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# Pressure-dependent inelastic neutron scattering studies of $CePt_2$

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**Abstract**
Inelastic neutron scattering measurements were made on $CePt_2$ at pressures up to 5 kbar. At ambient pressure spin wave excitations were observed in an antiferromagnetic phase that transformed into a quasi-elastic response above $T_N$. At low temperatures ($T \leqslant T_N$), the magnetically ordered state appears to show some instability at a pressure of around 4 kbar where the dynamic susceptibility determined from the neutron data exhibits distinctive features that resemble those of heavy fermion compounds showing non-Fermi liquid behaviour.

## 1. Introduction

The formation of a magnetically ordered ground state in heavy fermion compounds depends on a delicate balance between two competing interactions; one is the single-ion Kondo interaction and the other the inter-site RKKY interaction [1]. In some compounds one can continuously change the magnetically ordered ground state into a non-magnetic one with a modest variation in parameters such as doping, pressure, and magnetic field. For systems in the non-magnetic ground state, the predictions of Fermi liquid theories hold reasonably well; well below the Fermi temperature, $T_F$, the resistivity exhibits quadratic temperature dependence, the heat capacity is linear in temperature, and the magnetic susceptibility becomes temperature independent. At a crossover point at which a system transforms from a magnetically ordered state to a non-magnetic ground state, however, the bulk properties are often observed to deviate significantly from the predictions of the Fermi liquid theories [2]. These phenomena, which are now called non-Fermi liquid behaviour, have since been discovered in several systems and their number is growing.

Although the bulk properties of non-Fermi liquid systems are very interesting and are currently the subject of intensive studies, the dynamic susceptibility obtained from inelastic neutron scattering techniques appears to be the most informative. Of particular interest is the energy ($E$) and momentum ($Q$) dependence of the spin dynamics, in particular the $E/T$ scaling behaviour in the dynamic susceptibility of non-Fermi liquid systems, as reported for $U(Cu, Pd)_5$ [3], $Ce(Cu, Au)_6$ [4], and Ce(Rh, Pd)Sb [5].

$CePt_2$ forms in the cubic Laves phase with cerium 4f electrons of some localized character [6]. Early heat capacity and susceptibility measurements confirmed that it undergoes an antiferromagnetic transition with $T_N = 1.6$ K [7]. It is also a heavy fermion compound with an extremely large mass enhancement, $\gamma \simeq 1.5$ J mol$^{-1}$ K$^{-2}$ at 2 K [8]. This transition temperature is rapidly suppressed by doping Ir or Rh on the Pt sites of $CePt_2$ as shown in the recent heat capacity measurements [8]. For example, $T_N$ is decreased to around 0.55 K for $Ce(Pt_{0.75}Ir_{0.25})_2$. With such a low transition temperature, it would be an ideal candidate for studies of magnetic-to-nonmagnetic crossover behaviour induced by pressure as demonstrated recently for several Ce compounds [9]. However, recent pressure-dependent measurements of the resistivity of $CePt_2$ did not reveal any significant changes with increasing pressure [10].

In the present study, we have performed inelastic neutron scattering measurements of $CePt_2$ at ambient pressure as well as at pressures up to 5 kbar. Unlike the resistivity measurements, we find that spin excitations arising from the ordered Ce moments show a clearly visible change with increasing pressure. We also observe unusual temperature dependence of the linewidth of quasi-elastic scattering. Furthermore, we observe an $E/T$ scaling behaviour, albeit over a limited energy range, in the dynamic susceptibility at 4 kbar and higher, implying that $CePt_2$ may well be located close to an unconventional critical point at around 4 kbar.

## 2. Experimental details

Samples of $CePt_2$ and the non-magnetic reference $LaNi_2$ were prepared by induction melting appropriate amounts of constituent elements at Grenoble. The resultant ingots were annealed at 800°C for 48 h under vacuum. We measured inelastic neutron scattering of $CePt_2$ and $LaNi_2$ from 200 down to 1.5 K using the cold neutron time-of-flight spectrometer IN6 at the ILL. All our measurements were made with an incident neutron energy of 3.1 meV. Pressure-dependent measurements were performed using a He gas pressure cell capable of producing pressures up to 5 kbar.

