NMR Study of Layered Transition Metal Ditelluride (Ir,Pt)Te$_2$

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Abstract. We report the results of $^{125}$Te and $^{195}$Pt NMR measurements on (Ir,Pt)Te$_2$ in order to elucidate the characteristic electronic states. For PtTe$_2$, the NMR spectrum exhibits a sharp line, which shows the uniaxially symmetric powder pattern due to the anisotropic Knight shift. The Knight shift is almost independent of temperature, and is larger than that for IrTe$_2$. Also, the nuclear spin-lattice relaxation rate $1/T_1$ of PtTe$_2$ is proportional to the temperature in a wide temperature range, that is, obeys the Korringa relation as expected for simple metallic systems. From the analyses of the Knight shift and $1/T_1$, it is suggested that the antiferromagnetic correlations slightly exist.

1. Introduction

The layered transition metal dichalcogenides have attracted considerable attention because of the various physical properties, which may be reflected by the low-dimensionality. In general, these compounds show anomalies in the electronic, magnetic and thermal properties.

$M$Te$_2$ ($M=\text{Ir}, \text{Pt}$) has the layered CdI$_2$ structure with the trigonal symmetry in high temperature region (see Fig.1(a)), where $M$ ions form an equilateral triangular lattice so that three $M-M$ bonds are equivalent. IrTe$_2$ is a paramagnetic metal. But, the anomalous behavior with a hysteresis in temperature has been found around 270 K in electrical and magnetic properties, indicative of the first-order transition[1]. The electrical resistivity exhibits a hump-shaped maximum, and the magnetic susceptibility reveals a step-like anomaly. It suggests the reduction of the density of states at the Fermi surface $N(E_F)$. The NMR measurements suggest that a simple metallic state realizes in both high- and low- temperature region and $N(E_F)$ is reduced below the transition temperature[2]. Also, by the X-ray diffraction experiments[1], it is clarified that the crystal structure deforms into the monoclinic symmetry gradually with temperature (see Fig.1(b)), where an Ir-Ir bond along $b$-axis shrinks and Ir ions form an isosceles triangular lattice, called bond ordering[3]. This structural transformation may cause the anomalous physical properties in electrical and magnetic measurements. In contrast, PtTe$_2$ in the trigonal symmetric structure shows metallic behavior and does not exhibit any anomaly down to 4.2 K.

Quite recently, it has been reported that the superconductivity exhibits in Ir$_{1-x}$Pt$_x$Te$_2$ with the maximum of $T_C = 3.1$ K for $x = 0.04[3]$. Thus, further studies are necessary to clarify the microscopic electronic states related with the superconductivity.
In this paper, we present the results of $^{125}\text{Te}$ and $^{195}\text{Pt}$ NMR measurements on polycrystalline samples of PtTe$_2$ and IrTe$_2$ in order to investigate the microscopic electronic states, related with the anomalous behaviors in electrical and magnetic properties of IrTe$_2$ and the superconductivity in Ir$_{1-x}$Pt$_x$Te$_2$. It is quite helpful to make use of the NMR method for a study of local electronic properties from the microscopic point of view.

2. Experimental

The polycrystalline specimens used were prepared with a solid state reaction method, as described elsewhere[1]. Powder X-ray diffraction patterns at room temperature confirm that the sample was confirmed to be single phase. The NMR measurements were performed at temperatures between 1.4 K and 300 K in an external magnetic field of 1.2 T using a standard phase-coherent-type pulsed spectrometer. The NMR spectra were obtained by Fast-Fourier-Transformation (FFT) of the spin-echo signals. The nuclear spin-lattice relaxation time $T_1$ was measured by the saturation recovery method, where the spin-echo signals are measured after the application of a comb of rf pulses and waiting a variable delay.

3. Results and Discussion

In PtTe$_2$, there are two NMR active nuclei, $^{125}\text{Te}$ ($I=1/2$, $^{125}\gamma_N/2\pi = 13.454 \text{ MHz/T}$) and $^{195}\text{Pt}$ ($I=1/2$, $^{195}\gamma_N/2\pi = 9.1525 \text{ MHz/T}$), where $I$ and $\gamma_N$ are the nuclear spin and gyromagnetic ratio, respectively.

Figures 2(a) and 2(b) show the $^{125}\text{Te}$ and $^{195}\text{Pt}$ NMR spectra for PtTe$_2$ at $T = 4.2$ K, which were obtained as Fourier transforms of the spin-echo signals at $\mu_0H = 1.1814$ T. Each spectrum exhibits a sharp line, where the line shape shows the uniaxially symmetric powder pattern due to the anisotropic Knight shift.
Figure 3. Temperature dependence of the isotropic Knight shift component of $^{125}\text{Te}$ for PtTe$_2$ and IrTe$_2$. In IrTe$_2$, the spectrum undergoes an appreciable broadening and has shoulder peaks below transition[2].

Figure 4. Temperature dependence of the nuclear spin-lattice relaxation rate $1/T_1$. It is in contrast to the results for IrTe$_2$, where the spectrum exhibits a very sharp peak and a full-width at the half maximum is about $\sim 2$ kHz. It indicates that the anisotropy of the Knight shift component is quite small for IrTe$_2$. Also, the spectrum undergoes an appreciable broadening and splits into three lines below transition temperature[2].

