ELSEVIER

Journal of Alloys and Compounds 464 (2008) 23–27

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

The change of some magnetic properties of Cr_2O_3 in the vicinity of T_N for the paramagnetic state

Z. Latacz, M. Baster*

Institute of Physics, Pedagogical University, Podchorazych 2, 30-084 Cracow, Poland

Received 15 July 2007; received in revised form 30 September 2007; accepted 1 October 2007
Available online 22 October 2007

Abstract

The observed anomaly of the parallel magnetic susceptibility of $Cr_2O_3$is discussed as the effect of the changes of the crystal lattice parameters with temperature and the changes of the exchange parameters in some range above T_N connected with it. It is shown that the temperature changes of the crystal parameters have an important influence on the behavior of the magnetic specific heat with temperature too. The results of the calculations based on the correlation theory and the modified Oguchi theory are presented.

Keywords: Chromium oxide; Antiferromagnetism; Magnetic susceptibility; Magnetic specific heat

1. Introduction

The idea of the correlation theory is to calculate both the static and dynamic properties self-consistently, including correlation effects in a mode–mode coupling approximation [1,2]. In other words, short order effects are taken into account by including pair correlations self-consistently in both static and dynamic properties. The calculations for an ideal two-sublattice Heisenberg ferromagnet Gd (which is a non-Bravais crystal, where the lattice consists of two identical hexagonal Bravais lattices) showed [3] that the correlation effects significantly modify the magnetic susceptibility $\chi(T)$ in the whole experimental temperature range of the paramagnetic state, even if the exchange parameters are constants. However, it should be noted that gadolinium is a Heisenberg magnet intermediate between the localized EuO and the itinerant Fe and Ni, and that gadolinium is an ideal Heisenberg ferromagnet governed by the relatively long-ranged Ruderman–Kittel exchange interactions. It should be interesting to perform similar calculations on the basis of the correlation theory, for a localized Heisenberg two-sublattice antiferromagnet Cr_2O_3 (which is a non-Bravais crystal with rhombohedral symmetry), both if the exchange parameters are constants and if these are dependent on temperature. Finally, these results can be compared with the results of similar calculations on the modified Oguchi method [4,5] and also with the available experimental data. In the present paper, the results for two physical quantities which describe the static properties of the system: the magnetic susceptibility and the magnetic specific heat $C^M(T)$ are considered.

2. Correlation Functions for Cr_2O_3

The two-sublattice Hamiltonian can be assumed in the following form [6,7]:

$$H = -\sum_{l\neq m,} J_{ll,} \mathbf{S}_l \cdot \mathbf{S}_{l,} - \sum_{m\neq m,} J_{mm,} \mathbf{S}_m \cdot \mathbf{S}_{m,} - 2\sum_{l\neq m} J_{lm} \mathbf{S}_l \cdot \mathbf{S}_m, \quad (1)$$

where in the first and second summation each interaction is taken twice. $J_{ll,}$ are the interaction constants between spins in the first sublatice, $J_{mm,}$ are the interaction constants between spins in the second sublattice, J_{lm} are the interaction constants between spins in the first and second sublattice. $\mathbf{S}_l, \mathbf{S}_{l,}, \mathbf{S}_m, \mathbf{S}_{m,}$ are the respective spin operators. Agree with (1) the Hamiltonian of the system for Cr_2O_3 in the case of the modifield Oguchi theory for paramagnetic state will be used in the following form:

$$H = -N(J_1\mathbf{S}_0 \cdot \mathbf{S}_1 + 3J_2\mathbf{S}_0 \cdot \mathbf{S}_2) \quad (2)$$

---

* Corresponding author. Fax: +48 12 637 22 43.
E-mail address: sfbaster@cyf-kr.edu.pl (M. Baster).