## 3. Results and analysis

Three representative examples of the data for $CePt_2$, taken at ambient pressure, are shown in figure 1. As one can see from the figure, the lower the temperature the narrower is the linewidth of the quasi-elastic contribution. As we noted in the introduction, $CePt_2$ is a strongly correlated electronic system with a high value of $\gamma \simeq 1.5$ J mol$^{-1}$ K$^{-2}$ at 2 K [8]. It also shows a crystal field excitation around 20 meV [11], observation of which in many heavy fermion compounds is generally regarded as a signature of the localized character of f electrons [12].

In order to determine the resolution function of the instrument, we used the data for $LaNi_2$ which has a strong incoherent cross-section and hence gives a strong resolution limited elastic scattering. The resolution function (elastic peak function) was found to be composed mainly of a Gaussian component with about 4–6% mixture of a Lorentzian component, i.e. a pseudo-Voigt function. The data for $LaNi_2$ at 130 K is given in the inset of figure 1. Analysis of the data of a vanadium standard measured subsequently under similar conditions also yields a similar value for the mixing parameter ($\mu$) between the Gaussian and Lorentzian components. We then fixed $\mu$ to the average value of the results for the $LaNi_2$ and vanadium data and have used the same $\mu$ for the analysis of all our $CePt_2$ data. However, we warn that the

