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Excitation spectrum of $PrOs_4Sb_{12}$ under a magnetic field

S Raymond[1], K Kuwahara[2], K Kaneko[3], K Iwasa[4], M Kohgi[5], A Hiess[6], J Flouquet[1], N Metoki[3], H Sugawara[7], Y Aoki[5] and H Sato[5]

[1] CEA-DSM/INAC/SPSMS, F-38054 Grenoble, France
[2] Institute of Applied Beam Science, Ibaraki University, Mito 310-8512, Japan
[3] ASRC, Japan Atomic Energy Agency, Ibaraki 319-1195, Japan
[4] Department of Physics, Tohoku University, Sendai 980-8578, Japan
[5] Department of Physics, Tokyo Metropolitan University, Tokyo 192-0397, Japan
[6] Institut Laue Langevin, F-38042 Grenoble Cedex, France
[7] Faculty of Integrated Arts and Science, University of Tokushima, Tokushima 770-8502, Japan

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Abstract

The evolution of the magnetic excitation spectrum of the heavy fermion superconductor $PrOs_4Sb_{12}$ was studied by inelastic neutron scattering on crossing the critical field H_{c2} for superconductivity at low temperature. The peak positions in energy and the peak intensities of the modes of the triplet split by magnetic field confirm the known crystal field parameters for $PrOs_4Sb_{12}$ in T_h symmetry. A selective broadening of the lineshape occurs on increasing the magnetic field: the linewidth of the upper mode of the triplet increases while the one of the middle mode does not.

$PrOs_4Sb_{12}$ is a heavy fermion (HF) superconductor ($T_c = 1.85$ K) that has recently attracted a lot of attention due to the strong interplay between electronic, multipolar and lattice properties and the related different possible mechanisms of electron pairing [1]. The possibility of unconventional superconductivity stems from the observation of time reversal symmetry breaking in μSR [2], the absence of the coherence peak in NQR [3] and the double superconducting transition, the intrinsic nature of which is still under debate [4]. More recently thermal conductivity [5] and NQR measurements [6] strongly suggest the possibility of multiband superconductivity with a fully open superconducting gap. Thus the debate on the nature of the superconducting state of $PrOs_4Sb_{12}$ is quite open. In contrast, the crystal field (CF) and the associated multipolar degrees of freedom are now well characterized with unique features arising from the T_h symmetry [7]. Above 4 T, a field-induced ordered phase (FIOP) occurs with a primary quadrupolar order parameter [8, 9]. The occurrence of an ordered phase in the vicinity of the superconducting pocket of the (H–T) phase diagram leads to the suggestion that quadrupolar fluctuations participate in the pairing [10] by analogy with the scenario of magnetic-mediated pairing for the Ce and U HF compounds. Another common suggestion is that a conventional electron–phonon mechanism assisted by quadrupolar fluctuations is in play [11].

In this context, the characterization of the magnetic excitation spectrum by inelastic neutron scattering (INS) provides important information, especially for studying its interplay with the superconductivity. Compared to Ce and U HF compounds, the 4f electrons in $PrOs_4Sb_{12}$ are well localized. The low lying excitations consist in a singlet–triplet crystal field transition modulated by primarily quadrupolar interactions [12]. The minimum in the dispersion of this so-called quadrupolar exciton occurs at the Brillouin zone boundary $\mathbf{Q} = (1, 0, 0)$, this wavevector being also the propagation vector of the FIOP. The interplay between this exciton and the superconductivity is known from the temperature dependence of the peak linewidth that shows an anomaly at T_c [12]. The aim of the present experiment was to investigate the quadrupolar exciton under a magnetic field on both sides of the upper critical field $H_{c2} = 2.3$ T for superconductivity at low temperature. In this paper, we show that the line broadening occurs for the upper level and not for the middle mode of the triplet when increasing the magnetic field.

