Inelastic neutron scattering from $PrFe_4P_{12}$ at low temperatures and under high magnetic fields

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$PrFe_4P_{12}$ exhibits a unique transition at 6.5 K with an order parameter whose nature is still controversial. In order to elucidate the origin of the transition, we have carried out inelastic neutron scattering experiments in a range of temperatures and magnetic fields. Our data reveal a different type of low-lying excitations centered at 1.5 and 3.5 meV in the ordered phase with an unusual Q dependence. With increasing field or temperature, the well-defined excitations develop into a quasielastic response, while the ordered phase becomes a paramagnetic state with a very large characteristic energy scale of $\Gamma \simeq 0.8$ meV, consistent with a large Sommerfeld coefficient $\gamma = 1.4$ J/mol K^2.

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I. INTRODUCTION

When many interactions of different origins compete with one another within a single compound, the ground state of such a system is very subtle and often hard to understand initially. More often than not, the true nature of the so-called hidden order is only unearthed after many theoretical scenarios have been carefully examined from every angle and all possible experimental techniques have been brought to the system under examination.

Quadrupole ordering is a case in point. Quadrupole ordering and its underlying mechanism in rare-earth intermetallic compounds have been a long-standing puzzle.[1] Although the importance of the quadrupole, or orbital, degrees of freedom was first noticed in rare-earth intermetallic systems some 30 years ago,[2] it has recently become a more popular and challenging research topic for transition metal oxides, where direct evidence of orbital ordering is well established now and its effects on bulk properties are intensively studied.[3] While quadrupole ordering has now been firmly established as an important order parameter of rare-earth materials, alongside the usual dipole moment, more recent investigations into new rare-earth compounds suggest that more exotic order parameters such as hexadecapoles may also be relevant and need to be considered in some cases.[4]

Despite the growing awareness of the importance of multipolar order parameters, their existence is often elusive, in particular, when expected to occur in materials with rare-earth elements that are known to have other order parameters. Pr intermetallic compounds are a good example. As Pr ions have a nonmagnetic Γ_3 doublet with nonzero quadrupole moments under cubic symmetry, Pr compounds have been considered as natural candidates in the search for quadrupole-related phenomena. In fact, unusual features of some Pr skutterudite systems have been ascribed to the quadrupole degrees of freedom. $PrFe_4P_{12}$ is one such example where the identification of the order parameter has been hampered probably by the received wisdom of Pr having a quadrupole order parameter until a more careful analysis of the data, together with theoretical studies, revealed the truer nature of the ground state.

$PrFe_4P_{12}$ exhibits very clear anomalies at 6.5 K in its bulk properties such as resistivity, magnetic susceptibility, and heat capacity.[5–7] However, neutron diffraction studies did not show any evidence of magnetic ordering, suggesting that the phase transition at 6.5 K should be of a nonmagnetic origin; a similar conclusion was also drawn from Pr nuclear specific heat data.[6] With the possibility of the nonmagnetic Γ_3 ground state with nonzero quadrupole moments, one of the natural explanations for the phase transition was to attribute it to the quadrupole ordering of the Γ_3 ground state doublet. In fact, an O_2^0 quadrupole model seemed to be consistent with the then available experimental data such as elastic constants[8] and x-ray diffraction studies.[9] However, more recent examinations of the experimental data, in particular, the latest nuclear magnetic resonance (NMR) results,[10] have been shown to be incompatible with the quadrupole scenario. Of particular note is that the quadrupole model fails to explain two key experimental observations: (i) the isotropic susceptibility in the ordered phase and (ii) the field-induced staggered moment only parallel to the field direction. New theoretical proposals have since been put forward for the experimental results including the latest NMR data; one of the most promising explanations is a scalar order model.[11]

Another equally interesting point about $PrFe_4P_{12}$ is that the heat capacity measurements show that the Sommerfeld coefficient γ is as large as 1.4 J/mol K^2. This extremely large γ value for Pr compounds corresponds to a heavy cyclotron mass $m^* = 81 m_o$, where m_o is the mass of bare electrons. This is larger than for most well-known heavy fermion compounds.[12] In spite of the experimental evidence, the origin of this heavy fermion state is also poorly understood. In

summary, $PrFe_4P_{12}$ has a strongly correlated electron ground state and a phase transition with a hidden order.