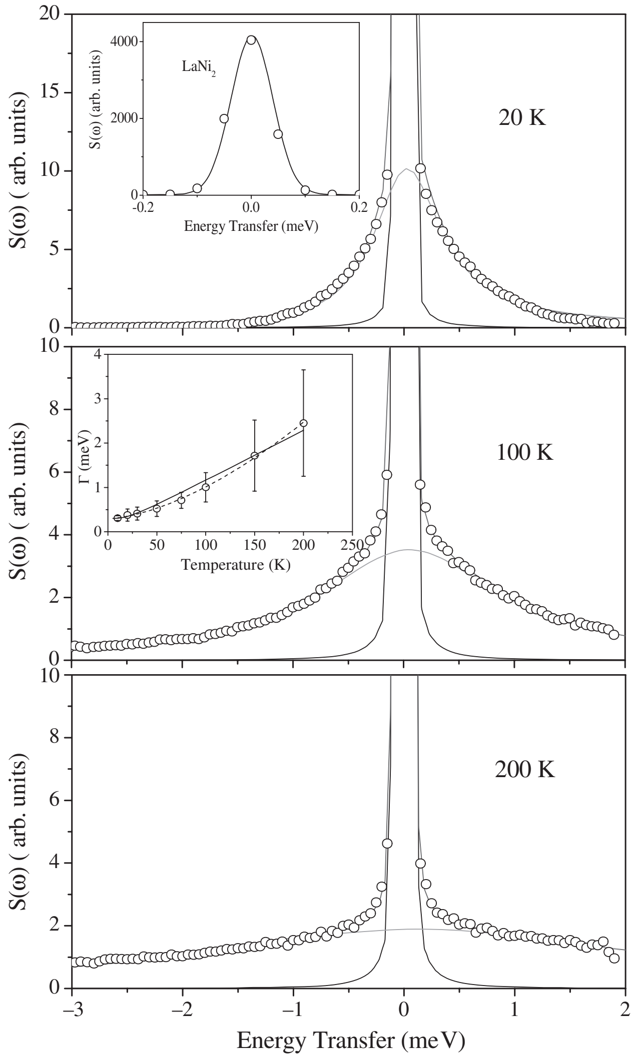


**Figure 1.** The measured scattering function $S(\omega)$ integrated over a wide angular range for $CePt_2$ at 20, 100, and 200 K. The fitted function consists of two components; a broad quasi-elastic Lorentzian and a sharp resolution (elastic) function. The resolution function was determined by fits to the $LaNi_2$ and V data (see the text). The inset in the top frame shows the data for $LaNi_2$. The inset in the middle frame displays the temperature dependence of the quasi-elastic linewidth ($\Gamma$). The dotted curve represents a fit using the formula $\Gamma(T) = \Gamma_0 + \alpha T^{\beta}$ with $\Gamma_0 = 0.30 \pm 0.02$ meV, $\alpha = 4.4(\pm 1.6) \times 10^{-4}$, and $\beta = 1.6 \pm 0.1$ while the solid curve is for the fitting results using the following equation [18]: $\Gamma(T) = I \coth(I/Gk_B T)$ with $I = 0.30 \pm 0.09$ meV and $G = 9.8(\pm 0.4) \times 10^{-4}$ meV $K^{-1}$ (see the text).

determined $\mu$ value should be taken with caution as it can be varied without disrupting the fitting too much. Nevertheless, we stress that the selection of a particular peak function would not interfere with our following discussion. For temperatures above $T_N$ we have included one quasi-elastic component to fit our data for $CePt_2$. The curves in figure 1 show the elastic peak and the quasi-elastic components fitted to the observed scattering integrated over a wide angular range.

In figure 1 we have also plotted the linewidth ($\Gamma$) of the quasi-elastic peak as a function of temperature (inset of figure 1). As one can see, the linewidth decreases with decreasing temperature. The dotted curve through the data points in the inset represents the result of a fit to the following function: $\Gamma(T) = \Gamma_0 + \alpha T^{\beta}$ with $\Gamma_0 = 0.30 \pm 0.02$ meV, $\alpha = 4.4(\pm 1.6) \times 10^{-4}$, and $\beta = 1.6 \pm 0.1$. The small $\Gamma_0$ value and thus small Kondo temperature $T_K \sim 2.2$ K ($T_K = (2\Gamma_0)/(\pi k_B)$) is consistent with the fact that Ce 4f electrons of $CePt_2$ are well localized [8] which is further substantiated by the observation of crystal field excitation in the inelastic neutron scattering [11]. Of particular interest in the plot is that it has a very unusual temperature dependence, different from what has often been observed in other heavy fermion compounds. For example, many heavy fermion compounds show $\beta = 1/2$ when fitted against the same function [13]. However, there are some examples showing non-$\sqrt{T}$ dependence such as $CePb_3$ [14], CeAg [15], and $CeRu_2Si_2$ [16]. There are also some Yb systems showing similar temperature dependence [17]. Although there is no microscopic theory known to us that explains the observed anomalous temperature dependence, nonetheless we note that this behaviour can be explained using an empirical expression [18]: $\Gamma(T) = I \coth(I/Gk_BT)$, where $I$ is the $\Gamma$ value at $T = 0$ K and $G$ the gradient. This function has been successfully used to describe the experimental data obtained on $CeRu_2(Si, Ge)_2$ series [18]. The solid curve in the inset of figure 1(b) represents our fitting results using the same function with $I = 0.30 \pm 0.09$ meV and $G = 9.8(\pm 0.4) \times 10^{-4}$ meV $K^{-1}$. It is worth noting that we have the same value for $\Gamma$ at $T = 0$ K as above, i.e. the same Kondo temperature. Finally, we comment that in both $CePt_2$ and $CeRu_2(Si, Ge)_2$ the crystal field excitation energy, $\Delta_{CEF}$, is much larger than the characteristic energy of Kondo temperature, $T_K$: $\Delta_{CEF} = 20$ meV and $T_K = 2.2$ K for $CePt_2$ while $\Delta_{CEF} \geqslant 35$ meV and $T_K \leqslant 22$ K for $CeRu_2(Si, Ge)_2$ [11, 18]. For heavy fermion compounds, $\Delta_{CEF}$ is usually comparable to $T_K$. This large difference in the two energies may be a reason for the unusual temperature dependence of the quasi-elastic linewidth observed in $CePt_2$ and $CeRu_2(Si, Ge)_2$ [19].

