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# Pressure effects of susceptibility and specific heat in $PrCu_2$

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**Abstract**

The pressure effects on magnetic susceptibility, specific heat and the co-operative Jahn–Teller transition temperature, $T_{JT}$, in the single crystalline sample are presented. At $P = 1.2\,GPa$ susceptibility shows remarkable maximum at $T = T_{max}$ while specific heat shows no anomalous change around $T_{max}$. However, at $T \ll T_{JT}$ the upturn of nuclear specific heat is developed with pressure. Our findings imply that a magnetic ordering is induced by pressure. © 2001 Published by Elsevier Science B.V.

*Keywords:* High pressure; Jahn–Teller effect; Magnetization; Specific heat—low temperature

The Van Vleck paramagnet $PrCu_2$ having the orthorhombic $CeCu_2$-type structure exhibits the co-operative Jahn–Teller transition at $T_{JT} = 7.8\,K$ and the nuclear magnetic ordering at 54 mK [1]. Recently, a new type of metamagnetic transition in $PrCu_2$ has been investigated intensively under high field. It is remarkable that the hard (*c*-axis) magnetization is switched to the easy (*a*-axis) one through the metamagnetic transition at $H = 100\,kOe$ [1]. It is pointed out by the analysis of magnetization and magnetostriction that the metamagnetic transition is attributed to the reorientation of the quadrupole moment. In the case of $Pr^{3+}$-ion in a crystal, a singlet orbital ground state is established by crystal field effects. The susceptibility approaches a temperature-independent value at low temperature. Nevertheless, some Pr intermetallic compounds undergo a magnetic ordering attributed to an inter-ionic exchange interaction [2–4]. It is expected in the orthorhombic $PrCu_2$ that the small crystal field splitting [1] is competitive with the exchange interaction between the Pr ions. Generally, the compression of a crystal modifies the electronic state through a one-ion magneto-elastic interaction and/or a two-ion interaction, such as, quadrupolar coupling and the RKKY interaction with conduction band electrons [5]. In this paper we present a pressure-induced magnetic anomaly in $PrCu_2$ that seems to be attributable to a magnetic transition at around $P > 1.2\,GPa$.

The magnetization at high pressure was measured using the Faraday method up to $P = 1.8\,GPa$ above $T = 1.7\,K$. The single crystals made by a Czochralski pulling method in an induction furnace under helium gas atmosphere [1] were filled into a Teflon cell with Fluorinert as pressure media and pressurized in a piston–cylinder-type clamp made by CuBe alloy. We have measured the specific heat with a quasi-adiabatic method under pressure up to 1.4 GPa between 0.7 and 15 K. A mixture of the powdered sample and the pressure media of AgCl was used in specific heat measurement. The details of the measurement procedure have been previously described elsewhere [6,7].

Fig. 1 shows the pressure dependence of susceptibility along the *a*-axis, $\chi_a(T)$. Since at lower pressure $\chi_a(T)$ approaches a constant value, a singlet orbital ground state seems to be realized in $PrCu_2$. $\chi_a(P)$ increases at lower pressure and attains a maximum at $P_{max} = 1.2\,GPa$ and then decreases rapidly with pressure. Above $P_{max}$ $\chi_a$–$T$ curves show obviously a maximum at $T = T_{max}$. $T_{max}$ increases with pressure. It is noteworthy that $T_{max}$ decreases with the ratio of $dT_{max}/dH \approx -0.15\,K/kOe$ that is comparable with that of antiferromagnetic transition temperature for electronic spin systems, $T_c$: $dT_c/dH \approx -\mu_e/k_B$, where $\mu_e$ and $k_B$ are the ordered 4f moment and the

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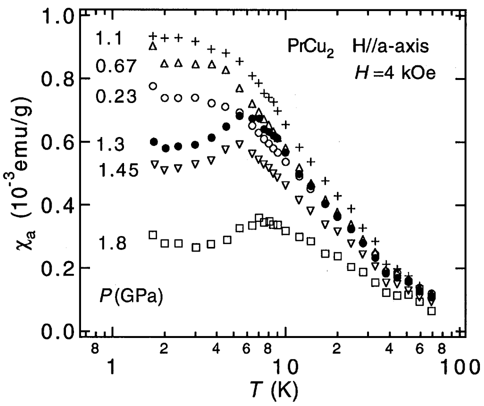


Fig. 1. Magnetic susceptibility of $PrCu_2$ at $H = 4\,kOe$ as a function of temperature and pressure.

