Electrical Resistivity Anomaly at the Nuclear Magnetic Ordering in a Van Vleck Paramagnet $PrCu_6$

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We have measured the electrical resistivity through the magnetic phase transition of the nuclear spins in the Van Vleck paramagnet $PrCu_6$ with varying frequencies and applied magnetic fields. The electrical resistivity starts to decrease at the transition temperature T_c and becomes constant well below T_c. This resistivity decrease is found to be caused by the ferromagnetic alignment of the nuclear spins. We have also observed a critical phenomenon just above T_c. This critical phenomenon can be explained by a simple calculation.

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In the system consisting of non-Kramers rare-earth ions with a crystal-field singlet ground state, electron spins are paramagnetic (Van Vleck paramagnet) and have no magnetic phase transition. If the hyperfine interaction which polarizes the electron spin is dominant, a nuclear magnetic phase transition occurs through the exchange interaction. This phase transition and the enhanced nuclear cooling of the Van Vleck paramagnet have been studied intensively as a coupled system of electron spins and nuclear spins [1,2]. However, the effect of nuclear ordering on conduction electrons in this system has not been well investigated. The electrical resistivity measurement is considered one of the suitable methods to observe this effect. The conduction electrons are only scattered by the nuclear spin degree of freedom through the *s-f* exchange interaction and the hyperfine interaction, so nuclear spin ordering leads to a decrease of the scattering rate and may affect the electrical resistivity.

In metals, there are several experiments which show resistivity anomalies at electronic magnetic phase transitions. These anomalies are characterized by the temperature derivative of the resistivity in the vicinity of T_c [3,4]. However, in the Van Vleck paramagnet, there are few experiments that have studied the effect of the nuclear ordering on the resistivity. The first observation of the resistivity anomaly in the Van Vleck paramagnet $PrCu_6$ near the nuclear ordering was done by Kiely, Manley, and Weyhmann [5]. They derived the complex susceptibility above T_c by analyzing their data obtained at a finite magnetic field of about 4 mT. The intermetallic compound $PrCu_6$ is a typical enhanced nuclear magnetic system. The nuclear magnetic ordering in $PrCu_6$ occurs at 2.45 mK and the ordered state is ferromagnetic [6].

In the present experiment, we extended the temperature range to well below T_c and measured an ac impedance with the magnetic field varying from 0 to 40 mT at several frequencies. Then, we could discriminate the resistivity in the impedance measurement from the susceptibility, and found that the temperature derivative of the resistivity changed abruptly at T_c. The present experiment gives clear evidence of the resistivity decrease caused by the ferromagnetic alignment of the nuclear spins.

A single crystal of $PrCu_6$ was grown by the Czochralski pulling method using a tungsten crucible in a helium atmosphere where helium gas was purified by a liquid-nitrogen trap [7]; it was cut into a rectangular shape of $1\times3\times23$ mm. $PrCu_6$ crystallizes in the orthorhombic $CeCu_6$ type of structure. The residual resistivity ratio (RRR) of our sample was 70, which was much higher than the value of 15 obtained in the previous experiment [5]. This indicates the high quality of the present sample. We also measured the susceptibilities above 1 K for three symmetrical axes to check the crystallization. The temperature dependence of the susceptibility agreed well with that obtained in a previous experiment [8].

One end of the sample was polished and was attached to a sample holder which was connected to a thermal link from a copper nuclear demagnetization stage. The thermal contact was made by gold-plated copper screws. A good thermal contact is very important to ensure the accurate temperature measurement of the sample, because the electrical resistivity measurement is followed by an unavoidable self-heating in the sample. In this experiment, this heat dissipation was less than a few picowatts, depending on the current amplitude. Therefore, the estimated temperature difference between the sample and the thermometer attached to the thermal link was less than 0.01 mK at 1 mK [9].

We used a SQUID system to measure the resistance with a conventional four-terminal method that is similar to the previous experiment [5]. The ground of the SQUID system was isolated from the cryostat to avoid a ground loop, since one end of the sample was attached to the cryostat to get a good thermal contact as described above. The expected resistance change at the nuclear magnetic transition was so small that an alternating current (ac) method was used to obtain a high resolution. Therefore, it was not the resistance but the impedance that we measured. In this impedance measurement, we had to take into account a frequency-dependent part which came from the self-inductance of the sample. The calculated value of the self-inductance was 0.6 nH. The frequency range in this measurement was from 100 to 1000 rad/s.

