

Strange-metal behavior without fine tuning in $\text{PrV}_2\text{Al}_{20}$ Marvin Lenk^{1,*}, Fei Gao², Johann Kroha^{1,3,†} and Andriy H. Nevidomskyy^{2,‡}¹Physikalisches Institut, Universität Bonn, Nussallee 12, 53115 Bonn, Germany²Department of Physics and Astronomy, Rice University, Houston, Texas 77005, USA³School of Physics and Astronomy, University of St. Andrews, North Haugh, St. Andrews KY16 9SS, United Kingdom

(Received 26 December 2023; revised 29 April 2024; accepted 12 August 2024; published 8 October 2024)

Strange-metal behavior observed in the praseodymium-based heavy-fermion material $\text{PrV}_2\text{Al}_{20}$ has been tentatively interpreted in the framework of proximity to a quantum critical point (QCP) associated with quadrupolar ordering. Here, we demonstrate that an alternative, natural explanation exists without invoking a QCP, in terms of the unconventional nature of the quadrupolar Kondo effect taking place in non-Kramers ions. Using a combination of *ab initio* density-functional theory calculations and analytical arguments, we construct a periodic Anderson model with realistic parameters to describe $\text{PrV}_2\text{Al}_{20}$. We solve the model using dynamical mean-field theory preserving the model symmetries and demonstrate the non-Fermi liquid strange-metal behavior stemming from the two-channel nature of the quadrupolar Kondo effect. Our calculations provide an explanation for the puzzling temperature dependence in the magnetic susceptibility and provide a basis for analyzing future photoemission experiments.

DOI: 10.1103/PhysRevResearch.6.L042008

In the past several decades, so-called strange metals and associated phenomena have garnered much attention. As the name suggests, these metals defy the standard Landau Fermi liquid paradigm [1] in that they lack well-defined quasiparticles, as demonstrated by the unusual, non-Fermi liquid (NFL) power-law exponents in the electrical resistivity, specific heat, magnetic susceptibility, and other thermodynamic and transport quantities. A large number of strange metals have been found in the heavy-fermion materials when magnetic ordering is suppressed to zero temperature by pressure, doping, or magnetic field, resulting in a quantum critical point (QCP) [2–6]. In some instances, strong quantum fluctuations prevent the textbook Nozières’ Fermi liquid state from forming, resulting in Kondo breakdown [7–9].

In the vast majority of heavy-fermion metals, the conduction electrons act to screen a spin- $\frac{1}{2}$ Kramers ion, such as Ce^{3+} [2,3] or Yb^{3+} [4,6], resulting in a competition between the Kondo effect and incipient magnetic ordering of the localized moments [10,11]. The situation is less clear in materials with non-Kramers ions, like praseodymium (Pr)-based PrPb_3 and PrPtBi . There, the $4f^2$ electrons are well localized but only very weakly coupled to conduction electrons, making the Kondo scale T_K too small to be observed.

The status quo has changed dramatically with the discovery of $\text{PrT}_2\text{Al}_{20}$ ($T = \text{Ti, V}$) about a decade ago, where the

large values of the Seebeck coefficient [12,13] and hyperfine coupling [14], much enhanced effective masses ($m^*/m_e \sim 20$ in $\text{PrTi}_2\text{Al}_{20}$ and ~ 120 in $\text{PrV}_2\text{Al}_{20}$ [15]), and direct evidence for the Abrikosov-Suhl resonance in photoemission [16] all indicate that Kondo hybridization plays an important role in these compounds. Intriguingly, non-Fermi liquid behavior is observed in $\text{PrV}_2\text{Al}_{20}$ [17,18], prompting the suggestions that this material may sit close to a putative QCP. Although a precedent for a stoichiometric material perched in close proximity to a QCP exists, notably $\beta\text{-YbAlB}_4$ [6], this is certainly unusual. A possible alternative to explaining the strange metal behavior in $\text{PrV}_2\text{Al}_{20}$ is based on the notion that the Kondo effect is unconventional in that no dipole moment is present in the $|\Gamma_3\rangle$ crystal-electric-field (CEF) ground state of the Pr^{2+} ion. The conduction electrons instead screen the quadrupolar moment of the non-Kramers doublet so that the conduction-electron spin represents two conserved screening channels protected by Kramers degeneracy. The resulting two-channel Kondo (2CK) effect is known to harbor non-Fermi liquid physics [19] and this option has previously been investigated generally for quadrupolar Kondo materials and specifically in the context of $\text{PrV}_2\text{Al}_{20}$ [20–24]. However, measurements of the magnetic susceptibility $\chi(T)$ [17,25] are at odds with the expected behavior for a standard 2CK effect [19]. Thus, the question remains open whether the 2CK effect alone can explain the strange-metal behavior or if it is necessary to involve competing quadrupolar order and the QCP scenario.

In this Letter, we use *ab initio* band-structure calculations combined with analytical insight to formulate a two-channel Anderson lattice model that captures the salient features of 1-2-20 compounds, focusing in particular on $\text{PrV}_2\text{Al}_{20}$. We then solve the model using dynamical mean-field theory (DMFT), finding that the spectral density shows a clear Abrikosov-Suhl resonance while the quasiparticle decay rate exhibits NFL scaling that approaches 2CK behavior. Surprisingly, we find

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that the magnetic susceptibility is dominated by the first excited CEF multiplet (the magnetic Γ_5 triplet) since it couples linearly to the magnetic field, in contrast to the quadratic coupling of the quadrupolar Γ_3 ground-state doublet. The Kondo physics of the triplet contribution is fundamentally different in that it gives rise to an intermediate impurity spin-1 Kondo effect, where the conduction-electron spin- $\frac{1}{2}$ is actively participating in the screening instead of acting as a channel. This crossover [26] between an impurity spin-1 Kondo effect at intermediate temperatures and 2CK behavior at low temperatures is responsible for the experimentally observed strange-metal behavior in $\text{PrV}_2\text{Al}_{20}$ [17] without involving the notion of it being fine tuned to a QCP.