INS experiments were performed on the cold three-axis spectrometer IN14 located at ILL, Grenoble. The incident

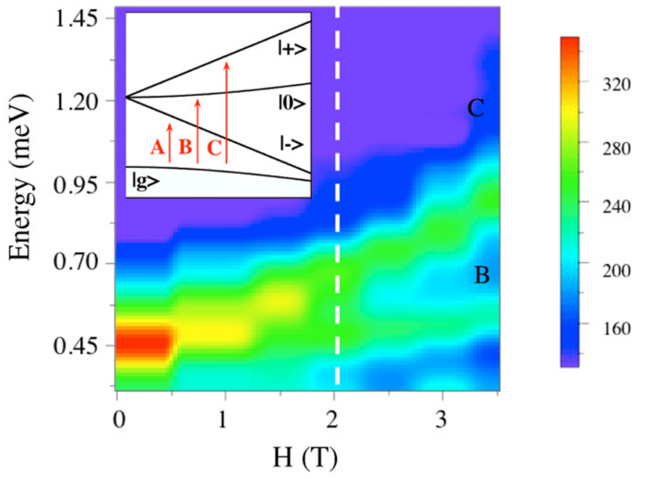

Figure 1. Contour plot of the neutron intensity measured on IN14 (approx. 40 min per point) at $\mathbf{Q} = (1, 0, 0)$ for $T = 100$ mK as a function of magnetic field and energy transfer. The white dotted line delimits the superconducting and normal regions. The inset is a scheme of the singlet–triplet energy levels.

(This figure is in colour only in the electronic version)

beam was provided by a vertically focusing pyrolytic graphite (PG) monochromator. A liquid-nitrogen-cooled Be filter was placed after the monochromator in order to cut down higher-order contamination of neutrons. A horizontally focusing PG analyzer was used. Energy scans at constant $\mathbf{Q}$ along the [1, 0, 0] direction were performed with a fixed final neutron energy of 2.7 meV and collimations open–60′–open–open. The energy resolution determined by the full width at half-maximum (FWHM) of the incoherent signal was 0.07 meV. However, the incoherent tail extends up to 0.4 meV. Therefore in the present paper, data are presented only for energy transfer above 0.4 meV. Preliminary data were taken on IN12 with a similar configuration, except for the fixed final neutron energy of 3.0 meV giving an incoherent FWMH of 0.09 meV. The assembly of two single crystals with a total mass of 6.8 g is the same as in [12]. It was attached to a dilution insert inside a 4 T vertical magnet with the field applied along the [0, 0, 1] direction.

Figure 1 shows a contour plot for the neutron intensity measured as a function of energy transfer and magnetic field at $\mathbf{Q} = (1, 0, 0)$ and $T = 100$ mK. The energy level scheme shown in the inset of figure 1 defines the notations used in this paper. Under magnetic field, the triplet mode splits. The lowest energy mode (transition A) is not observed due to its position near the incoherent tail and its weak intensity (see the calculation below). Representative spectra from which the map in figure 1 is built are shown in figure 2 for $H = 1, 2$ and 3.5 T. As for $H = 0$ T [12], the peaks exhibit substantial broadening and the energy spectra are therefore described by Lorentzian lineshapes rather than by resolution-limited Gaussian lineshapes. The high energy mode (transition C) is broader under magnetic field than at zero field and the middle mode (transition B) has a similar energy width to that at zero field. Concerning the peak magnitudes, the intensity of the upper mode (C) increases while the one of the middle mode (B) decreases when the magnetic field increases.