To measure the magnetic-field dependence of the resistance, the sample was mounted in a niobium cylinder which could trap a static magnetic field. Since the cylinder was thermally connected to the nuclear stage with a low-thermal-conductivity material, we could change the trapped field without warming the mixing-chamber temperature of the dilution refrigerator up to 1 K. The trapped fields were measured with a calibrated Hall sensor [10].

A platinum NMR thermometer was used to measure the temperature of the thermal link attached to the copper nuclear stage. Calibration of the thermometer was made against the superfluid transition temperature of liquid ^{3}He [11]. We cooled the sample by a small temperature step, and maintained the temperature until thermal equilibrium was established. Near T_c, it took more than several hours to get the thermal equilibrium. After the thermal equilibrium was established, we measured the real part (resistance) and the imaginary part (reactance) of the impedance, varying the frequency at several applied magnetic fields. We also took the entire trace of the impedance during cooldown and warmup.

Figure 1 shows the resistance and the reactance near the transition temperature T_c (2.15 mK) as a function of temperature at zero field (<0.03 mT). The points were obtained in thermal equilibrium, and the lines were obtained during warmup and cooldown. The zeros were taken to be the high-temperature (8 mK) limit. The small deviation of the points from the line was caused by the time drift of our bridge system. The data show that both resistance and reactance are quite constant from 100 mK down to 5 mK and the resistance was 19 $\mu\Omega$. (A broad dip of the resistance was observed around 40 mK when we used a sample of RRR=22 before. It could be the effect of magnetic impurity.) Near T_c, large peaks are observed in both resistance and reactance as seen in Fig. 1. Both resistance and reactance peak heights decrease as the frequency decreases, and the peak seems to disappear at the zero-frequency limit. This strong frequency-dependent peak is attributed to the susceptibility as discussed below. On the other hand, the resistance at any frequency starts to decrease near T_c and becomes nearly constant well below T_c. These resistance decreases are about 20 nΩ, which corresponds to 7 pΩ m of resistivity. Since in general the resistivity is independent of the frequency, this resistance decrease is considered a resistivity decrease. The ordered state below T_c is ferromagnetic [6]. This experimental result is consistent with the theoretical expectation that the ferromagnetic alignment of the nuclear spins causes the resistivity decrease. A recent mean-field calculation shows the two

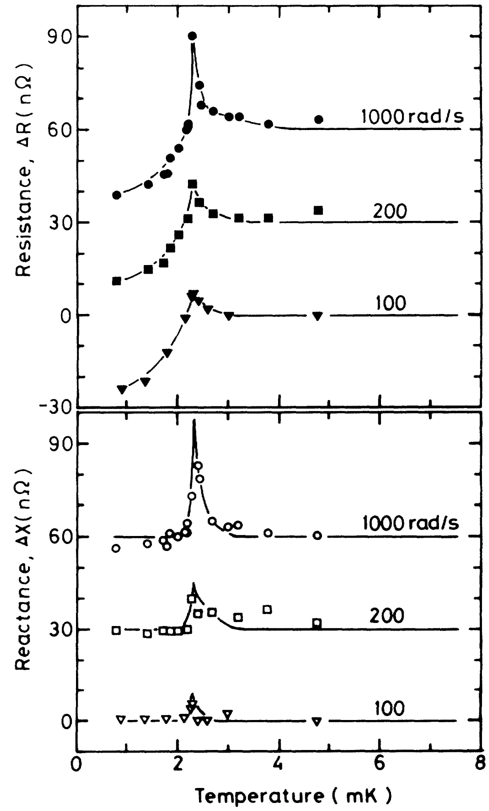

FIG. 1. Resistance ΔR and reactance ΔX at different frequencies in zero magnetic field as a function of temperature. The zeros were taken to be the high-temperature (8 mK) limit. In this picture, the zeros for the data at 200 rad/s and at 1000 rad/s have been shifted upwards by 30 and 60 nΩ, respectively. The points were obtained in thermal equilibrium, and the solid lines were taken during warmup or cooldown. The total resistance of the sample at 8 mK is about 19 $\mu\Omega$ and the reactance is about 0.4 $\mu\Omega$ at the frequency of 1000 rad/s.

plateaus of resistivity well above and below the transition temperature T_c of the nuclear spins, and the resistivity decrease at T_c [12]. Here, we can define the transition temperature T_c as the onset temperature at which the resistance starts to decrease at the zero-frequency limit. The obtained transition temperature is 2.15 mK. This value is consistent with the heat-capacity measurement, if we adopt the new temperature scale [13].