Valence fluctuations and CEF excitations. The experimentally determined CEF ground state of the Pr^{2+} ion is the $|\Gamma_3 \pm\rangle$ doublet of the cubic T_d point group (see the Supplemental Material [27] and Refs. [28–31] cited therein for details). The valence fluctuations are between this $4f^2$ ground state and the excited $4f^3$ and $4f^1$ Kramers states. In order to determine the dominant channel, we have performed *ab initio* density functional theory (DFT) calculations using the full-potential augmented plane-wave method implemented in the Wien2k package [32], with a fixed number of f electrons in the core (the “open core” method, see [27] for details). We found the energy difference to the $|4f^3\rangle$ state, $\Delta E_{2 \leftrightarrow 3} = E_{4f^3} - E_{4f^2} \approx 2.2$ eV, to be significantly lower than the energy of the excited $|4f^1\rangle$ level ($\Delta E_{2 \leftrightarrow 1} \approx 3.8$ eV). We, therefore, adopt $4f^2 \leftrightarrow 4f^3$ as the dominant Kondo hybridization channel, in contrast to the previous studies that have assumed $4f^1$ to be the lowest excited valence state. According to Cox and Zawadowski [33], Γ_6 is the ground-state Kramers doublet (due to spin degeneracy) of the $4f^3$ multiplet, which we adopt for our calculation [34].

The aforementioned transitions are mediated via conduction electrons projected onto Γ_8 states at the Pr atom positions, since the product of the irreducible representations (irreps) $\Gamma_6 \otimes \Gamma_3 = \Gamma_8$. Note that the Γ_8 state is fourfold degenerate, labeled by the isospin $\alpha = \pm$ and the Kramers spin $\mu = \uparrow, \downarrow$. The valence fluctuations and the corresponding multiplets are shown schematically in Fig. 1(a). As we demonstrate below, the first excited $4f^2$ CEF state (Γ_5 triplet) plays a crucial role in the magnetic susceptibility. Experimental data suggest a CEF splitting of $\Delta/k_B \approx 40$ K [17].

Two-channel nature of the Kondo effect. The non-Kramers Γ_3 doublet has no magnetic dipole but a nonvanishing quadrupole [33,35] and octupole moment [35,36]. The conduction electrons screen this state in an unconventional manner, dubbed the quadrupolar Kondo effect, which was first studied by Cox and Zawadowski [33,37] in the context of uranium-based heavy-fermion materials. They made the crucial observation that the Kramers degeneracy inherent in the spin- $\frac{1}{2}$ conduction electrons provides two degenerate channels for Kondo screening, thus resulting in a two-channel Kondo (2CK) effect which is known to exhibit non-Fermi liquid behavior below the Kondo temperature T_K [38,39]. In this Letter, we build a microscopically faithful model of this effect in $\text{PrV}_2\text{Al}_{20}$, introduced below, and use DMFT to solve it numerically.

Model. We construct a periodic Anderson model (PAM) to capture the $4f^2 \leftrightarrow 4f^3$ valence fluctuations. It is most natu-

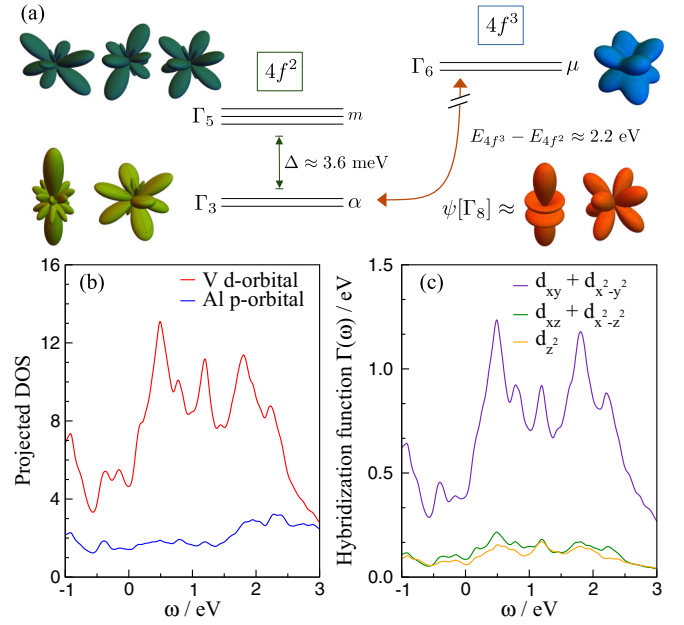


FIG. 1. (a) CEF-Scheme of the states involved in the valence fluctuations of the Pr atoms. (b) Projected density of states (DOS) of V d orbitals and Al p orbitals normalized to the number of Pr neighbors instead of the number of atoms in a unit cell. (c) Contribution to the hybridization function from different V d orbitals [c.f. Eq. (7)].

rally written in terms of Hubbard operators [40], which we conveniently express by auxiliary particle operators [41–44],

$$a_{j,\mu}^\dagger |0\rangle = |j; 4f^3 \Gamma_6, \mu\rangle, \quad (1)$$

$$f_{j,\alpha}^\dagger |0\rangle = |j; 4f^2 \Gamma_3, \alpha\rangle, \quad d_{j,m}^\dagger |0\rangle = |j; 4f^2 \Gamma_5, m\rangle, \quad (2)$$

$$\hat{Q}_j = \sum_{\alpha} f_{j,\alpha}^\dagger f_{j,\alpha} + \sum_m d_{j,m}^\dagger d_{j,m} + \sum_{\mu} a_{j,\mu}^\dagger a_{j,\mu}, \quad (3)$$

subject to the constraint that the locally conserved auxiliary-particle number $\hat{Q}_j$ be fixed to unity. Here, j labels the position of a Pr ion, and $\alpha = \pm$, $m = 0, \pm 1$, $\mu = \pm \frac{1}{2}$ are the quantum numbers of the respective CEF states. Written in terms of these operators, the Pr $4f$ part of the PAM reflects the energies of $4f^2$ and $4f^3$ states in Fig. 1(a):

$$H_{\text{Pr } 4f} = E_{4f^2} \sum_{j,\alpha} f_{j,\alpha}^\dagger f_{j,\alpha} + (E_{4f^2} + \Delta) \sum_{j,m} d_{j,m}^\dagger d_{j,m} + E_{4f^3} \sum_{j,\mu} a_{j,\mu}^\dagger a_{j,\mu} + \sum_j \lambda_j (\hat{Q}_j - 1). \quad (4)$$

