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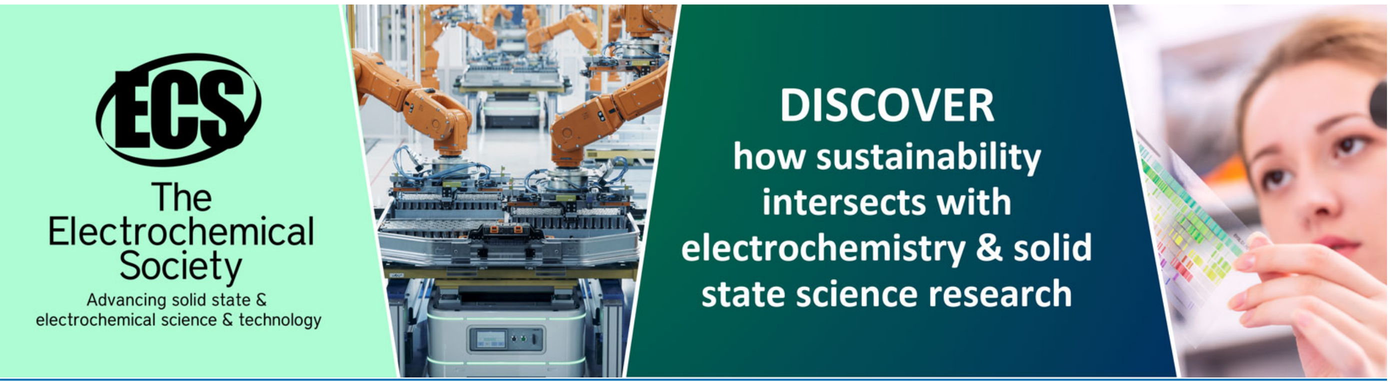


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# Magnetic susceptibility of $PrMg_3$ at ultra low temperatures

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**Abstract.** We report susceptibility measurements of $PrMg_3$ with a non-magnetic $\Gamma_3$ crystalline-electric-field ground state down to 0.3 mK. The susceptibility shows the Van-Vleck-like behavior with an additional logarithmic temperature dependence below 10 K. Furthermore, the susceptibility exhibits a small peak around 0.1 K, which is attributed to the quenching of the multipole degrees of freedom. Below 30 mK, the susceptibility is almost constant down to the lowest temperature without showing the contribution of Pr nuclear spins. The disappearance of nuclear spin contribution suggests that the quenching of the multipole degrees of freedom strongly affects the nuclear spin system.

## 1. Introduction

Recently, there has been growing interest in Pr-based compounds with the $f^2$ configuration of $Pr^{3+}$ as a possible strongly correlated electron system arising from the hybridization between the conduction and $f$ electrons ($c$-$f$ hybridization). A $Pr^{3+}$ ion in the crystalline-electric-field (CEF) of cubic symmetry can have the non-magnetic $\Gamma_3$ ground state (GS) with electric quadrupole moments $O_{20}$ and $O_{22}$, and magnetic octupole $T_{xyz}$. Therefore, in $\Gamma_3$ GS systems such as $PrPb_3$ and $PrAg_2In$, an observation of nature due to the coupling between the $c$-$f$ hybridization and $4f^2$ multipole degrees of freedom is strongly expected at low temperatures. Indeed, neutron diffraction measurements for $PrPb_3$ have revealed a modulated antiferro-quadrupole phase, which suggests a partial quenching of local quadrupole by a quadrupole Kondo effect [1]. In addition, a broad peak in the specific-heat of $PrAg_2In$ at 0.4 K is caused by not a cooperative phase transition including a quadrupole order but a quadrupole Kondo effect [2, 3]. However, the mechanism for suppression of a quadrupole order has not yet been established. It is intriguing to investigate $\Gamma_3$ GS systems as strongly correlated electron systems with the multipole degrees of freedom.

