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Magnetic fluctuations in the magnetically ordering heavy-fermion compounds UCu_5 and U_2Zn_{17}

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We report on the first direct observation of magnetic fluctuations in magnetically ordering heavy-fermion U systems above T_N. They were measured for UCu_5 and U_2Zn_{17} via the quasielastic lines by inelastic neutron scattering. Both compounds behave very similarly. We observe a common broad Lorentzian quasielastic line with width of order $\Gamma_{QE} \simeq 10$ meV, virtually unaffected by the magnetic phase transition, but also a narrow Gaussian quasielastic line, which we attribute to critical spin fluctuations. We discuss the occurrence of magnetic order in spite of the fast valence-fluctuation rate as in contrast to similar magnetically ordering $4f$ systems.

I. INTRODUCTION

Among the heavy-fermion U compounds (Ref. 1) only U_2Zn_{17} ($T_N = 9.7$ K) (Refs. 2 and 3), UCd_{11} ($T_N = 5.0$ K) (Ref. 4), and more recently UCu_5 ($T_N = 15$ K) (Refs. 5–7) are known to exhibit magnetic phase transitions at low temperatures. Magnetic ordering, however, is only one possible alternative for a system to reduce unaffordable degeneracy of the f electrons in their ground state when approaching zero temperature. Also nearly ideal Fermi-liquid states ($CeAl_3$,$CeCu_6$) and even charge- or spin-density waves [URu_2Si_2 (Ref. 8)] are features observed in heavy-fermion systems. Quite unexpectedly, the transition to a superconducting state may also occur despite the presence of competitive magnetic U moments.[1] As recent theories indicate this might be due to a novel coupling mechanism between the conduction electrons mediated by the instability (magnetic fluctuation) of the $5f$ electrons, in analogy to liquid ^{3}He. The instability was confirmed in UPt_3 (Ref. 9) and UBe_{13} (Ref. 10), for example, by inelastic neutron scattering.

Unlike these superconducting compounds, the systems forming magnetically ordered or charge- or spin-density-wave phases are expected to exhibit a fluctuation energy of the order $k_B T_N$ or even less than that. This condition would imply a sufficiently stable magnetic moment for either local or itinerant $5f$ electrons and therefore allow the formation of a coherent magnetic state. In the following we show with inelastic neutron scattering that this is not the case in UCu_5 and U_2Zn_{17}, but that the quasielastic (QE) linewidth Γ_{QE}, as a direct measure of the fluctuation rate, is much larger than $k_B T_N$ and even seems to be unaffected by the magnetic phase transition.

II. EXPERIMENTAL PROCEDURE

76 g (80 g) of polycrystalline UCu_5 (U_2Zn_{17}) has been prepared by the arc-melting technique. The magnetic response was measured over a wide range of energy transfers by inelastic neutron scattering at the time-of-flight spectrometer HRMECS ($E_0 = 60$ and 250 meV) at the Argonne National Laboratory and the IN6 spectrometer ($E_0 = 3.1$ meV) at the Institute Laue Langevin, Grenoble. We will first discuss the results of thermal and hot neutron scattering (HRMECS) and only later turn to those derived from cold neutrons (IN6). We mention that the Q dependence of the scattering function as given in the following is one related only to $Q \equiv |\mathbf{Q}|$ and not to $\mathbf{Q}$. This is because polycrystalline material was used.

A. Thermal neutron scattering

According to the results of other metallic U compounds[8–10] an incoming energy of $E_0 = 60$ meV was chosen in the first place to cover the expected magnetic-energy transfer in these systems. In order to distinguish between magnetic scattering and phonon contributions, spectra were taken at small and large scattering angles separately, corresponding to a low ($Q = 0.5-1.5$ Å^{-1}) and high ($Q = 8-10$ Å^{-1}) momentum transfer Q, respectively. The vanishing form factor of uranium above $Q \simeq 5$ Å^{-1} therefore leads at high Q to phonon spectra only, i.e., free of any magnetic contributions. As a result of the polycrystalline average in our experiments, the weakly dispersive optical phonons contribute primarily to these high-Q spectra. Therefore, the assumption is often made,[11] confirmed so far for nonmagnetic reference compounds, that this high-Q phonon structure also resembles the one at low Q. This is why the magnetic response function of arbitrary shape can be extracted by subtracting the phonon spectrum, as being scaled from the high Q.