The magnetic-field dependence of the resistance at the frequency of 1000 rad/s is shown in Fig. 2. The peak height in the resistance decreases with increasing magnetic field. At 4 mT, the peak disappears, but the onset temperature of the resistance decrease is still the same as T_c. Above 4 mT, the transition temperature becomes broad and goes up. From the result that the resistance decreases at different fields are nearly the same, 20 nΩ, we can conclude that high magnetic fields induce a ferromagnetic alignment of the nuclear spins and that this alignment causes the resistance decrease.

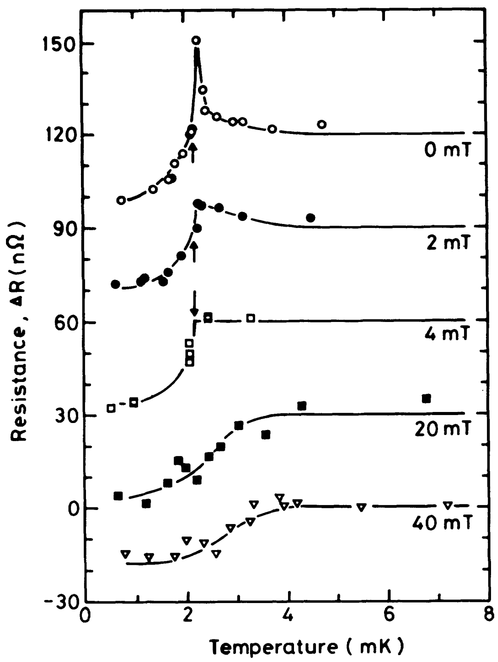

FIG. 2. Resistance at the frequency of 1000 rad/s in various magnetic fields as a function of temperature. The points and solid lines were obtained with the same procedure as that in Fig. 1, and the zeros for the upper curves have been shifted upwards by 30 and 60 nΩ. The arrows show the transition temperature T_c which we defined. In high fields, the transition becomes broad.

Next, we discuss the data above T_c in Fig. 1. The impedance Z obtained in this ac measurement can be described as follows:

$$Z=\frac{l}{S}\rho+i\omega(L+M)$$
$$=\frac{l}{S}\rho+L_0\epsilon\omega(\chi''+i\chi')+i\omega(L_0+M)\,. \qquad (1)$$

The first term in Eq. (1) is the dc resistance, where ρ, l, and S are the resistivity, the length between the voltage terminals, and the cross-sectional area of the sample, respectively. The second term is related to the nuclear complex susceptibility $\chi=\chi'-i\chi''$ and the self-inductance $L=L_0(1+\epsilon\chi)$, where L_0 is the empty inductance, ϵ is a geometric factor, and ω is the angular frequency of the alternating current. The last term is related to L_0 and a mutual inductance M between the voltage and the current leads. Note that the first two terms have temperature dependence.

We assume a single spin-relaxation time τ. Then the susceptibility can be expressed by a Debye form as follows:

$$\chi'=\chi_0\frac{1}{1+\omega^2\tau^2},\quad \chi''=\chi_0\frac{\omega\tau}{1+\omega^2\tau^2}\,, \qquad (2)$$

where χ_0 is the static susceptibility. Second, in general, the spin-relaxation time and the static susceptibility are expected to diverge at the critical point and these can be expressed as $\tau=at^{-\Delta}$ and $\chi_0=bt^{-\gamma}$, where $t=(T-T_c)/T_c$, and a and b are constant values [4]. Substituting Eqs. (2) in Eq. (1) and calculating differentials of the resistance and reactance, we obtained the reduced temperature at which each resistance and reactance has a maximum value. The condition that both resistance and reactance have maximum values at $t>0$ leads to $\Delta/\gamma>1$. Comparing the ratio of these maximum values with that of the peak heights of the experimental data at higher frequencies, we obtained $\Delta/\gamma=4\pm1$, independent of a and b. We used a and b as fitting parameters for the absolute values of the experimental data.