In this work, we present our inelastic neutron scattering results of $PrFe_4P_{12}$ measured from 300 to 2 K and up to 7 T. Although some of our early results were previously reported in a short paper,[13] we present here a complete analysis of our results.

II. EXPERIMENTAL DETAILS

For our experiments, we prepared 8.5 g of $PrFe_4P_{12}$ polycrystalline sample by using a tin flux method.[5] In order to have a reliable estimate of the phonon scattering in our data for $PrFe_4P_{12}$, we also made 9.3 g of $LaFe_4P_{12}$ that has the same filled skutterudite crystal structure. Our subsequent x-ray powder diffraction confirmed that our samples form in the cubic structure with the *Im*-3 space group at room temperature. Inelastic neutron scattering measurements were made with the HET chopper spectrometer at the UK ISIS spallation neutron source from 300 to 7 K without a magnetic field. We also made low temperature measurements from 12 to 2 K using a He^4 cryostat equipped with a 7 T superconducting magnet. At the HET spectrometer, neutrons are scattered from the sample into two forward detector banks: one at low scattering angles $\phi=2.6° \rightarrow 7.2°$ at a distance of 4 m from the sample position, and a second bank covering slightly higher scattering angles $\phi=9.3° \rightarrow 28.7°$ at a distance of 2.5 m from the sample. Two high-angle detector banks are located at $\phi=110.4° \rightarrow 138.7°$. The scattering function $S(Q,E)$ is then obtained in absolute units of mb meV^{-1} sr^{-1} $f.u.^{-1}$ from the raw time-of-flight data by normalizing them to the incoherent scattering from a flat vanadium standard sample, weighing 20.14 g, for each chosen incident energy, and then to the incident flux using the integral of the incident beam monitor.

III. RESULTS AND ANALYSIS

Figure 1(a) shows the magnetic field (B)-temperature (T) phase diagram of $PrFe_4P_{12}$ obtained from bulk measurements.[14] As one can see, there occurs a field-induced transition from the paramagnetic phase to the hidden order phase at finite magnetic fields, whose exact values depend on the direction of the magnetic field. It is also clear from the figure that the ordered phase is quenched at all temperatures when a magnetic field as large as 7 T is applied in any direction, so for polycrystalline samples, like ours, 7 T is sufficient to induce the paramagnetic phase even at 2 K. In order to study the field and temperature dependence of the scattering function $S(Q,E)$ in the ordered phase, we have measured the inelastic neutron scattering of $PrFe_4P_{12}$ at six different points in the B-T phase below 8 K as marked by the symbols: three points in the ordered phase and three others in the paramagnetic phase.

Before presenting the low temperature and high field data, let us first discuss a strong temperature dependence seen in the data. In order to obtain the magnetic response, S_{mag}, we used a so-called direct subtraction method using $LaFe_4P_{12}$ as a phonon blank material.[15] As shown in Figs. 1(b) and 1(c),

