critical slowing down at quantum criticality. (Data taken from (Seiro *et al.*, 2018)).

effect has been identified with the feature at a particular bias, -6meV . The initial onset of this feature takes place near 25K , which corresponds to T_0^{en} , an estimate of T_0 based on the (spin) entropy S and defined through $S(T_0^{\text{en}}/2) = 0.4R \ln 2$, where R is the ideal gas constant, see Table I. At $B = 0$, the measurements down to $T = 0.3\text{K}$ show an increase in the spectral weight [Fig. 7(a)]. This is compatible with the dynamical Kondo effect at non-zero temperatures.

The STM experiments have also determined the isothermal B -dependence of the peak width, at the lowest measured temperature $T = 0.3\text{K}$. It shows a minimum near $B^*(T = 0.3\text{K})$, as shown in Fig. 7(b). This observation is consistent with a critical slowing down associated with the Kondo-destruction energy scale that was implicated by magnetotransport and thermodynamic measurements (Friedemann *et al.*, 2010; Gegenwart *et al.*, 2007; Paschen *et al.*, 2004). As such, it represents the most direct evidence so far for the Kondo-destruction quantum criticality based on a single-particle measurement in YbRh_2Si_2 .

Summary of Section IV

We now summarize the salient results on YbRh_2Si_2 discussed in this section.

High-energy features: STM experiments for YbRh_2Si_2 at $B = 0$ clearly observe the initial onset of dynamical Kondo correlations around T_0 , a comparatively high temperature, as expected for any Kondo-lattice system regardless of the nature (Kondo-screened or Kondo destruction) of its ground state. This is consistent with the observation of a hybridization gap in the optical spectrum (Kimura *et al.*, 2006). As temperature is further lowered below T_0 , $4f$ electron spectral weight is expected to develop, and this has also been clearly observed.

Low-energy isotherms: STM experiments for YbRh_2Si_2 have been carried out as a function of magnetic field at $T = 0.3\text{K}$. The Kondo-lattice spectral peak shows a critical slowing-down feature at B^* , the Kondo-destruction scale previously determined from magnetotransport and thermodynamic measurements. As such, the STM results are consistent with local quantum criticality.

V. THE CERIUM-BASED 115 FAMILY: PHOTOEMISSION VS. TUNNELING SPECTROSCOPY

The Cerium-based 115 family is comprised of compounds CeMIn_5 where $M = \text{Co}, \text{Rh}, \text{or Ir}$. These compounds are stoichiometric and can be grown in a very clean form. All three compounds crystallize in the HoCoGa_5 structure type and thus possess tetragonal unit cells. Due to their proximity to quantum criticality, they have contributed considerably to a global understanding of quantum critical heavy-electron materials (Park and Thompson, 2009; Si, 2006). While CeCoIn_5 and CeIrIn_5 under ambient conditions are low-temperature superconductors, CeRhIn_5 is an antiferromagnet (Movshovich *et al.*, 2001; Petrovic *et al.*, 2001b). In addition, substituted variants, *e.g.* by Cd substitution on the In site or by substitution of Ce, have also been investigated. For a review, see (Thompson and Fisk, 2012).

A. CeIrIn_5

CeIrIn_5 at ambient pressure is a heavy-electron superconductor with a transition temperature $T_c = 0.40\text{K}$ (Petrovic *et al.*, 2001a). After almost two decades of study, the origin of superconductivity remains controversial, though there is growing support for a magnetically-driven mechanism (Chen *et al.*, 2015). In spite of this controversy, superconductivity in CeIrIn_5 has attracted recent attention because of its unusual strain tunability (Bachmann *et al.*, 2019).

In contrast to CeCoIn_5 or even CeRhIn_5 , CeIrIn_5 has been comparatively less studied by STM and ARPES.

Early ARPES studies led to different conclusions concerning the formation of $4f$ -derived flat bands (Fujimori *et al.*, 2006, 2003). More recently, a high-resolution ARPES study by Chen *et al.* mapped out the full band-structure of CeIrIn_5 (Chen *et al.*, 2018a). Interestingly, this study was able to resolve the complete fine structure of both the $4f_{5/2}^1$ and $4f_{7/2}^1$ peaks in the measured energy-distribution curves (EDCs) and momentum-distribution curves (MDCs), which may be a reflection of the comparatively stronger $4f-c$ hybridization than in CeCoIn_5 (Chen *et al.*, 2018a).

To the best of our knowledge, no scanning tunneling spectroscopy of CeIrIn_5 is available, apart from an STM investigation that focuses on the structural properties of CeIrIn_5 surfaces (Ernst *et al.*, 2010; Wirth *et al.*, 2014). Our main focus in this section will therefore be on CeCoIn_5 and CeRhIn_5 .

B. CeCoIn_5

CeCoIn_5 has attracted interest not only for its comparatively high superconducting transition temperature $T_c \sim 2.3\text{K}$ but also for an overall phenomenology that resembles that of the underdoped cuprates.

The strong interest in CeCoIn_5 includes early photoemission studies which, however, have led to contradictory results concerning the localized vs. itinerant nature of the $4f$ electrons (Koitzsch *et al.*, 2008, 2013, 2009). Optical conductivity measurements of CeCoIn_5 show the existence of a hybridization gap at high energies which starts forming at comparatively high temperatures (Burch *et al.*, 2007; Singley *et al.*, 2002) and recent STM studies of CeCoIn_5 are in line with these findings (Allan *et al.*, 2013; Aynajian *et al.*, 2012; Zhou *et al.*, 2013). De Haas-van Alphen (dHvA) studies performed at low temperatures indicate that the Fermi surface of CeCoIn_5 includes the $4f$ electrons and that therefore the Fermi surface of CeCoIn_5 is large (Settai *et al.*, 2001; Shishido *et al.*, 2002). This conclusion is further corroborated by band-structure calculations that treat the $4f$ electrons as fully itinerant (Haule *et al.*, 2010).

