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Geometrically Frustrated Kagome Lattice with Chemical Disorder

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The effect of nonmagnetic Mg^{2+} and magnetic Co^{2+} doping in Kagome compounds was investigated using DC magnetic susceptibility. The main effect of doping is to change the crystal field parameters. It was shown that Co^{2+} doping increases considerably crystal field parameters in $\text{Ni}_3\text{V}_2\text{O}_8$ while doping with nonmagnetic Mg^{2+} ions decreases considerably crystal field parameters in $\text{Co}_3\text{V}_2\text{O}_8$ crystals.

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1. Introduction

One of the most studied geometrically frustrated systems is the two-dimensional kagome lattice. Recently, the isostructural compounds $\text{M}_3\text{V}_2\text{O}_8$ ($\text{M} = \text{Ni}, \text{Co}, \text{Cu}, \text{Mg}$) were shown to be new variants of the kagome lattice [1–11]. Among them the most frequently studied of the $\text{M}_3\text{V}_2\text{O}_8$ systems have been $\text{Ni}_3\text{V}_2\text{O}_8$ and $\text{Co}_3\text{V}_2\text{O}_8$. Despite these materials having identical crystal symmetry and similar structural parameters, their magnetic properties are quite different. The difference between the two materials may, in part, be attributed to the magnetocrystalline anisotropy, which is found to be far larger in $\text{Co}_3\text{V}_2\text{O}_8$ than in $\text{Ni}_3\text{V}_2\text{O}_8$ [6]. The magnetic behavior of these materials may be described in terms of two inequivalent magnetic ion sites, known as spine sites and cross-tie sites. The ordering in both materials involves spin component aligned along the a -direction. The magnetic structure always involves ordering of the spine spins. Magnetic susceptibility, neutron diffraction and specific heat measurements revealed that both $\text{Ni}_3\text{V}_2\text{O}_8$ and $\text{Co}_3\text{V}_2\text{O}_8$ undergo a series of magnetic phase transitions versus temperature and magnetic field. Recently, we have studied [12] the phase diagram of cobalt-doped geometrically frustrated $\text{Ni}_3\text{V}_2\text{O}_8$ single crystals. This paper is a continuation of the studies presented in [12] focused on crystal field effects in pure and doped kagome systems.

2. Experimental results and discussion

The $\text{Ni}_{3-3x}\text{Mg}_{3x}\text{V}_2\text{O}_8$ ($x = 0$ and 0.03) and $\text{Co}_{3-3x}\text{Mg}_{3x}\text{V}_2\text{O}_8$ ($x = 0, 0.05,$ and 0.1) single crystals were grown by a method of spontaneous crystallization. The samples were found to be single phase by X-ray diffraction measurements with the crystal structure of orthorhombic space group ($Cmca$). Magnetic measurements were performed in temperature range from 2 K to 300 K using a commercial SQUID magnetometer (MPMS-5, Quantum Design).

Figures 1 and 2 display two examples of the inverse magnetic susceptibility $\chi^{-1}(T)$ at the magnetic field $H = 6$ kOe applied along the a , b , and c -axes.

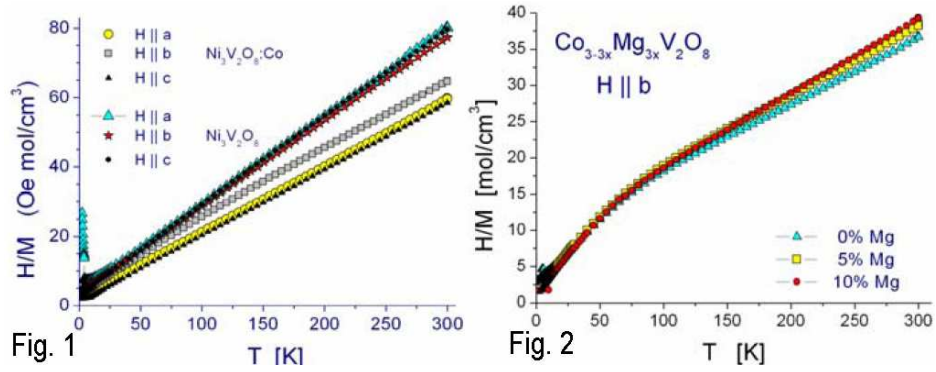


Fig. 1. $\chi^{-1}(T)$ for magnetic field applied along each of the crystallographic directions of $\text{Ni}_{3(1-x)}\text{Co}_{3x}\text{V}_2\text{O}_8$ ($x = 0, 0.03$).