Another interesting aspect of the linewidth ($\Gamma$) of the quasi-elastic peak is that it exhibits a distinct $Q$-dependence: the linewidth increases with $Q$. Data taken at 20 K for $Q = 0.6$ and 1.5 Å$^{-1}$ are shown in figure 2. These data were obtained in the constant $Q$ format by interpolation of the data taken at constant scattering angle shown in figure 1. As shown in figure 2, the linewidth of the quasi-elastic peak increases with increasing $Q$ value. This kind of $Q$ dependence of the linewidth ($\Gamma$) is observed up to 50 K, above which temperature the $Q$-dependence of the linewidth ($\Gamma$) becomes less clear. The summary of the $Q$-dependence of the linewidth ($\Gamma$) is given in the inset of figure 2 for 10, 20, 30, and 50 K. It is also interesting to note that there is a small anomaly in the $Q$-dependence around $Q \simeq 0.9$ Å$^{-1}$, close to $2\pi/a = 0.81$ Å$^{-1}$.

As the temperature is decreased below 10 K, the quasi-elastic component begins to evolve into an inelastic peak. It is thus difficult for us to fit the 5 K data with a quasi-elastic distribution plus the central (resolution-limited) elastic peak. A better fit of the 5 K data is obtained assuming an inelastic peak instead of a quasi-elastic distribution. This change in the character of the scattering with decreasing temperature, we believe, is due to the development of short range magnetic correlations, that is also borne out by a strong increase in the heat capacity below about the same temperature [8]. On cooling further, it becomes clear that the low energy part of the data is dominated by an inelastic process. In order to verify the $Q$-dependence of the inelastic peaks, we converted the data to the constant $Q$ form. Two examples are shown for $Q = 0.7$ and 1.5 Å$^{-1}$ for the data at 1.5 K shown in figure 3. The curves are results of fits using the same elastic peak function with two inelastic peaks. The presence of the inelastic peaks is more pronounced in the lower $Q$ data. The centres of the peaks in the data for $Q = 0.7$ Å$^{-1}$ are located at 0.17 and 0.99 meV, respectively. We note that the lower energy inelastic peak

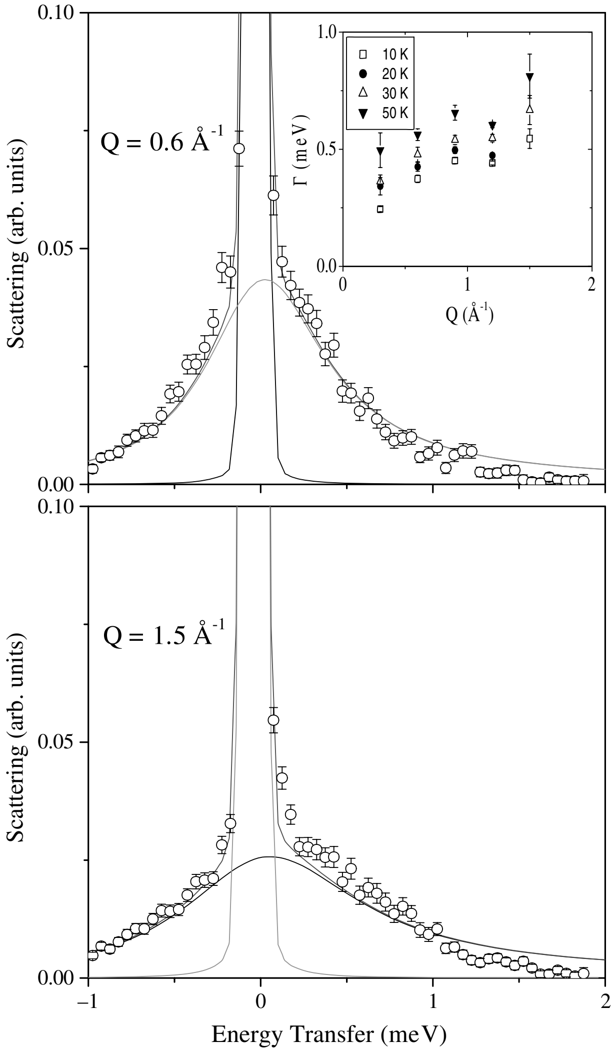


**Figure 2.** Constant $Q$ data at 20 K, obtained by constant $Q$ interpolation of the data shown in figure 1. Curves represent fits obtained using the elastic (resolution-limited) and quasi-elastic spectral components. The inset shows the $Q$ dependence of the quasi-elastic linewidth ($\Gamma$) for four different temperatures; 10, 20, 30, and 50 K.