CeCoIn_5 under ambient conditions is believed to be located close to an antiferromagnetic QCP of the SDW type and can be tuned to a quantum phase transition by applying a magnetic field (Ronning *et al.*, 2005; Singh *et al.*, 2007; Zaum *et al.*, 2011). The STM study by Aynajian *et al.* also reported an interesting energy-over-temperature (ω/T) scaling of the local conductance of CeCoIn_5 which sets in around 60K (Aynajian *et al.*, 2012). It is worth recalling that STM probes the single-electron response while the dynamic spin susceptibility measures the magnetic fluctuation spectrum. As ω/T scaling is not expected in the dynamical spin susceptibility at a QCP of the SDW type, the observation of dynamical scaling in the local conductance suggests that

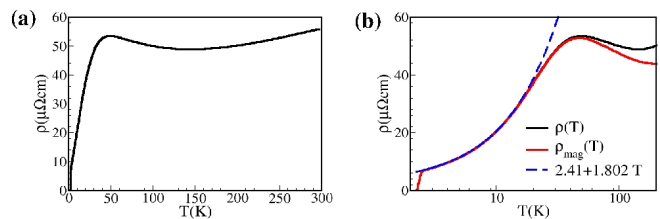


FIG. 8 Temperature dependence of the resistivity ρ of CeCoIn_5 (Color online): (a) $\rho(T)$ in the temperature range from 2.2K to 300K. ρ has a smooth maximum around $T_{\text{coh}} \approx 40\text{K}$, which defines the coherence temperature T_{coh} . (b) ρ (black continuous line) and the magnetic resistivity ρ_m (red continuous line) in a semi-log plot for temperatures from 2K to 200K. $\rho_m(T)$ is defined as the difference between the resistivity of CeCoIn_5 and that of its non-magnetic reference compound LaCoIn_5 at temperature T . The dashed line represents a linear law fit to $\rho_m(T)$ and shows that $\rho_m(T)$ is linear in T from T_c to approximately 20K.

the SDW nature applies, at least at ambient conditions, only at asymptotically low energies. In any case, the observation of ω/T scaling does appear to be in line with the linear-in-temperature behavior of the resistivity below 20K (Petrovic *et al.*, 2001b) as shown in Fig. 8. Further support in favor of such an ω/T scaling in CeCoIn_5 for the single-particle excitations near the Γ point has come from a recent high-resolution ARPES study (Chen *et al.*, 2017).

The ARPES study by Chen *et al.* reported the first 3D Fermi-surface mapping of CeCoIn_5 and provided a measurement of the full band structure of this heavy-electron system (Chen *et al.*, 2017). Due to the large temperature range of the study from 14K to 310K, Chen *et al.* were able to demonstrate that the formation of the $4f$ -derived flat band sets in at temperatures far above the coherence temperature. This finding is significant, although not entirely unexpected. It demonstrates not only the slow evolution of the Kondo screening process but also the likely role of a Kondo effect on the excited crystal field levels (Chen *et al.*, 2017; Pal *et al.*, 2019). These results contrast with a temperature-independent Fermi surface in YbRh_2Si_2 that was inferred from the state of the art ARPES measurements in a temperature window from 1K to 100K (Kummer *et al.*, 2015). An earlier laser-based ARPES study of YbRh_2Si_2 reported a T -dependent bandstructure below 100K (Mo *et al.*, 2012). In this regard, we note that Chen *et al.* suggested that ARPES at temperatures larger than 100K may be required in YbRh_2Si_2 due to the large effect of the crystal field levels (Chen *et al.*, 2017). This is consistent with $T_0^{\text{hyb}} \approx 160\text{K}$ in this compound, see Table I.

High-resolution ARPES results on CeCoIn_5 that are largely compatible with those of Chen *et al.* (Chen *et al.*, 2017) have also been reported by Jang *et al.* (Jang *et al.*, 2017). Though ARPES measurements on heavy-electron compounds have been a major experimental achievement,

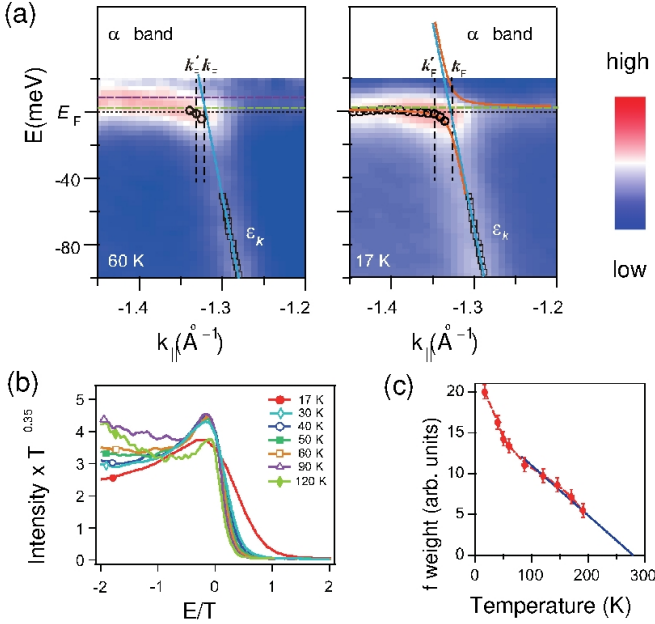


FIG. 9 ARPES view of the $4f$ electron weight near the Fermi energy in CeCoIn_5 (Color online): (a) Evidence for the initial development of hybridization between $4f$ and conduction band α at $T = 60$ and at $T = 17$ K after dividing the EDCs by the Fermi-Dirac function. (b) ω/T scaling of the EDCs near the Γ point in an intermediate temperature range and energy range around the Fermi energy E_F . (c) Background subtracted $4f$ electron spectral weight transfer near the Γ point versus temperature. The EDCs have been integrated over an energy window from -40 meV to 2 meV. (Data taken from (Chen *et al.*, 2017)).

care has to be taken when extrapolating to the high-temperature region where the $4f$ electrons have to be localized across the phase diagram, as argued above. In Fig. 9(a) we reproduced the EDCs from (Chen *et al.*, 2017) for the α -band, one of three bands that are part of the (high-temperature) Fermi surface, in the vicinity of its Fermi crossing both for $T = 60$ K and $T = 17$ K. The data have been divided by the Fermi-Dirac function to access the region (slightly) above the Fermi energy. The dashed lines in Fig. 9(a) indicate the positions of maxima of the main and the first excited CEF-related Kondo-resonance-like features, both of which are taken to be dispersionless. Here, k_F is the Fermi momentum of the conduction electrons without $4f$ participation, *i.e.*, at high temperatures.