Fig. 2. $\chi^{-1}(T)$ for magnetic field applied along the b -axis of $\text{Co}_{3(1-x)}\text{Mg}_{3x}\text{V}_2\text{O}_8$ ($x = 0, 0.05, 0.1$).

Important information concerning magnetocrystalline anisotropy and crystal field parameters may be obtained from the high temperature susceptibility tensor, whose components are given asymptotically at high temperatures ($T \gg |\theta_i|$) as

$$\chi_{ii} = C_i / (T + \theta_i), \quad (1)$$

where i labels the Cartesian components in orthorhombic coordinate system. For high temperatures, a fit of the measured susceptibilities to Eq. (1) yields the θ_i values gathered in Table. The experimental data will be analyzed using the Hamiltonian of the form

$$\mathcal{H} = \sum_i H_{\text{cr}}^i - \frac{1}{2} \sum_{i \neq j} I_{ij} J_i J_j + g_j \mu_B \mathbf{H} \sum_i J_i, \quad (2)$$

where

$$H_{\text{cr}}^i = \sum_{k,q} B_{kq} \mathbf{O}_k^q, \quad (3)$$

describes the crystal field potential; \mathbf{J} is the angular momentum; I_{ij} is the exchange integral; μ_B is the Bohr magneton, \mathbf{O}_k^q are the Stevens operators.

Following [13], the Hamiltonian \mathcal{H} may be considered in molecular field approximation which results in the following expressions for crystal field and molecular field parameters and their relation with magnetic susceptibility χ_{ii} :

$$\chi_{ii}^{-1} = 3T / [g_j^2 \mu_B^2 J(J+1)(1 + \theta_i/T)] - \lambda, \quad (4)$$

$$\lambda = \text{const}(\theta_x + \theta_y + \theta_z), \quad \xi B_{20} = (1/3)(2\theta_z - \theta_x - \theta_y), \quad (5)$$

$$\xi B_{22} = \theta_x - \theta_y, \quad \xi = (1/5)(2J-1)(2J+3), \quad (6)$$

λ is the molecular field parameter proportional to the exchange integral I_{ij} . The results of the calculations of B_{20} , B_{22} parameters are presented in Table. It results from the Table that:

TABLE
Paramagnetic Curie–Weiss temperatures and crystal field parameters for $\text{Ni}_{3-3x}\text{Mg}_{3x}\text{V}_2\text{O}_8$ ($x = 0$ and 0.03) and $\text{Co}_{3-3x}\text{Mg}_{3x}\text{V}_2\text{O}_8$ ($x = 0, 0.05$, and 0.1).

Materials	NiV_2O_8	CoV_2O_8	NiV_2O_8	CoV_2O_8	CoV_2O_8
	θ [K]	θ [K]	+3%Co θ [K]	+5%Mg θ [K]	+10%Mg θ [K]
$H \parallel a$	15.8	-6.3	11.4	-4.9	-2.7
$H \parallel b$	17.6	114.0	31.1	99.0	86.0
$H \parallel c$	15.2	7.2	7.1	11.9	8.3
$\sum \theta_i$	48.4	114.9	49.6	106.0	91.6
B_0^2	-1.0	-13.0	-9.4	-9.8	-9.4
B_2^2	-1.8	-50.1	-19.7	-43.3	-33.5

— Chemical disorder introduced by low level doping has rather weak effect on exchange interactions in kagome system. This effect is weaker for the Ni doping than for Mg doping.

— Crystal field is considerably weaker in $\text{Ni}_3\text{V}_2\text{O}_8$ than in $\text{Co}_3\text{V}_2\text{O}_8$ crystals. Co doping increases considerably crystal field parameters in $\text{Ni}_3\text{V}_2\text{O}_8$ and induces an easy axis along c -direction in agreement with experimental data presented in [12]. In contrast, doping with nonmagnetic Mg^{2+} ions decreases considerably crystal field parameters in $\text{Co}_3\text{V}_2\text{O}_8$ crystals. From the presented results it is seen that the a -axis is an easy axis for pure and Mg-doped $\text{Co}_3\text{V}_2\text{O}_8$ crystals. The specific behavior of Co^{2+} ions is, without any doubt, related to the Jahn–Teller character of these ions.

Acknowledgments

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