ELECTRONIC STRUCTURE AND MAGNETISM OF INTERMETALLIC $NdAl_2$ *

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Magnetic and electronic properties of $NdAl_2$ have been found to be well described within the crystal-field approach. A λ-type peak observed at the magnetic ordering temperature in the temperature dependence of the heat capacity is related to the time-reversal symmetry breaking in the atomic scale. The good description proves the existence of the discrete atomic-like states, in the meV energy scale, in this intermetallic compound.

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1. Introduction

The aim of this contribution is to investigate magnetic properties of $NdAl_2$. It crystallizes in the cubic structure $MgCu_2$, is an intermetallic compound and exhibits a ferromagnetic order below T_c=65 K [1,2]. We have attributed the magnetism and low-temperature properties to Nd atoms.

2. Theoretical outline

The Nd^{3+} ion is assumed to have three f electrons in the incomplete $4f$ shell, which form a highly-correlated atomic-like $4f^3$ system. We approximate the strong correlation within the incomplete $4f$ shell by means of the three Hund rules. This enables us to describe the $4f^3$ system by quantum numbers S=3/2 and L=6. The third Hund's rule, realized due to a strong spin–orbit coupling, allows for J to be a good quantum number. In this way, the multiplet $^4I_{9/2}$ becomes the ground multiplet for the $4f^3$ system of Nd^{3+} ion.

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The Nd^{3+} ion is a Kramers f^3 system. Its ground multiplet is 10-fold degenerate. The crystal field of the cubic symmetry removes only partly this degeneracy producing a doublet and two quartets. In $NdAl_2$ the ground state is the doublet. The effect of the charge interactions of the cubic CEF is shown in Fig. 1, in the paramagnetic region, *i.e.* above 65 K. The doublet and the quartets are split in the magnetic state, *i.e.* below T_c.

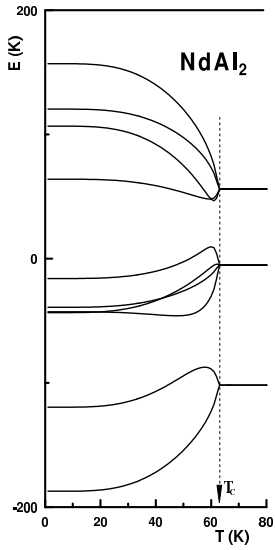

Fig. 1. Calculated temperature dependence of 10 states of the ground multiplet in the paramagnetic and the magnetic states. In a magnetic state the Kramers degeneracy is lifted.

For the description of electronic and magnetic properties we have applied a single-ion-like Hamiltonian [3]:

$$H = H_f + H_{f-f} = \sum\sum B_n^m \hat{O}_n^m + n g_{\mathrm{L}}^2 \mu_{\mathrm{B}}^2 \left(-J\langle J\rangle + \tfrac{1}{2}\langle J\rangle^2\right) + g_{\mathrm{L}}\mu_{\mathrm{B}} J \cdot B_{\mathrm{ext}}\,, \tag{2.1}$$

where g_{L} is the Lande factor (8/11) and the magnetic moment of the Nd ion is expressed as m=-$g_{\mathrm{L}} J \mu_{\mathrm{B}}$. The cubic CEF interactions are described by two terms only, with parameters B_4 and B_6. The second term accounts for the intersite spin-dependent interactions approximated in this paper by the molecular-field approximation with the molecular-field coefficient n. The self-consistent calculations are performed in the same manner as those presented in Ref. [3] for $ErNi_5$. Having obtained the electronic structure, for both the magnetic and paramagnetic states, we can calculate the Helmholtz free energy and the resulting thermodynamical properties by means of the statistical physics.

3. Results and discussion

The calculated temperature dependence of the fine low-energy electronic structure of the Nd^{3+} ion is shown in Fig. 1. The formation of the magnetic state in the atomic scale at T_c=65 K is accompanied by the lifting of the Kramers degeneracy. In Fig. 2 we show the calculated temperature dependence of the ordered magnetic moment and of the heat capacity. For the calculations we have used the cubic CEF parameters: B_4=−11.48 mK and B_6=+0.464 mK [1,2]. The self-consistent calculations, in order to reproduce T_c of 65 K, yield the molecular-field coefficient n = +24.8 T/μ_B. It means that at 0 K the molecular field on the Nd moment has the magnitude of 65 T. In the ordered state there appears a spin gap, that amounts to 6 meV at 0 K. By ordering magnetically the system gains the energy of 90 K/f.u. = 750 J/mol.

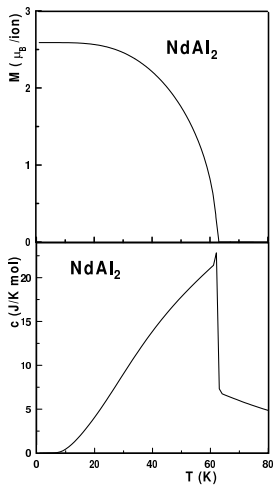

Fig. 2. Magnetic phase transition in $NdAl_2$. (a) Calculated temperature dependence of the magnetic moment of the Nd^{3+} ion; (b) temperature variation of the f-subsystem contribution to the heat capacity.

The λ-type peak observed at the magnetic ordering temperature is related to the time-reversal symmetry breaking in the atomic scale. We also obtain the magnetic moment of 2.62 μ_B, in good agreement with experimental datum of 2.5 μ_B. This moment is composed of the orbital and spin parts of +4.58 μ_B and −1.96μ_B, respectively.

The minimum of the free energy has been found for the magnetic moment aligned along the [100] direction, *i.e.* along the cubic edge, Fig. 3. This result is in agreement with the experimental observations [1,2]. The hard magnetic axis is along the cube diagonal, with the energy difference of 5.5 K. We have

calculated also the magnetocrystalline-anisotropy parameters according to the Ref. [4] using the free energy expressions for the main cubic directions [100], [111] and [110]. We obtain the values K_1^c=+3.95 J/mol and K_2^c=+1.2 kJ/mol.

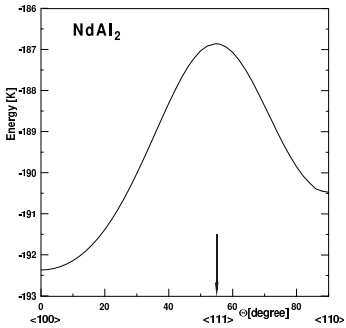

Fig. 3. Angular dependence of the ground-state energy for the Nd^{3+}-ion moment in $NdAl_2$. This dependence is the microscopic origin of the magnetocrystalline anisotropy. The easy axis is along the cube edge [100], whereas [111] is the hard axis.

4. Conclusions

We have presented the microscopic, atomic-scale, description of the magnetic and electronic properties, including the magnetic transition at 65 K, for $NdAl_2$. A good description of the intermetallic compound proves the existence of the discrete atomic-like states, on the meV energy scale, confirming a*posteriori* the basic assumption of our approach the electronic and magnetic properties of this $4f$ compound.

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