From a viewpoint of the nuclear magnetism, the magnetic field at the Pr nuclear site can be dramatically enhanced by the hyperfine interaction when the conduction electrons are polarized by the applied field in a system with the non-magnetic CEF GS such as a $\Gamma_1$ singlet or a $\Gamma_3$ doublet. At millikelvin temperatures, hyperfine enhanced nuclear spin orders have been observed in several materials with the singlet CEF GS, such as $PrNi_5$, $PrCu_6$, $PrIn_3$ and $PrBe_{13}$ [4]. The hyperfine nuclear spin contribution in the $\Gamma_3$ CEF GS has been also exhibited in $PrAg_2In$ as

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the enhancement of specific-heat below 200 mK [3], and in $PrPb_3$ as the peak of susceptibility at 5 mK [5].

An intermetallic compound $PrMg_3$ with the cubic $Fe_3Al$-type structure, in which $Pr^{3+}$ occupies the cubic site, has been reported as the $\Gamma_3$ GS system with energy splitting 56 K to the first excited state $\Gamma_4$ [6, 7]. As well as $PrAg_2In$, $PrMg_3$ shows the broad peak in the specific-heat at 0.9 K and no evidence for a cooperative phase transition has been detected down to 0.54 K. The enhancement factor $K = 6.1$ is evaluated from the Van-Vleck susceptibility and the hyperfine coupling constant [8]. In order to investigate the strongly correlated electron system with multipole degrees of freedom and the hyperfine nuclear magnetism in the $\Gamma_3$ GS system, we have measured the susceptibility of $PrMg_3$ down to submillikelvin temperatures.

## 2. Experiment

Two single crystal samples of $PrMg_3$ were prepared by the Bridgman method for the dc and ac susceptibility measurements. The dc susceptibility was measured by using a Quantum Design SQUID magnetometer above 2 K. Below 2 K, the ac susceptibility was measured by a mutual inductance method similar to the arrangement used by Herrmannsdörfer *et al.* [9]. A sample for the ac susceptibility measurement had cylindrical shape with 4 mm in diameter and 5 mm in length, and was silver epoxied to the copper cold finger bolted to the copper nuclear demagnetization stage. The primary coil and the static field coil were placed inside a Nb shield, and thermally anchored to the mixing chamber of a dilution refrigerator. The whole assembly was surrounded by a mu metal shield to suppress external stray fields. The modulated excitation field of 16 Hz, was less than 1.5 $\mu$T and parallel to the external static field up to 10 mT.

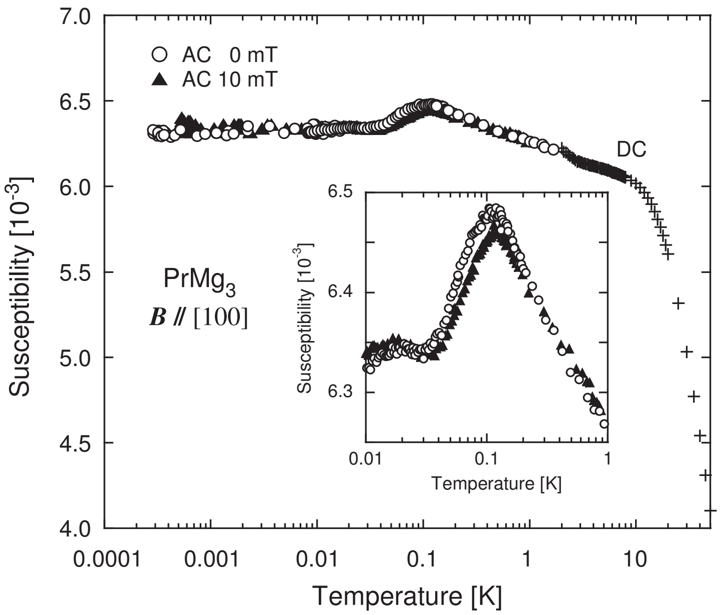


**Figure 1.** Temperature dependence of the dc and real part of ac susceptibility of $PrMg_3$ along the [100] direction on a logarithmic temperature scale. The inset displays behaviors near the peak of the ac susceptibility.