Energies are measured relative to the Fermi energy E_F and $\lambda_i \rightarrow \infty$ enforces the constraint $\hat{Q} = 1$ [44]. The first contribution to the hybridization is the $4f^2$, Γ_3 isospin screened by the conduction electrons projected onto the Γ_8 states, via valence fluctuations with the Pr $4f^3$ state,

$$H_{\text{VF}}^{\Gamma_3} = \frac{V_0}{\sqrt{2}} \sum_{j\alpha\mu} \text{sgn}(\mu) a_{j,\mu}^\dagger f_{j,\alpha} \psi_{j,-\alpha,\mu} + \text{H.c.}, \quad (5)$$

$$\psi_{j,\alpha\mu} = \sum_{\mathbf{k}} \sum_{a\sigma} e^{i\mathbf{k} \cdot \mathbf{R}_j} \Phi_{a\mu,\sigma}^a(\mathbf{k}) c_{a\sigma}^a(\mathbf{k}). \quad (6)$$

Note that (modulo adopting $4f^3$ rather than $4f^1$ as the lowest-energy valence excitation) this is analogous to the hybridization used by other authors [22,45]. Here, $\psi_{j,\mu\alpha}$ represents the Wannier function of the conduction electrons projected onto the set of $|\Gamma_8, \alpha\mu\rangle$ states of the Pr atom at position $\mathbf{R}_j$, and the sum is over the conduction-electron orbitals a . These Wannier functions depend on both the quadrupolar isospin α and the Kramers index μ . The Wannier projection is captured by the $\mathbf{k}$ -dependent form factor $\Phi_{\alpha\mu;\sigma}^a(\mathbf{k}) = \sum_{j,i} e^{-i\mathbf{k}(\mathbf{R}_j - \mathbf{r}_i)} \langle \Gamma_8, j\alpha\mu | i a \sigma \rangle$, which projects the atomic orbital a from site $\mathbf{r}_i$ onto the $|\Gamma_8, \alpha\mu\rangle$ states of a given Pr atom at position $\mathbf{R}_j$ [46].

Taking the orbital, energy, and $\mathbf{k}$ dependence of the hybridization form factors $\Phi^a(\mathbf{k})$ into account beyond the standard Anderson model, the local contribution to the Γ_3 hybridization function using Eq. (5) reads,

$$\Gamma^0(\omega) = |V_0|^2 \sum_{ka} \Phi^a(\mathbf{k}) \text{Im}\{\mathbf{G}_{\text{eff}}^{c,0}\}(\omega, \mathbf{k}) \Phi^{+a}(\mathbf{k}), \quad (7)$$

where the term under the sum is understood as a matrix product in all the relevant orbital indices. It incorporates the projection of all bands onto the relevant orbitals, where $\mathbf{G}_{\text{eff}}^{c,0}$ represents a weighted sum of the Green functions over the conduction bands as defined below. The form-factor $\Phi(\mathbf{k})$ defined in Eq. (6) includes the $\mathbf{k}$ -dependent projection of those Wannier states onto the local Pr Γ_8 states. This nontrivial $\mathbf{k}$ dependence drastically alters the DMFT procedure, where it manifests as a $\mathbf{k}$ -dependent hybridization. For a more detailed discussion, see [27]. The trace of $\Gamma^0(\omega)$ is plotted in Fig. 1(c).

The second contribution to the hybridization is the $4f^2, \Gamma_5$ triplet. We couple the Γ_5 operators $d_{j,m}$ to a linear superposition of isospins so as not to break the α degeneracy guaranteed by the CEF symmetry [47],

$$H_{\text{VF}}^{\Gamma_5} = V_1 \sum_{j\alpha m \mu \mu'} \Xi_{m\mu;\alpha\mu'} a_{j,\mu}^\dagger d_{j,m} \psi_{j,\alpha\mu'} + \text{H.c.} \quad (8)$$

Details of the hybridization matrix Ξ are discussed in the Supplemental Material [27]. The resulting hybridization function is given analogously to Eq. (7) with Ξ incorporated and a prefactor of V_1 instead of V_0 . While it is very difficult to compute the values of the hybridization strength V_1 from first principles, for concreteness, we set $V_1 = V_0$.

The projection of conduction electrons onto the local Γ_8 states generally contains contributions from multiple conduction bands, originating from different orbital degrees of freedom labeled by a in Eq. (6). Previous treatments of $\text{PrV}_2\text{Al}_{20}$ have used geometric arguments to select the relevant conduction bands [22,45,48]. Our DFT analysis shows that mostly aluminum p orbitals and vanadium d orbitals hybridize with Pr $4f$ states, and, given the stoichiometric composition, there are many (140, not counting spin) of such conduction orbitals per unit cell. A full treatment of the orbital-dependent projection is numerically challenging. We therefore justify the following approximations, which significantly improve the feasibility of solving the model.

(i) The conduction-electron density of states (DOS) near the Fermi level is dominated by V $3d$ electrons, as shown by DFT calculations, see Fig. 1(b). This motivates us to ignore

the hybridization with Al states, which reduces the number of orbitals considerably.