shows a similar $Q$-dependence to the quasi-elastic peak observed at higher temperatures. On the other hand, the higher energy peak is almost $Q$ independent within our $Q$ range from 0.3 to 1.5 Å$^{-1}$. The inelastic peak at higher energy is clearly due to spin wave excitations of Ce moments in the antiferromagnetic phase, as its temperature dependence demonstrates clearly its association with the low temperature antiferromagnetic phase; the peak at lower energy is probably a residual fraction of the quasi-elastic scattering we observed at higher temperatures. At the lowest temperature, 1.5 K, which is not far below $T_N$, the linewidth of the higher energy inelastic peak is significantly larger than the instrumental resolution. The linewidths of the peaks at $Q = 0.7$ Å$^{-1}$ are 0.30 and 0.47 meV, respectively, while the instrumental resolution at the elastic position is about 0.10 meV. This large linewidth of the inelastic peak results from the powder averaging and perhaps also the short lived nature of the spin wave excitations at 1.5 K.

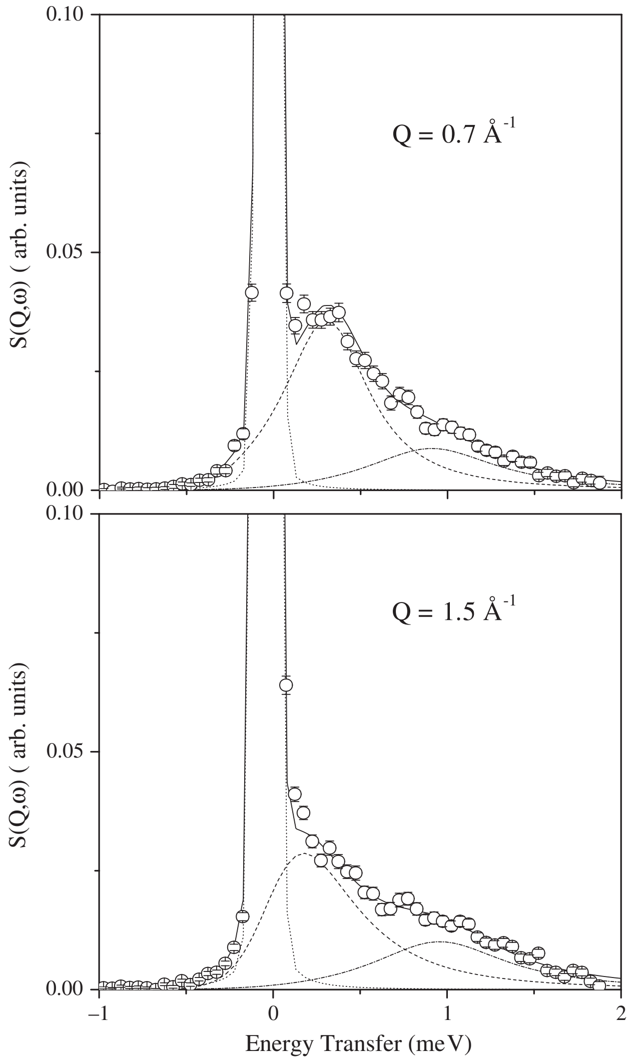


**Figure 3.** Constant $Q$ scattering at 1.5 K for 0.7 and 1.5 Å$^{-1}$, obtained within a $Q$-width of 0.2 Å$^{-1}$. The fits were obtained assuming two inelastic peaks (see the text).