The procedure outlined above was used for UCu_5 and U_2Zn_{17} and is demonstrated in Figs. 1 and 2, where the corresponding spectra are shown for $T = 10$ and 50 K and for both low scattering angles (low Q) and high scattering angles (high Q, $T=50$ K). In each case the solid line through the measured spectra (solid dots) gives the result of a fit to the spectra. At high Q (lower part) the phonon spectra of both samples can best be reproduced by assuming a number of inelastic Gaussian lines for the one-phonon scattering contributions (UCu_5: $\Delta=8.5$, 13.6, 15.8, 21.0, 27.1, and 35.5 meV; U_2Zn_{17}: $\Delta=9.5$, 15.2, 22.4, and 30.2 meV) and a surplus, broad quasielastic Gaussian line for the expected multiple-phonon scattering. When turning to forward scattering angles (low Q, two upper parts) the overall phonon shape was kept constant and only the intensity varied by means of single intensity parameter. Obviously, additional scattering appears around 8 meV in these low-Q spectra, which we attribute to the magnetic $5f$ electron scattering. From our data we cannot find any indication that this magnetic scattering should be fitted to a more complicated spectral function than a single broad quasielastic (QE) Lorentzian line. Thus we have taken into account only one QE Lorentzian line, the underlying shape of which is also indicated in Figs. 1 and 2 by the heavy solid line. Spectra taken at $T=10$ K display the same characteristics and thus lead to comparable fitting results for UCu_5 as well as for U_2Zn_{17}.

In Table I we give a summary of the measured QE linewidths Γ_{QE} [half-width at half maximum (HWHM)] and static susceptibilities χ_{QE} as derived from the magnetic QE intensities compared to the experimental bulk values χ_{expt}.[2,12] The agreement between the calculated and measured static susceptibilities is very good for