(ii) The CEF effect on V sites, with the local D_{3d} point-group symmetry, splits the d -electron levels into three irreps: the singlet (ignoring spin) A_{1g} irrep consisting of a d_{z^2} orbital, and two E_g doublets. Of these, the largest contribution to the hybridization function comes from the $\{d_{xy}, d_{x^2-y^2}\}$ doublet, as can be seen in Fig. 1(c). We thus limit the orbital index a to only run over this doublet, resulting in the total of $N = 4 \times 2 = 8$ orbitals (because there are 4 V atoms per unit cell).

(iii) While the DFT operates in the Bloch basis rather than the N vanadium orbitals discussed above, for the DMFT calculation we use an effective V electron propagator obtained by a sum of projected Bloch bands n , where the weight factor $w_n^a(\mathbf{k})$ projects onto the $a = Vd_{xy,x^2-y^2}$ Wannier orbital, and the individual Bloch propagators $G_n^{c,0}(\omega, \mathbf{k})$ are extracted from DFT. This reduces the DMFT computational effort from treating a large matrix propagator at each discretized $\mathbf{k}$ point to a single-valued one. In order to extract the unhybridized electron bands entering $G_n^{c,0}(\omega, \mathbf{k})$, we need to remove the effect of V-Pr hybridization already present at the DFT level to avoid double counting. We achieve this by performing the so-called “open core” calculation that places Pr $4f$ electrons into the core, effectively removing their effect on the conduction bands (see Sec. III in the Supplemental Material [27]).

Results. The lattice system is solved via DMFT [49–51], utilizing the noncrossing approximation (NCA) [44,52,53] as the impurity solver. The NCA is known to correctly describe the low-energy scales, frequency dependence, and infrared exponents in the 2CK case [54]. It is capable of efficiently treating complex multi-orbital systems with satisfactory accuracy while reaching temperatures well below T_K [55]. For the numerical evaluations we used $V_0^2 = V_1^2 = 1.5 \text{ eV}^2$ and a bare hybridization such that the renormalized CEF splitting of $\Delta_{\text{eff}}/k_B \approx 40 \text{ K}$, in agreement with the experiment.

The spectral function $A(\mathbf{k}, \omega) = \frac{1}{\pi} \text{Tr}[\text{Im}\mathbf{G}(\mathbf{k}, \omega)]$, extracted from DMFT, is shown in Fig. 2. The accumulation of spectral weight near E_F is observable already at $T_K^{(0)} \approx 50 \text{ K}$ which marks the perturbative onset of the Kondo effect, involving both Γ_3 and Γ_5 multiplets. Their CEF splitting into Stokes and anti-Stokes satellites [55] at $\omega \approx -3.2 \text{ meV}$ and $\omega \approx +2 \text{ meV}$ [arrows in Fig. 2(c)] becomes discernible below $T \approx 20 \text{ K}$, defining the (renormalized) CEF energy Δ_{eff} . Note that in Pr the $4f^2\Gamma_5 \rightarrow 4f^3 \rightarrow 4f^2\Gamma_3$ transition process is virtual only, so that the anti-Stokes CEF resonance at $+2 \text{ meV}$ appears merely as a shoulder in Fig. 2(c) [55]. Finally, the non-Fermi liquid shape of the 2CK Abrikosov-Suhl resonance starts to emerge below $T_K \approx 5 \text{ K}$, which we identify as the 2CK strong-coupling Kondo scale T_K . This is confirmed by the imaginary part of the Γ_3 component of the self-energy $\text{Im}\Sigma(\tilde{\omega})$ (quasiparticle decay rate) shown in Fig. 3 for a sequence of temperatures, where $\tilde{\omega} = \omega - \omega_0$ [56] is the frequency relative to its minimum near E_F . For $T < T_K \approx 5 \text{ K}$ it approaches the universal scaling form [19,57]

$$\text{Im} \Sigma(\tilde{\omega}) \sim \sqrt{T/T_K} F(x = \tilde{\omega}/k_B T), \quad (9)$$

where $F(x) \propto \sqrt{x}$ for $\sqrt{x} > 1$ in the $T/T_K \rightarrow 0$ case. This is consistent with the experimentally observed temperature dependence of the Pr $4f$ contribution to electrical resistivity

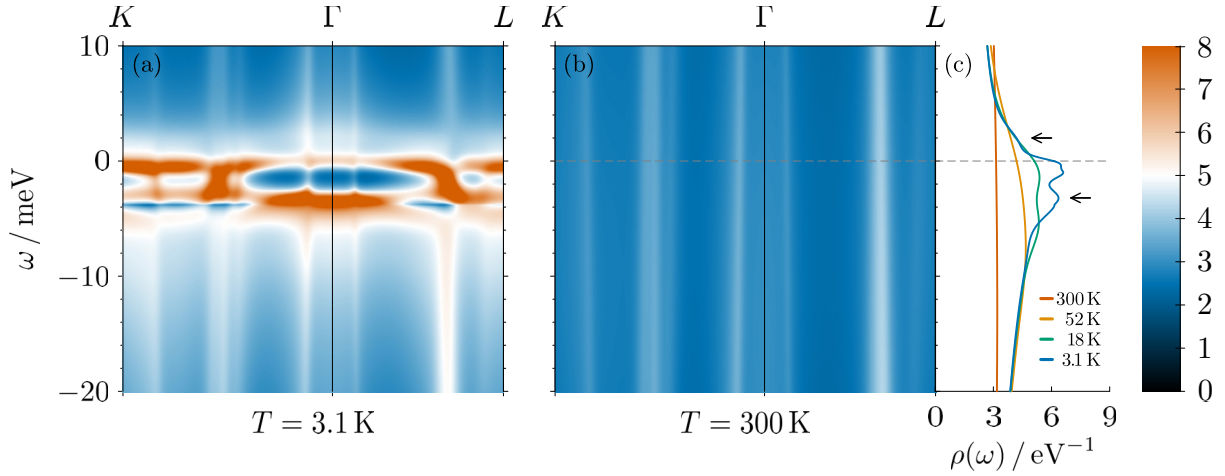


FIG. 2. (a), (b) Momentum-dependent, renormalized spectral functions as obtained from DMFT, traced over the V $d_{xy+x^2-y^2}$ and Pr $4f$ bands, for temperatures $T = 3.1 \text{ K}$ and $T = 300 \text{ K}$. (c) Momentum-integrated density of states for different temperatures. Arrows indicate features at $\omega = -3.2 \text{ meV}$ and $\omega = 2 \text{ meV}$.