In Fig. 9(c), the building-up of spectral weight near the Fermi energy is shown as a function of temperature. This is calculated by integrating the EDCs near the Fermi energy, *i.e.*, from -40 meV to 2 meV, and after subtracting a flat, temperature-independent overall background. It is worth recalling that the majority of the Kondo-resonance-like features of a Cerium-based system is located above the Fermi energy, a region which is, especially at low T , inaccessible to ARPES.

It is instructive to analyze the high-resolution ARPES data of Chen *et al.* for the temperature-dependent band structure of CeCoIn_5 in light of the expectation that the Fermi surface of this compound should contain the $4f$ electrons at sufficiently low temperatures. In other words, in terms of Fig. 2, CeCoIn_5 is located on the $\delta > \delta_c$ side of the E_{loc}^* line. Note, however, that Fig. 2 presents one type of specific cut through the global heavy-electron phase diagram (Si, 2006) and does not contain CeCoIn_5 which is believed to be in close proximity to an SDW QCP.

The red continuous line in the right-hand panel of Fig. 9(a) is a fit of the data to the mean field expression for the single level, single band Anderson lattice model. The circles in Fig. 9(a) are obtained from the maximum in the EDCs and interpreted as the dispersion of the quasiparticle band. This leads to the value of k'_F , where k'_F is the *projected* zero-temperature Fermi momentum. Such a fit should not be taken too literally. As mentioned above, mean field approaches may in principle be suitable to address the conduction bands at comparatively high energies and temperatures or the low-energy behavior on either the small or the large Fermi volume side in a limited energy range. They do, however, generically fail to describe the crossover from the high to the low energy/temperature behavior. In addition, there is the general difficulty of constructing the correct mean field theory. The effective model for a system like CeCoIn_5 should not be the single level, single band Anderson lattice model. Nonetheless, the mean field construction provides an estimate for the change in Fermi wavevector from its high-temperature value k_F to k'_F . If $k'_F = k_F$, the $4f$ electrons remain localized and do not contribute to the Fermi volume. As discussed above and also briefly mentioned in Ref. (Chen *et al.*, 2018b), if the Fermi surface of CeCoIn_5 expands from k_F to k'_F as the zero-temperature fixed point is approached, the bandstructure in the vicinity of E_F should resemble that sketched in Fig. 4(a) and the spectral weight close to k_F needs to vanish as $T \rightarrow 0$ so that the incoherent spectral weight at the Fermi energy is gapped out. The detection of such a, possibly very small, gap is challenging in view of the limited energy resolution and k_z broadening effects of ARPES experiments as discussed in Section III. Note that, although it is expected that $k'_F \neq k_F$ in CeCoIn_5 , results shown in Fig. 9(a) are indicative of a spectral weight increase near and at k_F as the temperature is lowered from $T = 60$ K to $T = 17$ K. This is most likely not an artifact due to the limited energy resolution of the measurement, indicating that the single-particle excitations are not of the form depicted in Fig. 4(a). This is also corroborated by the strange metal behavior, encoded in an approximately linear-in-temperature dependence of the resistivity over a wide temperature window above the superconducting transi-

tion temperature ($T_c \sim 2.3\text{K}$) (Petrovic *et al.*, 2001b). In Fig. 8, the temperature dependence of the resistivity ρ of CeCoIn_5 is shown together with the magnetic resistivity, *i.e.* the difference between the resistivities of CeCoIn_5 and its non-magnetic reference compound LaCoIn_5 .

The ARPES study of Chen *et al.* also indicated the presence of ω/T scaling in the EDCs near the Γ point in an intermediate temperature range (Chen *et al.*, 2017). This is reproduced in Fig. 9(c). Already at around 90K, the EDCs multiplied by $T^{x_{EDC}}$ (with $x_{EDC} \approx 0.36$) collapses on a function depending only on ω/T . This, however, should not be interpreted as reflecting an ω/T scaling of all single-particle excitations, which would imply a strict linear-in- T behavior of the resistivity. Indeed, this scaling seems to be confined to the vicinity of the Γ point and is absent in the angle-integrated EDCs. Moreover, this peculiar scaling exists only in an intermediate T range and fails below 20K, as shown in Fig. 9(c). This conclusion appears to be compatible with the findings reported in (Aynajian *et al.*, 2012), taking into account that tunneling into states with small lattice momenta is favored over tunneling into large-momentum states (Huang *et al.*, 2015; da Silva Neto *et al.*, 2013; Tersoff and Hamann, 1985). This demonstrates that ARPES and STM indeed provide information on the single-particle Green function that can be directly compared to each other. It is, however, noteworthy that the temperature exponents accompanying this ω/T scaling in the intermediate temperature range from 20K to around 70K differ somewhat depending on the measurement technique. While the STM-derived exponent is $x_{STM} \approx 0.53$, the best fit of the ARPES data was obtained for $x_{EDC} \approx 0.36$. The difference between the ARPES and STM results is most likely due to the dependence of the STM current on the degree of tunneling into $4f$ and c electron states. This dependence is encoded in the Fano parameter.

