Excess heat capacity in magnetically ordered Ce heavy-fermion metals

A. Scheie,[1,*] Yu Liu,[1] E. A. Ghioldi,[2] S. Fender,[1] P. F. S. Rosa,[1] E. D. Bauer,[1] Jian-Xin Zhu,[3] and F. Ronning[1,†]
[1]*MPA-Q, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA*
[3]*Theoretical Division and Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

(Received 18 January 2024; revised 21 June 2024; accepted 19 July 2024; published 9 August 2024)

We study the magnetic heat capacity of a series of magnetically ordered Ce-based heavy-fermion materials, which show an anomalous T^3 heat capacity in excess of the phonon contribution in many materials. For compounds for which magnon models have been worked out, we show that the local-moment magnon heat capacity derived from the measured magnon spectra underestimates the experimental specific heat. The excess heat capacity reveals increasing density of states with increasing energy, akin to a pseudogap. We show that this anomalous temperature-dependent term is not associated with proximity to a quantum critical point, but is strongly correlated with T_N, indicating the anomalous excitations are governed by the magnetic exchange interaction. This insight may hold key information for understanding magnetically ordered heavy fermions.

DOI: 10.1103/PhysRevB.110.085123

I. INTRODUCTION

First discovered in the 1970s [1,2], heavy fermions are a prototypical problem of strongly correlated electron systems [3–8]. Deriving their name from an anomalously large effective electron mass at low temperatures, these materials display a variety of strongly correlated quantum phases, including non-Fermi liquids [8,9], unconventional superconductivity [7], volume collapse [10], topological Kondo insulators [11], and hidden order [12]. Many share similar phenomenology of quantum criticality, summarized by the famous Doniach phase diagram [13]. It is known that heavy-fermion behavior arises from the interactions between local and itinerant electrons. Yet, despite decades of work, there is no microscopic model able to account for their behavior. This signifies key gaps in our understanding of superconductivity, nonquasiparticle transport, and fundamental many-body quantum physics.

As the list of heavy-fermion materials continues to grow but theory is still lacking, one route to explaining heavy fermions is looking for trends across materials families [14,15]. In this paper, we focus on the heat capacity of magnetically ordered Ce heavy-fermion materials [16]. Beginning with $CeIn_3$, we show that a common feature of these compounds is an anomalous density of states at low energies (in addition to a T-linear Sommerfeld coefficient), often taking the form of an approximate T^3 term in heat capacity. In certain cases, where a rigorous magnon model has been worked out, we show that the experimental heat capacity far exceeds the bosonic magnon heat capacity at low temperatures. Correlation analysis shows this density of states to be uncorrelated with proximity to quantum criticality but strongly correlated with the ordering temperature. Thus, these anomalous excitations are related to the magnetic exchange interaction.

II. EXPERIMENTS AND ANALYSIS

A. $CeIn_3$

By way of introduction, let us begin by examining the heat capacity of the magnetically ordered heavy-fermion system $CeIn_3$. This compound magnetically orders at $T_N = 10.23(1)$ K [17], and has superconducting and non-Fermi-liquid properties under pressure [18,19]. We measured its heat capacity using a Quantum Design physical property measurement system (PPMS) from 0.4 to 20 K that utilizes a quasiadiabatic thermal relaxation method, and the data are shown in Fig. 1. Plotting the data on a C/T vs T^2 graph shows a nearly straight line below T_N, indicating T^3 heat capacity (slope) with a T-linear term (y-axis offset). The T-linear term is explicable (at least phenomenologically) as a Sommerfeld term from the enhanced fermion mass [20]. The T^3 term, however, is much larger than the phonon heat capacity (approximated by the nonmagnetic $LaIn_3$ [21]) and is more of a challenge.

In theory, gapless linear dispersive magnons in three dimensions in the low-temperature limit give T^3 heat capacity

$$c_{\mathrm{mol}} = N_A v_0 \frac{4\pi^2}{15} k_B \left(\frac{k_B T}{h v}\right)^3, \tag{1}$$

where v_0 is the volume of the unit cell, h is Planck's constant, and v is the velocity of the modes [20]. Recent $CeIn_3$ neutron scattering studies have shown gapless linear dispersive magnons with a velocity of $v \approx 600$ m/s [22]. However, the calculated heat capacity from such modes via Eq. (1), shown in Fig. 1(a), underestimates the specific heat by two orders of magnitude. (The slope of heat capacity suggests a magnon

