Magnetic excitations in the high-temperature phase of $\alpha'$-Na$_2$V$_5$O$_5$

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Abstract

We measured the $^{51}$V nuclear spin relaxation rate in the high-temperature phase of the quarter-filled ladder compound $\alpha'$-Na$_2$V$_5$O$_5$. We compare the results with theories for the spin-$\frac{1}{2}$ Heisenberg chain and find no serious discrepancies between them.

Key words: $\alpha'$-Na$_2$V$_5$O$_5$; NMR; charge ordering; spin excitation

$\alpha'$-Na$_2$V$_5$O$_5$ is known to show a charge ordering at $T_C \sim 34$ K which involves valence change of V ions as $2V^{4.5+} \rightarrow V^{4+} + V^{5+}$ [1]. This charge ordering is an insulator-insulator transition [2]; the insulating behavior in the high-temperature phase was explained by the anisotropic electronic hopping amplitude ($t_{\perp} \gg t_{\parallel}$) in a quarter-filled ladder [3,4]. In the anisotropic limit, one electron occupies a V-O-V molecular orbital on a rung, and the system becomes equivalent to one-dimensional chains. Indeed, the magnetic susceptibility has temperature dependence similar to that of the spin-$\frac{1}{2}$ Heisenberg chain with $J \sim 560$K [5].

In $\alpha'$-Na$_2$V$_5$O$_5$, the anisotropy of the hopping amplitude is not very large, and the inter-site Coulomb repulsion, which is responsible for the charge ordering, is important for the optical properties even in the high-temperature phase [6,7]. Thus the charge fluctuations on the rungs are not small, and it is an open question why the magnetic properties is similar to the spin-$\frac{1}{2}$ Heisenberg chain. In this paper, we report the $^{51}$V nuclear spin relaxation rate ($1/T_1$) in the high-temperature phase. We compare the experimental result with the theoretical calculation for the spin-$\frac{1}{2}$ Heisenberg chain, and discuss the difference in the magnetic excitations between $\alpha'$-Na$_2$V$_5$O$_5$ and the spin-$\frac{1}{2}$ Heisenberg chain.

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Fig. 1. Temperature dependence of the anisotropy of $1/T_1$ for $B = 36, 78, 111$ kG along $a$ axis.

The relaxation rate was measured with a magnetically-aligned powder sample for $B||c$ and a single-crystalline sample for the magnetic field $B||a$. We show the temperature dependence of $1/T_1$ for $B||c$ and the anisotropy of $1/T_1$ in Fig. 1 and 2, respectively.

As shown in Fig. 2, the ratio of $1/T_1$ for $B||a$ to that for $B||c$ is almost temperature-independent (~ 5.0). If the off-diagonal elements of the hyperfine coupling tensor $A_{\alpha\beta}$ and the supertransferred hyperfine coupling from the nearest neighbor V sites are negligible, this ratio is given by
The normalized dimensionless relaxation rate as the experimental results with the theories, we define

\[
\left( \frac{1}{T_1} \right)_{a} / \left( \frac{1}{T_1} \right)_{c} = \frac{A_b^2 + A_c^2}{A_a^2 + A_b^2}. \tag{1}
\]

The experimental result of the hyperfine coupling, \(A_a = -4.4 \times 10^{-19}\), \(A_b = -2.9 \times 10^{-19}\), and \(A_c = -14.8 \times 10^{-19}\) erg [1,8], gives the ratio 8.2. This disagreement may indicate that the assumption on the hyperfine coupling is wrong. Since \(A_c\) is much larger than \(A_a\) and \(A_b\), and is dominated by the diagonal contact interaction, it is reasonable to assume that \((1/T_1)_{a} \propto (A_b^2 + A_c^2)\) in the comparison with the theories.

The dynamics of the spin-1/2 Heisenberg chain has been well understood recently with field theory and numerical calculation. At high temperatures, the dynamical structure factor grows sharply as \(k \rightarrow 0\) and \(\omega \rightarrow 0\). At low temperatures, on the other hand, spectral weight around \(k \sim \pi\) is dominant [9]. To compare the experimental results with the theories, we define the normalized dimensionless relaxation rate as

\[
\left( \frac{1}{T_1} \right)_{\text{norm}} = \frac{2hJ}{A_a^2 + A_b^2} \left( \frac{1}{T_1} \right)_{a}\tag{2}
\]

with \(J \sim 560\)K. The \(k \sim \pi\) contribution to \(1/T_1\), which is dominant at low temperatures, is given by

\[
\left( \frac{1}{T_1} \right)_{\text{norm}} \approx 2D \sqrt{\ln \frac{A}{T} + \frac{1}{2} \ln \left( \ln \frac{A}{T} \right)}, \tag{3}
\]

where \(D = (2\pi)^{-3/2}, A = 2\sqrt{2\pi}e^{C+1}J\), and \(C\) is Euler’s constant [10]. This result contains no adjustable parameters.

In Fig. 3, the experimental result is shown with the theoretical one. Below \(T \sim 0.2J\), \(1/T_1\) is almost temperature independent and near the theoretical value. At higher temperatures, \(1/T_1\) increases noticeably, although the theoretical \(1/T_1\) decreases slowly. This discrepancy can be due to the \(k \sim 0\) contribution to \(1/T_1\).

This ferromagnetic mode often has diffusive behavior, which leads to strong magnetic field dependence of \(1/T_1\). However, as shown in Fig. 1, at high temperatures above 150K, where \(1/T_1\) increases remarkably, it depends only weakly on the magnetic field. If the supertransferred coupling is significant, the couplings for the \(k \sim 0\) and \(\sim \pi\) modes become different. Then the anisotropy of \(1/T_1\) should change, when the \(k \sim 0\) mode grows with temperature. But this is not the case. Quantum Monte Carlo calculations of \(1/T_1\) show only weak temperature dependence below \(T \sim 0.5J\) [9]. Only from the present results, it is unclear that the observed dynamics is understood as that of the spin-1/2 Heisenberg chain.

Another possible origin of the discrepancy at high temperatures is the effect of charge fluctuation. Recently, a theoretical study on quarter-filled ladders suggests that strong charge fluctuation modifies the magnetic excitations from those of the Heisenberg chain [11], but it is unknown how it does in detail.

In summary, we measured \(1/T_1\) in high-temperature phase of \(\alpha’-\text{NaV}_2\text{O}_5\) and compared the results with theories for the spin-1/2 Heisenberg chain. We found no serious discrepancy between them, but further experimental and theoretical studies are desirable to obtain clear conclusions.

References