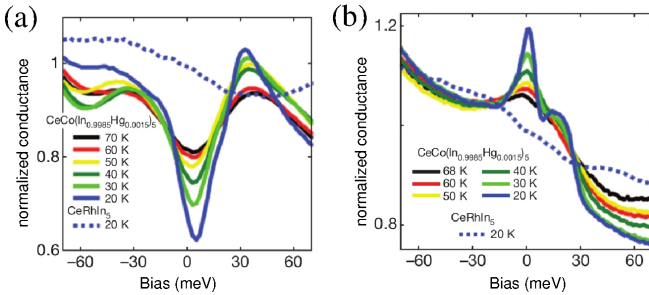


FIG. 10 Tunneling spectroscopy of CeCoIn_5 and CeRhIn_5 (Color online): local conductance vs applied bias voltage for different temperatures on (a) Ce-terminated surfaces and (b) Co (respectively Rh)-terminated surfaces. The peak-dip-peak structure in conductance of CeCoIn_5 (panel a) is typical of a hybridization gap that is not obvious in CeRhIn_5 , even at the lowest temperature. (Data taken from (Aynajian *et al.*, 2012)).

STM studies on CeCoIn_5 (and to a much lesser extent, on CeRhIn_5 and CeIrIn_5) have been performed by several groups (Allan *et al.*, 2013; Aynajian *et al.*, 2012, 2014; Ernst *et al.*, 2010; Haze *et al.*, 2018; Zhou *et al.*, 2013). In Fig. 10(a) and (b), results are shown for the local tunneling conductance of CeCoIn_5 very lightly doped with mercury (Hg) as well as CeRhIn_5 at different temperatures and on two different surfaces (Aynajian *et al.*, 2012). The Hg-doping induced disorder in CeCoIn_5 generates impurity scattering at the dopant sites which in turn can be systematically used to obtain lattice momentum-resolved information of the local DOS through QPI (Derry *et al.*, 2015). This use of QPI to extract the band structure near E_F in the low-temperature limit, however, also has potential shortcomings that were already alluded to in Section III.

C. CeRhIn_5

CeRhIn_5 is an antiferromagnet with a Néel temperature of $T_N = 3.8\text{K}$ at ambient pressure and has predominantly localized moments (Hegger *et al.*, 2000). Under pressure, T_N can be suppressed to zero, thus tuning the system to a QCP at a critical pressure p_c . De Haas-van Alphen studies of CeRhIn_5 across the QCP display a clear jump of the dHvA frequencies at p_c , see Fig. 11(a), which implies that the Fermi surface changes discontinuously at the QCP (Shishido *et al.*, 2005). This compound therefore likely hosts a Kondo-destruction QCP at $\delta_c = p_c$ (δ was defined in Section II). This conclusion is further corroborated by an effective mass that tends to diverge on approach to p_c , Fig. 11(b). The latter reflects the vanishing of the wavefunction renormalization factor z , depicted in Fig. 2(b), as the QCP is reached from either above or below p_c . In addition, transport measurements

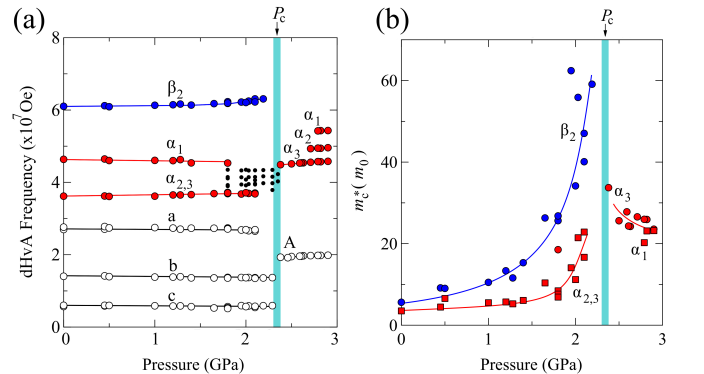


FIG. 11 De Haas-van Alphen measurements on CeRhIn_5 (Color online): (a) Jump of the dHvA frequencies at p_c indicating a reconstruction of the Fermi surface as the QCP is crossed. (b) Diverging effective mass upon approaching p_c from above and below (Data taken from (Shishido *et al.*, 2005)).

provide evidence for the Kondo-destruction QCP (Park *et al.*, 2006, 2008). These low-energy quantum critical features are accompanied by experiments measuring high energy properties. The optical conductivity of CeRhIn₅ has been reported in (Mena *et al.*, 2005) and shows the formation of a weak hybridization gap at high frequencies as temperature is lowered below the crossover scale T_0 .

Despite evidence for the existence of a QCP featuring critical reconstruction of the Fermi surface in CeRhIn₅ under pressure, APRES and STM investigations of this compound are comparatively rare. This is largely due to difficulties in preparing a suitable surface and to the present impossibility of making these measurements under applied pressure. Early non-resonant ARPES investigations of CeRhIn₅ reported that the $4f$ electrons in this compound are predominantly itinerant (Moore *et al.*, 2002), whereas a second non-resonant ARPES study argued that the $4f$ electrons are nearly localized (Fujimori *et al.*, 2003).

Scanning tunneling spectroscopy data on Ce- and on Rh-terminated surfaces of CeRhIn₅ show no clearly discernible Fano resonances, at least at around 20K (Aynajian *et al.*, 2012), see Fig. 10(a) and (b). Interestingly, these results seem incompatible with high-resolution resonant ARPES data, Fig. 12, which point to the development of the $4f$ -electron spectral weight near the Fermi energy, although the weight transfer is much weaker than in CeCoIn₅, Fig. 12 (Chen *et al.*, 2018b). The spectral weight transfer depicted in Fig. 12 for the three bands crossing the Fermi surface also show that, in the temperature range studied, spectral weight transfer occurs mainly near the γ band crossing. The difference between the ARPES measurements of (Chen *et al.*, 2018b) and the STM investigation of (Aynajian *et al.*, 2012) is likely due to the increased surface sensitivity of STM. One possibility is that the Kondo temperature at the surface is