We acknowledge that it is rather unusual to have both the quasi-elastic and the inelastic components in the ordered phase as in figure 3. An alternative explanation would be that the high energy excitation may arise from some other sources such as phonons. However, this scenario is at variance with the experimental facts. First, there is no visible sign of the peak in the data on the energy gain or loss side with increasing temperatures. Second, the $Q$-dependence of the peak is not consistent with the usual $Q^2$ dependence of phonon-related excitations.

Having established that $CePt_2$ shows clear spin wave excitations in the inelastic neutron scattering data taken at ambient pressure, we have studied the pressure dependence of the spectral response. Using a gas pressure cell, we measured the spin dynamics of $CePt_2$ at three different pressures: 1, 4, and 5 kbar. The summary of our data is contained in figure 4.

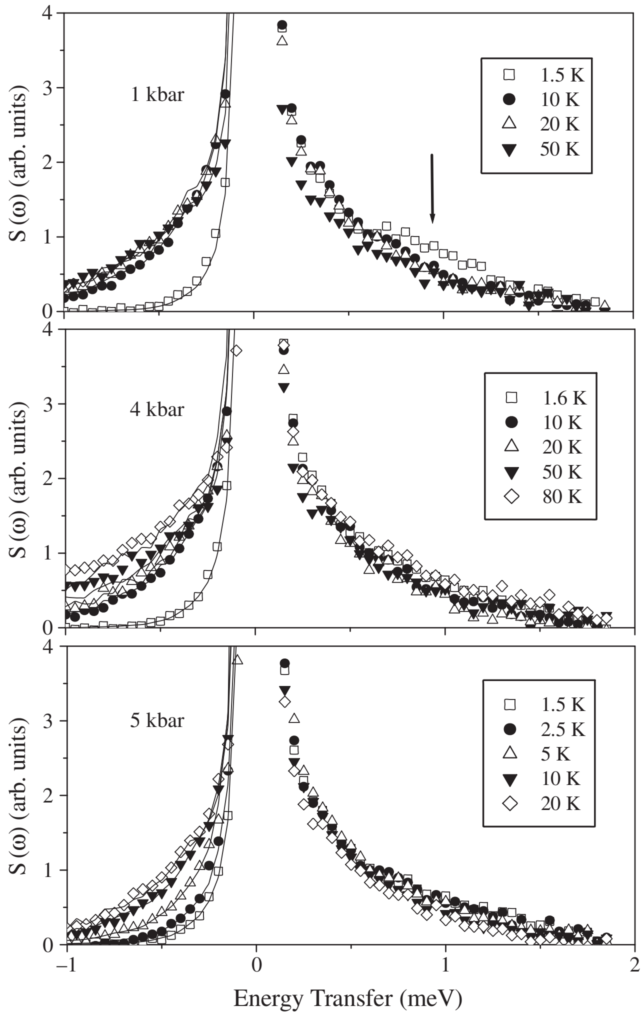


**Figure 4.** Pressure dependence of the scattering at several temperatures. The curves through the data on the energy gain side represent the scattering expected via the detailed balance factor from the measured data on the energy loss side (see the text). The arrow in the top panel indicates where we observed the inelastic peak of spin wave for the ambient pressure data.

Since the pressure cell has a large size and thermal mass, it was important for us to check whether the temperature obtained from the thermometer placed at the top of the cell represented the real temperature of the sample inside the cell. To do this we used the detailed balance principle. The curves through the data points on the energy gain side are the results of our calculations using the data on the energy loss side. We found that there was little difference between the readings from the thermometer and the temperatures determined by this procedure. The temperatures shown in figure 4 are therefore the nominal values read from the thermometer.