[17]. Note that the lattice model obeys the same scaling behavior as the impurity model [19,58] since the k -integrated spectral density of the Kondo flat band resembles that of the single-impurity case. Around $T \approx 5 \text{ K}$, a shoulder at $x \approx 3$ appears and can be attributed to the presence of the Γ_5 triplet. The shape of $\Sigma(\tilde{\omega})$ matches also qualitatively the numerical renormalization group results for a generic 2CK model [59].

The magnetic susceptibility $\chi(T)$ is the key quantity to compare theory and experiment, and to interpret the experimental results on $\text{PrV}_2\text{Al}_{20}$. Experiments [17,25] reported non-Fermi liquid behavior, $[\chi(T) - \chi(0)] \sim -\sqrt{T}$ seemingly in accordance with the theoretically conjectured temperature dependence of the van Vleck susceptibility by Cox and Makivic [60], at intermediate temperatures, $2 \text{ K} \lesssim T \lesssim 30 \text{ K}$ where, however, a precise functional dependence is hard to extract from the finite temperature range, and which is at odds with the log divergence expected for a 2CK system [19]. Theoretically, we calculated $\chi(T)$ within DMFT in a

small magnetic field B coupled to the conduction spins and all the magnetic local Pr states, numerically taking the derivative with respect to B . This includes quantum and thermal fluctuations on the level of DMFT. The theoretical results, displayed in Fig. 4(a), show that at all temperatures, the total susceptibility χ_{tot} is dominated by the magnetic $4f^2 \Gamma_5$ CEF excitation. This is expected since the ground-state Γ_3 doublet, being nonmagnetic, provides only a second-order van Vleck susceptibility, and the $4f^3 \Gamma_6$ doublet is only a virtually occupied state. All contributions show a logarithmic increase down to $T \approx 20 \text{ K}$. By comparison with the spectra of Fig. 2(c) we identify this as the perturbative onset [61] of the Kondo effect which occurs in the 2CK symmetry-breaking Γ_5 triplet as well as in the channel-conserving Γ_6 doublet. Below about $T \approx 11 \text{ K}$, separated by an intermediate maximum which may be attributed to the freezing-out of the Γ_5 CEF excitation, we find a second log increase of $\chi(T)$. Since at these low T , the conduction-electron channel (magnetic spin) is conserved by the $4f^2 \Gamma_3$ and $4f^3 \Gamma_6$ transitions, this may be understood

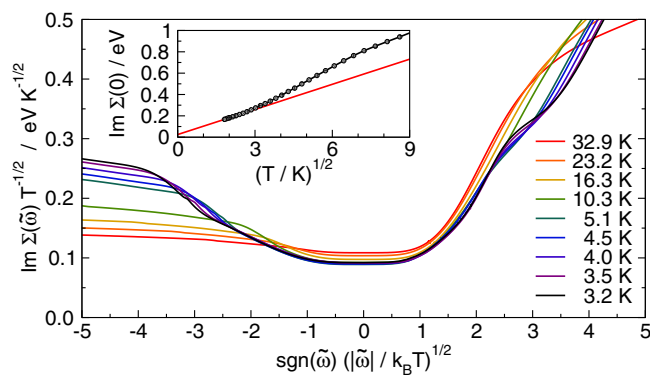


FIG. 3. Imaginary part of the Γ_3 contribution to the impurity self-energy divided by $\sqrt{T}$ plotted against the expected 2CK universal scaling function $\sqrt{\tilde{\omega}/k_B T}$ for different temperatures. The inset shows $\text{Im} \Sigma(\tilde{\omega} = 0)$ plotted against the expected $\sqrt{T}$ dependency and a line as a guide to the eye for the asymptotic behavior.

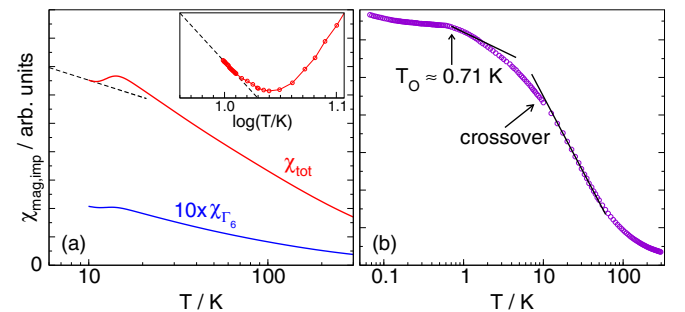


FIG. 4. Magnetic impurity susceptibility from DMFT. (a) The red curve represents the total susceptibility. The blue curve shows the contribution of the $4f^3 \Gamma_6$ doublet (i.e., without the Γ_5 contribution), multiplied by ten for better visibility. The inset shows a blowup of the low-temperature region. The dashed line is an extrapolation by fitting logarithmic behavior to the low-temperature results. (b) Experimental data extracted from Ref. [25]. The solid lines are linear guides to the eye.

as the log divergence characteristic of the 2CK effect [19]. Comparing now these findings with the experimental results of Ref. [25] shown in Fig. 4(b), all the described features can be identified in the experiments, including the high- and low- T log behaviors, where the former is terminated by a crossover point near 10 K that possibly corresponds to the Γ_5 freezeout. Intriguingly, the 2CK low- T log behavior seemingly persists through T_O .