*Contact author: scheie@lanl.gov
†Contact author: fronning@lanl.gov

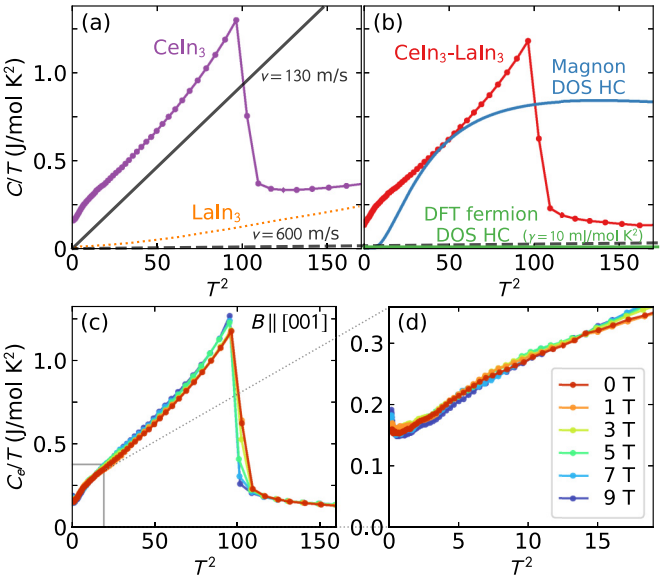

FIG. 1. $CeIn_3$ heat capacity compared to magnon models. (a) shows the simplistic heat capacity from Eq. (1), with the low-energy velocity from the neutron-derived magnon model $v \approx 600$ m/s [22]. This is two orders of magnitude smaller than experiment. Also shown is the heat capacity of nonmagnetic analog $LaIn_3$ from Ref. [21]. (b) shows the calculated heat capacity from a more sophisticated model, integrating over the whole Brillouin zone for the magnon band structure (blue) and the electron band structure (green). Both significantly underestimate the low-energy density of states compared to the electronic specific heat C_e. (c) shows the [001] field-dependent heat capacity, with (d) as a closer view of the low-T behavior. Application of a 9 T field makes almost no difference to the T^3 heat capacity, contrary to expected magnon behavior.

velocity $v \approx 130$ m/s, inconsistent with the neutron results.) We can improve this calculation by instead integrating over the full magnon band structure rather than just the bottom of the dispersion. Taking the $CeIn_3$ magnon dispersion from Ref. [22], one can more rigorously calculate the heat capacity by numerically integrating over the entire Brillouin zone,

$$c_v = k_B \sum_s \int dk \left(\frac{\hbar\omega_s(k)}{k_B T}\right)^2 \frac{e^{\hbar\omega_s(k)/k_B T}}{(e^{\hbar\omega_s(k)/k_B T} - 1)^2}, \qquad (2)$$

summing over s magnon modes where $\omega_s(k)$ are the mode dispersions [20]. The results of these calculations are shown in Fig. 1(b), where C_e is the electronic (phonon-subtracted) specific heat. The calculated heat capacity comes close to the $C_e(T)/T$ data near 5 K (where the validity of the theory is questionable—the expansion is only valid when the moment size is near saturation, likely $T \lesssim \frac{T_N}{2}$), but the calculated heat capacity is far too small below ~3 K. (For magnetic materials where this calculation matches well, cf. $RbMnF_3$ [23,24] or $Nd_2Zr_2O_7$ [25].) Clearly, the anomalously large T^3 heat capacity cannot be explained by the derived local-moment magnon model.

Further evidence against the T^3 heat capacity being magnons is found in the field-dependent data, shown in Figs. 1(c) and 1(d). Ordinarily, a magnetic field shifts gapless magnon bands up in energy, decreasing the low-energy density of states and suppressing the low-temperature Eq. (2) heat capacity [26,27]. However, the heat capacity below T_N is barely affected by a magnetic field, indicating this density of states is not from local moment magnons.

As a final attempt to explain the $CeIn_3$ T^3 heat capacity, we calculate the electron band structure with density functional theory (DFT). Using the $CeIn_3$ experimental crystal structure, we performed DFT calculations by using a full-potential linearized augmented plane wave (FP-LAPW) as implemented in the WIEN2K code [28]. On top of the generalized gradient approximation (GGA) [29] for the exchange-correlation functional, we used a value of Hubbard $U_{\text{eff}} = 6.0$ eV on Ce-$4f$ electrons for a G-type antiferromagnetically ordered state with the magnetization imposed along the (111) direction. The spin-orbit coupling was included in a second variational way. A plane-wave cutoff $RK_{\max} = 8$ was taken with $12 \times 12 \times 12$ **k** points. The resulting heat capacity, calculated via Eq. (2) but with fermionic statistics, is shown as the green line in Fig. 1(b). Not only does it vastly underestimate the Sommerfeld γ term ($\gamma = 9.88$ mJ/mol K^2), it has virtually no T^3 dependence with a T^3 prefactor $2.37(3) \times 10^{-5}$ J/mol K^4 for $T^2 < 50$ K^2, six orders of magnitude smaller than $CeIn_3$'s fitted T^3 prefactor 10.31(11) J/mol K^4. [If we renormalize the DFT band-structure energy to a yield larger density of states (DOS) near the Fermi energy and match the empirical $\gamma = 130$ mJ/mol K^2, this still falls far short with a T^3 prefactor $1.76(2) \times 10^{-2}$ J/mol K^4 for $T^2 < 10$ K^2, still three orders of magnitude too small.] Thus, DFT electronic band structures are unable to explain the T^3 heat capacity. This is not so surprising, as DFT often struggles to capture strong correlations between electrons.

Clearly, there is some significant density of states at low energy that pure magnon and pure electron band theory fails to capture. The strong correlations in $CeIn_3$ produce a substantial energy-dependent density of states (i.e., T^3 specific heat), not merely an enhanced electron mass (which would give T-linear specific heat).