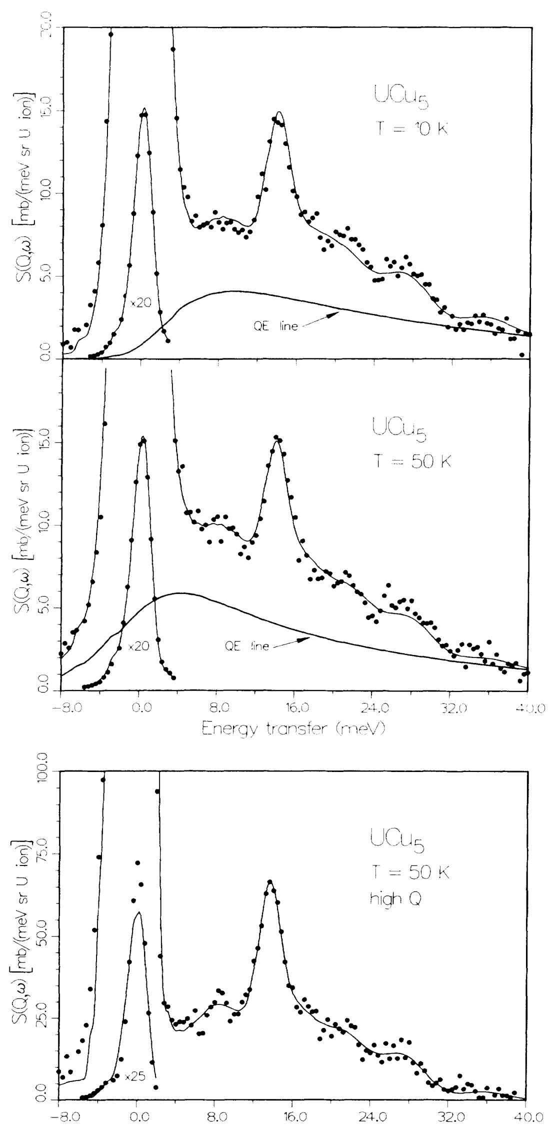

FIG. 1. The scattering function (solid circles) of UCu_5 as measured with thermal neutrons ($E_0=60$ meV) at low scattering angles (low Q: $Q=0.5-1.5$ Å^{-1}) (two upper panels) and high scattering angles (high Q: $Q=8-10$ Å^{-1}) (lower panel). The solid line is a fit to the spectra. At low Q the underlying Lorentzian QE line is indicated by the heavy solid line.

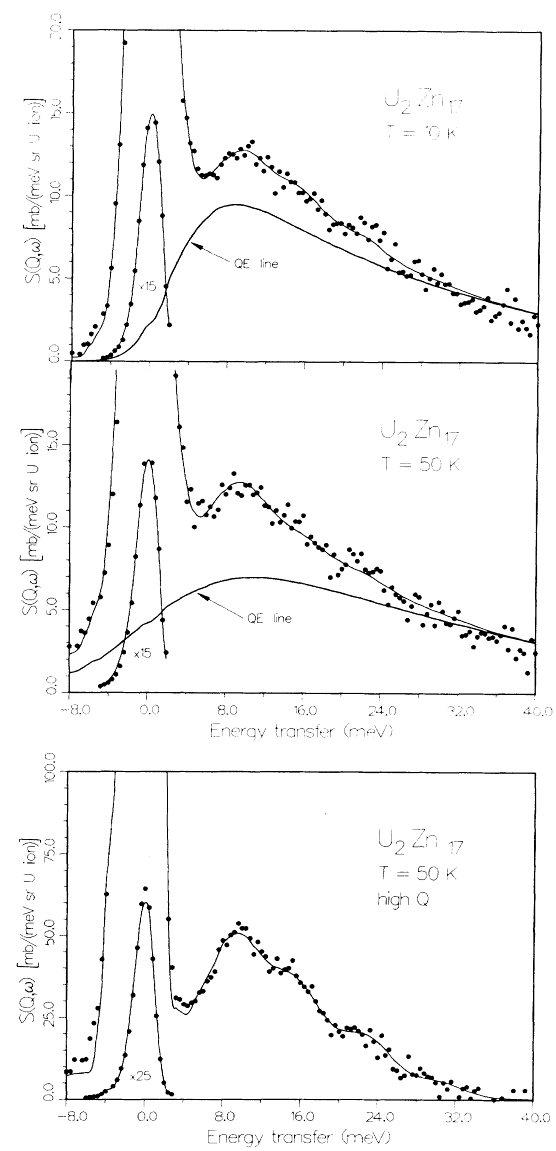

FIG. 2. The scattering function of U_2Zn_{17} as measured with thermal neutrons. Same conditions as Fig. 1.

TABLE I. The Lorentzian QE linewidth Γ_{QE} (HWHM) and the static susceptibility as calculated from the QE intensity (χ_{QE}) compared to the susceptibility as measured (χ_{expt}) for UCu_5 and U_2Zn_{17}.

		Γ_{QE} (meV)	χ_{QE} (10^{-3}emu/mol)	χ_{expt} (10^{-3}emu/mol)
UCu_5	$T=10$ K	9.8 ± 0.8	5.7 ± 0.4	7.2
	$T=50$ K	8.2 ± 0.8	6.9 ± 0.4	6.1
U_2Zn_{17}	$T=10$ K	8.9 ± 0.6	24 ± 2	24
	$T=50$ K	14.1 ± 2.0	20 ± 3	21

U_2Zn_{17}. It is still satisfactory for UCu_5, however, their tendencies in temperature are reverse. An interesting feature is the slowly varying linewidth in dependence of temperature. In U_2Zn_{17} the width shows a familiar drop for decreasing temperature from 14 meV to about 9 meV thereby revealing a nearly identical shape of $S(Q,\hbar\omega)$ at $T=10$ and 50 K (see Fig. 2). On the other hand, the width in UCu_5 increases from roughly 8 meV at $T=50$ K to 10 meV at 10 K.

