INVESTIGATION OF THE MAGNETIC PROPERTIES OF NEODYMIUM MONOCHALCOGENIDES AT 4.2–150°K

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It is reported that neodymium chalcogenides (NdS, NdSe, and NdTe) exhibit a transition to the antiferromagnetic ordered state. This is in agreement with the Kramers theorem, which predicts that if a partly filled atomic shell contains an odd number of electrons the lower energy level in the crystal field remains degenerate, and that this, at sufficiently low temperatures, may cause a transition to a magnetically ordered state. Confirmation was obtained for neodymium Nd^{3+} $(4f^3)$ and cerium $(4f^1)$ monochalcogenides, which were taken as examples.[9]

THE magnetic and electrical properties of rare-earth metal compounds have been investigated intensively in the last few years. Thus, for example, Busch et al.[1] investigated the magnetic properties of phosphides, arsenides, and antimonides of rare-earth metals from 1.5 to 300° K in strong fields (up to 160 kOe). Holtzberg et al.[2] carried out investigations of the magnetic and electrical properties of the rare-earth compounds $Gd_4(Sb_xBi_{1-x})_3$, $(Eu_{1-x}Gd_x)Se$, and others. An extensive review is given in the monograph by Belov et al.[3]

Rare-earth metals may form compounds with elements in the sixth group of the periodic system (S, Se, Te) in the stoichiometric ratio 1:1. These compounds, known as monochalcogenides, have the rocksalt (NaCl) structure. The magnetic properties of the monocompounds of rare-earth metals with metalloids of the fifth and sixth groups of the periodic system were investigated by Iandelli,[4] but his measurements were carried out over a relatively narrow range of temperatures (90–473° K) and in a constant magnetic field of 8200 Oe. The magnetic properties of monochalcogenides have not yet been investigated at low temperatures. The only exceptions are Eu monochalcogenides,[5] of which EuS and EuSe are ferromagnetic below 16 and 6° K, respectively, while EuTe is antiferromagnetic with a Néel point $T_N = 11°$ K. The present investigation is one of a series dealing with the magnetic properties of rare-earth monochalcogenides. Earlier, we investigated the magnetic properties of the same compounds at temperatures of 100–1300° K.[6]

Compound	a, kX	T_N, °K	Θ, °K	$\mu_{\text{eff}}^{\text{exp}}/\mu_B$
NdS	5.63	8	—24	3.62
NdSe	5.90	14	—9	3.40
NdTe	6,26	13	—14	3.54

The present communication reports the results of an investigation of the magnetic properties of neodymium monochalcogenides in the temperature range 4.2–150° K. Measurements were carried out using a pendulum magnetometer in fields of 6–13 kOe. The investigated compounds were synthesized from elements by the method described in the paper of Zhuze et al.[7] Phase analysis of the samples was carried out by x-ray diffraction and it was found that the samples consisted of practically one phase and had well-established structure of the NaCl type. The results of the magnetic measurements are given in the table [which includes the lattice parameter a, the antiferromagnetic transition temperature T_N, the constant Θ in the Curie–Weiss law $\chi = C/(T + \Theta)$, and the experimentally determined effective number of

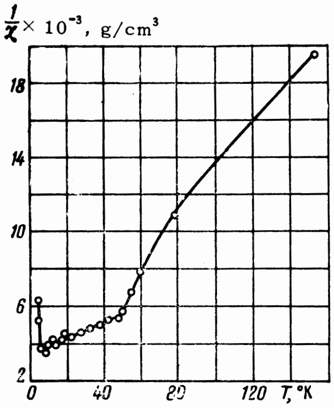

FIG. 1. Temperature dependence of the reciprocal of the specific magnetic susceptibility of NdS.

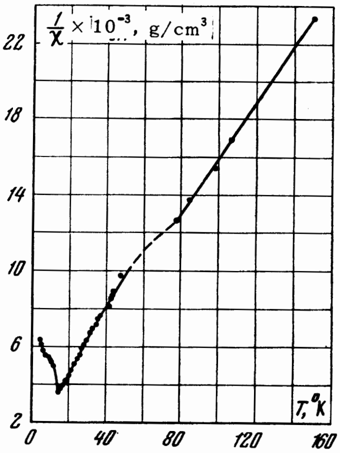

FIG. 2. Temperature dependence of the reciprocal of the specific magnetic susceptibility of NdSe.