B. Other compounds

Having observed such behavior in one magnetically ordered heavy-fermion material, a natural question is how general is this behavior. In Fig. 2, we compare experimental lattice-subtracted heat capacity to magnon heat capacity for five magnetically ordered heavy-fermion materials for which a magnon model exists: $CeRhIn_5$ [31], $CePd_2Si_2$ [32], $CeCu_2Ge_2$ [33], and $CePt_3Si$ [34] (see the Supplemental Material [35], and also Refs. [36,37] therein). The data from these compounds, and the calculated magnon specific heat [Eq. (2)], are shown in Figs. 2(a)–2(e). (For $CeCu_2Ge_2$ a proper magnon model does not exist, and the calculated magnon heat capacity is from an Einstein mode with the energy of the flat band measured in Ref. [33].)

In every case, there is a large temperature-dependent specific heat term in the experimental data that can be not accounted for by the magnon model. This is made more evident by the lower row of Figs. 2(f)–2(j), where the magnon calculated specific heat has been subtracted from the experimental heat capacity. In all compounds, the residual specific heat has a peak at low temperatures, which vaguely resembles a Schottky anomaly (indicated by the red lines). This is true

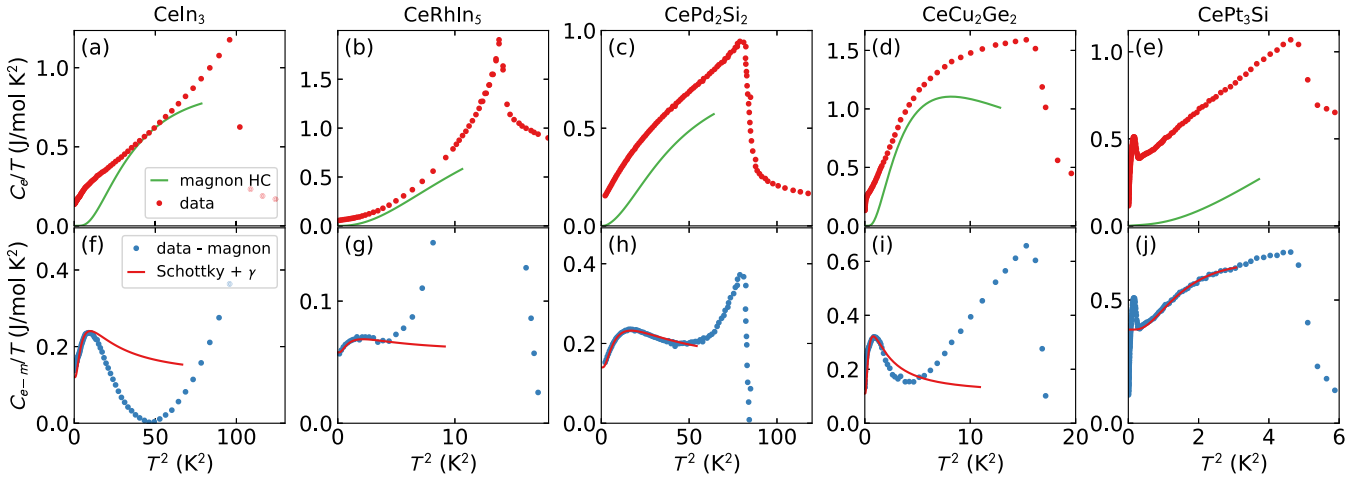

FIG. 2. Electronic heat capacity of five different magnetically ordered heavy-fermion materials for which a magnon model is available. (a)–(e) show the raw data, which has had the lattice contribution subtracted (red) compared to the calculated magnon heat capacity (green). The bottom row (f)–(j) shows the data with the magnon model subtracted (C_{e-m}), compared to a Schottky anomaly offset by a γ term. In each case the extra density of states has the character of a (pseudo)gapped density of states. Contrast this with the Kondo effect heat capacity, where C/T monotonically decreases with temperature [30].

even for $CeRhIn_5$, which has the smallest γ value of the five compounds. This suggests some kind of (pseudo)gap in the density of states, wherein the density of states increases with increasing energy—or equivalently, a depletion of density of states at the lowest energies. In each compound, the excess heat capacity rises to 20%–50% of the γ value ($T \to 0$ K), by no means a small contribution. Furthermore, the pseudogap energy is consistently the same order as T_N (see the Supplemental Material [35]), suggesting an energy scale governed by the magnetic order.

The five compounds in Fig. 2 had the luxury of a fitted magnon model, but we gain more insight by extending this analysis to a broader set of compounds. In Table I we examine 17 different magnetically ordered Ce-based heavy-fermion materials. Taking their heat capacities below T_N from the literature, we fit the lowest-temperature data to

$$c = \gamma T + N_a k_B \left(\frac{T}{T_\beta}\right)^3, \tag{3}$$

where N_a is Avogadro's number and T_β serves as a magnetic analog of the Debye temperature. (This is not meant to imply that the true nonmagnon specific heat is T^3 over many decades, but is meant to capture the lowest-temperature behavior which, as Fig. 2 shows, is mainly preserved when the local-moment magnon contribution is subtracted.) The fits are shown in the Supplemental Material [35], and the fitted values are listed in Table I.