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## 3. Experimental results and discussions

Figure 1 displays the temperature dependence of the dc susceptibility and the real part of ac susceptibility. The dc susceptibility increases monotonically with cooling at high temperatures and starts to saturate around 20 K, indicating the Van-Vleck-like behavior. Both the dc and ac susceptibility indicate $-\log T$ dependence in the temperature range from 10 K to 0.1 K. These results extend the temperature region for the behavior, which has been previously limited to 2 K [8], and are similar to that observed in $PrAg_2In$ and $(Pr_xLa_{1-x})Ag_2In$ [2, 10]. As shown in the inset of Fig. 1, the ac susceptibility shows a peak at 0.1 K and is almost temperature independent below 30 mK. This peak is slightly suppressed by applying the magnetic field 10 mT. Any hysteresis has not been observed between cooling and warming, and between field-cooling and zero-field-cooling around 0.1 K. It is unlikely that this peak is due to a nuclear magnetic order, since the enhancement factor $K = 6.1$ for $PrMg_3$ [8], is much smaller than $K = 30$ for $PrPb_3$ which orders at much lower temperature 5 mK [5]. Therefore, we consider that the peak following $-\log T$ dependence in the susceptibility is due to the quenching of the multipole degrees of freedom in the $\Gamma_3$ GS.

Figure 2 displays a comparison between the observed ac susceptibility and the expected contribution of Pr nuclear spins. The contribution of Pr nuclear spins is calculated by the Curie law including enhancement factors, $K = 0$ (bare nuclear spin) and $K = 6.1$. The observed constant susceptibility down to 0.3 mK cannot be explained even by bare Pr nuclear spins. One of the possible explanation for the disappearance of the nuclear spin contribution is that around 0.1 K the electron system exhibits a crossover to the state which screens the Pr nuclear spin and forms a nuclear spin-singlet. Another is that the nuclear spins are unresponsive to the modulated excitation field 16 Hz owing to the enhancement of the nuclear spin-spin relaxation time below 0.1 K. In either case, the quenching of multipole degrees of freedom is considered to play an important role in the disappearance of the nuclear spin contribution.

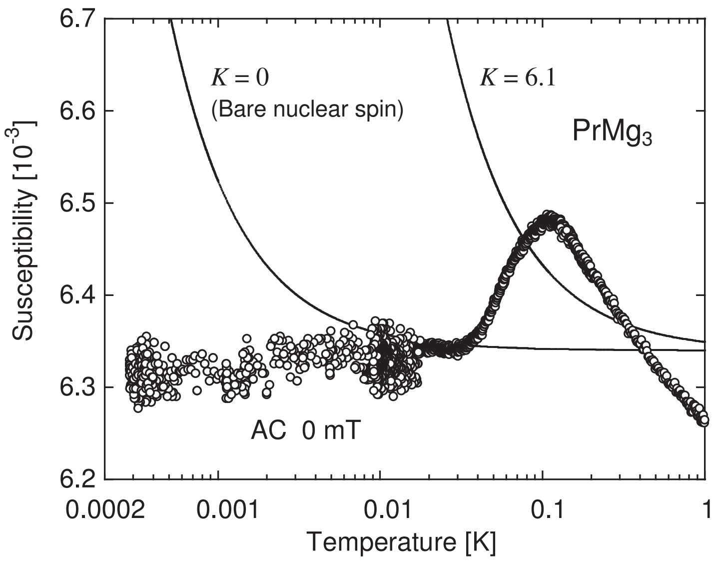


**Figure 2.** Comparison between the observed ac susceptibility and the expected contribution of Pr nuclear spins in $PrMg_3$. The calculation of nuclear contribution is explained in the text. A small signal-to-noise ratio below 10 mK was due to the reduced excitation field to suppress eddy current heating.

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