Conclusion. The rich physics of the 1-2-20 family of Pr-based heavy-fermion materials has provided a canvas for several intriguing observations. In the present work, we deliberately ignored the quadrupolar (and possibly octupolar) ordering observed below $T_O \approx 0.71$ K in $\text{PrV}_2\text{Al}_{20}$ [17,62], because this phase has been studied extensively in other works [22,35,63–65]. Instead, we focused on the non-Fermi liquid strange-metal aspect of these materials, whose interpretation in terms of quantum-critical fluctuations near a quantum-critical point is problematic as it does not appear to require fine-tuning $T_O \rightarrow 0$. Our key conclusion is that instead, the observed non-Fermi liquid features naturally stem from the two-channel nature of the quadrupolar Kondo effect. We showed that the qualitative shape of the magnetic susceptibility is faithfully reproduced on all temperature scales above T_O only by including the excited, channel-symmetry-breaking Γ_5 CEF states, which, in turn, mask the pure two-channel Kondo behavior at elevated temperatures. The non-Fermi liquid behavior is clearly visible in the self-energy which below 5 K approaches the $\sqrt{\omega/T}$ scaling behavior expected

of the two-channel Kondo model in the absence of excited Γ_5 hybridization. We thus extract $T_K \approx 5$ K as the two-channel Kondo scale in $\text{PrV}_2\text{Al}_{20}$, which differs from the perturbative Kondo scale $T_K^{(0)} \approx 50$ K. We conclude that the two-channel Kondo scenario with crystal-field excitations correctly describes the susceptibility in accordance with experiments over several different temperature regimes. This provides strong support for this scenario to be realized in $\text{PrV}_2\text{Al}_{20}$. Our findings open up several avenues for future research, including comparing computed spectral functions with future photoemission experiments. Possible future extension of the theory include a detailed calculation and analysis of the specific heat including the Γ_5 CEF state and investigation of the magnetic susceptibility below T_O .

Acknowledgments. The authors thank Y.-B. Kim, R. Flint, and T. Onimaru for discussions. The work at Rice University (F.G. and A.H.N.) was supported by the U.S. National Science Foundation Division of Materials Research under the Award DMR-1917511. The work at the University of Bonn (M.L. and J.K.) was supported in part by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy – Cluster of Excellence “Matter and Light for Quantum Computing,” ML4Q (390534769), and through the DFG Collaborative Research Center CRC 185 OSCAR (277625399). A.H.N. acknowledges the hospitality of the Aspen Center for Physics, supported by the National Science Foundation Grant No. PHY-1607611, where this collaboration was initiated.

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- [1] L. D. Landau, The theory of a fermi liquid, *ZhETF* **30**, 1058 (1956)[*J. Exp. Theor. Phys.* **3**, 920 (1956)].
 - [2] H. V. Löhneysen, Non-Fermi-liquid behaviour in the heavy-fermion system, *J. Phys.: Condens. Matter* **8**, 9689 (1996).
 - [3] J. Paglione, M. A. Tanatar, D. G. Hawthorn, E. Boaknin, R. W. Hill, F. Ronning, M. Sutherland, L. Taillefer, C. Petrovic, and P. C. Canfield, Field-induced quantum critical point in CeCoIn_5 , *Phys. Rev. Lett.* **91**, 246405 (2003).
 - [4] J. Custers, P. Gegenwart, H. Wilhelm, K. Neumaier, Y. Tokiwa, O. Trovarelli, C. Geibel, F. Steglich, C. Pépin, and P. Coleman, The break-up of heavy electrons at a quantum critical point, *Nature (London)* **424**, 524 (2003).
 - [5] P. Gegenwart, Q. Si, and F. Steglich, Quantum criticality in heavy-fermion metals, *Nature Phys* **4**, 186 (2008).
 - [6] Y. Matsumoto, S. Nakatsuji, K. Kuga, Y. Karaki, N. Horie, Y. Shimura, T. Sakakibara, A. H. Nevidomskyy, and P. Coleman, Quantum criticality without tuning in the mixed valence compound b-YbAlB_4 , *Science* **331**, 316 (2011).
 - [7] Q. Si, S. Rabello, K. Ingersent, and J. L. Smith, Locally critical quantum phase transitions in strongly correlated metals, *Nature (London)* **413**, 804 (2001).
 - [8] P. Coleman, C. Pépin, Q. Si, and R. Ramazashvili, How do Fermi liquids get heavy and die? *J. Phys.: Condens. Matter* **13**, R723 (2001).
 - [9] T. Senthil, M. Vojta, and S. Sachdev, Weak magnetism and non-Fermi liquids near heavy-fermion critical points, *Phys. Rev. B* **69**, 035111 (2004).
 - [10] S. Doniach, Kondo lattice and weak antiferromagnetism, *Physica B+C* **91**, 231 (1977).
 - [11] A. Nejati, K. Ballmann, and J. Kroha, Kondo destruction in RKKY-coupled Kondo lattice and multi-impurity systems, *Phys. Rev. Lett.* **118**, 117204 (2017).
 - [12] T. Kuwai, M. Funane, K. Tada, T. Mizushima, and Y. Isikawa, Thermoelectric power anomaly of $\text{PrTi}_2\text{Al}_{20}$ and $\text{PrV}_2\text{Al}_{20}$ with non-Kramers Γ_3 ground state, *J. Phys. Soc. Jpn.* **82**, 074705 (2013).
 - [13] Y. Machida, T. Yoshida, T. Ikeura, K. Izawa, A. Nakama, R. Higashinaka, Y. Aoki, H. Sato, A. Sakai, S. Nakatsuji, N. Nagasawa, K. Matsumoto, T. Onimaru, and T. Takabatake, Anomalous enhancement of seebeck coefficient in Pr-based 1-2-20 system with non-Kramers doublet ground states, *J. Phys.: Conf. Ser.* **592**, 012025 (2015).
 - [14] Y. Tokunaga, H. Sakai, S. Kambe, A. Sakai, S. Nakatsuji, and H. Harima, Magnetic excitations and $c-f$ hybridization effect in $\text{PrTi}_2\text{Al}_{20}$ and $\text{PrV}_2\text{Al}_{20}$, *Phys. Rev. B* **88**, 085124 (2013).
 - [15] Y. Shimura, M. Tsujimoto, B. Zeng, L. Balicas, A. Sakai, and S. Nakatsuji, Field-induced quadrupolar quantum criticality in $\text{PrV}_2\text{Al}_{20}$, *Phys. Rev. B* **91**, 241102(R) (2015).
 - [16] M. Matsunami, M. Taguchi, A. Chainani, R. Eguchi, M. Oura, A. Sakai, S. Nakatsuji, and S. Shin, Kondo resonance in $\text{PrTi}_2\text{Al}_{20}$: Photoemission spectroscopy and single-impurity Anderson model calculations, *Phys. Rev. B* **84**, 193101 (2011).
 - [17] A. Sakai and S. Nakatsuji, Kondo effects and multipolar order in the cubic $\text{PrTr}_2\text{Al}_{20}$ ($\text{Tr} = \text{Ti}, \text{V}$), *J. Phys. Soc. Jpn.* **80**, 063701 (2011).
 - [18] M. Tsujimoto, Y. Matsumoto, and S. Nakatsuji, Anomalous specific heat behaviour in the quadrupolar Kondo system $\text{PrV}_2\text{Al}_{20}$, *J. Phys.: Conf. Ser.* **592**, 012023 (2015).