Interestingly, we find many Ce-based heavy-fermion materials with a large low-temperature T^3 heat capacity. In some

TABLE I. Experimental properties of various magnetically ordered Ce heavy-fermion materials. γ (Sommerfeld coefficient), P_c (critical pressure), and T_N (Néel temperature) are taken from the literature, but T_β is fitted to the data found in the reference indicated. *Note that $CeRh_6Ge_4$ is a ferromagnet with T_c rather than T_N.

Compound	γ (mJ/mol K^2)	P_c (GPa)	T_N (K)	T_β (K)
$CeIn_3$	130	2.65 [19]	10.23(1) [17]	9.31(3)
$CeRhIn_5$	70 [38]	2.3 [39]	3.8 [40]	7.41(7) [41]
Ce_2RhIn_8	400 [42]	1.36 [43]	2.8 [42]	4.445(14) [41]
$CePt_2In_7$	50 [44]	3.5 [44]	5.5 [44]	7.02(7) [44]
$CePd_5Al_2$	56 [45]	10.8 [45]	2.87 [45]	3.274 [46]
$CePd_2Si_2$	131 [47]	2.87 [48]	9.3 [47]	8.072(15) [47]
$CeRh_2Si_2$	22.8[49]	0.97[48]	36[50]	56(5) [51]
$CeCu_2Ge_2$	77 [52]	7.7 [53]	4.15(5) [52]	3.51(2) [52]
$Ce_2Ni_3Ge_5$	90 [54]	3.9 [55]	4.3 [54]	3.551(7) [54]
$CeNiGe_3$	76 [56]	5.5 [57]	5 [56]	5.166(9) [56]
$CePt_3Si$	335 [58]	0.6 [58]	2.25 [58]	3.659(9) [58]
$CeRhSi_3$	110 [59]	2.36 [60]	1.6 [59]	3.44(2) [59]
$CeIrSi_3$	105 [61]	2.63 [62]	5 [61]	9.5(3) [61]
$CeCoGe_3$	32 [63]	5.5 [63]	21 [64]	28.0(9) [64]
CePdAl	250 [65]	0.92 [66]	2.7 [65]	2.862(7) [65]
$CeRh_6Ge_4$	250 [67]	0.85 [68]	2.5 [67]*	4.19(5) [67]

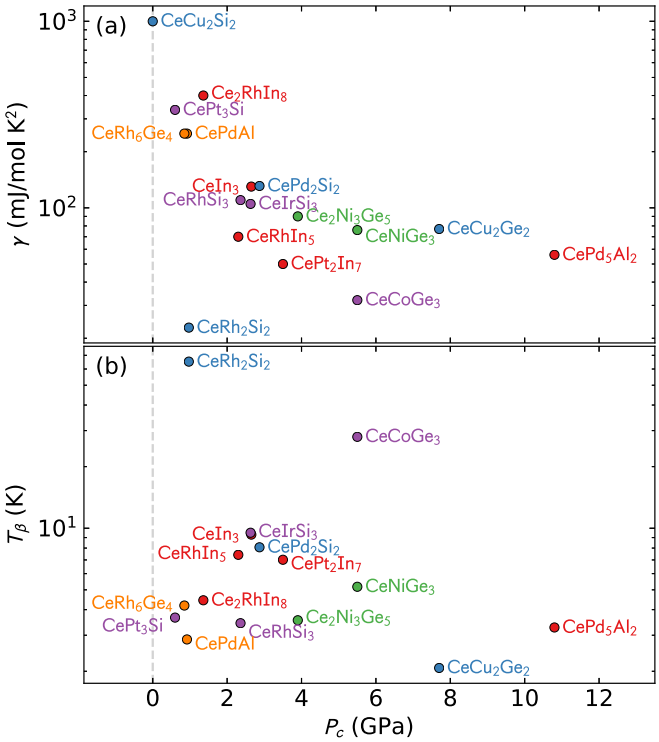

FIG. 3. Correlations between physical properties for various magnetically ordered heavy-fermion materials. (a) Sommerfeld coefficient γ vs critical pressure P_c, showing a clear trend of increasing γ as P_c decreases. (b) T^3 term β vs P_c, showing no clear correlation. Colors indicate families of materials. Data for this plot are shown in Table I.

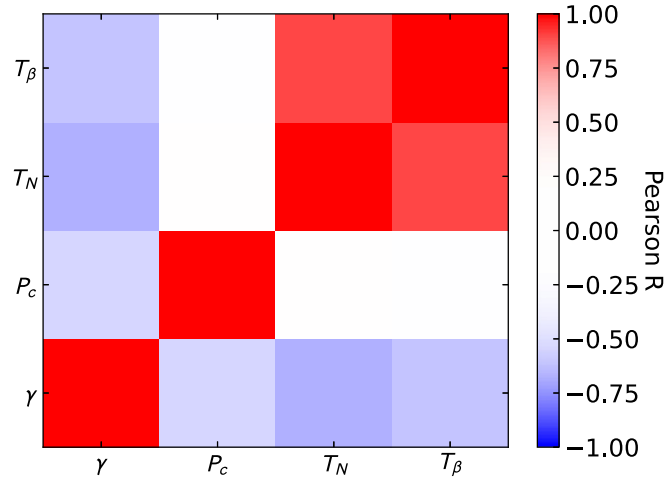

FIG. 4. Correlation matrix between physical properties as indicated by the Pearson R coefficient. Red indicates positive correlation, and blue indicates negative correlation. T_β is strongly correlated only with T_N, and weakly correlated with γ, but essentially uncorrelated with P_c, indicating that it is not a function of proximity to a quantum critical point (QCP).

cases this exists over a full decade in temperature. One might wonder if this is correlated with how "close" the system is to quantum criticality. If we take the critical pressure P_c (at which magnetic ordering temperature goes to $T = 0$) as a measure of this, we can answer this question empirically.