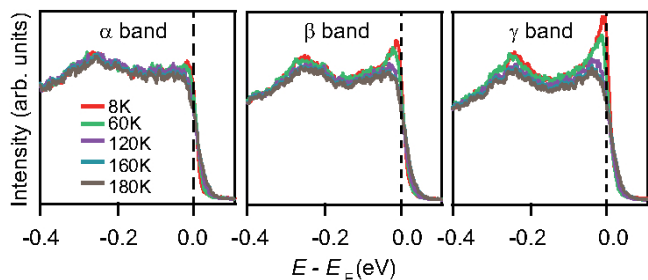


FIG. 12 EDCs of CeRhIn₅ vs temperature (Color online): The energy distribution curves show the evolution of spectral weight with temperature near the Fermi energy E_F for the three bands that cross E_F , labelled α , β and γ -band. Data were taken at along the ΓM direction at $k_{\parallel} = -0.57\text{\AA}^{-1}$ (α -band), $k_{\parallel} = -0.3\text{\AA}^{-1}$ (β -band), and $k_{\parallel} = -0.124\text{\AA}^{-1}$ (γ -band) and with an uncertainty of $\delta k_{\parallel} \sim 0.03\text{\AA}^{-1}$ for each of the three k_{\parallel} -values. (From (Chen *et al.*, 2018b)).

reduced due to the reduced hybridization; a second is that the cleaving process to obtain suitable surfaces appears to be more problematic for CeRhIn₅ and CeIrIn₅ than for CeCoIn₅. In fact, very recent STM results (Haze *et al.*, 2019) on epitaxially grown CeRhIn₅ with well-defined surfaces are very much in line with the ARPES measurements of (Chen *et al.*, 2018b). As in the case of the STM images of YbRh₂Si₂ [see Fig. 7(a)], these data are consistent with the dynamical Kondo effect taking place near the small Fermi surface.

D. Further considerations

We now turn to several additional points that cut across specific Ce-115 families. First, the connection between the different Ce-115 families deserves further studies. As already discussed, isothermal dHvA measurements in CeRhIn₅ provide evidence for a sudden Fermi surface reconstruction at p_c . Intriguingly, frequencies of dominant α orbits at $p > p_c$ for CeRhIn₅ are very similar to those found for the large Fermi surface of CeCoIn₅ at atmospheric pressure (Shishido *et al.*, 2005). No Fermi-surface reconstruction has been found in CeCoIn₅. It is possible that a sudden Fermi-surface reconstruction can still be found in CeCoIn₅ under a new tuning parameter, such as negative pressure. But it may also be that such an effect simply does not exist in CeCoIn₅, reflecting its inherent difference from CeRhIn₅. For example, the $4f - c$ hybridization is much larger in CeCoIn₅ than in CeRhIn₅, as evident in their STM spectra (Fig. 10). As argued recently, this difference in overall hybridization can be traced to an anisotropic spatial extent of their $4f$ orbitals that is set by details of the crystal electric field wavefunction (Sundermann *et al.*, 2019; Willers *et al.*, 2015).

Second, an alternative explanation for the jump of the dHvA measurements across p_c in CeRhIn₅ has been proposed in Ref. (Watanabe and Miyake, 2010). It was suggested that the $4f$ -valence fluctuations lead to a rapid valence change near p_c and a strongly first order antiferromagnetic transition. The latter implies a large jump of the order parameter and, thus a large reconstruction of the Fermi surface. So far, however, all experimental evidence points to a continuous transition at p_c . In addition, canonical valence-fluctuating systems such as CeSn₃ (CePd₃) have specific-heat coefficients of 53 (37) mJ/(mol K²), an effective Kondo temperatures of 770 (1120) K (Lawrence *et al.*, 1981). In those cases, the $4f$ -occupancy n_f will be far from 1 or 0 and, consequently, the entropy in the valence-fluctuation sector, which one can estimate by $R [n_f \ln n_f^{-1} + (1 - n_f) \ln (1 - n_f)^{-1}]$, will be a sizable fraction of $R \ln 2$. By contrast, in the quantum critical regime of CeRhIn₅, the specific heat coefficient is very large [$\gamma \approx 1.25$ J/(mol K²)] (Park and Thompson, 2009), implying that n_f is exceedingly close

to 1. Thus, the valence-fluctuation sector will have a tiny entropy compared to the nearly $R \ln 2$ entropy in the spin sector, and can hardly be the main driver of the critical fluctuations. In other words, the quantum criticality should primarily be driven by physics of the Kondo limit (Park *et al.*, 2006, 2008). Similar arguments apply to CeCoIn₅, CeIrIn₅, YbRh₂Si₂, and CeCu_{5.9}Au_{0.1}.

E. Summary of Section V

We close this section by summarizing the status of ARPES and STM investigations in Cerium-based 115 systems, as discussed in this section.

By and large, the existing STM and ARPES results on the Cerium-based 115 family are consistent with each other, given the requirements of surface quality and the associated difficulties. The recent high-resolution ARPES investigation of these 115 materials also shows that none of the three compounds follows the low-temperature band-structure expectations of the heavy-electron phenomenology, encoded in Fig. 4(a). This is in line with other measurements, in particular transport measurements, which suggest that none is in a Fermi liquid regime in the range where the ARPES measurements were made. Further, the limited energy resolution

VI. PROGRESS, CHALLENGES AND PROSPECTS

A. High energy Kondo features

We have stressed that the initial onset of dynamical Kondo correlations or hybridization is expected, at the T_0 scale of Fig. 2, for all heavy-electron systems regardless of the nature of their ground states.

This scale is evident in YbRh₂Si₂ by STM and optical conductivity. Similarly, the formation of a hybridization gap was manifested in CeRhIn₅ by optical conductivity measurements (Mena *et al.*, 2005) and, recently, by STM measurements (Haze *et al.*, 2019). Also for CeCu₆, which is near a QCP that is accessed by introducing Au-substitution for Cu, a hybridization gap has been observed by optical conductivity (Marabelli and Wachter, 1990). This captures the high-energy T_0 scale for the onset of hybridization-gap formation [Fig. 2(a)], and indeed evolves smoothly across the critical substitution $x_c = 0.1$ based on photoemission measurements (Klein *et al.*, 2008). The T_0 scale is also evidenced by recent time-resolved measurements in the critical substitution range (Pal *et al.*, 2019; Wetli *et al.*, 2018). Here, a terahertz irradiation pumps the system and disturbs the correlations between the local moments and conduction electrons. We can expect the underlying Kondo coupling

of state-of-the-art ARPES is still posing a major challenge in the heavy-electron materials class in which the associated energy scales are typically very small.

