Evidence for a frustrated square lattice with ferromagnetic nearest-neighbor interaction in the new compound Pb$_2$VO(PO$_4$)$_2$

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Abstract

We present the magnetic susceptibility of a new frustrated square lattice system, Pb$_2$VO(PO$_4$)$_2$, and compare it with that measured on our new samples of Li$_2$VOSiO$_4$ and Li$_2$VOGeO$_4$, the only known examples of such magnetic lattice. The analysis of the data using high-temperature series expansion gives a ferromagnetic nearest-neighbor exchange $J_1 \approx 6$ K and an antiferromagnetic next-nearest-neighbor exchange $J_2 \approx 9.8$ K for Pb$_2$VO(PO$_4$)$_2$.

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PACS: 75.50.-y; 75.40.Cx; 75.30.Et

Keywords: $J_1$–$J_2$ model; Vanadium oxide; Low-dimensional magnetic systems; Frustrated lattice; New compound

The frustrated $S = \frac{1}{2}$ square lattice Heisenberg antiferromagnet ($J_1$–$J_2$ model) has attracted strong theoretical attention since an interesting phase diagram [1] with unusual ground states is predicted as a function of the ratio $z = J_2/J_1$ between the nearest neighbor, $J_1$, and next-nearest neighbor (in the diagonal), $J_2$, magnetic interactions ($z$ quantifies the degree of frustration on the system). For $0 \leq z \leq 0.34$ the ground state is the normal Néel order while for $z > 0.6$ a collinear order should set in. At intermediate values $0.34 \leq z \leq 0.6$, where the effects of the frustration should be stronger, a disordered ground state with spin liquid properties has been predicted [1].

However, there was no experimental verification of these theoretical predictions until Melzi et al. [2] recently suggested two compounds: Li$_2$VOSiO$_4$ (LVSiO) and Li$_2$VOGeO$_4$ (LVGeO) as the first realizations of such magnetic lattice. From magnetic susceptibility ($\chi(T)$) and specific heat ($C_p(T)$) measurements, they deduced a ratio $z \approx 1.1$ with $J_1 + J_2 = 8.2$ K for LVSiO. Analysis of NMR data gave strong evidence that the magnetic order below $T_N = 2.8$ K is of the Collinear type.

Later on, Rosner et al. [3] concluded, from LDA calculations of the electronic structure, that in these compounds $J_2$ should be much larger than $J_1$. Indeed, they developed high-temperature series expansions (HTSE) for the $J_1$–$J_2$ model [4] and used them for fitting the same $\chi(T)$ and $C_p(T)$ data of Melzi et al. [2]. They obtained $J_2 \approx 9$ K and $z \approx 10$ (in LVSiO) putting these systems well into the large $J_2$ regime in contrast to the claim of Melzi et al. Also, Rosner et al. [3] predicted a quite strong interlayer exchange in these materials.

Recently, we found a new low-dimensional compound Pb$_2$VO(PO$_4$)$_2$ [5] (PbVPO) whose structure is closely related to Li$_2$VOSi(Ge)O$_4$. The main differences being a commensurate modulation of the magnetic layers and a much larger separation between them. This should result in an enhancement of the 2D character of the magnetic properties in PbVPO. Here, we present the results of a preliminary investigation of the magnetic susceptibility of PbVPO and compare it with our new $\chi(T)$ data of the...
Li-based compounds. A detailed description of the PbVPO structure and magnetic properties will be presented in a forthcoming paper [6].

In PbVPO, \(\chi(T)\) (Fig. 1) presents a clear maximum at \(T_{\text{w Max}}\) \(=\) 5.34 K, expected for a low-dimensional spin system. At \(T_N\approx 2.7\) K a kink in \(\chi(T)\) points to a transition into an ordered state. This kink is very small in low magnetic field but increases and shifts to higher temperatures with increasing field. In Fig. 1 we also plot the \(\chi(T)\) of LVSiO and LVGeO measured on our samples. Our data for LVSiO do not show the departure from a Curie–Weiss law observed at high temperatures by Melzi et al. This makes a fit of the high temperature data more reliable. We therefore fit our \(\chi(T)\) data for all three compounds using the HTSE developed by Rosner [4]. In Table 1 our results are summarized and compared. Our results for LVSiO and LVGeO agree very well with those of Rosner and the recent work of Misguich et al. [7] confirming that these compounds correspond to the large \(J_2\) limit with a negligible \(J_1\). In contrast, our best fit for the PbVPO data was obtained for \(J_1 \approx -6\) K and \(J_2 \approx 9.8\) K meaning that the nearest exchange \(J_1\) on the square lattice is small and ferromagnetic, while the diagonal exchange \(J_2\) is stronger and antiferromagnetic. A direct evidence for a ferromagnetic \(J_1\) is given by the large ratio \(T_{\text{w Max}}/\theta_{\text{CW}}\) (\(\theta_{\text{CW}}\) is the Curie–Weiss temperature). PbVPO is thus the first experimental example for a ferromagnetic frustrated square lattice. Further, the larger \(T_{\text{w Max}}/T_N\) value observed on PbVPO confirm the more pronounced 2D character on the magnetic behaviour of this compound.

We thank Dr. K. Penc, Dr. B. Schmidt and Dr. P. Thalmeier for fruitful discussions.

References