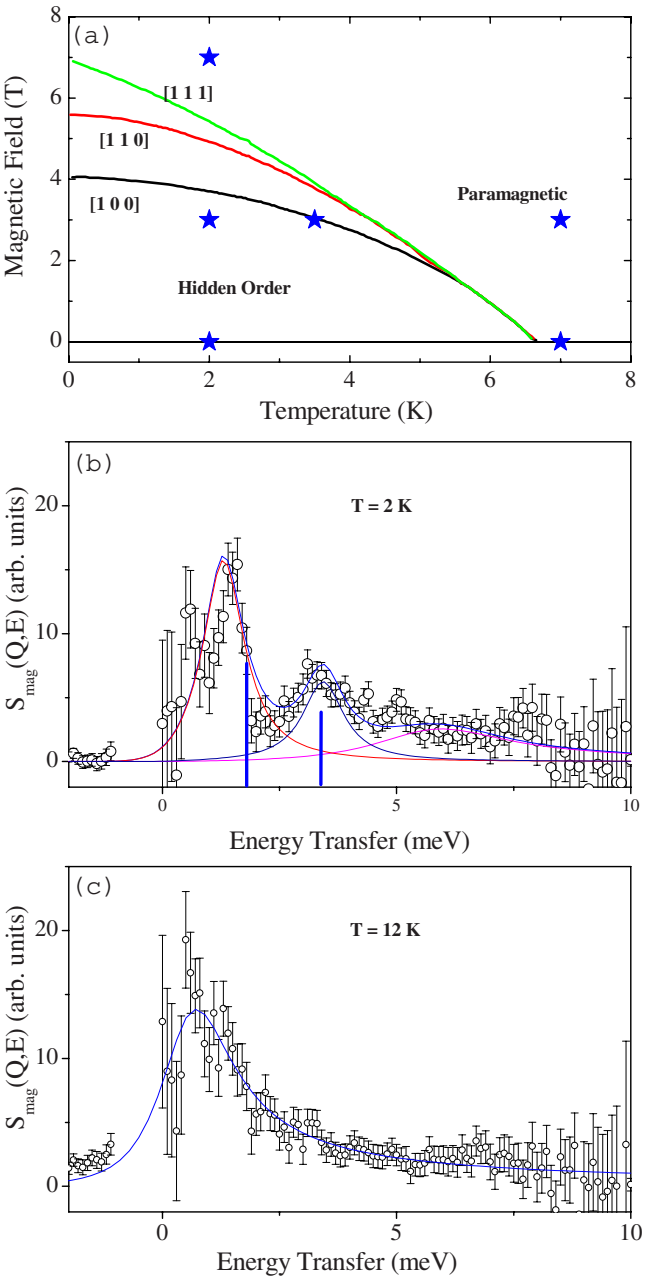

FIG. 1. (Color online) (a) B-T phase diagram of $PrFe_4P_{12}$ obtained from bulk measurements (Ref. 14) with fields along three main symmetry axes. The symbols indicate where in the B-T phase our measurements have been carried out. Magnetic scattering is shown for (b) 2 K and (c) 12 K after subtracting phonon contributions using a phonon blank material of $LaFe_4P_{12}$ measured under identical conditions. The blue line in (b) is the sum of three theoretical localized excitations while the line in (c) is the fitting result of quasielastic scattering. The two vertical bars represent the allowed excitations from a single ion model as discussed in the text.

upon cooling below the transition temperature, clearly localized modes emerge out of a quasielastic line shape of the inelastic response seen in the paramagnetic phase. Without any analysis, it is clear that there are at least two well-defined excitations centered at 1.44 and 3.22 meV, respectively, and probably another very broad peak above 5 meV. Due to lack

of information about a correct crystal field Hamiltonian, we fitted the data as best as we could using a minimum number of peaks to reach the final fitting results as shown in Fig. 1(b). The vertical bars represent allowed transitions between the ground state and two excited states, which were obtained using a single ion model with $Zt=1.8$ as described in Ref. 16. Although four more excitations are possible from this single ion model, they are much weaker than the two shown in the figure. Clearly, this single ion model, without any term accounting for the order parameter or any interaction term, fails to reproduce the experimental results. We can imagine that the agreement would improve once one carries out more elaborate calculations with several terms including a hybridization term as discussed in Ref. 17. For comparison, an O_2^0 quadrupole model Hamiltonian, having both an interaction term and an O_2^0 quadrupole term, produced excitations at similar positions with comparable intensity. The line in Fig. 3(a) shows the results of the theoretical calculations convoluted with the experimental resolution function of the HET spectrometer. Horizontal bars below the data points in Fig. 3(a) indicate the experimental resolution at two energies: the elastic position and the position of the inelastic peak, i.e., $\Delta E=3.5$ meV.