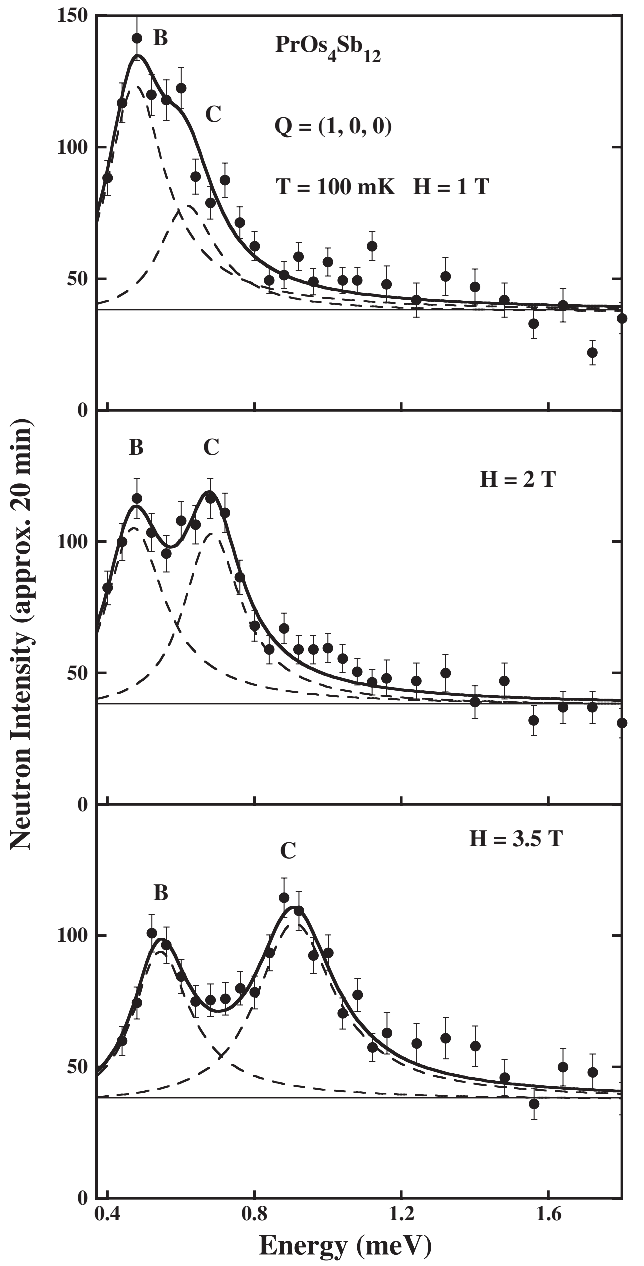

Figure 2. Excitation spectrum measured on IN14 at $\mathbf{Q} = (1, 0, 0)$ for $H = 1, 2$ and 3.5 T and $T = 100$ mK. The dashed lines are Lorentzian fits as described in the text. The horizontal solid line indicates the background.

At zero field and for the highest applied fields ($H \geqslant 2$ T), the data analysis is straightforward because there is either one peak or two well-separated peaks. The FWHM determined for the Lorentzian peaks is shown in figure 3. The linewidth of the central peak is constant under magnetic field with an average FWHM of 0.18 meV while the one of the highest peak increases and reaches an upper value of FWHM of 0.31 meV at 3.5 T. For intermediate magnetic fields, the split peaks overlap. We therefore cannot determine independently all the parameters (two peak intensities, positions and widths). To overcome this difficulty, we fix the peak linewidths for $H = 1, 1.25, 1.5$ and 1.75 T according to the lines shown in figure 3 drawn from the $H = 0$ and $H \geqslant 2$ T data. The values of the

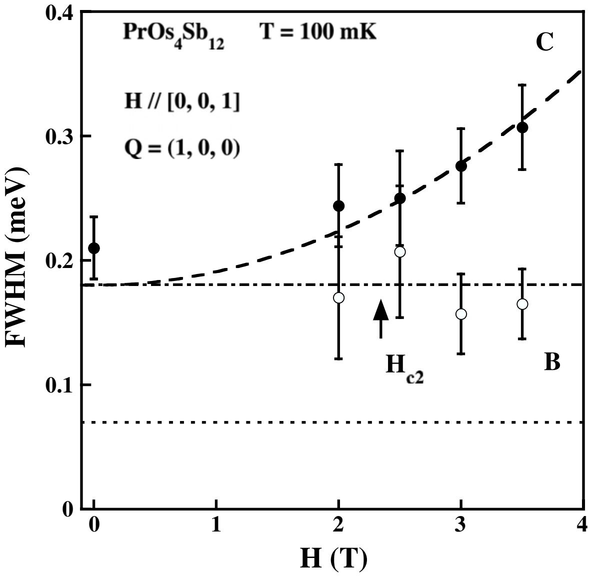

Figure 3. FWHM of the peaks. The dashed line is a quadratic fit, the dotted–dashed line is the average value of the low energy mode linewidth and the dotted line is the incoherent signal FWHM.

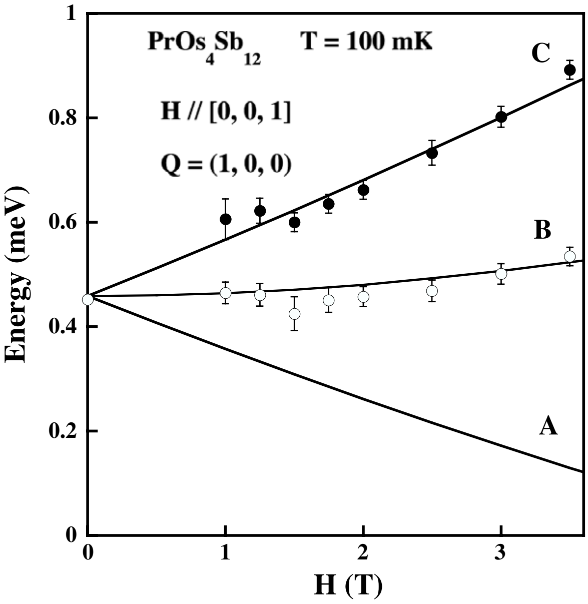

Figure 4. Peak positions as a function of the magnetic field. The lines correspond to the calculation explained in the text.

remaining parameters weakly depend on this hypothesis for the FWHM.