- [19] I. Affleck and A. W. W. Ludwig, Exact conformal-field-theory results on the multichannel Kondo effect: Single-fermion Greens function, self-energy, and resistivity, *Phys. Rev. B* **48**, 7297 (1993).
- [20] A. Tsuruta and K. Miyake, Non-fermi liquid and fermi liquid in two-channel Anderson lattice model: Theory for $\text{PrA}_2\text{Al}_{20}$ ($A = \text{V, Ti}$) and $\text{PrIr}_2\text{Zn}_{20}$, *J. Phys. Soc. Jpn.* **84**, 114714 (2015).
- [21] K. Inui and Y. Motome, Channel-selective non-Fermi liquid behavior in the two-channel Kondo lattice model under a magnetic field, *Phys. Rev. B* **102**, 155126 (2020).
- [22] A. S. Patri, I. Khait, and Y. B. Kim, Emergent non-Fermi-liquid phenomena in multipolar quantum impurity systems, *Phys. Rev. Res.* **2**, 013257 (2020).
- [23] A. S. Patri and Y. B. Kim, Critical Theory of Non-Fermi liquid fixed point in multipolar kondo problem, *Phys. Rev. X* **10**, 041021 (2020).
- [24] D. J. Schultz, A. S. Patri, and Y. B. Kim, Rise and fall of non-Fermi liquid fixed points in multipolar Kondo problems, *Phys. Rev. Res.* **3**, 013189 (2021).
- [25] K. Araki, Y. Shimura, N. Kase, T. Sakakibara, A. Sakai, and S. Nakatsuji, Magnetization and specific heat of the cage compound $\text{PrV}_2\text{Al}_{20}$, in *Proceedings of the International Conference on Strongly Correlated Electron Systems (SCES2013)* (Journal of the Physical Society of Japan, Tokyo, Japan, 2014).
- [26] This crossover scale turns out to be only marginally larger than the quadrupole-ordering temperature in $\text{PrV}_2\text{Al}_{20}$, such that pure two-channel Kondo physics is obstructed by the quadrupolar order.
- [27] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevResearch.6.L042008> for further details and additional data regarding the computation of the magnetic susceptibility.
- [28] T. Onimaru and H. Kusunose, Exotic quadrupolar phenomena in non-Kramers doublet systems - the cases of $\text{PrT}_2\text{Zn}_{20}$ ($T = \text{Ir, Rh}$) and $\text{PrT}_2\text{Al}_{20}$ ($T = \text{V, Ti}$), *J. Phys. Soc. Jpn.* **85**, 082002 (2016).
- [29] K. Momma and F. Izumi, *VESTA 3* for three-dimensional visualization of crystal, volumetric and morphology data, *J. Appl. Crystallogr.* **44**, 1272 (2011).
- [30] A. Schiller, F. B. Anders, and D. L. Cox, Mixed-valent regime of the two-channel Anderson impurity as a model for UBe_{13} , *Phys. Rev. Lett.* **81**, 3235 (1998).
- [31] H. Kusunose and H. Ikeda, Interplay of crystal field structures with f^2 configuration to heavy fermions, *J. Phys. Soc. Jpn.* **74**, 405 (2005).
- [32] P. Blaha, K. Schwarz, F. Tran, R. Laskowski, G. K. H. Madsen, and L. D. Marks, WIEN2k: An APW+lo program for calculating the properties of solids, *J. Chem. Phys.* **152**, 074101 (2020).
- [33] D. L. Cox and A. Zawadowski, Exotic Kondo effects in metals: Magnetic ions in a crystalline electric field and tunnelling centres, *Adv. Phys.* **47**, 599 (1998).
- [34] Adopting the Γ_7 doublet as the lowest $4f^3$ state, as done, e.g., in Ref. [45] will not qualitatively change any of our conclusions, since in either case the hybridization with the conduction electrons will proceed in the $\Gamma_8 = \Gamma_7 \otimes \Gamma_3$ channel.
- [35] K. Hattori and H. Tsunetsugu, Antiferro quadrupole orders in non-Kramers doublet systems, *J. Phys. Soc. Jpn.* **83**, 034709 (2014).
- [36] K. Hattori and H. Tsunetsugu, Classical Monte Carlo study for antiferro quadrupole orders in a diamond lattice, *J. Phys. Soc. Jpn.* **85**, 094001 (2016).
- [37] D. L. Cox, Quadrupolar Kondo effect in uranium heavy-electron materials? *Phys. Rev. Lett.* **59**, 1240 (1987).
- [38] N. Andrei and C. Destri, Solution of the multichannel kondo problem, *Phys. Rev. Lett.* **52**, 364 (1984).
- [39] V. J. Emery and S. Kivelson, Mapping of the two-channel kondo problem to a resonant-level model, *Phys. Rev. B* **46**, 10812 (1992).
- [40] J. Hubbard and B. H. Flowers, Electron correlations in narrow energy bands. II. The degenerate band case, *Proc. R. Soc. London A* **277**, 237 (1964).
- [41] P. Coleman, New approach to the mixed-valence problem, *Phys. Rev. B* **29**, 3035 (1984).
- [42] S. E. Barnes, New method for the Anderson model. II. The $U=0$ limit, *J. Phys. F* **7**, 2637 (1977).
- [43] A. A. Abrikosov, Electron scattering on magnetic impurities in metals and anomalous resistivity effects, *Phys. Phys. Fiz.* **2**, 5 (1965).
- [44] T. A. Costi, J. Kroha, and P. Wölfle, Spectral properties of the Anderson impurity model: Comparison of numerical-renormalization-group and noncrossing-approximation results, *Phys. Rev. B* **53**, 1850 (1996).
- [45] J. S. Van Dyke, G. Zhang, and R. Flint, Field-induced ferromagnetic phase in cubic non-Kramers doublet systems, *Phys. Rev. B* **100**, 205122 (2019).
- [46] This is very similar to the structure introduced in Ref. [45], with the only difference that those authors have assumed the lowest excited valence state to be $|4f^1, \Gamma_7\rangle$, different from the state $|4f^3, \Gamma_6\rangle$ that our *ab initio* calculations have found, see Fig. 1(a).
- [47] Note that a non-conservation in the scattering does not necessarily lead to a broken degeneracy in the conduction electrons. This is because the physical conduction electron self-energy is given by a sum over α and the projected electrons are calculated from the physical electrons.
- [48] S. E. Han, D. J. Schultz, and Y. B. Kim, Non-Fermi liquid behavior and quantum criticality in cubic heavy fermion systems with non-Kramers multipolar local moments, *Phys. Rev. B* **106**, 155155 (2022).
- [49] A. Georges and G. Kotliar, Hubbard model in infinite dimensions, *Phys. Rev. B* **45**, 6479 (1992).
- [50] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions, *Rev. Mod. Phys.* **68**, 13 (1996).
- [51] Y. Kuramoto and T. Watanabe, Theory of momentum-dependent magnetic response in heavy-fermion systems, *Physica B+C* **148**, 80 (1987).
- [52] Y. Kuramoto, Self-consistent perturbation theory for dynamics of valence fluctuations: I. Single-site theory, *Z. Phys. B* **53**, 37 (1983).
- [53] H. Keiter and J. C. Kimball, Diagrammatic approach to the Anderson model for dilute alloys, *J. Appl. Phys.* **42**, 1460 (1971).
- [54] D. L. Cox and A. E. Ruckenstein, Spin-flavor separation and non-Fermi-liquid behavior in the multichannel Kondo problem: A large- N approach, *Phys. Rev. Lett.* **71**, 1613 (1993).
- [55] D. Ehm, S. Hüfner, F. Reinert, J. Kroha, P. Wölfle, O. Stockert, C. Geibel, and H. V. Löhneysen, High-resolution photoemission