Figure 3 plots the γ and T_β terms of the various compounds against P_c. For γ, there is a clear trend: The closer to criticality, the larger is the γ (with one outlier, $CeRh_2Si_2$, which also has an anomalously large T_N). This is as expected for mass renormalization driven by quantum criticality. For T_β, however, there is no apparent trend: The T^3 specific heat appears to be uncorrelated with P_c.

We can be more precise about these trends by using Pearson's R correlation coefficient. Applying this to the logarithm of the data (to account for nonlinear trends) in Table I yields a correlation matrix, plotted in Fig. 4. This reveals a very strong correlation between T_β and T_N, weak correlation between T_β and γ, and virtually no correlation between T_β and P_c. Therefore, the T^3 heat capacity is not dependent on proximity to quantum criticality, but instead seems to be closely related to T_N (indeed, in the Supplemental Material [35], we show this relationship is essentially linear). Thus, this excess density of states seems to be governed by magnetic exchange interactions.

This is consistent with the behavior of $CeRhIn_5$ under hydrostatic pressure: As this compound approaches the QCP, the γ value grows but T_β shrinks as T_N is suppressed [38]. This was interpreted as "decreasing spin-wave stiffness," but our results here indicate that it is not spin waves at all, but of some other origin.

One weakness of the above correlation analysis is that it does not consider how much of the T^3 heat capacity comes from magnons alone. However, the examples of $CeIn_3$ and other compounds in Fig. 2 show the local-moment magnons come nowhere near explaining the heat capacity in the magnetically ordered state, suggesting it holds across the heavy-fermion family.

One additional example, not included in Fig. 2, is $CeRh_6Ge_4$. This material is a ferromagnet [67], which should have $T^{3/2}$ heat capacity at low temperatures because of its quadratic magnon dispersion [20], but the magnetic specific heat is also definitively T^3 below T_N (see the Supplemental Material [35]). This alone signals a significant discrepancy, but because there is no magnon model it is difficult to say how severe is the difference between the measured and magnon heat capacity.

III. DISCUSSION

At this point, we are left with a quandary. We have shown that a large number of magnetically ordered Ce heavy-fermion materials have an anomalous temperature-dependent heat capacity which often approximates T^3 as $T \to 0$, and this term is not related to QCP proximity. It is tempting to invoke heretofore unobserved Dirac fermions to explain these density of states. After all, a Dirac cone dispersion (linear dispersing bands) generically produces T^3 specific heat, and proposed Weyl-Kondo semimetal states in heavy fermions predict precisely such a density of states at the Fermi energy [69–71]. If this explanation is correct, it indicates that such behavior is far more common in the heavy fermions than previously thought. However, this explanation does not readily explain the correlation with T_N. Furthermore, because the velocity

would have to be very small, it constrains the linear crossing to be close to the Fermi energy, for which no mechanism is known.

Generically, coupling to bosonic fluctuations (e.g., electron-hole pairs, magnons, or phonons) will also create a $T^3 \ln T$ contribution to the specific heat of a Fermi liquid [72]. However, such a Fermi-liquid correction from electron-hole pairs can be ruled out because the correction is the wrong sign from that which is observed. A correction due to coupling to magnons can also be ruled out on the basis that a magnetic field will gap out the magnons, while the $CeIn_3$ experimental heat capacity is essentially unchanged up to 9 T [Fig. 1(c)] (a similar low-field independence is observed in the other Fig. 2 compounds [47,52,58,73]). Finally, a correction from coupling to phonons appears inconsistent with the observed correlation between T_β and T_N, which suggests a magnetic origin; and the size of the excess heat capacity relative to the electronic term implies something beyond a perturbative correction to the Sommerfeld term.

This observation of a pseudogapped density of states in magnetically ordered heavy fermions begs for an explanation. As it cannot be explained by electrons, magnons, or phonons alone, it suggests an entanglement between various degrees of freedom. For instance, it could be that the excess T^3 heat capacity arises from coherent spin waves in the itinerant electron bands that lie below the particle-hole continuum [74]. If such physics could be produced by a staggered field of magnetic order (which remains to be seen), one could have density of states governed by magnetic exchange but in the itinerant electron bands—but this is speculation at this point.

An interesting question, but beyond the scope of this paper, is how common the pseudogap feature is in other types of compounds. A similar pseudogapped density of states has been observed in nonmagnetically ordered $Ce_3Bi_4Pd_3$ [75] and elemental plutonium [76], indicating generic heavy-fermion behavior even beyond magnetically ordered systems. Another interesting question which may be addressed with a broader survey of compounds is whether the pseudogapped density of states correlates with the sharpness of the magnetic transition.