The peak positions and intensities can be calculated by using the CF parameters deduced from a diffraction experiment [8, 9] and neutron spectroscopy [13]. With the CF Hamiltonian for T_h symmetry written in the form

$$H_{CF} = W\left[x\frac{O_6^0 + 5O_4^4}{60} + (1-|x|)\frac{O_6^0 - 21O_6^4}{1260} + y\frac{O_6^2 - O_6^6}{30}\right] \qquad (1)$$

where O_q^k are Stevens' operators, the following values are found: $W = 3.08$ K, $x = 0.460$ and $y = 0.105$ ($y \neq 0$ distinguishes T_h from O_h symmetry) [8, 13]. For $\mathbf{Q}$ along the x axis, the neutron probes fluctuations along the y and z axes. The neutron intensity is proportional to the dynamic structure factor which, in this case, is $S(\mathbf{Q}, \omega) = S^{yy}(\mathbf{Q}, \omega) + S^{zz}(\mathbf{Q}, \omega)$. It is proportional to the sum of the squares of the matrix elements between the singlet ground state $|g\rangle$ and the excited states of the triplet $|m\rangle$ ($m = +, 0, -$): $S(\mathbf{Q}, \omega) \propto |\langle m|J_y|g\rangle|^2 + |\langle m|J_z|g\rangle|^2$. In order to compare the calculation with the measurement, we consider the experimental energy integrated intensities in order to take into account the observed lineshape broadening. The comparison between the measured and calculated energies and intensities are shown in figures 4 and 5, respectively. The calculated intensity of the lowest mode is weak and decreases when the field increases. For both quantities, there is only a single scaling factor between the calculation and the experimental data for $H = 0$ T and there are no other adjustments. Such a normalization is needed because the calculation does not take into account the quadrupolar interactions that renormalize the energy of the mode (see below) and because the neutron data are not calibrated into absolute units. There is a fairly good agreement between the calculation and the experiment. This validates the known CF level scheme and the associated parameters. While the present calculation only takes into account the local non-dispersive CF Hamiltonian (equation (1)), there is a substantial $\mathbf{q}$ dependence of the exciton as the bandwidth is of about 1/3 of the average energy [12]. In order to study the $\mathbf{q}$ dependence of the field effect on the magnetic excitation spectra, data were collected at $\mathbf{Q} = (2.1, 0, 0)$ in the vicinity of the zone centre. Such a spectrum is shown in figure 6 for $H = 0$ and 2.5 T at 65 mK. The same difference as at the zone boundary for the field-induced broadening between the middle mode $|0\rangle$ and the upper mode $|+\rangle$ is observed: only the upper mode linewidth substantially increases at high field. The difference in energy between the upper and middle modes, $\Delta_+ - \Delta_0$, is the same as at $\mathbf{Q} = (1, 0, 0)$ for the same field. Thus the field effect on the magnetic exciton for fields below the FIOP phase is '**q**-independent' except for the modulation in energy and intensity induced by the antiferroquadrupolar interactions.

The main mechanism of relaxation of the 4f electrons is, at low temperatures, the interaction with conduction electrons (f–c interaction) while at higher temperatures phonon scattering or exciton–exciton scattering can be active. In a previous paper, we showed a sharp increase in the temperature dependence of the relaxation rate of the triplet above T_c as the main effect of the interplay between exciton and superconductivity [12]. The change in the relaxation rate values between the superconducting and normal phase is associated with the depression of the conduction electron density of states related to the opening of the superconducting gap. This phenomenon is well known for rare earth ions dissolved in superconducting materials, both for conventional and unconventional ones [14, 15].