High-energy features: Existing ARPES and STM investigations of the 115 members show the initial onset of dynamical Kondo correlations around the T_0 temperature scale [Fig. 2(a)] and the concomitant onset of hybridization-gap formation. This is in line with optical conductivity measurements on these compounds (Chen and Wang, 2016). Comparing ARPES and STM data for the same compound gives complementary results that are compatible with each other, and provide evidence for the existence of the hybridization-gap onset scale T_0 .

Low-energy features: Neither in CeCoIn₅ nor CeRhIn₅ has ARPES been able to confirm unambiguously the existence of either k_F^L or k_F^S . While this may not be surprising due to the limited energy and momentum resolution currently available to ARPES, this finding is also compatible with the absence of Fermi liquid signatures in the investigated temperature range in these compounds; in this range, Fermi liquid signatures are absent as well in transport and thermodynamic properties. Isothermal measurements of dHvA have shown a sudden reconstruction of the Fermi surface across the pressure-induced QCP in CeRhIn₅, which provides strong evidence for a Kondo-destruction QCP.

to produce an initial echo at a time corresponding to $\hbar/(k_B T_0)$. Such a finite time scale is indeed observed both away from and at the QCP. Note that the Fermi liquid scale of CeCu₆ is 0.2K (see Table I), which is not accessible by current experiments done at temperature above 1.5K. Nonetheless, it is conceivable that future experiments may probe not only the echo effect at $\hbar/(k_B T_0)$, but also the scaling time regime much beyond $\hbar/(k_B T_0)$.

Table I compiles high-temperature and Fermi liquid energy scales of the heavy-electron compounds discussed in this Colloquium. This table lists both T_0^{hyb} , the initial onset of the hybridization gap, and T_0^{en} , based on the (spin) entropy S . These two high-energy scales can differ by as much as an order of magnitude, which is not too surprising given that the crossover of Kondo lattice systems from the high-temperature incoherent regime towards the low-temperature coherent/quantum critical/ordered regime is rather broad. This regime can be made even broader when the excited crystal field levels are involved. In practice, we propose to use

$$T_0 = \sqrt{T_0^{\text{hyb}} T_0^{\text{en}}} \quad (7)$$

as a measure of the crossover Kondo scale. Defined in this way, we can infer from Table I that T_0 is ~ 62 K in YbRh₂Si₂, ~ 50 K in CeCoIn₅, $\gtrsim 25$ K in CeRhIn₅, and $\gtrsim 13$ K in CeCu₆.

	T_{FL}/K	$T_0^{\text{hyb}}/\text{K}$	T_0^{en}/K	Reference
YbRh ₂ Si ₂	0.07	~ 160	≈ 24	(Gegenwart <i>et al.</i> , 2006; Kimura <i>et al.</i> , 2006; Trovarelli <i>et al.</i> , 2000)
YbRh ₂ Si ₂ ($B=B_c$, $B\parallel c$)	< 0.008 (LMT)	~ 160	≈ 24	(Gegenwart <i>et al.</i> , 2006; Kimura <i>et al.</i> , 2006; Taupin <i>et al.</i> , 2015)
YbRh ₂ Si ₂ ($B=2T$, $B\parallel c$)	0.135	~ 160	≈ 24	(Gegenwart <i>et al.</i> , 2002, 2006; Kimura <i>et al.</i> , 2006)
CeCoIn ₅	0.14 ($B=6T$)	$\gtrsim 100$	≈ 25	(Mena <i>et al.</i> , 2005; Paglione <i>et al.</i> , 2007; Petrovic <i>et al.</i> , 2001b)
CeRhIn ₅	< 0.15 ($p_c = 2.35\text{GPa}$, $\mu_0H=10T$)	$\gtrsim 60^{(\dagger)}$	≈ 10	(Chen <i>et al.</i> , 2018b; Park <i>et al.</i> , 2008; Park and Thompson, 2009)
CeCu ₆	0.2	$\gtrsim 40$	≈ 4	(Amato <i>et al.</i> , 1987; Fischer <i>et al.</i> , 1987; Marabelli and Wachter, 1990)
CeCu _{6-x} Au _x ($x_c = 0.1$)	< 0.02 (LMT)	$\gtrsim 40$	≈ 4	(Fischer <i>et al.</i> , 1987; Löhneysen <i>et al.</i> , 1994; Marabelli and Wachter, 1990)

TABLE I Characteristic high- and low-temperature scales for several heavy-electron compounds located in the vicinity of quantum criticality. Here, T_{FL} is a temperature scale below which the Landau Fermi liquid T^2 resistivity is observed. T_0^{hyb} is a ‘high temperature’ estimate for the onset of the hybridization gap, and is estimated from the optical conductivity $\sigma(\omega, T)$ (Figs. 3,13) with the exception of CeRhIn₅, where existing $\sigma(\omega, T)$ data only indicate that $8\text{K} < T_0^{\text{hyb}} < 300\text{K}$ (Mena *et al.*, 2005). T_0^{en} is a ‘low-temperature’ estimate of T_0 based on the (spin) entropy S , using a procedure for the single-impurity Kondo model with constant conduction electron density of states (for which $T_0=T_0^{\text{en}}=T_K^0$): $S(T_0^{\text{en}}/2) = 0.4R \ln 2 \approx 0.277R$, where $R = 8314.5 \text{ mJ}/(\text{mol K})$ is the ideal gas constant. (LMT) designates the lowest measured temperature for the electrical resistivity ρ .

For YbRh₂Si₂ at the critical field, T_{FL} has been estimated from $\rho(T)$ and using the result that $\rho(T) \sim T$ down to the LMT of 8mK (Taupin *et al.*, 2015), making the listed value to be an upper bound. The hybridization gap onset in $\sigma(\omega < T)$ is assumed to be the same for $0 \leq B \leq 2T$. Similarly, changes of T_0^{en} are assumed to be small for fields $0 \leq B \leq 2T$, where the specific heat at around 20K is only weakly field-dependent for $B \leq 2T$, see (Gegenwart *et al.*, 2006).