IV. SUMMARY AND CONCLUSION

In summary, we have shown that a large number of magnetically ordered Ce heavy fermions display anomalous, substantial T^3 specific heat inside their magnetic ordered phases. Comparison to the few materials for which magnon models exist shows that this heat capacity is not due to local-moment magnons. The T^3 term is not correlated with the critical pressure, indicating this effect is not due to QCP proximity, but is strongly correlated with the ordering temperature T_N, indicating the effect is governed by the magnetic exchange interaction. These results highlight a behavior: A Sommerfeld $\sim\gamma T$ term is insufficient to capture the density of states of the magnetically ordered Ce materials. Although it is perhaps not surprising that simplistic local-moment models fail to describe strongly correlated systems such as magnetically ordered heavy fermions, this paper highlights exactly how such models fail, and shows the fruitfulness of examining correlations across materials families. More importantly, the identified pseudogap will hopefully sharpen the theoretical studies of this fascinating class of strongly correlated materials.

ACKNOWLEDGMENTS

We gratefully acknowledge the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Science and Engineering under project "Quantum Fluctuations in Narrow-Band Systems." DFT calculations were done with support from LANL LDRD Program. The authors also acknowledge helpful discussions with Shizeng Lin and Cristian Batista.

[1] K. Andres, J. E. Graebner, and H. R. Ott, Phys. Rev. Lett. **35**, 1779 (1975).
[2] F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz, and H. Schäfer, Phys. Rev. Lett. **43**, 1892 (1979).
[3] G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).
[4] Z. Fisk, J. Sarrao, J. Smith, and J. Thompson, Proc. Natl. Acad. Sci. USA **92**, 6663 (1995).
[5] E. Pavarini, P. Coleman, and E. Koch, Many-body physics: From Kondo to Hubbard, Tech. Rep. Theoretische Nanoelektronik, 2015, http://hdl.handle.net/2128/9255 (unpublished).
[6] S. Wirth and F. Steglich, Nat. Rev. Mater. **1**, 16051 (2016).
[7] F. Steglich and S. Wirth, Rep. Prog. Phys. **79**, 084502 (2016).
[8] V. R. Shaginyan, A. Z. Msezane, and G. S. Japaridze, Atoms **10**, 67 (2022).
[9] Q. Si and F. Steglich, Science **329**, 1161 (2010).
[10] M. Lavagna, C. Lacroix, and M. Cyrot, Phys. Lett. A **90**, 210 (1982).
[11] M. Dzero, K. Sun, V. Galitski, and P. Coleman, Phys. Rev. Lett. **104**, 106408 (2010).
[12] J. Mydosh and P. Oppeneer, Philos. Mag. **94**, 3642 (2014).
[13] S. Doniach, Phase diagram for the Kondo lattice, in *Valence Instabilities and Related Narrow-Band Phenomena*, edited by R. D. Parks (Springer, Boston, MA, 1977), pp. 169–176.
[14] K. Kadowaki and S. Woods, Solid State Commun. **58**, 507 (1986).
[15] K. G. Wilson, Rev. Mod. Phys. **47**, 773 (1975).
[16] Z. F. Weng, M. Smidman, L. Jiao, X. Lu, and H. Q. Yuan, Rep. Prog. Phys. **79**, 094503 (2016).
[17] J. M. Lawrence and S. M. Shapiro, Phys. Rev. B **22**, 4379 (1980).
[18] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. W. Haselwimmer, and G. G. Lonzarich, Nature (London) **394**, 39 (1998).
[19] G. Knebel, D. Braithwaite, P. C. Canfield, G. Lapertot, and J. Flouquet, Phys. Rev. B **65**, 024425 (2001).
[20] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Cengage Learning, Boston, MA, 1976).

[21] N. Berry, E. M. Bittar, C. Capan, P. G. Pagliuso, and Z. Fisk, Phys. Rev. B **81**, 174413 (2010).

[22] W. Simeth, Z. Wang, E. A. Ghioldi, D. M. Fobes, A. Podlesnyak, N. H. Sung, E. D. Bauer, J. Lass, S. Flury, J. Vonka, D. G. Mazzone, C. Niedermayer, Y. Nomura, R. Arita, C. D. Batista, F. Ronning, and M. Janoschek, Nat. Commun. **14**, 8239 (2023).

[23] C. G. Windsor and R. W. H. Stevenson, Proc. Phys. Soc. London **87**, 501 (1966).

[24] L. J. D. Jongh and A. R. Miedema, Adv. Phys. **50**, 947 (2001).

[25] A. Scheie, M. Sanders, X. Gui, Y. Qiu, T. R. Prisk, R. J. Cava, and C. Broholm, Phys. Rev. B **104**, 134418 (2021).

[26] S. M. Rezende, A. Azevedo, and R. L. Rodríguez-Suárez, J. Appl. Phys. **126**, 151101 (2019).

[27] B. C. Passenheim, D. C. McCollum, Jr., and J. Callaway, Phys. Lett. **23**, 634 (1966).

[28] P. Blaha, K. Schwarz, G. K. Madsen, D. Kvasnicka, J. Luitz, *et al.*, *WIEN2k: An Augmented Plane Wave+ Local Orbitals Program for Calculating Crystal Properties* (Karlheinz Scwartz, TU Vienna, 2001).

[29] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).

[30] H.-U. Desgranges and K. Schotte, Phys. Lett. A **91**, 240 (1982).