In the present work, we report an increase of the exciton linewidth in the normal state (above H_{c2}) when increasing the magnetic field at very low temperature. Due to the

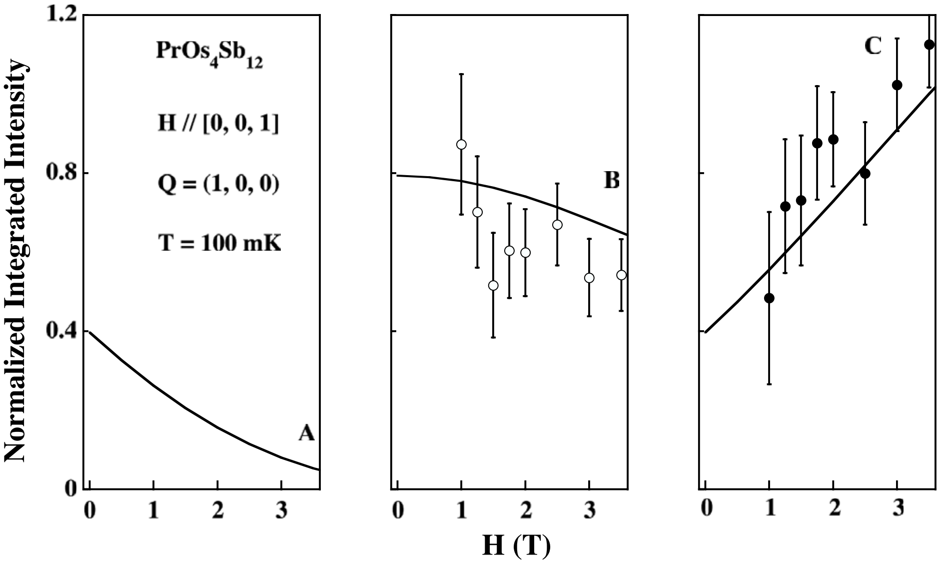

Figure 5. Energy integrated intensities as a function of magnetic field. Each panel shows one mode of the triplet. The lines correspond to the calculation explained in the text.

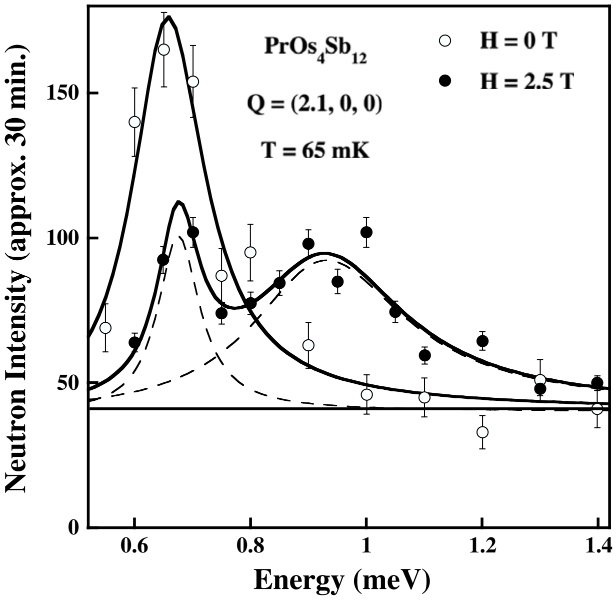

Figure 6. Excitation spectrum measured on IN12 at $\mathbf{Q} = (2.1, 0, 0)$ for $H = 0$ and 2.5 T for $T = 65$ mK. Lines are Lorentzian fits.

overlap of the Zeeman split peaks, we cannot discuss the field dependence of the linewidth inside the superconducting phase. The presence of vortices in the mixed state will certainly induce a smooth field dependence of the peak width in contrast to the sharp effect observed at T_c [12]. Our main result is that the line broadening is selective. Indeed, a significant broadening is observed only for the upper mode of the triplet while no broadening is observed for the middle mode within the accuracy of the experiment. This selective broadening of the peaks contains microscopic information that allows us, in principle, to better characterize the coupling between f and conduction electrons in $PrOs_4Sb_{12}$. Within the pseudo-quartet considered, the f–c interaction can be of exchange type (magnetic origin) or from the aspherical part of the 4f distribution (quadrupolar origin). For each mechanism, the broadening of the triplet mode involves an inelastic transition between the singlet and the triplet at low temperatures (at higher temperatures, elastic scattering within the triplet is also to be taken into account). For the magnetic case, the relaxation rate is at low temperatures $\Gamma_{\mathrm{mag}} \propto g_{\mathrm{mag}}^2 \times \Delta \times M_{\mathrm{dip}}^2$, where $g_{\mathrm{mag}} = J_{\mathrm{ex}}\rho$ with J_{ex} the f–c exchange interaction and ρ the conduction electron density of states, Δ is the excitation energy and M_{dip} is a dipolar matrix element related to the singlet–triplet transition [17, 18]. We assume that a similar formula applies for the non-magnetic mechanism defining a relaxation rate $\Gamma_{\mathrm{quad}} \propto g_{\mathrm{quad}}^2 \times \Delta \times M_{\mathrm{quad}}^2$ with another coupling constant g_{quad} related to the quadrupolar interaction between f and c electrons and M_{quad} corresponding to quadrupolar matrix elements for the singlet–triplet transition.