B. Hot neutron scattering

The inelastic response at higher energy transfers was also routinely checked by means of hot neutrons ($E_0=250$ meV). Neither additional phonon nor magnetic scattering was found at $T=10$ K between 60 and 225 meV in both compounds.

C. Cold neutron scattering

In order to also resolve possible magnetic response in the range of the correlation energy $k_B T_N \simeq 1$ meV, experiments with cold neutrons ($E_0=3.1$ meV) were performed between $T=2$ and 50 K (Figs. 3 and 4). As a general rule, cold neutrons give only limited access ($\Delta E=2$ meV) to the energy-loss side of the neutrons. On the other hand, owing to the smaller incoming wave vector, one-phonon scattering as well as multiple scattering is nearly totally suppressed compared to thermal or hot neutrons. The result is a virtually pure magnetic scattering in the energy range of interest (-6 meV $< \Delta E < 2$ meV).

Interestingly enough, an additional QE line with *Gaussian* line shape could be observed in UCu_5 as well as in U_2Zn_{17}. Figures 3 and 4 show the magnetic scattering functions deconvoluted into the underlying broad Lorentzian QE line (heavy solid line) as described above and the new Gaussian QE line (dashed line). We point out that in UCu_5 this Gaussian QE line is not negligible, but its integrated intensity at $T=20$ K is 41% of the Lorentzian one. On the other hand, in U_2Zn_{17} the ratio between Gaussian and Lorentzian intensity at $T=50$ K is about 1:20 and hence much smaller than that of UCu_5.

We note that at $T=50$ K, where spectra exists from thermal and cold neutron scattering, the derived broad Lorentzian QE lines are identical in linewidth and absolute intensity within the error limits. After having recognized the additional Gaussian line the HRMECS data were refitted and found consistent with it, although this line could barely be distinguished from the huge, resolution-broadened elastic line. This is why it is not shown in Figs. 1 and 2.

III. DISCUSSION

The occurrence of a Gaussian QE line as just described is not absolutely new, though, since it also has been observed in magnetically ordering, unstable Yb compounds like $YbBe_{13}$ (Ref. 13) and the Yb-Pd systems YbPd, Yb_3Pd_4 (Ref. 14). In contrast, rare-earth (RE) compounds, which do not order magnetically such as $CeCu_6$ (Ref. 15) or $PrCu_6$ (Ref. 16) definitely do not show such a Gaussian line. In the Yb systems it could be shown that this line can be associated with spin fluctuations caused by critical fluctuations, which occur as precursors of the magnetic ordering. In UCu_5 and U_2Zn_{17} the coherent critical fluctuations (Gaussian QE lines) are therefore ex-