To complete our discussion, we comment on some important differences between the two aforementioned model Hamiltonians. As is well known, skutterudite materials with T_h symmetry do not have umklappung and fourfold rotation axis unlike O_h symmetry.[18] Therefore, a full single ion crystalline-electric-field (CEF) Hamiltonian is given as follows: $H_{\mathrm{CEF}}=A_4(O_4^0+5O_4^4)+A_6^c(O_6^0-21O_6^4)+A_6^t(O_6^2-O_6^6)$. We found that the A_6^c term is larger than the A_6^t term in the O_2^0 quadrupole model, whereas the single ion model produces the A_6^t term much larger than A_6^c. For $Zt=1.8$ used above, we have the following set of CEF parameters from the single ion model: $A_4=-3.04\times10^{-3}$ meV, $A_6^c=-6.68\times10^{-6}$ meV, and $A_6^t=-1.29\times10^{-4}$ meV, while the O_2^0 quadrupole model requires $A_4=-1.36\times10^{-4}$ meV, $A_6^c=-2.44\times10^{-5}$ meV, and $A_6^t=-2.00\times10^{-6}$ meV.

Let us turn to the field dependence of the data at low temperatures. In Fig. 2, we present contour plots for five sets of the data obtained from the low temperature measurements as marked in Fig. 1(a) with the incident neutron energy of 11 meV. In these figures, we only show the data measured by the two low-angle detector banks: one at 4 m with low scattering angles $\phi=2.6°\rightarrow7.2°$, and the other at 2.5 m covering slightly higher scattering angles $\phi=9.3°\rightarrow28.7°$. Magnetic scattering in the data obtained from two other high angle banks is very weak compared with phonon contributions. The narrower strip of the data on the lower momentum transfer side is for the data taken by the 4 m bank, while the thicker one on the right represents the data taken with the 2.5 m detector bank. For the incident energy of 11 meV, the magnetic form factor of Pr ions drops by less than 4% by increasing the momentum transfer from 0 to 1.2 $Å^{-1}$, which corresponds to the Q range of the two detector banks located at low angles. It is noticeable in Fig. 2(a) that the data taken at 2 K without a magnetic field exhibit two well-defined excitations in addition to strong elastic nuclear peaks. This is in striking contrast with the total absence of such scattering in the $LaFe_4P_{12}$ data taken under the same condition [see Fig. 2(f)]. On the other hand, there is a strong quasielastic response in the $PrFe_4P_{12}$ data taken at 7 K and zero field, which is in the temperature-induced paramagnetic heavy fermion phase [Fig. 2(d)]. Some indications of such a change in the low energy excitations can also be found in a previous report.[19] However, with increasing magnetic field, the well-defined inelastic peaks broaden and disappear without much sign of a quasielastic response from the data taken at 2 K and 7 T, which is an indication of a field-induced paramagnetic phase [see also Fig. 3(b)]. Therefore, two paramagnetic points in the B-T phase diagram: (i) a *field-induced* paramagnetic point [Fig. 2(c)] and (ii) a *temperature-induced* paramagnetic point [Fig. 2(d)], exhibit apparently different low energy responses. Our measurements at another point (7 K and 3 T) in the paramagnetic phase display a low-lying response [Fig. 2(e)], which sits somehow in between the 2 K–0 T data [Fig. 2(a)] and the 7 K–0 T data [Fig. 2(d)].

