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PRESSURE DEPENDENCE OF THE CRYSTAL FIELD STATE OF A LOW CARRIER SYSTEM CeP

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Abstract

Pressure dependence of the crystal field splitting in CeP has been studied by inelastic neutron scattering up to 0.8 GPa. It was found that the Γ_7 - Γ_8 crystal field splitting in CeP decreases by about 10 % from that at ambient pressure by applying pressure of 0.8 GPa at the temperatures below 60 K. An evidence for the temperature dependence of the pressure effect is seen.

Keywords: high pressure, crystal field, neutron scattering, low carrier, CeP

Running title: PRESSURE DEPENDENCE OF CRYSTAL FIELD IN CeP

1. Introduction

Ce-monopnictide, CeX (X=P, As, Sb, Bi), is a low carrier system which exhibits unusual magnetic and transport properties with Kondo-like anomalies. So far the studies of this system were mainly concentrated on the heavier Ce-monopnictides CeSb and CeBi because these compounds exhibit highly unusual properties such as a very small crystal field splitting and a complex magnetic phase diagram. Recently, the lighter compounds CeP and CeAs have also attracted attention since it was revealed that the carrier numbers in CeP and CeAs are very small, the order of 10^{-3} carriers per a formula[1,2], which is a factor ten smaller than that of CeAs or CeBi, and that these compounds also exhibit unusual magnetic phase diagrams. Both compounds show the type I antiferromagnetic ordering below about 10 K at ambient pressure. With applying a pressure above about 0.3 GPa(CeP) or 1.1 GPa(CeAs), a new magnetic phase with a ferromagnetic component is seen just above the antiferromagnetic phase in each compound[3]. The obtained P-T phase diagram is quite similar for both compounds, but CeP shows the anomaly at lower pressure. These facts indicate that the 4f electronic state in these compounds is quite similar and very sensitive to the atomic distance.

The purpose of the present paper is to report a part of the neutron scattering study on the pressure dependence of the crystal field splitting in CeP using a chopper spectrometer. The same kind of study on CeAs using a triple axis spectrometer has been performed in parallel with the present study. The result of CeAs is reported in a separate paper[4].

2. Experimental details

polycrystalline samples of CeP and its phonon reference material LaP were prepared by synthesizing initial materials of the required composition in quartz tubes at 800°C for 45 days. X-ray powder diffraction analysis showed that there is no appreciable other phases than the target material in the samples.

The neutron scattering experiments were performed on the chopper spectrometer INC at the pulsed neutron source KENS in

the National Laboratory for High Energy Physics. A pressed CeP polycrystalline sample of about 10 g was sealed in a thin aluminum cell(the sample space is 1 cm diameter and 5 cm long) with the non-hydrogenous pressure media(Fluorinert, Sumitomo 3M Co., Ltd.). The cell was inserted in a clamp-type high pressure sample container made by aluminum. Then, the sample was pressurized up to 0.8 GPa. The outer diameter of the sample container is 8 cm, however, two cuts with the depth of 2 cm are made on the wall along the neutron incident beam(1 cm by 5 cm) and the scattered neutron paths with the scattering angle less than 12° in order to decrease the scattering from the aluminum wall. All the surfaces of the high pressure container except those in the neutron paths were covered by cadmium sheets of 0.5 mm thick. The container was mounted within a closed cycle refrigerator cryostat on the INC spectrometer. The neutron scattering measurements were done at 20, 40, 60 and 80 K. The incident neutron energy was chosen at 40 meV. The resolution of the spectrometer for this energy is about 2.2 meV(FWHM) at $\hbar\omega = 0$.

The measurement on LaP was also done at 20 and 80 K with the same condition but only at ambient pressure in order to estimate the phonon background originated from the sample itself as well as from the wall of the high pressure sample container. The ammount of the LaP sample was chosen so that its total nuclear scattering cross section becomes equivalent to that of the CeP sample.

3. Results and Discussion

Fig. 1 shows spectra of CeP at 1 atm(open circles) and 0.8 GPa(closed circles) at (a) 20 K and (b) 80 K taken in the low angle bank of detectors(2θ = 5° - 12°). The broken lines represent the result of the measurement on LaP sample at 1 atm at the same temperatures, which can be used as the phonon background in the present experimental condition. Note that the LaP data at 20 K and 80 K are quite similar. Clearly, the peaks around 13 meV in the CeP data correspond to the Γ_7 - Γ_8 crystal field excitations in CeP.