Bohr magnetons $\mu_{\text{eff}}^{\text{exp}} = 2.84\,[\chi_M(T - \Theta)]^{1/2}\mu_B$, which is in good agreement with the theoretical value $\mu_{\text{eff}}^{\text{theor}} = g_J[J(J + 1)]^{1/2}\mu_B = 3.62\,\mu_B$, where J is the quantum number of the lower level of the multiplet of the Nd^{3+} ion, and g_J is the Landé factor]. The electrical measurements showed that all the investigated samples were metals at room temperature.[7]

Figures 1–3 give the dependences of the quantity χ^{-1} on T for neodymium monochalcogenides. In the temperature range 150–70° K, the dependence $\chi^{-1}(T)$ for NdS obeys the Curie–Weiss law but below these temperatures they are departures from the linear dependence $\chi^{-1}(T)$ and the susceptibility (within the limits of accuracy of measurements of χ, which was 1.5%) is independent of the intensity of the external magnetic field H_0.

Below 50° K the $\chi^{-1}(T)$ curve has a kink and a spontaneous magnetic moment $\sigma_S(T)$ appears. In this range of temperatures the magnetization depends linearly on the external magnetic field intensity: $\sigma(T, H_0) = \sigma_S(T) + \chi(T)H_0$. The dependence of the spontaneous magnetic moment on temperature is given in Fig. 4.

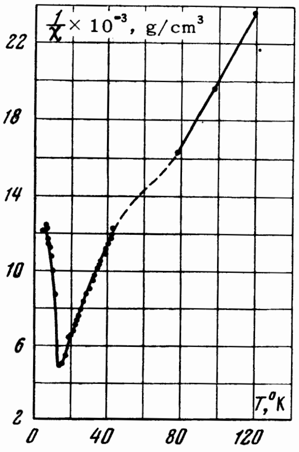

FIG. 3. Temperature dependence of the reciprocal of the specific magnetic susceptibility of NdTe.

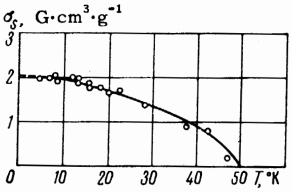

FIG. 4. Temperature dependence of the specific spontaneous magnetic moment of NdS.

The spontaneous magnetic moment per formal unit of NdS ($n = 0.067\,\mu_B$) represents only a small fraction (2%) of the magnetic moment of the Nd^{3+} ion, calculated from Hunds formula ($n = g_J J \mu_B = 3.28\,\mu_B$). It would seem that this can explain the transition to the weakly ferromagnetic state. In the light of Turov's work,[8] the appearance of weak ferromagnetism of relativistic origin is possible in structures of the NaCl type (space symmetry group O_h^5). However, it is also likely that the transition at T = 50° K is due to the presence of ferromagnetic compounds or solid solutions of europium and gadolinium. The neodymium, used in the synthesis of the NdS, contained large amounts of other rare-earth elements as impurities, compared with the neodymium used to prepare the NdSe and NdTe. This was also supported by the fact that the transition temperature of 50° K was unusually high compared with other neodymium compounds. Therefore, we concluded that the sharp change in χ at 8° K corresponded to a phase transition to the antiferromagnetic state.

From a consideration of the dependence $\chi^{-1}(T)$ for NdSe and NdTe, it was evident that these compounds satisfied the Curie–Weiss law up to 80° K, but below this temperature there were deviations from the linear dependence, which were evidently due to the influence of the crystal field. At temperatures below 14° K in the case of NdSe and below 13° K in the case of NdTe, there was a transition to the antiferromagnetic ordered state. The susceptibility of these compounds was independent of the external magnetic field intensity over the whole investigated temperature range.

The Nd^{3+} ion in the investigated monochalcogenides has the electron configuration $4f^3$. According to Kramers' theorem, if an ion has an odd number of electrons in its partly filled shell, the lowest energy level of the ion in the crystal field remains degenerate. Then, below some definite temperature (T_N), at which kT becomes less than the exchange energy, there is a transition to a magnetically ordered state, as observed for neodymium monochalcogenides. In this connection, it is useful to note that, as discovered by us earlier,[9] cerium monochalcogenides are also transformed to a magnetically ordered state at low temperatures.

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