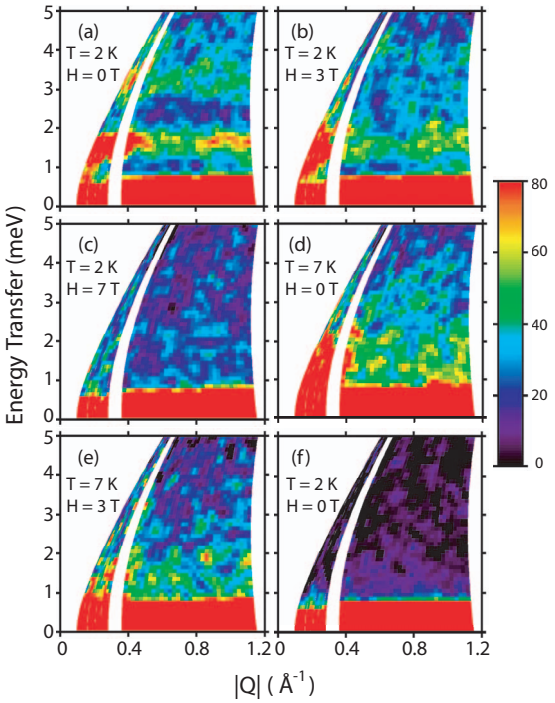

FIG. 2. (Color) Contour plots are given for the momentum (Q) and energy (E) transfer of the total scattering function, $S(Q,E)$, of $PrFe_4P_{12}$ at low temperatures. They were measured at (a) 2 K and 0 T, (b) 2 K and 3 T, (c) 2 K and 7 T, (d) 7 K and 0 T, and (e) 7 K and 3 T. Data of $LaFe_4P_{12}$ taken at 2 K and 0 T are also given in (f). The scale on the right is given in absolute units of mb meV^{-1} sr^{-1} $f.u.^{-1}$.

The field and temperature dependences of the low-lying excitations in Fig. 2 can be better compared in the plots of the scattering function $S(E)$, which were obtained by averaging the raw data over the two low-angle detector banks (see Fig. 3). In Fig. 3(a), we compare zero-field data taken at 2 and 7 K together with the data of $LaFe_4P_{12}$ taken at 2 K. As one can see in Fig. 3(a), except for the nuclear Bragg peaks, there is almost no scattering at all above 1 meV for $LaFe_4P_{12}$. In contrast, there is a strong additional scattering in the 7 K–0 T data for $PrFe_4P_{12}$ that evolves into two well-defined inelastic peaks centered at 1.5 and 3.5 meV upon cooling below the transition temperature. These two inelastic