It is clear from the top frame in figure 4 that at 1 kbar the spin dynamics of $CePt_2$ has changed little compared with the ambient pressure data. For example, the spin wave excitation near 1 meV is clearly present at 1 kbar and shows a similar temperature dependence to that found at ambient pressure. However, this excitation at 1 meV is not so clealy seen in either the 4

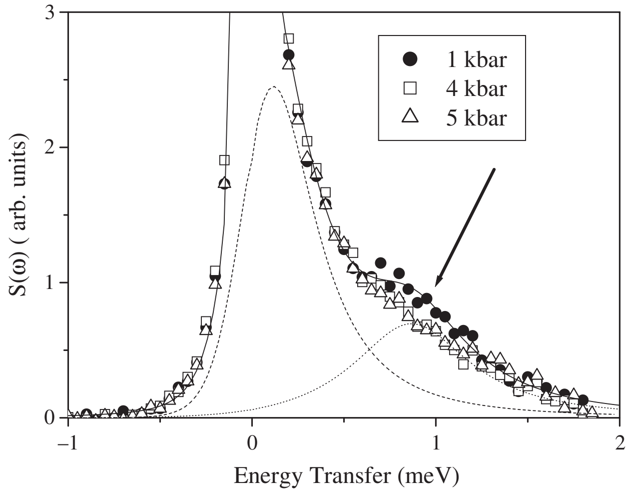


**Figure 5.** Scattering integrated over a wide angular range measured at 1.5 K for 1, 4, and 5 kbar. The arrow indicates the position of the magnetic peak centred around 1 meV.

or the 5 kbar data. In order to illustrate more clearly the effect of pressure on the spin dynamics, we have plotted in figure 5 the data for the three pressures taken at the lowest temperature. The curve through the 1 kbar data points is the results of fits using two inelastic Lorentzian peaks. In the region around 1 meV (indicated by the arrow) the spin wave excitation shows a clear pressure dependence, becoming overdamped around 4 and 5 kbar.

Another interesting point about figure 4 is that at 4 kbar all the data on the energy loss side collapse on top of each other from 1.6 to 80 K while the spectra are strongly temperature dependent on the energy gain side. To further illustrate this point, we plot the data on the energy loss side at 4 kbar in a double-logarithmic plot (see figure 6). It is clear that all the data follow a single straight line of the form $S(\omega) \simeq \omega^{-1}$. This kind of scaling behaviour has previously been observed in other heavy fermion compounds near a magnetic-to-nonmagnetic crossover [3–5]. Unlike these previous studies, however, $CePt_2$ shows the scaling behaviour in a rather limited range of energy, which prohibits detailed analysis of energy ($E$) versus temperature ($T$) relation in the dynamic susceptibility as successfully done in the other systems. Nonetheless, we show a $\chi''T$ versus $E/k_BT$ plot in figure 7. In order to compare our data with two phenomenological functions successfully used previously in [3, 4], we have plotted the two functions together with the data. The solid curve represents the function used in the analysis of $U(Cu_{5-x}Pd_x)$ [3]: $\chi''T^\alpha \approx (E/k_BT)^{-\alpha}\tanh(E/\beta k_BT)$, using $\alpha = 1.0$ and $\beta = 1.2$. We found that for $1.1 \leqslant \beta \leqslant 1.3$ agreement remains almost equally good. The dashed curve is a function used for $Ce(Cu_{6-x}Au_x)$ [4]: $\chi''T^\alpha \approx \sin[\alpha\tan^{-1}(E/k_BT)]/[(E/k_BT)^2+1]^{\alpha/2}$, using $\alpha = 1.0$. It is noticeable that this latter function does not describe our data well. The data points deviating from the fitted function are those that were seen not to follow the linear behaviour in figure 6. This deviation clearly indicates that the characteristic energy scale, above which the scaling behaviour is expected to break down, is quite low for $CePt_2$. This low energy scale is consistent with the small Kondo energy obtained from the bulk properties and our inelastic neutron scattering experiments. As in the case of $Ce(Rh_{0.8}Pd_{0.2})Sb$ but different from $U(Cu_{5-x}Pd_x)$ and $Ce(Cu_{6-x}Au_x)$, a clear CEF excitation is observed for $CePt_2$ at energies higher than the characteristic energy scale. In order to understand a possible microscopic mechanism of the $E/T$ scaling behaviour, we have studied the $Q$-dependence