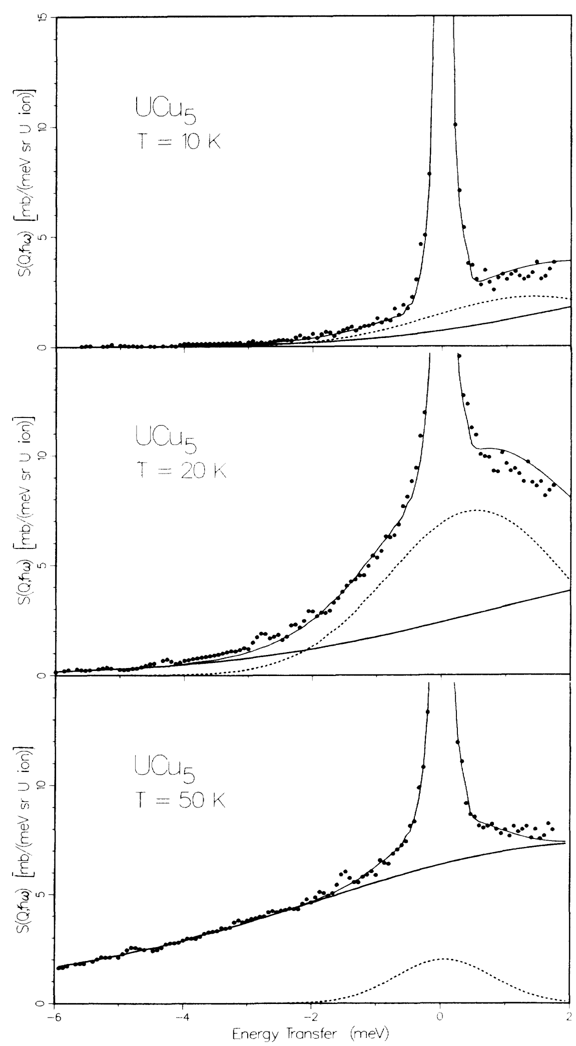

FIG. 3. The scattering function UCu_5 as measured with cold neutrons ($E_0=3.1$ meV). The solid line is a fit to the spectra deconvoluted into the underlying broad Lorentzian QE line (heavy solid line) and the Gaussian QE line (dashed line).

pected to show a strong Q dependence unlike the broad Lorentzian QE line, where no significant Q dependence could be observed. We indeed found a strong temperature and Q dependence of the QE intensity (shown in Figs. 5 and 6, respectively) but only a weak dependence for the QE linewidth. From Fig. 5 we conclude that in the cubic UCu_5 antiferromagnetic spin correlations exists between adjacent (111) planes owing to the coincidence of the two intensity maxima in Q with the Brillouin-zone boundaries along the $[\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2}]$ and [111] axes. This finding is in accordance with the magnetic structure of UCu_5 (Ref. 6) consisting of parallel (111) sheets of ferromagnetically coupled U ions but with antiferromagnetic coupling between neighboring sheets. The temperature dependence (Fig. 6) of the Gaussian QE intensity confirms this interpretation in UCu_5 because it peaks just above the ordering temperature and vanishes at $T=0$ and above $T\simeq 60$ K. As a result of the much weaker Gaussian QE intensity of U_2Zn_{17} around T_N no Q dependence could be obtained in this case. The Q-integrated temperature dependence (bump around T_N), however, indicates that the critical fluctuation interpretation in general also holds for U_2Zn_{17}. However, because it shows an unreasonable overall temperature behavior, we suggest it to be of a different nature than that in UCu_5.

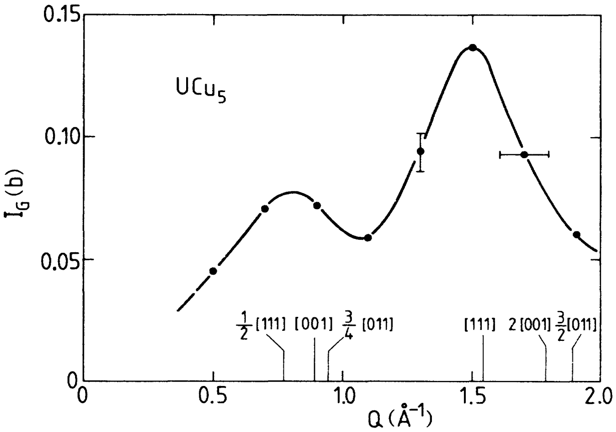

FIG. 5. The $|\mathbf{Q}|$ dependence of the scattering intensity I_G of the Gaussian QE line at $T=10$ K. The Brillouin-zone boundaries along the main crystal axis of the fcc lattice are given by the square brackets.

