ANISOTROPIC MAGNETIC SUSCEPTIBILITY OF NEODYMIUM SUBSTITUTED $SrLaAlO_4$ CRYSTALS*

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The uniaxial anisotropy of magnetic properties of $SrLa_{1-x}Nd_xAlO_4$ single crystals ($x = 0.01$ and 0.05) was found from the measurements of temperature dependencies of magnetic susceptibility. Results of measurements, with magnetic field along a- and c-axis, are compared with the similar data obtained for $CaNdAlO_4$ crystal. The successful description of experimental data was done in frames of the crystal field approximation. The anisotropy of magnetic susceptibility appears due to crystal field acting on magnetic neodymium ions in a system without exchange interactions.

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1. Introduction

Single crystals of neodymium substituted $SrLaAlO_4$ may be of interest as a laser active material. $SrLaAlO_4$ belongs to the group of perovskite-like crystals with K_2NiF_4 structure. The crystal is built up, along the c direction, as a sequence of layers containing AlO_6 octahedra separated by layers consisting of Sr^{2+} and La^{3+} ions more or less randomly distributed in nine-coordinated sites of C_{4v} symmetry. In investigated crystals a part of La ions are substituted by neodymium ions.

The strong uniaxial anisotropy of magnetic susceptibility has been found in $CaNdAlO_4$ crystal with the same structure [1, 2]. This anisotropy is introduced by Nd^{3+} ions. As it was shown by Fink-Finowicki et al. [2], it can appear due to

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anisotropic exchange interactions between neodymium ions or due to low symmetry crystal field acting on the neodymium ions. Therefore, studies of magnetic properties of neodymium substituted crystals with the same crystallographic structure are very useful to understand deeper the nature of magnetic properties in this class of materials. The aim of this paper is to discuss the origin of magnetic anisotropy of Nd ions in perovskite-like crystals with K_2NiF_4 structure.

2. Experimental procedure

Single crystals of $SrLa_{1-x}Nd_xAlO_4$ (with $x = 0.01$ and 0.05) and $CaNdAlO_4$ were grown by Czochralski method [3]. Samples with dimensions of few millimeters were X-ray oriented for magnetic measurements performed with magnetic field applied along various crystallographic directions.

Temperature dependence of magnetization of $SrLa_{0.95}Nd_{0.05}AlO_4$, as well as $CaNdAlO_4$, has been measured using vibrating sample magnetometer (VSM–PAR Model 155) at temperatures 4.2–160 K in fixed external magnetic field of 10 kOe. The accuracy of magnetic moment measurements was better than 10^{-4} emu and the accuracy of temperature control better than 0.2 K. In the case of $SrLa_{0.99}Nd_{0.01}AlO_4$, because of lower contents of magnetic neodymium ions, the magnetization measurements were performed with Quantum Design SQUID magnetometer with resolution of about 10^{-7} emu.

3. Results and discussion

Magnetization measurements of $SrLa_{1-x}Nd_xAlO_4$ in the (a–b) plane indicate that susceptibility is isotropic in this plane, similarly as it was observed for $CaNdAlO_4$ [2]. Therefore, the temperature measurements were done for two external magnetic field orientations: along the a- and c-axis. The results of these measurements show the uniaxial anisotropy of magnetic susceptibility. The similar result was earlier obtained for $CaNdAlO_4$ [2].

Temperature dependencies of magnetic susceptibility χ multiplied by temperature T for $CaNdAlO_4$ are presented in wide temperature range up to 760 K in Fig. 1. The product of χT was used in order to get better presentation of data at low temperatures. Anisotropic susceptibility of paramagnetic ions can be described in the frames of the crystal field theory, as it was done e.g. by Elliott and Stevens [4]. In this formalism magnetic susceptibility is given by

$$\chi = \frac{N\mu_{\mathrm{B}}^2}{4k_{\mathrm{B}}T}\frac{g_0^2+\sum_i g_i^2\exp(-\Delta_i/k_{\mathrm{B}}T)}{1+\sum_i\exp(-\Delta_i/k_{\mathrm{B}}T)}+(\chi_{\mathrm{VV}}+\chi_{\mathrm{dia}}), \tag{1}$$

where g_0 and g_i denote g-factors and Δ_i are crystal field energy levels. In the above formula terms χ_{VV} and χ_{dia} represent temperature independent Van–Vleck and diamagnetic susceptibilities. The values of Δ_i for the lowest levels of $^4I_{9/2}$ ground state of Nd^{3+} ions in $CaNdAlO_4$ are known [5]. One can expect the anisotropy of g_i-factors, because of the low symmetry crystal field acting on Nd ion surrounded by 9 oxygen ions. Fitting the formula (1) to the experimental data, good agreement (see Fig. 1) was obtained for g_i values listed in Table and

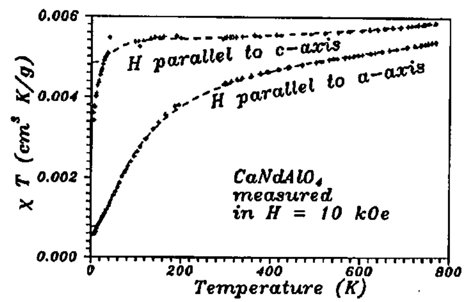

Fig. 1. Temperature dependence of magnetic susceptibility χ multiplied by temperature T for $CaNdAlO_4$; points (+) — experimental data; dashed lines — crystal field approximation.