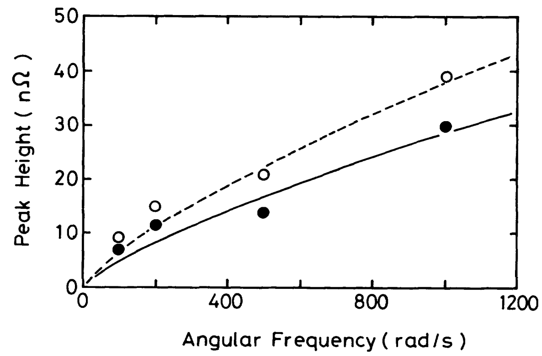

FIG. 3. The peak heights of resistance (solid circles) and reactance (open circles) just above T_c vs frequency at zero magnetic field. The solid and broken lines are calculated values with the critical index ratio $\Delta/\gamma=4$.

The frequency dependence of the peak heights is shown in Fig. 3. The calculated curves agree relatively well with the experimental data of both resistance and reactance, considering the relatively larger uncertainty of the data at lower frequencies. If we choose 2 for the ratio, both heights become the same. The obtained ratio $\Delta/\gamma=4$ indicates that the relaxation time diverges more rapidly than the susceptibility near T_c in the ferromagnetic transition. According to the theory of critical phenomena [14], the ratio $\Delta/\gamma>1$ is expected but our value of the critical index ratio seems considerably large. Our calculation also predicts that the maximum of the reactance always appears at higher temperature than that of the resistance. This feature is clearly seen at higher frequencies. The precise data near T_c and the calculated curves are shown in Fig. 4. The calculated curves of the resistance and the reactance reasonably reproduce the experimental data except for the behavior of the reactance at higher temperatures.

In this experiment, both alternating currents and external magnetic fields are applied along the same direction close to the c axis of the orthorhombic structure. As the crystal axes of $PrCu_6$ are not symmetric, the threshold field to depress the spontaneous ordering may change if

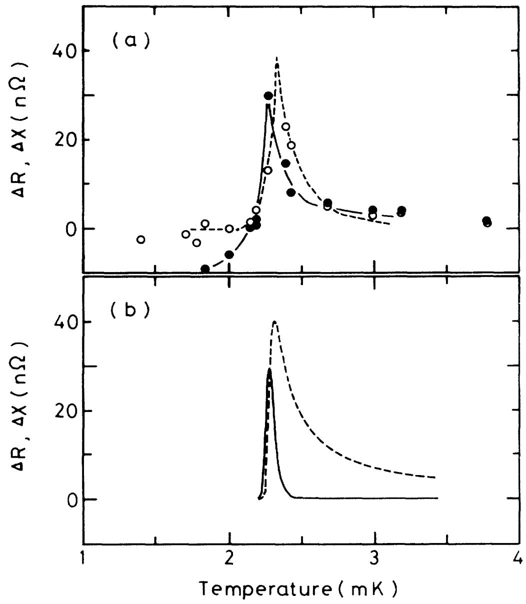

FIG. 4. (a) The experimental data at zero field with the frequency of 1000 rad/s. Solid circles and solid line are resistance ΔR, and open circles and broken line are reactance ΔX. (b) The calculated values of resistance (solid line) and reactance (broken line) vs temperature above T_c. We can see that the peak of the reactance appears at a higher temperature than that of the resistance. At low frequencies, the peaks become closer together.

the magnetic field is applied in a different direction; for example, along the b axis we have observed the largest Van Vleck susceptibility. For a more detailed discussion of the resistivity anomaly below T_c, simultaneous measurement of the resistance and the susceptibility is necessary. The theory expects that the resistivity is proportional to $1-(M/M_{\text{sat}})^2$, where M is the magnetization and M_{sat} is the saturated magnetization [4].

In summary, the present experiment shows the first clear evidence of the resistivity decrease by the polarization of the nuclear spins. We also show that the spontaneous ordering in $PrCu_6$ may be depressed by a small magnetic field. Moreover, the whole behavior of the impedance above T_c can be reproduced by our simple calculation.

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