[31] P. Das, S.-Z. Lin, N. J. Ghimire, K. Huang, F. Ronning, E. D. Bauer, J. D. Thompson, C. D. Batista, G. Ehlers, and M. Janoschek, Phys. Rev. Lett. **113**, 246403 (2014).

[32] N. H. van Dijk, B. Fåk, T. Charvolin, P. Lejay, and J. M. Mignot, Phys. Rev. B **61**, 8922 (2000).

[33] G. Knopp, A. Loidl, K. Knorr, L. Pawlak, M. Duczmal, R. Caspary, U. Gottwick, H. Spille, F. Steglich, and A. P. Murani, Z. Phys. B **77**, 95 (1989).

[34] B. Fåk, S. Raymond, D. Braithwaite, G. Lapertot, and J.-M. Mignot, Phys. Rev. B **78**, 184518 (2008).

[35] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.110.085123 for more details of the experiments and calculations.

[36] H. Hodovanets, S. L. Bud'ko, W. E. Straszheim, V. Taufour, E. D. Mun, H. Kim, R. Flint, and P. C. Canfield, Phys. Rev. Lett. **114**, 236601 (2015).

[37] W. Knafo, S. Raymond, B. Fåk, G. Lapertot, P. C. Canfield, and J. Flouquet, J. Phys.: Condens. Matter **15**, 3741 (2003).

[38] R. A. Fisher, F. Bouquet, N. E. Phillips, M. F. Hundley, P. G. Pagliuso, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. B **65**, 224509 (2002).

[39] F. Ronning, T. Helm, K. Shirer, M. Bachmann, L. Balicas, M. K. Chan, B. Ramshaw, R. D. Mcdonald, F. F. Balakirev, M. Jaime *et al.*, Nature (London) **548**, 313 (2017).

[40] H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).

[41] A. L. Cornelius, P. G. Pagliuso, M. F. Hundley, and J. L. Sarrao, Phys. Rev. B **64**, 144411 (2001).

[42] T. Ueda, H. Shishido, S. Hashimoto, T. Okubo, M. Yamada, Y. Inada, R. Settai, H. Harima, A. Galatanu, E. Yamamoto, N. Nakamura, K. Sugiyama, T. Takeuchi, K. Kindo, T. Namiki, Y. Aoki, H. Sato, and Y. Ōnuki, J. Phys. Soc. Jpn. **73**, 649 (2004).

[43] M. Yashima, S. Taniguchi, H. Miyazaki, H. Mukuda, Y. Kitaoka, H. Shishido, R. Settai, and Y. Ōnuki, J. Phys.: Conf. Ser. **200**, 012238 (2010).

[44] E. D. Bauer, H. O. Lee, V. A. Sidorov, N. Kurita, K. Gofryk, J.-X. Zhu, F. Ronning, R. Movshovich, J. D. Thompson, and T. Park, Phys. Rev. B **81**, 180507(R) (2010).

[45] F. Honda, M.-A. Measson, Y. Nakano, N. Yoshitani, E. Yamamoto, Y. Haga, T. Takeuchi, H. Yamagami, K. Shimizu, R. Settai, and Y. Ōnuki, J. Phys. Soc. Jpn. **77**, 043701 (2008).

[46] T. Onimaru, Y. F. Inoue, K. Shigetoh, K. Umeo, H. Kubo, R. A. Ribeiro, A. Ishida, M. A. Avila, K. Ohoyama, M. Sera, and T. Takabatake, J. Phys. Soc. Jpn. **77**, 074708 (2008).

[47] I. Sheikin, Y. Wang, F. Bouquet, P. Lejay, and A. Junod, J. Phys.: Condens. Matter **14**, L543 (2002).

[48] A. Demuer, D. Jaccard, I. Sheikin, S. Raymond, B. Salce, J. Thomasson, D. Braithwaite, and J. Flouquet, J. Phys.: Condens. Matter **13**, 9335 (2001).

[49] R. Movshovich, T. Graf, D. Mandrus, M. Hundley, J. Thompson, R. Fisher, N. Phillips, and J. Smith, Phys. B: Condens. Matter **223-224**, 126 (1996).

[50] S. Quezel, J. Rossat-Mignod, B. Chevalier, P. Lejay, and J. Etourneau, Solid State Commun. **49**, 685 (1984).

[51] T. Graf, M. F. Hundley, R. Modler, R. Movshovich, J. D. Thompson, D. Mandrus, R. A. Fisher, and N. E. Phillips, Phys. Rev. B **57**, 7442 (1998).

[52] J. Klaasse, P. Veenhuizen, A. BöHM, C. Bredl, U. Gottwick, H. Mayer, L. Pawlak, U. Rauchschwalbe, H. Spille, F. Steglich *et al.*, in *Anomalous Rare Earths and Actinides* (Elsevier, Amsterdam, 1987), pp. 91–94.

[53] R. Fisher, J. Emerson, R. Caspary, N. Phillips, and F. Steglich, Phys. B: Condens. Matter **194-196**, 459 (1994).

[54] A. Thamizhavel, H. Nakashima, Y. Obiraki, M. Nakashima, T. D Matsuda, Y. Haga, K. Sugiyama, T. Takeuchi, R. Settai, M. Hagiwara, K. Kindo, and Y. Ōnuki, J. Phys. Soc. Jpn. **74**, 2843 (2005).