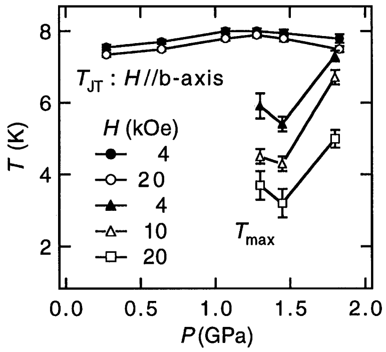


Fig. 2. Pressure dependence of $T_{JT}$ and $T_{max}$ at various fields.

Boltzmann constant, respectively. At $P = P_{max}$, the Jahn–Teller transition temperature, $T_{JT}$, exhibits a broad maximum while its magnetic field dependence is rather weaker than that of $T_{max}$ as shown in Fig. 2. $T_{JT}(P)$ is determined by the measurement of susceptibility along the *b*-axis, $\chi_b(T)$, which possesses a shoulder at $T_{JT}$ [1]. At ambient pressure $T_{JT}$ has a very weak field dependence with the field directions perpendicular to the *a*-, *b*- and *c*-axis below 20 kOe [8].

On the other hand, the results of specific heat measurements under zero-field depend slightly on pressure. Even at $P > 1.2\,GPa$ no additional anomaly is observed at around $T_{max}$ with that of the co-operative Jahn–Teller transition (Fig. 3). However, as shown in Fig. 3, a low temperature upturn is developed with pressure. In the magnetically ordered compounds, it is clearly indicated that the $T^{-2}$-term in nuclear specific heat is observed below $T_c$ [9].

Let us consider magnetic ordering in magnetic substances having a singlet ground state where exchange interaction is comparable with crystal field effects [2–4,10]. The electronic states can be categorized by

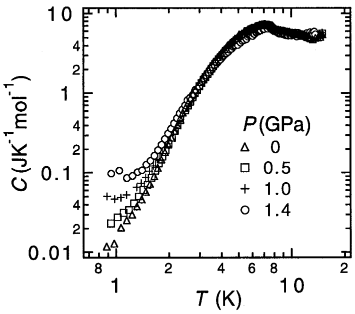


Fig. 3. Temperature dependence of specific heat, $C(T)$, at various pressures.

using a parameter, $\eta = 4K_0\langle 0|J_z|1\rangle^2/\Delta$, where $K_0$, $\langle 0|J_z|1\rangle$ and $\Delta$ are an exchange coupling constant between electronic moments on the Pr ions, a matrix element of the total angular moment and the energy difference between the ground and the first excited states, respectively [2–4]. The system undergoes an electronic magnetic ordering at $\eta \geqslant 1$ (ferromagnetic) or at $\eta \leqslant -1$ (antiferromagnetic), but no transition at $|\eta| < 1$ [2–4]. In such a system the anomaly in specific heat associated with the magnetic transition is rather smaller than that of degenerated ground state systems and a nuclear $T^{-2}$ term appears below $T_c$ [11]. It is possible, therefore, that the upturn in specific heat results from the developing spontaneous hyperfine field produced by the ordered 4f moment. To estimate the hyperfine field we can try to fit the data below 3.5 K to a fitting function, $C(T) = \gamma T + \beta T^3 + cT^{-2} + b\log(-\Delta_{JT}/k_B T)$, where the first, second and third terms are electronic, lattice and nuclear contributions, respectively. The last term is of the co-operative Jahn–Teller transition which is valid at $T \ll T_{JT}$. From the obtained value of $c = 0.078\,K^2$ at $P = 1.4\,GPa$ the hyperfine field is derived to be $H_{hf} = 950\,kOe$ which is comparable to that of the magnetically ordered states in rare earth elements and compounds [12,13]. The observed facts, the maximum of susceptibility at $T_{max}$ at $P > 1.2\,GPa$ and the upturn in specific heat at $T \ll T_{JT}$, indicate that an antiferromagnetic ordering is realized by applying pressure in $PrCu_2$.

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