The linewidths of these Gaussian lines exhibit a much weaker temperature dependence than their intensities. At $Q=1.5$ Å^{-1} and at $T=20$ K, UCu_5 displays a width (HWHM) of $\Gamma_{QE}=0.88$ meV, which slowly but constantly *increases* with decreasing temperature, until it rolls off at a constant width of $\Gamma_{QE}=2.2$ meV below $T\simeq T_N$. In contrast, in U_2Zn_{17} the reverse seems to happen, and so again indicates a different nature of spin fluctuations in this compound. Starting with a linewidth of $\Gamma_{QE}=1.1$

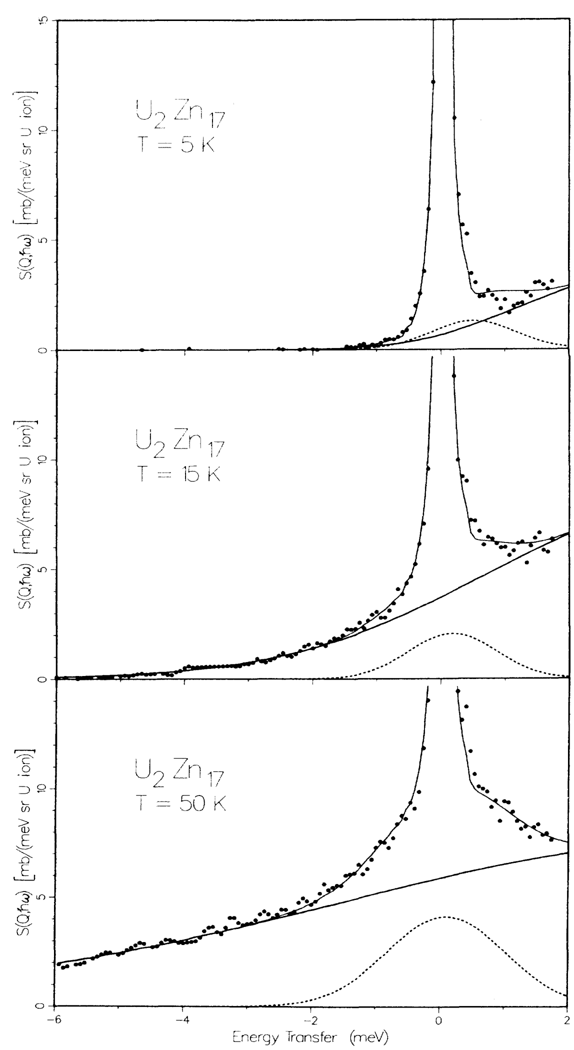

FIG. 4. The scattering function of U_2Zn_{17} as measured with cold neutrons. Same conditions as Fig. 3.

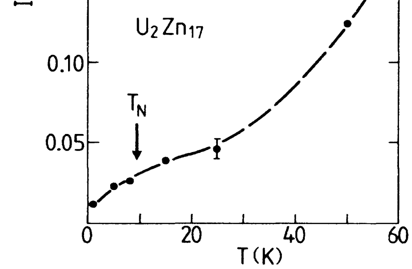

FIG. 6. The temperature dependence of the scattering intensity I_G of the Gaussian QE line integrated over $Q=0-1.5$ Å^{-1}.

meV at $T=50$ K it slowly but constantly *decreases* with decreasing temperature down to the limiting value of about $\Gamma_{QE}=0.75$ meV below $T \simeq T_N$. Anyway, in both cases the two energy scales Γ_{QE} and $k_B T_N$ nearly coincide, and because of the striking similarity to the magnetically ordering Yb compounds it now seems convincing that the Gaussian lines in both the U and Yb compounds owe their existence to the same mechanism, viz., antiferromagnetic critical spin fluctuations. The only difference seems to be the absence of any magnon excitation below T_N in UCu_5 and U_2Zn_{17}. However, from our data taken on a polycrystalline sample, we cannot exclude the existence of a broad inelastic excitation at a few meV as caused by the density of states of strongly overdamped spin waves. At present, the reason for the apparent deficiency of magnon excitations is still unknown, but we wish to refer to US and UAs,[17] where in spite of magnetic order, no magnon excitations were found as well.