For CeRhIn₅, the QCP is located at $p_c = 2.35\text{GPa}$ and H_c with $\mu_0H_c \lesssim 10T$, see (Park *et al.*, 2008).

^(†)This value for T_0^{hyb} has been estimated from the ARPES data of (Chen *et al.*, 2018b) for ambient conditions, see also Fig. 12. For CeCu_{6-x}Au_{0.1}, $\rho(T)$ is linear in T down to the LMT 20mK; hence, the listed value is also an upper bound. The estimate of T_0^{hyb} in CeCu_{6-x}Au_{0.1} is supported by the specific heat data of (Löhneysen *et al.*, 1994).

The references in this table are arranged such that in each row the first reference provides T_{FL} , the second contains estimates for T_0^{hyb} and the third provides results on the low-temperature (spin) entropy.

B. Isothermal evolution at low temperatures

We have discussed in Section II that, to assess the nature of quantum criticality (Kondo destruction vs. SDW), the isothermal evolution of quasiparticle spectral weight at low temperatures is particularly informative. In YbRh₂Si₂, this has been done through STM measurements as a function of magnetic field at $T = 0.3 \text{ K}$, and the results (Seiro *et al.*, 2018) support the Kondo-destruction scale that had been inferred from magnetotransport and thermodynamic measurements (Friedemann *et al.*, 2010; Gegenwart *et al.*, 2007; Paschen *et al.*, 2004). Further STM measurements at lower temperatures will clearly be instructive. Whether related STM studies can be carried out in 115 systems is at the present time unclear, because the QCP is realized at a relatively large pressure (CeRhIn₅) or possibly at negative pressure (CeCoIn₅ and CeIrIn₅) (Pham *et al.*, 2006; Sidorov *et al.*, 2002). In these latter two cases, applying uniaxial tension might open the possibility of both ARPES and STM studies in a regime that would access their

respective QCP. Similar isothermal studies by ARPES appears to be difficult, due to the low temperature that is needed, and also because ARPES cannot be performed in the presence of a magnetic field.

C. Outlook

As discussed above (see Section III), STM is a real space probe and thus generally lacks momentum resolution. It is, however, possible to extract information on the band structure near the Fermi energy using Friedel oscillations that occur near defects (Petersen *et al.*, 2000, 1998). Since STM is a surface probe, QPI only provides a projected bandstructure. Furthermore, the standard approach which is based on Born scattering is known to be insufficient in many cases (Toldin *et al.*, 2013). This limitation notwithstanding, it will be instructive to obtain bandstructure information through Fourier transform-STM on either side of the QCP to interpret QPI spectra in the quantum critical fan of the QCP.

Critical Kondo destruction is accompanied by a partic-

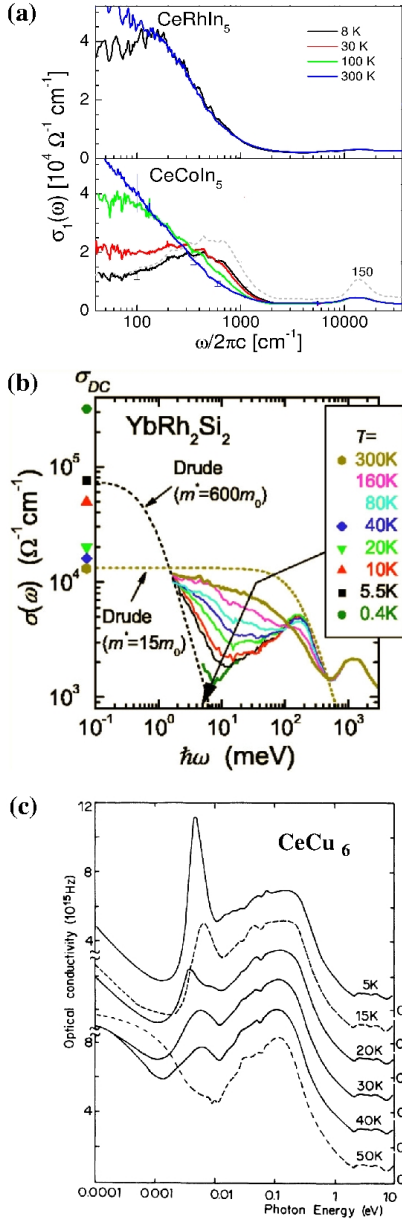


FIG. 13 Optical conductivity $\sigma(\omega, T)$ and evolution of the hybridization gap (Color online): (a) Although the hybridization gap in CeRhIn_5 (top) is overall less pronounced than that in the optical conductivity of CeCoIn_5 (bottom), the overall features for both compounds are in accordance with general expectations (see Fig. 3): at the highest measured T a broad Drude peak exists out of which a hybridization gap develops below $\sqrt{T_0 D}$ as T is lowered (Data taken from (Mena *et al.*, 2005)). (b) The hybridization gap in YbRh_2Si_2 evolves over a large T region, starting well above 100K. As the data are taken at zero external field, the system is located on the $\delta < \delta_c$ side (see section II) and a Drude peak is therefore expected in $\sigma(\omega, T)$ at small ω and sufficiently low T . (Data taken from (Kimura *et al.*, 2006)). (c) In CeCu_6 the optical conductivity develops a hybridization gap at around $\hbar\omega \approx 1\text{meV}$ below 50K which is flanked towards higher energies by a pronounced peak. (Data taken from (Marabelli and Wachter, 1990)).

ular kind of ω/T scaling. Recently, this type of scaling has been demonstrated for the optical conductivity of YbRh_2Si_2 thin films grown by molecular beam epitaxy, studied by time-domain THz-transmission spectroscopy (Prochaska *et al.*, 2020). This result would be surprising from the perspective of an SDW QCP, where only the spin dynamics is expected to be critical. However, it is in line with critical Kondo destruction (Prochaska *et al.*, 2020). Because the magnetic quantum phase transition is accompanied by the transition from a phase with asymptotically decoupled local-moment and conduction-electron degrees of freedom to one in which the entangling of the two turns the $4f$ local moments into composite quasiparticles, it is natural that both the single-particle and charge dynamics are critical. Indeed, calculations at the Kondo-destruction QCP in various large- N limits (Cai *et al.*, 2020; Kirchner *et al.*, 2005; Komijani and Coleman, 2019; Zhu *et al.*, 2004) and, more recently, in the physical $N = 2$ case (Cai *et al.*, 2020) have shown such a singular charge dynamics. Intriguingly, this type of charge dynamical scaling in models of the Kondo limit smoothly connects to the ω/T -scaling for the charge dynamics in the beyond-Landau-type quantum criticality in the mixed-valence regime (Pixley *et al.*, 2012).