0925-8388/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2007.10.044

and in the case of the correlation theory within the RPA method [4] in the form:

$$H = -N(J_1\mathbf{S}_0\cdot\mathbf{S}_1 + 3J_2\mathbf{S}_0\cdot\mathbf{S}_2 + 3J_3\mathbf{S}_0\cdot\mathbf{S}_3 + J_5\mathbf{S}_0\cdot\mathbf{S}_5), \tag{3}$$

where N is the number of the magnetic ions, $J_1,\ldots,J_5$ and $\mathbf{S}_0,\ldots,\mathbf{S}_5$ are the interaction constants and the spin operators, respectively as [6]. (Here we restrict to four spin interactions J_1, J_2, J_3 and J_5)

By the concept of the static correlation function, we shall understand an average value of the product of the two spins

$$\langle AB\rangle = \frac{\mathrm{Tr}\rho AB}{Tr\rho}, \tag{4}$$

where A and B are the z-components of the spin operators, Tr is denoted as trace and ρ is the density $\mathrm{e}^{-\beta H}$ where $\beta = 1/(k_BT)$, T is the absolute temperature, k_B is the Boltzmann constant.

In the present paper we shall consider the correlation functions of the type $\langle S_l^z S_m^z\rangle$. The correlation functions for the spins of a pair of the nearest neighbours, connected with the strongest interaction parameter J_1 will be denoted C^{J_1}. The correlation functions for the spins of a pair of the next nearest neighbours with the integration parameters $J_2,\ldots,J_5$ will be denoted C^{J_2}, $\ldots$, C^{J_5}, respectively.

In the case of the correlation theory for two-sublattice it is convenient to introduce the so-called uniform and straggered Fourier transformed variables $\mathbf{U}_q$ and $\mathbf{V}_q$ as [2,3]:

$$U_{\mathbf{q}}^{\alpha} = \frac{1}{\sqrt{2}}(S_A^{\alpha}(\mathbf{q}) + \eta_{\mathbf{q}}^{*}S_B^{\alpha}(\mathbf{q})),$$
$$V_{\mathbf{q}}^{\alpha} = \frac{1}{\sqrt{2}}(S_A^{\alpha}(\mathbf{q}) - \eta_{\mathbf{q}}^{*}S_B^{\alpha}(\mathbf{q})), \tag{5}$$

$$S_A^{\alpha}(\mathbf{q}) = \frac{1}{\sqrt{2}}(U_{\mathbf{q}}^{\alpha} + V_{\mathbf{q}}^{\alpha}), \qquad S_B^{\alpha}(\mathbf{q}) = \frac{1}{\sqrt{2}}\eta_{\mathbf{q}}^{*}(U_{\mathbf{q}}^{\alpha} - V_{\mathbf{q}}^{\alpha}), \tag{6}$$

where $\eta_{\mathrm{q}} = j(\mathrm{q})/\,|j(\mathrm{q})|$, $(\eta_{\mathbf{q}=0} < 0)$, then the intra-sublattice Fourier transformed interaction $J(\mathbf{q})$ is given by the formula:

$$J(\mathbf{q}) = \mathrm{e}^{\mathrm{i}\,\mathbf{q}\nu}[J_1 + J_2(\mathrm{e}^{-\mathrm{i}\,\mathbf{q}\mathbf{a}_1} + \mathrm{e}^{-\mathrm{i}\,\mathbf{q}\mathbf{a}_2} + \mathrm{e}^{-\mathrm{i}\,\mathbf{q}\mathbf{a}_3}) + J_3(\mathrm{e}^{-\mathrm{i}\,\mathbf{q}(\mathbf{a}_1+\mathbf{a}_2)} + \mathrm{e}^{-\mathrm{i}\,\mathbf{q}(\mathbf{a}_1+\mathbf{a}_3)} + \mathrm{e}^{-\mathrm{i}\,\mathbf{q}(\mathbf{a}_2+\mathbf{a}_3)}) + J_5\,\mathrm{e}^{-\mathrm{i}\,\mathbf{q}(\mathbf{a}_1+\mathbf{a}_2+\mathbf{a}_3)}], \tag{7}$$

where $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$ are the primitive lattice vectors for small unit cell, $\nu = 0.210(\mathbf{a}_1 + \mathbf{a}_2 + \mathbf{a}_3)$ [8,9], The correlation functions are defined by $\mathbf{U}_q$ and $\mathbf{V}_q$ operators as:

$$\langle S_l^{\alpha} S_{l,}^{\alpha}\rangle = \frac{2}{N}\sum_{\mathbf{k}}(\langle U_{\mathbf{k}}^{\alpha}U_{-\mathbf{k}}^{\alpha}\rangle + \langle V_{\mathbf{k}}^{\alpha}V_{-\mathbf{k}}^{\alpha}\rangle)\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}(\mathbf{l}-\mathbf{l}_{,})}, \tag{8}$$