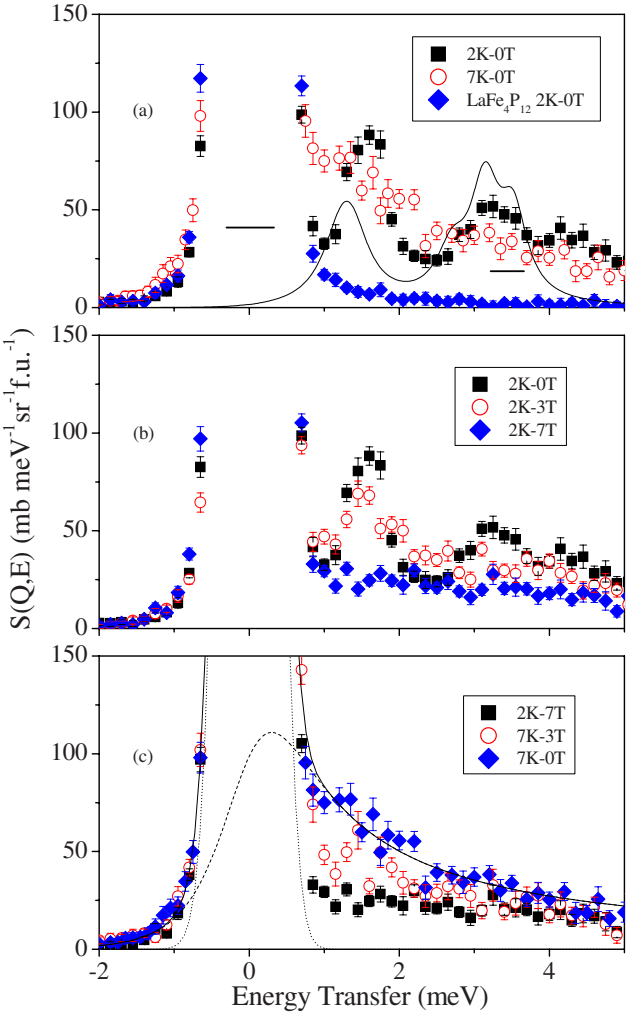

FIG. 3. (Color online) Total scattering function summed over the two low-angle detector banks is given for several values of magnetic field and temperature. (a) $PrFe_4P_{12}$ data measured at two settings of 2 K–0 T and 7 K–0 T are presented together with the data for $LaFe_4P_{12}$ measured at 2 K–0 T. The line is for the theoretical curve, obtained using an O_2^0 quadrupole model in addition to the crystal field Hamiltonian, convoluted with the experimental resolution function (see the text). The horizontal bars under the data points indicate the experimental resolution at the elastic position and $\Delta E=3.5$ meV. (b) $PrFe_4P_{12}$ data measured at three points in the *B-T* phase of 2 K–0 T, 2 K–3 T, and 2 K–7 T. (c) $PrFe_4P_{12}$ data measured at three points in the paramagnetic phase of the *B-T* phase shown in Fig. 1(a): 2 K–7 T, 7 K–3 T, and 7 K–0 T. The line is for the curve fitting results using one elastic peak (dotted line) and one quasielastic peak (dashed line) with $\Gamma=0.8$ meV. The solid line is for the sum of the two functions.

peaks are gradually suppressed with increasing magnetic fields from 0 to 7 T at 2 K as shown in Fig. 3(b). On the other hand, the broad response observed in the 7 K–0 T data also gets reduced while going from 7 K–0 T to 7 K–3 T to 2 K–7 T in the *B-T* phase diagram although all three measurements are supposedly done in the same paramagnetic phase. We comment that the quasielastic response in the paramagnetic phase is not uniformly suppressed by the magnetic field as shown in Fig. 3(c).

As we have noted, with increasing temperatures, the well-defined excitations become rapidly quasielastic-like. For example, we have fitted the 7 K–0 T data using one quasielastic term together with another term for the elastic peak, and the lines in Fig. 3(c) represent our fitting results. See also the line in Fig. 1(c) for our fitting results of the 12 K data. Upon further increasing the temperature, the quasielastic contributions become considerably wider. The broad response observed above the transition temperature is consistent with the fact that the paramagnetic phase is a heavy fermion state with $\gamma=1.4$ J/mol K^2, which is often the case with Ce and Yb heavy fermion compounds.[12] In fact, the quasielastic