Figure 3 shows the temperature dependencies of the isotropic Knight shift components $K_{\text{iso}} = (K_{\parallel} + 2K_{\perp})/3$ of $^{125}\text{Te}$ for PtTe$_2$, together with the results for IrTe$_2$. The components perpendicular $K_{\perp}$ and parallel $K_{\parallel}$ to the principal axis are derived from the peak maximum and the shoulder peak determined by the derivative of the spectrum, respectively. For PtTe$_2$, the Knight shift is nearly constant in a wide temperature range, which seems to be intrinsic and is different from the Curie-like upturn in $\chi(T)$ possibly originating from the paramagnetic impurities[1]. Also, the value of $K_{\text{iso}}$ is larger than that for IrTe$_2$, indicating that $N(E_F)$ is larger than that for IrTe$_2$.

On the other hand, the $^{195}\text{Pt}$ Knight shift $^{195}\chi(T)$ is negative ($\sim -0.51\%$), thus the sign is opposite to the value of $^{125}\chi(T)$. It indicates that the hyperfine field on $^{195}\text{Pt}$ nuclei is negative, which originates from the core polarization due to the charge transfer from $p$-electron on Te to $d$-electron on Pt.

Figure 4 shows the temperature dependencies of the nuclear spin-lattice relaxation rates $1/T_1$ of $^{125}\text{Te}$ and $^{195}\text{Pt}$ for PtTe$_2$, together with the results of $^{125}\text{Te}$ for IrTe$_2$[2]. For PtTe$_2$, $1/T_1$'s are practically linear in temperature, $1/T_1 = 23.8$ (sK)$^{-1}$ for $^{125}\text{Te}$ and $1/T_1 = 1.51$ (sK)$^{-1}$ for $^{195}\text{Pt}$. For IrTe$_2$, $1/T_1$ is proportional to the temperature in both high- and low-temperature regions. In the low temperature phase where the spectrum has shoulder peaks excited by the rf-field[2], $T_1$ measured at the central peak has a single component. Thus $T_1$ seems to be almost the same for shoulder peaks. The value of $1/T_1$ is reduced discontinuously by a factor $\sim 3$ between high- and low-temperature regions, which reflects the reduction of $N(E_F)$[2].

Generally, $1/T_1T$ is expressed as[4]

$$
\frac{1}{T_1T} = \left(\frac{\gamma_e^2 k_B}{(\gamma_e \hbar)^2}\right) \lim_{\omega \to 0} \sum_q |A_q|^2 \frac{\chi''(q, \omega)}{\omega}
$$
where $\gamma_e$ and $\chi''(q, \omega)$ are the electronic gyromagnetic ratio and the imaginary part of the dynamical susceptibility, respectively. When both a spin part of the Knight shift $K_s$ and $1/T_1$ are due to the hyperfine field from $s$ electrons for non-interacting electron systems such as simple metals, the Korringa relation is known to be satisfied as $T_1 T K_s^2 = (\gamma_e/\gamma_N)^2 (\hbar/4\pi k_B) \equiv S_0$. In the presence of spin correlations, the ratio $R \equiv S_0/(T_1 T K_s^2)_{\text{exp}}$ and the Korringa value of non-interacting electron system $S_0$ provides important information about magnetic correlations, where the value of $R$ smaller (larger) than unity is a signature for the presence of ferromagnetic (antiferromagnetic) correlations. Since $K_s$ and $1/T_1 T$ probes $\chi_s = \chi(0, 0)$ and $\sum_q \chi(q, \omega \to 0)$, $1/T_1 T$ is enhanced by either ferromagnetic or antiferromagnetic correlations, while $K_s$ is enhanced by only ferromagnetic correlations.

In order to estimate $K_s$, we have to subtract the orbital part of the Knight shift $K_{\text{orb}}$ from the observed Knight shift. Assuming that the observed Knight shift is dominated by the spin contribution $K_s$ ($K \sim K_s$), $R$ is estimated to be constant with $\sim 1.4$ for $^{125}$Te in PtTe$_2$ and $\sim 1.9$ for $^{125}$Te in IrTe$_2$ at high temperatures. Also, if the finite $K_{\text{orb}}$ value, which is positive, is considered, $K_{\text{orb}}$ becomes larger, the value of the ratio $R$ is larger. It indicates the antiferromagnetic correlations with finite $q$ vectors far from $q = 0$ slightly exist in PtTe$_2$ and IrTe$_2$.

4. Summary
We have carried out the NMR measurements for PtTe$_2$ and IrTe$_2$ in order to clarify the microscopic electronic states. For PtTe$_2$, the Knight shift is almost independent of temperature, and is larger than that for IrTe$_2$. Also, $1/T_1$ obeys the Korringa relation in a wide temperature range, as expected for simple metallic systems. From the analyses of the Knight shift and $1/T_1$, it is suggested that the antiferromagnetic correlations slightly exist in PtTe$_2$ and IrTe$_2$.

Acknowledgements
This work was partly supported by a Grants-in-Aid for Scientific Research from the Japan Society for the Promotion of Science.

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