Fig. 2 (a), (b), (c) and (d) show the pressure dependence of the magnetic response from CeP at 20, 40, 60 and 80 K, respectively,

which were obtained from the CeP data by subtracting the LaP data as the phonon background. For the phonon background at 40 and 60 K, the LaP data at 20 and 80 K, respectively, were used. This may be allowed since the temperature dependence of the LaP data is rather small. The pressure dependence of the phonon background was also assumed to be negligible. The solid and broken curves in the Fig. 2 show the results of least squares fit to the data at ambient pressure and at 0.8 GPa, respectively, where the spectral shape function was assume to be a sum of two Lorentzians, one for the quasi-elastic scattering and the other for the crystal field excitation. The peak positions of the crystal field excitations decrease by applying pressure. However, no big change of the spectral shape is seen.

The peak positions of the crystal field excitations and their linewidths obtained from the Lorentzian fitting are summarized in Fig. 3(a) and (b), respectively. The peak position at ambient pressure decreases linearly with temperature up to 60 K, and shows a bit faster decrease at 80 K. The values at 20 and 80 K are about 5 % smaller than those reported by Heer et al. at 15 and 77 K, respectively[5]. At 0.8 GPa, the peak position also decreases linearly with temperature up to 60 K but with about 10 % lower value than that at ambient pressure, whereas the value at 80 K is almost same as that at 60 K, thus the pressure dependence of the peak position becomes smaller(about 5 %) than that at lower temperature. The linewidth at ambient temperature increases slowly from 20 K to 40 K, then becomes to be almost proportional to the temperature above 40 K. At 0.8 GPa, the linewidth behaves nearly the same as that at ambient pressure but with about 10 to 20 % larger value than that at ambient pressure up to 60 K and again the pressure dependence becomes small(almost same linewidth) at 80 K.

The above features of the pressure dependence of the crystal field excitation seem to be correlated with the observed anomalies in the electrical resistivity[3]. At ambient pressure, the resistivity shows a sharp peak at the Néel temperature(about 10 K). After passing through a minimum at about 20 K, the resistivity shows a broad peak at around 70 K, above which it shows a Kondo-like decrease with increasing temperature. At the pressure of about 0.8 GPa, the broad

peak shifts to the lower temperature side by about 10 %. The observed decrease of the crystal field splitting at 0.8 GPa is also about 10 % at low temperatures as described above. This coincidence suggests that the broad peak of the resistivity is strongly related with the crystal field splitting. Another interesting feature is that, at 80 K which is the temperature just above the broad peak of the resistivity, the pressure dependence of the crystal field excitation is small compared with those at the lower temperatures. This indicates that the electronic state in CeP at the temperature region below the broad peak position of the resistivity is more sensitive to the pressure than that at higher temperature.

More detailed study including the experiment at higher pressure is under way.

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Figure captions

Fig. 1 Spectra of CeP at (a) 20 K and (b) 80 K measured at ambient pressure(open circles) and 0.8 GPa(closed circles). Broken lines represent the phonon background estimated from the LaP data.

Fig. 2 Pressure dependence of the magnetic response from CeP at (a) 20 K, (b) 40 K, (c) 60 K and (d) 80 K. Solid and broken lines are the results of a Lorentzian least squares fit to the data at ambient pressure and 0.8 GPa, respectively.

Fig. 3 Pressure dependence of (a) the excitation energy and (b) the linewidth(HWHM) of the crystal field excitation in CeP versus temperature.

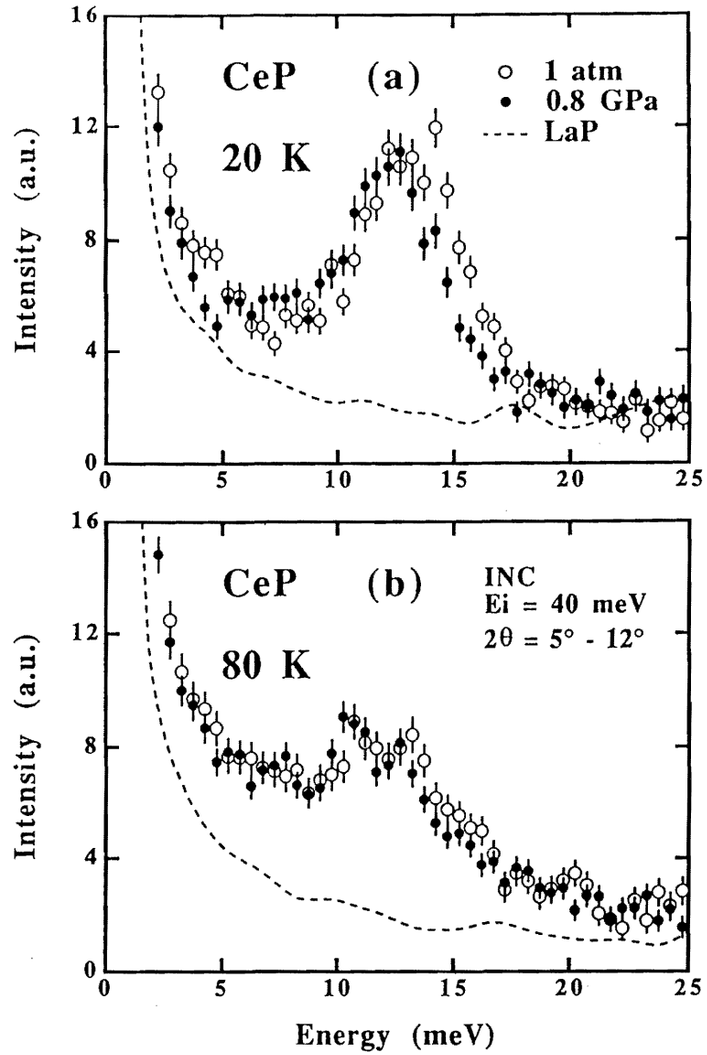
CeP (a)
20 K
1 atm
0.8 GPa
LaP
CeP (b)
80 K
INC
Ei = 40 meV
2θ = 5° - 12°
Intensity (a.u.)
Energy (meV)