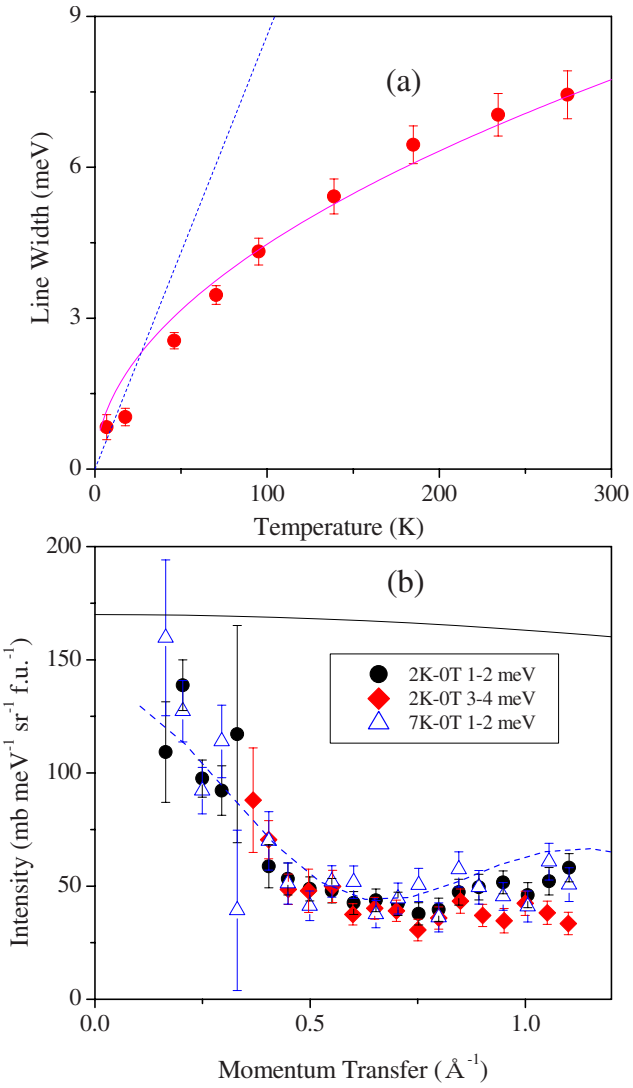

FIG. 4. (Color online) (a) Temperature dependence of the linewidth of the quasielastic peak. The solid line is for our curve fitting results using a $\sqrt{T}$ dependence and the dashed line for a conventional k_BT dependence (see the text). (b) Q dependence of the excitations centered at 1.5 and 3.5 meV that was obtained from the data taken at 2 K and 0 T. A similar plot is also made for the data taken at 7 K and 0 T for the energy range of 1–2 meV. The solid line is for $F(Q)^2$, where $F(Q)$ is the theoretical form factor of Pr ions (Ref. 22), while the dashed line is for the case in which we assumed that there is additional short-ranged ferromagnetic correlations among some Pr moments.

linewidth of $PrFe_4P_{12}$ follows roughly a $\sqrt{T}$ dependence [the solid line in Fig. 4(a)] as observed for other heavy fermion systems, although we notice some discrepancies between the data and the line for the $\sqrt{T}$ dependence at low temperatures.[20] It should also be noted that the linewidth of the quasielastic term is smaller than the thermal fluctuations of k_BT, as indicated by the dotted line in Fig. 4(a). Interestingly enough, the linewidth estimated at 0 K is $\Gamma \simeq 0.8$ meV, which is not far off from Γ estimated by using the following theoretical relationship: $\Gamma\gamma = 0.97$ meV J mol^{-1} K^{-2}. This relationship between Γ and γ is known to hold reasonably well for most heavy fermion compounds.[21]

It is also interesting to look at the Q dependence of the quasielastic response. As shown in Fig. 4(b), when integrated from 1 to 2 meV energy transfer, the intensity falls rapidly with increasing momentum transfer (Q): a similar Q dependence is also obtained for different E ranges. This is in marked contrast to the usual Q dependence of the magnetic form factor of Pr ions. In Fig. 4(b) we plot $F(Q)^2$ (solid line), where $F(Q)$ is the theoretical form factor of Pr ions.[22] It is worth noting that almost the same Q dependence is also observed in the ordered phase over the same energy range. In order to explain the observed Q dependence, we have assumed that there is short-ranged ferromagnetic correlation among Pr moments by adding an extra term, $\frac{\sin Qr_0}{Qr_0}$, to the usual Q dependence of the form factor, where r_0 is fixed at the nearest distance between Pr ions, $r_0 = 6.77$ Å.[23] The dashed line is for what we obtained from the sum of both terms. As we can see, our theoretical curve explains the experimental Q dependence reasonably well. Related to this observation, we note that magnetic susceptibility data show a positive Curie-Weiss temperature at low temperatures.[24]

Finally, let us comment on the nature of the excitations seen in our data. Although we measured the magnetic cross section in inelastic neutron scattering, what we have observed are the excitations of the system where the interplay of the hidden order and the crystal field (or spin) is anticipated to be very strong and probably plays an important role. Therefore, we believe that the spin and hidden order degrees of freedom are so strongly and inextricably interconnected in $PrFe_4P_{12}$ that it would be rather misleading to distinguish between these parts in the excitations we measured.

IV. CONCLUSIONS

To summarize, we have studied the inelastic neutron scattering of $PrFe_4P_{12}$, which shows a transition from a paramagnetic state with a very large Sommerfeld coefficient $\gamma = 1.4$ J/mol K^2, i.e., a heavy fermion state, to a hidden order state below 6.5 K. Above the transition temperature, the inelastic response is dominated by quasielastic scattering with a correct characteristic energy scale of $\Gamma \simeq 0.8$ meV consistent with the Sommerfeld coefficient. Upon cooling below 6.5 K, at least two well-defined localized excitations emerge out of the broad response in the paramagnetic phase. With increasing magnetic field or temperature, the well-defined excitations become significantly broadened and develop into a quasielastic response in the supposedly heavy fermion paramagnetic phase. Although we cannot presently distinguish between the available theoretical models based on our experimental observations, nevertheless, we believe that our results of the well-defined excitations in the ordered phase will prove useful for the further development of, and put a strict constraint on, a correct model, such as in another famous heavy fermion compound URu_2Si_2 with a hidden order.[25]

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