Epitaxial thin films of members of the 115 family and CeIn_3 have been available for some time (Shishido *et al.*, 2010) but STM measurements on these films of CeCoIn_5 and CeRhIn_5 have only been reported very recently (Haze *et al.*, 2019, 2018). The observed onset of the hybridization gap in the STM spectrum demonstrates the high-energy T_0 scale which, as we have emphasized, is consistent with a Kondo-destruction ground state in CeRhIn_5 . It will be interesting to see whether a lattice mismatch between substrate and thin film might be used as a substitute for pressure tuning and to establish the range of ω/T scaling both within the general phase diagram and with respect to the type of correlator, *i.e.*, single-particle excitations, two-particle correlators like the density-density, spin-spin, or current-current correlation functions, and their n -point ($n > 4$) counterparts.

We also briefly discussed in Section III that the underlying assumption in the interpretation of STM spectra in terms of the equilibrium local DOS is less justified at higher voltages. This may be particularly pertinent near the QCP, where the temperature of the measurement itself is expected to set the only relevant scale (Kirchner and Si, 2009). It would be interesting to explore the scaling of spectral density with bias voltage in the nonequilibrium regime which could be yet another way of unraveling the properties of the underlying QCP (Ribeiro *et al.*, 2015; Zamani *et al.*, 2016b).

We have so far focused on YbRh_2Si_2 and Ce-115 compounds. It will be instructive to carry out measurements of single-electron properties in other candidate heavy-electron materials for Kondo destruction (Gegen-

wart *et al.*, 2008; Kirchner *et al.*, 2013; Si and Paschen, 2013; Stewart, 2001). A case in point is CeNiAsO, a heavy-electron relative of the high- T_c Fe-based oxypnictides. Here, the recent neutron scattering experiments provide evidence for a local-moment antiferromagnetic order, whose ordering wavevector is determined by the RKKY interaction mediated by the conduction-electron states near the small Fermi surface (*i.e.*, the Fermi surface of the conduction electrons alone, with the $4f$ -electrons localized) (Wu *et al.*, 2019), and transport measurements have suggested the possibility of a Kondo-destruction QCP induced by either pressure or P-for-As doping (Luo *et al.*, 2014).

More broadly, there is the question of where to look for new examples of Kondo-destruction criticality. If the f - c hybridization is too strong, magnetic order would more likely be of the SDW type that, when tuned to $T = 0$, would result in a conventional QCP. Thus, weaker hybridization is expected to be a more favorable setting to access a possible Kondo-destruction QCP. Alternatively, a low carrier density gives a small Fermi surface in a Kondo lattice and delays the full development of a Kondo singlet state with decreasing temperature. CeNi₂As_{2- δ} appears to be an example of such a case with evidence of Kondo-destruction quantum criticality (Luo *et al.*, 2015). Finally, in the absence of tuning hybridization or carrier density, increasing frustration, whether through crystal structure or reduced dimensionality, offers an exciting opportunity for discovering new examples (Fritsch *et al.*, 2014; Si, 2006; Tokiwa *et al.*, 2015; Zhao *et al.*, 2019).

VII. CONCLUSION

We have reviewed and compared recent ARPES and STM investigations on heavy-electron materials close to magnetic instabilities with a focus on Kondo-destruction quantum criticality. Real-space and momentum-space spectroscopies combine the power of both methods (Crepaldi *et al.*, 2013; Nicoara *et al.*, 2006) which has proven to be useful in the study of complex materials such as the cuprate high-temperature superconductors (Markiewicz, 2004; Shen and Davis, 2008) and the Kondo insulator SmB₆ (Matt *et al.*, 2018). In the context of Cerium- and Ytterbium-based rare earth intermetallics as well as actinide-based compounds, such a combination seems particularly promising given that much of the excitement and interest generated by these materials derives from the interplay of local and itinerant degrees of freedom; while Kondo screening is primarily a local phenomenon, a possible Fermi-volume increase is best addressed in momentum space. Method-specific constraints, limited energy resolution and the need for very low temperatures in order to resolve a Fermi momentum change across a Kondo-destruction quantum critical point pose unique challenges to both ARPES and STM

investigations.

On the other hand, combining ARPES and STM results with other measurements, like resistivity and magnetotransport measurements, neutron scattering and optical conductivity investigations, can provide a consistent picture of Kondo-destruction quantum criticality that emerges as a function of some non-thermal tuning parameter and enables one to locate a specific compound in the general phase diagram of heavy-electron materials. This appears particularly relevant in the present context in order to aide a separation of bulk and surface contributions as both ARPES and STM are primarily surface sensitive. The change in symmetry and c - f hybridization that typically occurs at surfaces can in Kondo systems substantially modify low-energy scales as compared to their bulk value.

We have emphasized the distinction between the spectroscopic properties that reflect the high-energy Kondo physics, such as the formation of the hybridization gap, and those that are capable of probing the nature of quantum criticality, such as low-temperature isothermal measurements across the quantum critical point. The latter has become possible in the STM measurements of YbRh₂Si₂, which corroborates the Kondo-destruction energy scale that had been extracted by isothermal magnetotransport and thermodynamic measurements. In CeRhIn₅, strong evidence for Kondo destruction in the one-electron excitation spectrum has been provided by quantum oscillation measurements across the critical pressure. It will certainly be instructive to explore further signatures of beyond-Landau quantum criticality in these and other heavy-electron systems.

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