We examine below each parameter in play. The field dependence of the gap is shown in figure 4. Since the upper mode energy increases, the broadening of this mode is therefore expected to be higher than the one of the middle mode whatever the mechanism in play. The mixing of the 4f and conduction electrons occurs with the a_u and t_u orbitals of the Sb_{12} cage. The coupling with the a_u orbital is of magnetic exchange type and involves a transition within the triplet while the coupling with the t_u band has both magnetic and non-magnetic channels and involves transitions from the ground state to the triplet [16]. The coupling constants are found to follow $g_{\mathrm{quad}} \approx g_{\mathrm{dip}}/3$ [11, 16]. They are not expected to be magnetic-field-dependent. On the other hand, the dipolar (M_{dip}) and quadrupolar (M_{quad}) matrix elements change under magnetic field. The squares of the different matrix elements to be considered are given in table 1 for a field of 3 T. Our data clearly exclude the relaxation processes associated with the operators that have no matrix elements with the upper mode of the triplet. Therefore we conclude that J_z and O_{xy} are not involved in the lineshape broadening. The quadrupolar

Table 1. Squares of the multipolar transition matrix elements between the ground state $|g\rangle$ and the triplet states $|m\rangle$ ($m = +, 0, -$) at 3 T. The microscopic parameters of the CF Hamiltonian (equation (1)) are taken from [8].

	J_x	J_y	J_z	O_{yz}	O_{zx}	O_{xy}
$m = +$	0.70	0.90	0	10.50	17.39	0
$m = 0$	0	0	0.69	0	0	30.69
$m = -$	0.16	0.08	0	21.07	12.39	0

matrix elements are larger by one order of magnitude than the dipolar ones. However this effect on the relaxation rates Γ_{magn} and Γ_{quad} is counteracted by the differences in the coupling constants. Therefore definitive conclusions on the magnetic versus non-magnetic mechanism in play cannot be drawn.

Among the different possibilities, we suggest that one mechanism is favoured along the following lines. For $H \parallel [0, 0, 1]$, the quadrupole O_{yz} has the largest matrix element between the ground state and the lower $|-\rangle$ mode (the $|-\rangle$ mode reaches zero energy at 4.2 T). It has been experimentally determined that O_{yz} is the primary order parameter of the FIOP [8]. Thus, the fluctuations associated with this quadrupole should be strong in the non-ordered phase near the FIOP. In this case, it is worthwhile to note that the quadrupole O_{yz} has no matrix elements between the ground state and the middle mode of the triplet and a sizeable one between the ground state and the upper mode of the triplet (see table 1). Because O_{yz} is the order parameter of the FIOP and because its matrix elements are consistent with the observed selective broadening, we suggest that the quadrupolar fluctuations associated with O_{yz} are responsible for the broadening of the mode as the main source of scattering between 4f and conduction electrons. This suggestion deserves further theoretical and experimental work.

While our conclusions are not definitive, we stress the fact that we are not aware of any other neutron studies concerning exciton line broadening as a function of magnetic field. The interpretation of such data is certainly more complicated than the study of the exciton temperature dependence but it allows, in principle, to discuss in detail the relevance of microscopic models. Concerning the study of other bosons suspected to participate in the superconducting pairing, inelastic neutron scattering data are scarce. Changes in the bosons' excitation spectra are observed for phonon-mediated superconductivity (YNi_2B_2C [19]) and magnetic-fluctuation-mediated superconductivity (UPd_2Al_3 [20]). As opposed to these intermetallic compounds, the critical fields are too large in high-T_c superconductors to be able to observe sizeable effects on the excitation spectra [21].

Accurate measurements of the field effect on the magnetic excitation spectrum of $PrOs_4Sb_{12}$ when crossing the superconducting to normal transition at H_{c2} reveal the broadening of the exciton lineshape. It is not clear if the broadening is gradual or occurs at H_{c2}. Selective broadening is observed and concerns the upper mode of the triplet. This observation cannot distinguish between magnetic and non-magnetic relaxation processes between f and conduction electrons due to the combined effect of coupling constants and matrix elements. The interaction between the exciton and the conduction electrons is now confirmed in a magnetic field at low temperature in the normal phase.

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