Fig. 1

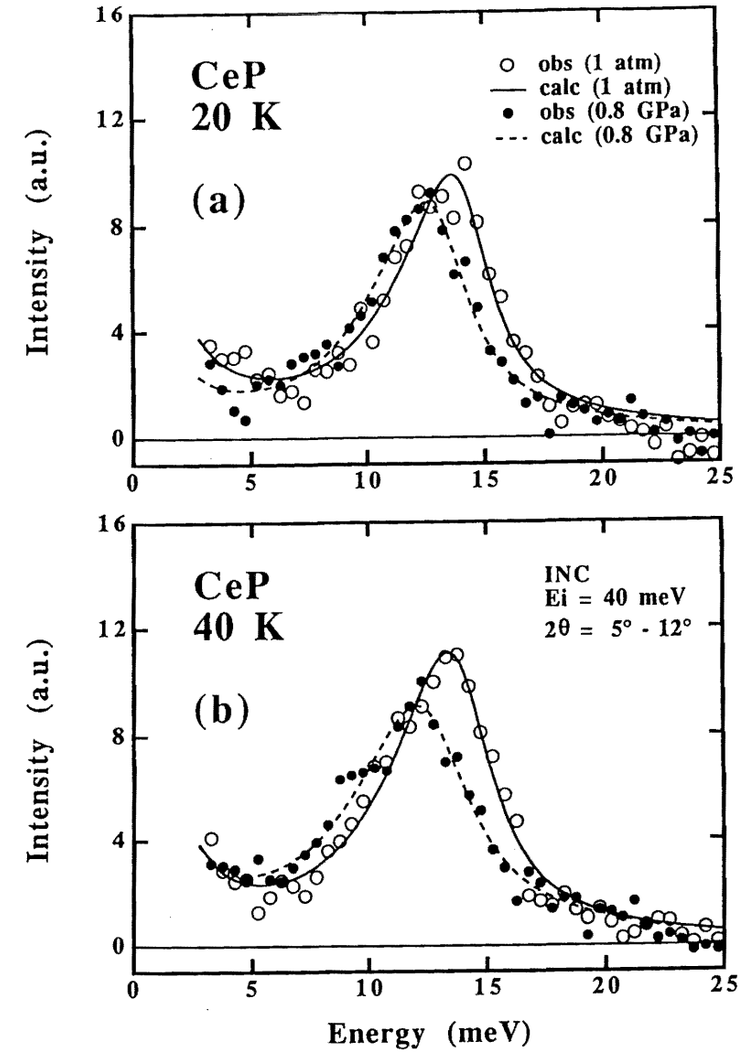
CeP
20 K
(a)
obs (1 atm)
calc (1 atm)
obs (0.8 GPa)
calc (0.8 GPa)
CeP
40 K
(b)
INC
Ei = 40 meV
2θ = 5° - 12°
Intensity (a.u.)
Energy (meV)

Fig. 2(a),(b)