[55] M. Nakashima, H. Kohara, A. Thamizhavel, T. D. Matsuda, Y. Haga, M. Hedo, Y. Uwatoko, R. Settai, and Y. Ōnuki, J. Phys.: Condens. Matter **17**, 4539 (2005).

[56] E. D. Mun, S. L. Bud'ko, A. Kreyssig, and P. C. Canfield, Phys. Rev. B **82**, 054424 (2010).

[57] M. Nakashima, K. Tabata, A. Thamizhavel, T. C. Kobayashi, M. Hedo, Y. Uwatoko, K. Shimizu, R. Settai, and Y. Ōnuki, J. Phys.: Condens. Matter **16**, L255 (2004).

[58] T. Takeuchi, T. Yasuda, M. Tsujino, H. Shishido, R. Settai, H. Harima, and Y. Ōnuki, J. Phys. Soc. Jpn. **76**, 014702 (2007).

[59] N. Kimura, Y. Muro, and H. Aoki, J. Phys. Soc. Jpn. **76**, 051010 (2007).

[60] J. Pásztorová, A. Howell, M. Songvilay, P. M. Sarte, J. A. Rodriguez-Rivera, A. M. Arévalo-López, K. Schmalzl, A. Schneidewind, S. R. Dunsiger, D. K. Singh, C. Petrovic, R. Hu, and C. Stock, Phys. Rev. B **99**, 125144 (2019).

[61] Y. Okuda, Y. Miyauchi, Y. Ida, Y. Takeda, C. Tonohiro, Y. Oduchi, T. Yamada, N. Duc Dung, T. D. Matsuda, Y. Haga, T. Takeuchi, M. Hagiwara, K. Kindo, H. Harima, K. Sugiyama, R. Settai, and Y. Ōnuki, J. Phys. Soc. Jpn. **76**, 044708 (2007).

[62] R. Settai, K. Katayama, D. Aoki, I. Sheikin, G. Knebel, J. Flouquet, and Y. Ōnuki, J. Phys. Soc. Jpn. **80**, 094703 (2011).

[63] G. Knebel, D. Aoki, G. Lapertot, B. Salce, J. Flouquet, T. Kawai, H. Muranaka, R. Settai, and Y. Ōnuki, J. Phys. Soc. Jpn. **78**, 074714 (2009).

[64] A. Thamizhavel, T. Takeuchi, T. D Matsuda, Y. Haga, K. Sugiyama, R. Settai, and Y. Ōnuki, J. Phys. Soc. Jpn. **74**, 1858 (2005).

[65] V. Fritsch, S. Lucas, Z. Huesges, A. Sakai, W. Kittler, C. Taubenheim, S. Woitschach, B. Pedersen, K. Grube, B. Schmidt, P. Gegenwart, O. Stockert, and H. v. Löhneysen, J. Phys.: Conf. Ser. **807**, 032003 (2017).

[66] M. Majumder, R. Gupta, H. Luetkens, R. Khasanov, O. Stockert, P. Gegenwart, and V. Fritsch, Phys. Rev. B **105**, L180402 (2022).

[67] E. Matsuoka, C. Hondo, T. Fujii, A. Oshima, H. Sugawara, T. Sakurai, H. Ohta, F. Kneidinger, L. Salamakha, H. Michor, and E. Bauer, J. Phys. Soc. Jpn. **84**, 073704 (2015).

[68] H. Kotegawa, E. Matsuoka, T. Uga, M. Takemura, M. Manago, N. Chikuchi, H. Sugawara, H. Tou, and H. Harima, J. Phys. Soc. Jpn. **88**, 093702 (2019).

[69] S. Dzsaber, L. Prochaska, A. Sidorenko, G. Eguchi, R. Svagera, M. Waas, A. Prokofiev, Q. Si, and S. Paschen, Phys. Rev. Lett. **118**, 246601 (2017).

[70] H.-H. Lai, S. E. Grefe, S. Paschen, and Q. Si, Proc. Natl. Acad. Sci. USA **115**, 93 (2018).

[71] L. Chen, C. Setty, H. Hu, M. G. Vergniory, S. E. Grefe, L. Fischer, X. Yan, G. Eguchi, A. Prokofiev, S. Paschen, J. Cano, and Q. Si, Nat. Phys. **18**, 1341 (2022).

[72] A. V. Chubukov, D. L. Maslov, and A. J. Millis, Phys. Rev. B **73**, 045128 (2006).

[73] S. Mishra, A. Demuer, D. Aoki, and I. Sheikin, Phys. Rev. B **103**, 045110 (2021).

[74] P. M. Platzman and P. A. Wolff, Phys. Rev. Lett. **18**, 280 (1967).

[75] S. K. Kushwaha, M. K. Chan, J. Park, S. M. Thomas, E. D. Bauer, J. D. Thompson, F. Ronning, P. F. S. Rosa, and N. Harrison, Nat. Commun. **10**, 5487 (2019).

[76] M. Wartenbe, P. H. Tobash, J. Singleton, L. E. Winter, S. Richmond, and N. Harrison, Phys. Rev. B **105**, L041107 (2022).