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Journal of Magnetism and Magnetic Materials 272–276 (2004) 922–923
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Evidence for a frustrated square lattice with ferromagnetic nearest-neighbor interaction in the new compound $Pb_2VO(PO_4)_2$

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Abstract

We present the magnetic susceptibility of a new frustrated square lattice system, $Pb_2VO(PO_4)_2$, and compare it with that measured on our new samples of Li_2VOSiO_4 and Li_2VOGeO_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 $J_1 \simeq -6$ K and an antiferromagnetic next-nearest-neighbor exchange $J_2 \simeq 9.8$ K for $Pb_2VO(PO_4)_2$.

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 $\alpha \equiv J_2/J_1$ between the nearest neighbor, J_1, and next-nearest neighbor (in the diagonal), J_2, magnetic interactions (α quantifies the degree of frustration on the system). For $0 \leqslant \alpha \lesssim 0.34$ the ground state is the normal Néel order while for $\alpha > 0.6$ a collinear order should set in. At intermediate values $0.34 \leqslant \alpha \lesssim 0.6$, 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_2VOSiO_4 (LVSiO) and Li_2VOGeO_4 (LVGeO) as the first realizations of a 2D frustrated square lattice. From magnetic susceptibility ($\chi(T)$) and specific heat ($C_p(T)$) measurements, they deduced a ratio $\alpha \simeq 1.1$ 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 $J_2 \simeq 9$ K and $\alpha \simeq 10$ (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 $Pb_2VO(PO_4)_2$ [5] (PbVPO) whose structure is closely related to $Li_2VOSi(Ge)O_4$. 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 preliminar investigation of the magnetic susceptibility of PbVPO and compare it with our new $\chi(T)$ data of the

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doi:10.1016/j.jmmm.2003.12.002

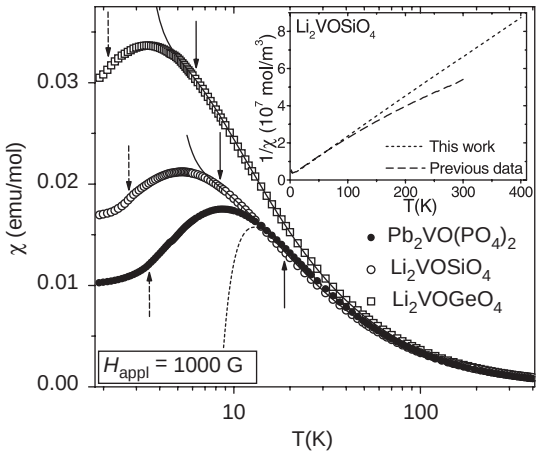

Fig. 1. Comparison of the $\chi(T)$ of $Pb_2VO(PO_4)_2$, Li_2VOSiO_4 and Li_2VOGeO_4. The continuous and dotted lines are the fit with HTSE [4]. Dashed arrows indicate the transition into an ordered state whereas the solid line arrows shows the low temperature limit used in the fit process. Inset: comparison between our $\chi(T)$ data and the data of Melzi et al. [2].

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 $T^{\chi}_{\mathrm{Max}} \simeq 8.7$ K, as expected for a low-dimensional spin system. At $T_{\mathrm{N}} \simeq 3.6$ 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 $J_1 \simeq -6$ K and $J_2 \simeq 9.8$ 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 $T^{\chi}_{\mathrm{Max}}/\theta_{\mathrm{CW}}$ (θ_{CW} is the Curie–Weiss temperature). PbVPO is thus the first experimental example for a ferromagnetic frustrated square lattice. Further, the larger $T^{\chi}_{\mathrm{max}}/T_{\mathrm{N}}$ value observed on PbVPO confirm the more pronounced 2D character on the magnetic behaviour of this compound.

Table 1
Summary of results obtained from the fits of the $\chi(T)$ measurements using the HTSE of Ref. [4]

	Li_2VOSiO_4	Li_2VOGeO_4	$Pb_2VO(PO_4)_2$
J_1 (K)	0.56	0.82	−6
J_2 (K)	6.3	4.1	9.8
J_2/J_1	11	5	1.64
$J_1 + J_2$ (θ_{CW}) (K)	6.9	4.9	3.8
T^{χ}_{max} (K)	5.34	3.5	8.67
T_{N} (K)	2.7	2.1	3.5
$T^{\chi}_{\mathrm{max}}/T_{\mathrm{N}}$	1.98	1.7	2.48

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

References

[1] O.P. Sushkov, et al., Phys. Rev. B 63 (2001) 104420.
[2] R. Melzi, et al., Phys. Rev. B 64 (2001) 024409.
[3] H. Rosner, et al., Phys. Rev. Lett. 88 (2002) 186405.
[4] H. Rosner, et al., Phys. Rev. B 67 (2003) 014416.
[5] E.E. Kaul, et al., Physics of Magnetism 2002, Talk O-1-12, July 1–5, 2002, Poznań, Poland.
[6] E.E. Kaul, et al., to be published.
[7] G. Misguich, et al., cond-mat/0302583, 2003.