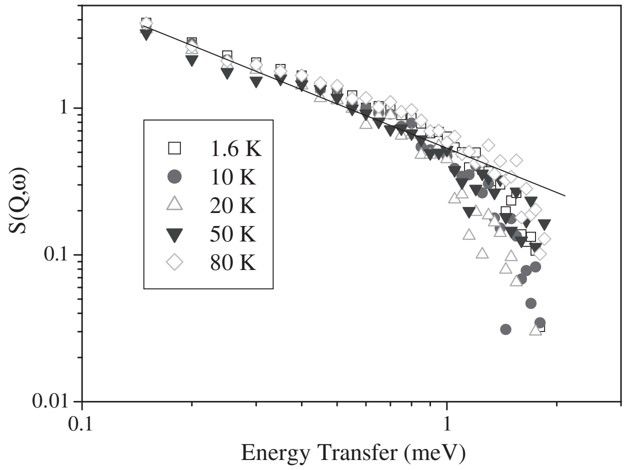


**Figure 6.** Double-logarithmic plot of the total scattering obtained at 4 kbar. The line represents a linear dependence on energy, $S(\omega) \sim \omega^{-\alpha}$ with $\alpha = 1.0$.

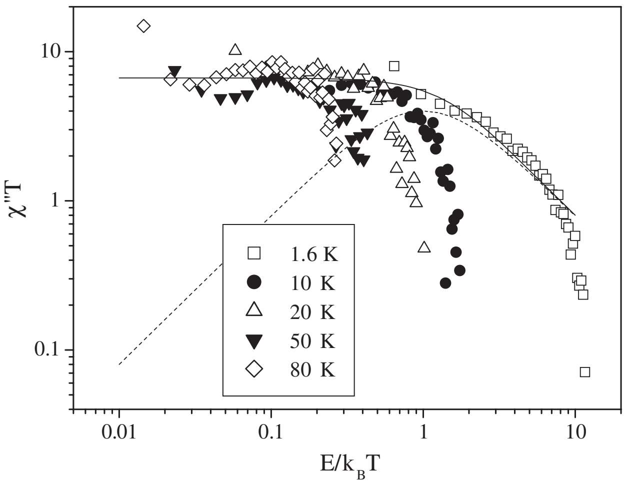


**Figure 7.** A $\chi''T$ versus $E/k_BT$ plot of the data taken at 4 kbar. The solid curve represents the function used in [3] with $\alpha = 1.0$ and $\beta = 1.2$, and the dashed curve the function used in [4] with $\alpha = 1.0$ (see the text).

of the dynamic susceptibility including the scaling plots for several $Q$-values. Our analysis failed to show any appreciable $Q$-dependence in the $E/T$ scaling behaviour. This indicates that the $E/T$ scaling behaviour is more likely to be due to spin fluctuations of local nature.

What distinguishes the present study from the previous investigations on the three heavy fermion compounds showing NFL behaviour [3–5] is that here we have mainly focused on the pressure-driven crossover behaviour without alloying. Although studies of pressure-driven crossover behaviour in the bulk properties have been performed quite frequently, there are very few such studies using the inelastic neutron scattering technique mainly because critical pressures for most magnetic heavy fermion compounds are too high and the available pressure cells operating at high pressures are inadequate for inelastic neutron scattering studies which require the use of large samples.

To summarize, we have studied the dynamic susceptibility of $CePt_2$, which in the antiferromagnetically ordered phase at low temperature and ambient pressure shows clear spin wave excitations. Above $T_N$ the inelastic excitations transform to yield the quasi-elastic response whose width in the limit of $T \rightarrow 0$ K corresponds well with the Kondo temperature estimated from the bulk measurements. With increasing pressure, this magnetically ordered state shows some marked instability at a pressure of around 4 kbar. At this critical pressure, the dynamic susceptibility obtained from the inelastic neutron scattering data exhibits features which resemble those of heavy fermion compounds showing NFL behaviour.

## Acknowledgments

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