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$Na_2V_3O_7$: An unusual low-dimensional quantum magnet

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Abstract

Results of present and previous measurements of the ^{23}Na NMR response, dc- and ac-magnetic susceptibilities and the specific heat of $Na_2V_3O_7$ at low temperatures suggest that this material is close to a quantum critical point (QCP) at $\mu_0 H = 0$ T. The experimental data can be explained by assuming that below 100 K the localized V magnetic moments ($S = \frac{1}{2}$) form a collection of dimers, with a broad distribution of singlet–triplet gaps. Most of the dimers adopt a singlet ground state with gaps between 0 and 350 K. A small fraction of them forms triplet ground states with gaps between 0 and 15 K. The degeneracy of the triplet ground states is lifted by a phase transition at an unusually low temperature of 0.086 K. Modest magnetic fields effectively quench this low-temperature state and the system is driven away from the QCP as the applied fields are enhanced to above 1 T.

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Keywords: $Na_2V_3O_7$; Quantum critical point; Magnetism

$Na_2V_3O_7$ is an insulating compound, first reported in 1999 by Millet and coworkers, with very unusual structural features [1]. The V ions (with spin $\frac{1}{2}$) are arranged on the walls of nanotube-type structural units with 5 Å diameter. During the last few years, various experimental results that reveal unusual behavior have been obtained for $Na_2V_3O_7$. One of the most interesting features is that upon varying the temperature T from well above to well below 100 K, most of the V moments seem to be gradually quenched [2,3]. Below 20 K, only $\frac{1}{9}$ of the V moments contribute to the magnetic susceptibility χ. At lower temperatures, the ^{23}Na NMR spin–lattice relaxation rate $T_1^{-1}(T)$ reveals a maximum at a field-dependent temperature T_a, which shifts to zero as the applied field H is reduced to zero. This was previously interpreted [2] as evidence for a quantum critical point (QCP) at $\mu_0 H = 0$ T. We present and discuss here a model to explain experimental results of the specific heat C_p, $T_1^{-1}(T)$ and $\chi(T)$, of $Na_2V_3O_7$ at low temperatures.

Results of extensive measurements of the magnetic, i.e., in excess of the lattice contribution, specific heat $C_m(T,H)$ for $Na_2V_3O_7$, shown in Fig. 1, revealed [4] the presence of broad maxima in $C_m(T,H)/T$ at $T_{\max}(H)$, which shift to zero as H decreases to zero. For comparison we also plot, in the same figure, the positions of the maxima in $T_1^{-1}(T)$ at T_a vs. H, using data from Ref. [2]. We note a close agreement between $T_a(H)$ and $T_{\max}(H)$, which seems to indicate some kind of phase transition at $T_{\max}(H)$. Because we also find a broad maximum in $C_m(T)$ at field-dependent temperatures $T_0(H)$, rather far from $T_{\max}(H)$ and with a completely different field dependence, another interpretation seems to be needed, however. A sharp anomaly in $\chi(T)$ at 0.086 K at $\mu_0 H = 0$ T indicates a phase transition at an unusually low temperature and therefore $Na_2V_3O_7$ should still be regarded as being close to a QCP.

Attempts to interpret our $C_m(T,H)$ data lead us to consider them as exhibiting Schottky-type anomalies due

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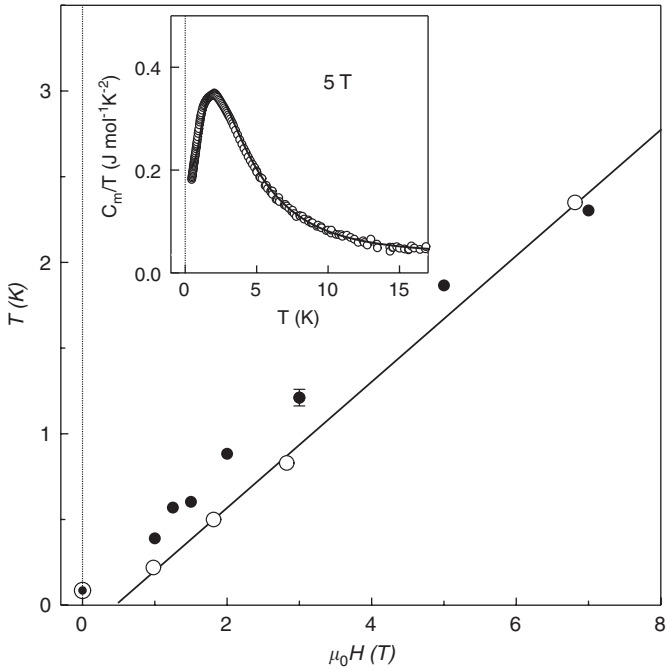

Fig. 1. Position of the maxima in $C_m(T)/T$ at T_{max} (filled circles) and of the maxima in $T_1^{-1}(T)$ at T_a (empty circles) as a function of H. The symbol at the lowest temperature corresponds to a sharp maximum in $\chi(T)$ for $\mu_0 H = 0$ T. The solid line represents the best fit to the $T_a(H)$ data using a linear function. Inset C_m vs. T. The solid line represents the best fit to the data using the model described in the text.