- study on low-TK Ce systems: Kondo resonance, crystal field structures, and their temperature dependence, *Phys. Rev. B* **76**, 045117 (2007).
- [56] The small frequency shift ω_0 is due to NCA overestimating potential scattering contributions [66].
- [57] D. C. Ralph, A. W. W. Ludwig, J. von Delft, and R. A. Buhrman, 2-channel kondo scaling in conductance signals from 2 level tunneling systems, *Phys. Rev. Lett.* **72**, 1064 (1994).
- [58] This has been explored on several occasions for the single-channel case, e.g. [55,67] and was also observed in the two-channel case for a wide temperature range in [20,21]. However, when non-local effects dominate, e.g. in an ordered phase, the behavior can deviate.
- [59] F. B. Anders, Renormalization-group approach to spectral properties of the two-channel Anderson impurity model, *Phys. Rev. B* **71**, 121101(R) (2005).
- [60] D. L. Cox and M. Makivic, Phenomenology of two channel Kondo alloys and compounds, *Phys. B: Condens. Matter* **199-200**, 391 (1994).
- [61] The perturbative onset of the Kondo effect involving both considered CEF multiplets at intermediate temperatures generically results in an initially logarithmic increase of the susceptibility and is *not* a signature unique to two-channel models.
- [62] L. Ye, M. E. Sorensen, M. D. Bachmann, and I. R. Fisher, Measurement of the magnetic octupole susceptibility of $\text{PrV}_2\text{Al}_{20}$, *Nat. Commun.* **15**, 7005 (2024).
- [63] T. Ishitobi and K. Hattori, Triple- q quadrupole-octupole order scenario for $\text{PrV}_2\text{Al}_{20}$, *Phys. Rev. B* **104**, L241110 (2021).
- [64] F. Freyer, J. Attig, S. B. Lee, A. Paramakanti, S. Trebst, and Y. B. Kim, Two-stage multipolar ordering in $\text{PrT}_2\text{Al}_{20}$ Kondo materials, *Phys. Rev. B* **97**, 115111 (2018).
- [65] F. Freyer, S. B. Lee, Y. B. Kim, S. Trebst, and A. Paramakanti, Thermal and field-induced transitions in ferroquadrupolar Kondo systems, *Phys. Rev. Res.* **2**, 033176 (2020).
- [66] S. Kirchner and J. Kroha, Self-consistent conserving theory for quantum impurity systems: Renormalization group analysis, *J. Low Temp. Phys.* **126**, 1233 (2002).
- [67] F. Reinert, D. Ehm, S. Schmidt, G. Nicolay, S. H fner, J. Kroha, O. Trovarelli, and C. Geibel, Temperature dependence of the Kondo resonance and its satellites in CeCu_2Si_2 , *Phys. Rev. Lett.* **87**, 106401 (2001).