$(\chi_{VV} + \chi_{dia}) = 10^{-6}$ cm^3/g. The g values for the lowest energy level are close to values of g_0 reported from ESR measurements [6] for $SrLaAlO_4$ doped with 5% of Nd, being equal to 1.55 and 3.67 along a- and c-axis, respectively. The values of g-factors fitted for higher energy levels are estimated with lower accuracy.

TABLE

The fitted values of g_i-factors along a- and c-axis for Δ_i energy levels of $CaNdAlO_4$ crystals [2]

Δ_i/k_B [K]	0	85.5	163	289
g_i || a-axis	1.50	3.93	3.39	5.65
g_i || c-axis	3.77	4.31	4.12	2.90

On the other hand, the observed dependence of susceptibility is similar to the susceptibility of isolated clusters of interacting magnetic ions (see for example paper by Smart [7]). The successful approximation describing the susceptibility along a-axis in the above exchange interaction model requires the assumption of the existence of various neodymium clusters (e.g. pairs and triplets of coupled ions in a–b plane) [2]. It can be realized in $CaNdAlO_4$ lattice with Ca^{2+} and Nd^{3+} ions placed randomly in crystallographically equivalent positions.

Experimental results of susceptibility of $CaNdAlO_4$ crystal can be described in crystal field approximation as well as by anisotropy of exchange interactions, and it is impossible to distinguish which approximation better describes experimental data. However, measurements of isostructural crystals with a part of neodymium ions substituted by diamagnetic ions could verify which model is valid. In frames of crystal field approximation the temperature behavior of susceptibility of non-interacting Nd ions should be similar for various concentrations, while in the anisotropic exchange description this behavior should be strongly dependent on neodymium concentration.

Magnetic susceptibility measured for $SrLa_{1-x}Nd_xAlO_4$ single crystals, besides temperature dependent χ_{Nd} and temperature independent paramagnetic χ_{VV} terms, contains diamagnetic contribution of host crystal χ_{dia}, which becomes significant at high temperature region, especially for samples with low Nd concentration. Diamagnetic term has been estimated as $\chi_{\mathrm{dia}} = -1.7 \times 10^{-7}$ cm^3/g.

Temperature dependent χ_{Nd} susceptibility was calculated by subtracting the temperature independent contribution from the measured susceptibility. Final re-

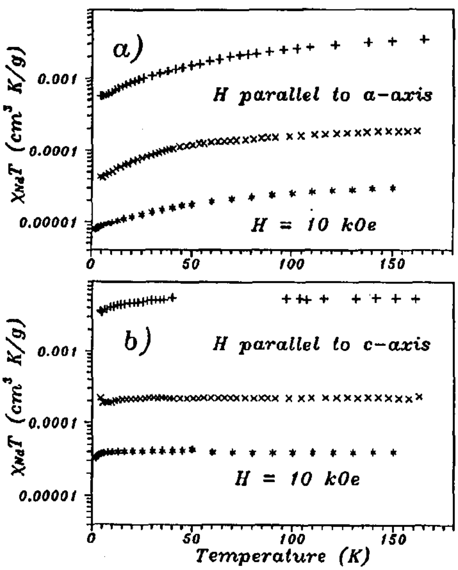

Fig. 2. Temperature dependent Nd^{3+} contribution to the total susceptibility presented in a logarithmic plot of $\chi_{\mathrm{Nd}}T$ vs. T (a) along a-axis and (b) along c-axis. Experimental points: $*****$ 1% Nd, $\times\times\times\times\times$ 5% Nd — substituted into $SrLaAlO_4$, and $+++++$ $CaNdAlO_4$ crystal.

sults of χ_{Nd} along a-axis and along c-axis are presented in Fig. 2a and 2b, respectively, where the values of $\chi_{\mathrm{Nd}}T$ are plotted in logarithmic scale vs. T. The lowest curves corresponding to 1% Nd, and the middle curves corresponding to 5% of Nd concentration in $SrLaAlO_4$ are compared with the highest curves for $CaNdAlO_4$. These plots show qualitatively the same temperature behavior for various Nd contents. Little differences can be due to: (1) experimental errors, especially for the lowest Nd concentration, (2) errors in estimated values of diamagnetic term χ_{dia}, (3) possible differences between energy levels and g-factors for Nd^{3+} ions in $SrLaAlO_4$ and in $CaNdAlO_4$.

It proves that the origin of observed anisotropic magnetic properties is the same, both in $SrLa_{1-x}Nd_xAlO_4$ and $CaNdAlO_4$ crystals. Finally, comparison of results obtained for the samples with different concentration of neodymium confirms that observed anisotropic effect, connected with magnetic behavior of Nd^{3+} ions, is a consequence of anisotropy of neodymium g-factor due to low symmetry crystal field. It indicates that the magnetic anisotropy even in the dense paramagnet

$CaNdAlO_4$ is also due to the same origin.

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