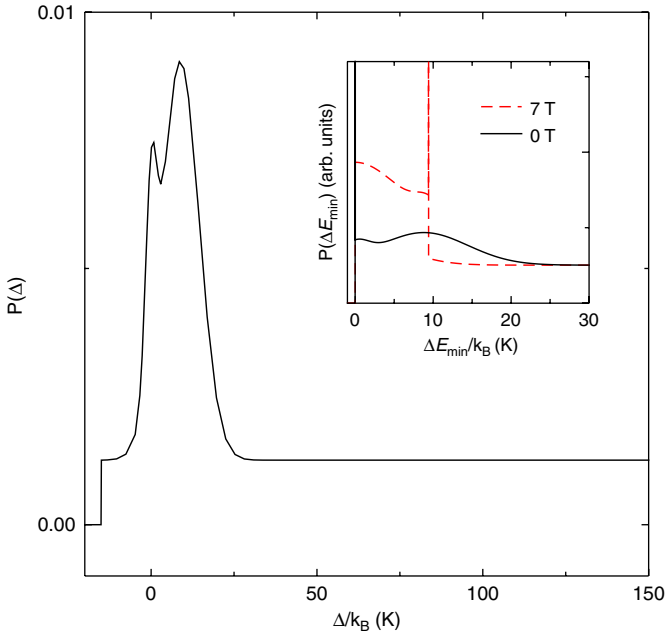

Fig. 2. Distribution $P(\Delta)$ of singlet–triplet gaps Δ. We note that in an applied magnetic field, H, Δ does not represent the excitation energy ΔE_{min} from the ground state. Even if $\mu_0 H = 0$ T, for the case that $\Delta < 0$, $\Delta E_{min} = 0$. Inset: Minimum excitation energy distribution $P(\Delta E_{min})$ for 0 and 7 T.

to an ensemble of singlet–triplet units. The corresponding singlet–triplet gaps Δ exhibit a broad distribution $P(\Delta)$. The low-temperature part of $P(\Delta)$ consistent with our experimental results is shown in Fig. 2. A broad range of gap distribution, from -15 K to up to above room temperature, is quite remarkable for any regular solid. The flat part of $P(\Delta)$ yields a linear-in-T term in $C_m(T, H)$ and the model also predicts a Curie-type behavior for $\chi(T)$, with a change of the Curie constant near 1 K. All these features are experimentally observed [2,4]. The fraction of dimers with negative gaps, i.e., adopting a triplet ground state, are believed to order at 0.086 K, as suggested by the entropy release at low temperatures [4].

To explain the anomaly in $T_1^{-1}(T, H)$ at $T_a(H)$, further assumptions have to be made. For instance, $T_a \propto H$ suggests a "slowing down" of the relevant V moments fluctuations with decreasing T [4]. The equality of the prefactors, however, i.e., $T_a(H) \approx T_{max}(H) = \alpha H$, is rather provocative and may point to the significance of the substantial spectral weight of $P(\Delta E_{min})$ at exactly $2\mu_0 H/k_B$ (see vertical lines at 0 and 9.4 K in the inset of Fig. 2). This weight arises from the dimers adopting a triplet ground state which is split in an external magnetic field. It is these dimers which influence T_{max} in $C_m(T)/T$. If the same dimers also play a central role in $T_1^{-1}(T, H)$, its behavior suggests that their associated moments are actually slowing down with decreasing T, leading to the phase transition at 0.086 K. The rest of the dimers, i.e., the majority, is of less importance for the low-temperature behavior.

We conclude that, well above 100 K, the system may be considered as paramagnetic with the V moments interacting via a very wide range of couplings. This is unusual for a well ordered solid, and points out to a delicate balance between the exchange interactions among the V moments and geometrical frustration imposed by the unusual structural features of $Na_2V_3O_7$. Near 100 K, the V moments undergo a dimerization process leading to a wide range of singlet–triplet gaps. The resulting system is very close to a QCP. The dimers with triplet ground states are involved in a phase transition at 0.086 K.

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