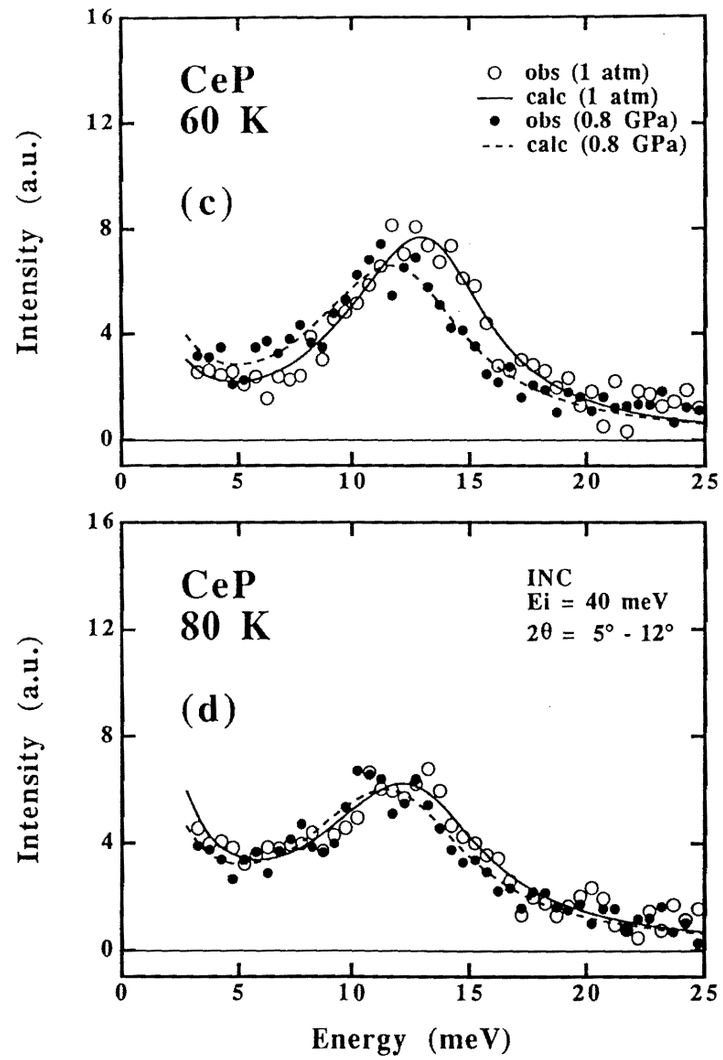
CeP
60 K
(c)
obs (1 atm)
calc (1 atm)
obs (0.8 GPa)
calc (0.8 GPa)
CeP
80 K
(d)
INC
Ei = 40 meV
2θ = 5° - 12°
Intensity (a.u.)
Energy (meV)

Fig. 2(c),(d)

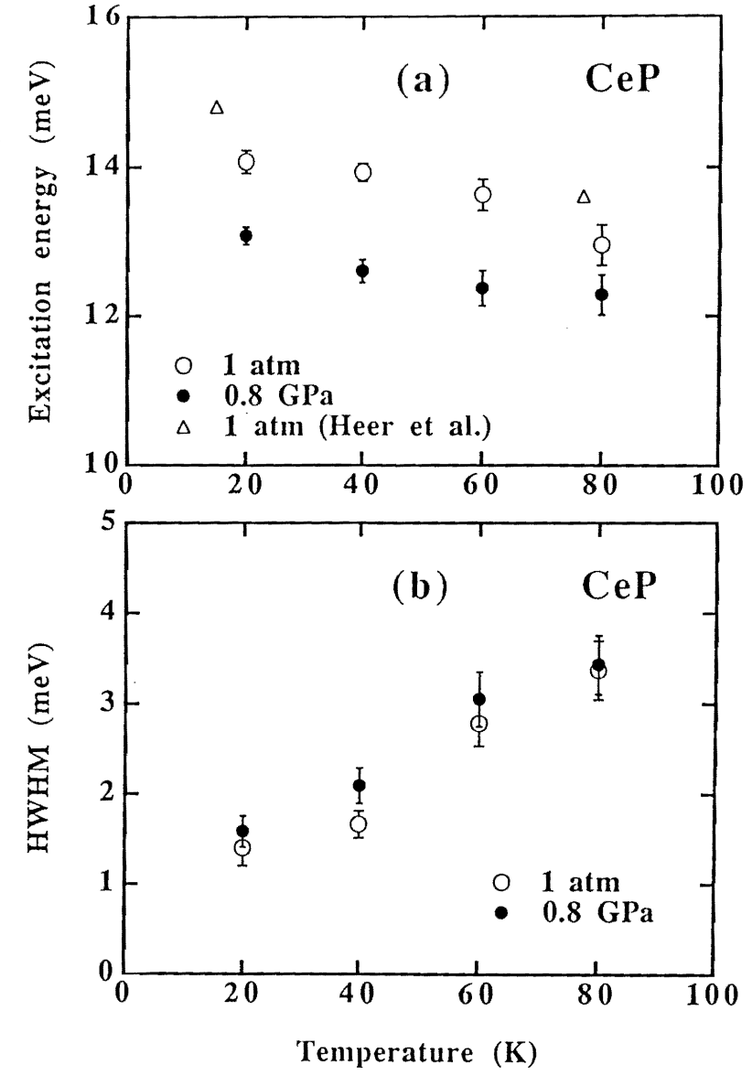
(a)
CeP
1 atm
0.8 GPa
1 atm (Heer et al.)
Excitation energy (meV)
(b)
CeP
1 atm
0.8 GPa
HWHM (meV)
Temperature (K)

Fig. 3