From the results given in Table I we see that the broad Lorentzian QE linewidth does not become smaller than the correlation energy $kT_N \simeq 1$ meV at or below the magnetic phase transition, but instead shows up as a more or less temperature- (and Q-) independent value of the order of 10 meV, i.e., 1 order of magnitude above the correlation energy. In addition, the close temperature sequences taken for UCu_5 ($T=2$, 5, 10, 13, 20, 25, and 50 K) and U_2Zn_{17} ($T=2$, 5, 8, 15, 25, and 50 K) showed that the QE line is not affected when passing through the ordering transition. We note that the magnetic entropy associated with the λ-like anomaly of the specific heat below the magnetic phase transition is only 60% in UCu_5 (Ref. 12) and 70% in U_2Zn_{17} (Ref. 2) of the expected value. This discrepancy might be due to the unaffected broad QE line, which implies a transfer of $5f$ states from the Fermi surface to areas inaccessible by temperature.

As only the magnitude of the broad Lorentzian QE line as caused by the magnetic fluctuation rate is concerned, there seems to be no qualitative difference between the superconducting heavy-fermion systems UPt_3 or UBe_{13} and the magnetically ordering heavy-fermion systems UCu_5 or U_2Zn_{17}. We point out that even the metallic nonheavy-fermion systems like UAl_2, USn_3, or UAs (Ref. 17) exhibit comparable linewidths and temperature dependences. Interestingly enough, in *all* these U systems the QE linewidth coincides with the Curie temperature as derived from the static susceptibility at high temperatures. The valence-fluctuation behavior of UCu_5 and U_2Zn_{17} is, however, totally different from that of URu_2Si_2 (Ref. 8) at temperatures below $T=50$ K. This becomes obvious not only from the temperature behavior of most macroscopic quantities (Ref. 18) at the magnetic phase transition, but especially from the breakdown of the Lorentzian QE line below $T=40$ K (Ref. 19) and the absence of any Gaussian QE line in URu_2Si_2.

IV. CONCLUSION

We conclude that $4f$ and $5f$ electrons may be subject to strong interactions either with conduction electrons or with other magnetic moments. The different nature of these interactions is revealed through the presence of QE lines of characteristic shapes and widths. The broad Lorentzian QE line, being the common signature of all unstable f ions, thereby seems to be a measure of the hybridization strength between a single f ion and the conduction electrons, whereas the relatively narrow Gaussian QE line is the result of the correlated interaction between many stable f moments. Our data of UCu_5 and U_2Zn_{17} indicate that the measured f hybridization by no means affects or is affected by the capability of "unstable" $5f$ moments to order antiferromagnetically. Support for this interesting statement comes from the observation of the strong depression of magnetic order in both UCu_5 (Ref. 20) and U_2Zn_{17} (Ref. 21) by the substitution of the nonmagnetic *host* atoms Cu(Zn) by only a few percent of Ni(Cu). This implies that the electronic band structure is mainly responsible for the ability of magnetic order and not the concentration of the $5f$ ions or their magnetic fluctuation rate.

In addition to the strong f hybridization, a precursor to magnetic ordering, i.e., correlated interactions between stable $5f$ moments, was observed in UCu_5 as well as in U_2Zn_{17}. This behavior of $5f$ systems is in striking contrast to prior findings in the unstable Ce, Tm, and Yb compounds.[14,22] Here, no RE compound could be found, so far, showing magnetic order and/or a narrow Gaussian QE line in coexistence with a broad Lorentzian QE line.[23] It was concluded that a necessary condition for magnetic ordering is a (in Tm at least one) Lorentzian linewidth, as generated by f hybridization, of the order of or below the correlation energy $k_B T_N$. The physical idea behind this is the loss of orientational memory and thus the ability of magnetic order, if every f moment is subject to too fast a valence-fluctuation rate. In closing we assert that, as it seems now, a fundamental difference exists between the nature of valene fluctuations in unstable $4f$ and $5f$ systems, at least from this point of view.

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