$$\langle S_l^{\alpha} S_m^{\alpha}\rangle = \frac{2}{N}\sum_{\mathbf{k}}\eta_{\mathbf{k}}^{*}(\langle U_{\mathbf{k}}^{\alpha}U_{-\mathbf{k}}^{\alpha}\rangle + \langle V_{\mathbf{k}}^{\alpha}V_{-\mathbf{k}}^{\alpha}\rangle)\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}(\mathbf{l}-\mathbf{m})}, \tag{9}$$

where the wave-vector variables $\mathbf{q}$ and $\mathbf{k}$ are general wave-vectors in the Brillouina zone for the reciprocal lattice of the magnetic structure.

3. Magnetic susceptibility of Cr_2O_3 in the correlation theory

As it was written in paper [1], the static equations in the correlation theory for the Hamiltonian of the type (1) and in the paramagnetic state give the following formulae for the static wave-vector-dependent susceptibilities: $\chi_u(\mathbf{q}) = (U_{\mathbf{q}}^z U_{-\mathbf{q}}^z)$- uniform mode, $\chi_v(\mathbf{q}) = (V_{\mathbf{q}}^z V_{-\mathbf{q}}^z)$-staggered mode, where the symbol (AB) is the static relaxation function:

$$(AB) = \int_0^{\beta}\langle \mathrm{e}^{\lambda H}U_{\mathbf{q}}^z\,\mathrm{e}^{-\lambda H}U_{-\mathbf{q}}^z\rangle \mathrm{d}\lambda.$$

(The theory has previously been reviewed in detail in Ref. [2].)

Performing the calculation, similarly as for a number of magnets [2,10], for a two-sublattice antiferromagnet Cr_2O_3we introduce in the above formulae, according to definition (7), $J(\mathbf{q}+\mathbf{k})$ and take the terms depending on $\mathbf{q}$ outside the summation over $\mathbf{k}$. Straightforward calculations show that we can express the above mentioned quantities by eight self-consistent parameters $\alpha_i (i = 1,\ldots,8)$:

$$\alpha_1 = \frac{J_1 2}{N}\sum_{\mathbf{k}}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle - \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle) = 2J_1C^{J_1}, \tag{10}$$

$$\alpha_2 = \frac{J_2 2}{N}\sum_{\mathbf{k}}\mathrm{e}^{-\mathrm{i}\,\mathbf{k}\mathbf{a}_1}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle - \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle) = 2J_2C^{J_2}, \tag{11}$$

$$\alpha_3 = \frac{J_3 2}{N}\sum_{\mathbf{k}}\mathrm{e}^{-\mathrm{i}\,\mathbf{k}(\mathbf{a}_1+\mathbf{a}_2)}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle - \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle) = 2J_3C^{J_3}, \tag{12}$$

$$\alpha_4 = \frac{J_5 2}{N}\sum_{\mathbf{k}}\mathrm{e}^{-\mathrm{i}\,\mathbf{k}(\mathbf{a}_1+\mathbf{a}_2+\mathbf{a}_3)}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle - \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle) = 2J_5C^{J_5}, \tag{13}$$

$$\alpha_5 = \frac{J_1 2}{N}\sum_{\mathbf{k}}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}\frac{|J(\mathbf{k})|}{|J(0)|}\times(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle + \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle), \tag{14}$$

$$\alpha_6 = \frac{J_2 2}{N}\sum_{\mathbf{k}}\mathrm{e}^{-\mathrm{i}\,\mathbf{k}\mathbf{a}_1}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}\frac{|J(\mathbf{k})|}{|J(0)|}\times(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle + \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle), \tag{15}$$

$$\alpha_7 = \frac{J_3 2}{N}\sum_{\mathbf{k}}\mathrm{e}^{-\mathrm{i}\,\mathbf{k}(\mathbf{a}_1+\mathbf{a}_2)}\eta_{\mathbf{k}}^{*}\,\mathrm{e}^{\mathrm{i}\,\mathbf{k}\nu}\frac{|J(\mathbf{k})|}{|J(0)|}\times(\langle U_{\mathbf{k}}^z U_{-\mathbf{k}}^z\rangle + \langle V_{\mathbf{k}}^z V_{-\mathbf{k}}^z\rangle), \tag{16}$$