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Magnetic g factor and magnetic anisotropy of Dy^{3+} ion in Dy_2BaNiO_5 determined from optical spectra of f-f transitions †

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The spectroscopic study of dysprosium chain nickelate, Dy_2BaNiO_5, was performed. New information on the crystal-field (CF) levels of the ground $^6H_{15/2}$ multiplet of Dy^{3+} ion was obtained. The evaluation of the magnitude of the Dy^{3+} magnetic moment using spectroscopic data gives a value of $9\,\mu_B$. Spectroscopic study of Dy_2BaNiO_5:Er (1 at.%) has shown that the g_x component of the magnetic g factor of dysprosium is zero. These findings are in agreement with predictions based on CF calculations performed by professor Boris Malkin [1].

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Keywords: chain nickelates, rare earth ions, phase transitions, magnetic g factor, magnetic anisotropy.

1. Introduction

The compounds R_2BaNiO_5 (R is a rare-earth (RE) element), so-called chain nickelates, are well known as model compounds for studying one-dimensional (1D) magnetism and crossover to three-dimensional (3D) magnetic ordering. Low-dimensional magnets are of great interest from the point of view of fundamental physics. It is worth mentioning that the 2016 Nobel Prize was given, among others, to Haldane for the discovery of new phase states of quantum magnetic chains [2, 3]. 1D magnets are also interesting for practical applications as materials with 1D thermal conductivity [4] and as a working media in adiabatic demagnetization cryostats [5]. Interest in RE chain nickelates is also related to the study of the magnetoelectric effect recently discovered in them [6–9].

In Y_2BaNiO_5 with a single magnetic system of Ni^{2+} chains interconnected by nonmagnetic Y^{3+} ions residing in C_{2v} symmetry positions, the most characteristic features of 1D magnetism are observed. (i) There is no 3D magnetic ordering [10]. (ii) The temperature dependence of magnetic susceptibility has a wide maximum at a temperature ~ 410 K due to antiferromagnetic (AFM) correlations within chains [11]. (iii) A Haldane gap is observed in the spectrum of magnetic excitations [11, 12]. Replacing non-magnetic yttrium with a magnetic RE element changes the behavior of R_2BaNiO_5. 3D magnetic ordering is observed [13–20]. At the same time, there is a significant reduction of spins and the Haldane gap is preserved at temperatures both above and below the Néel temperature [21–23]. This was interpreted as the coexistence of 1D and 3D magnetism.

Dy_2BaNiO_5 is AFM ordered at low temperatures. Data on the Néel temperature T_N vary slightly in the range 59–64 K [13, 14, 24, 25]. A study of the temperature-dependent magnetic susceptibility of Dy_2BaNiO_5 revealed two features [7, 24]. In addition to the relatively narrow peak at T_N, there is a broad maximum at a lower temperature around 40 K, which, as will be shown below, is associated with the splitting of the ground Kramers doublet of the Dy^{3+} ion.

To construct a theory explaining magnetic properties, the knowledge of the CF parameters and wave functions of RE ions is necessary. These data can be obtained from calculations using

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[†] This paper is dedicated to Professor Boris Z. Malkin, who made a significant contribution to the field of magnetic radio spectroscopy in Kazan University, on the occasion of his 85th birthday.

the CF theory, which, in turn, is based on experimental data on the CF energies of RE ions. Professor Boris Zalmanovich Malkin is a recognized classic in the field of CF theory and CF calculations. His exchange charge smodel (ECM) [26–28] gives excellent results [1,19,29–35] and is used by many other researchers [35–37]]. The main advantage of ECM is that the calculated initial CF parameters are physically grounded. These parameters take into account both point charges and the exchange interactions with the nearest neighbors. ECM helps to find the global minimum for a function of many variables and to avoid the falling into a local minimum.

B.Z. Malkin estimated the CF acting on the Dy^{3+} ion in Dy_2BaNiO_5 [1]. According to the assessment, (i) the ground state of the Dy^{3+} ion in Dy_2BaNiO_5 is an isolated Kramers doublet, the next sublevel of the ground multiplet $^6H_{15/2}$ is located at an energy distance of more than 120 cm^{-1}. (ii) The components of the magnetic moment of the Dy^{3+} ion in the ground state are (in units of Bohr magneton) $g_x/2 = 0.08$, $g_y/2 = 0.04$ and $g_z = 9.66$, which is in agreement with the neutron measurements [14]. On the other hand, it is possible to get information on the magnitude and anisotropy of Dy magnetic moment using optical spectroscopic data. The main goal of this work is to test the theoretical predictions regarding chain nickelate Dy_2BaNiO_5 [1] using optical spectroscopy methods. For this purpose, (i) we studied the structure of low-energy levels of the ground $^6H_{15/2}$ multiplet of the Dy^{3+} ion in Dy_2BaNiO_5, and (ii) we evaluated the magnitude of the magnetic g factor of the Dy^{3+} ion as well as its anisotropic properties.

2. Experimental details

Polycrystalline samples of Dy_2BaNiO_5 and Dy_2BaNiO_55:Er (1 at.%) were synthesized at the Lomonosov Moscow State University by B.V. Mill. The transmission spectra were measured in a wide range of frequencies and temperatures using a Bruker 125 HR spectrometer. To measure the transmission spectra, the tablets of a mixture of sample and optical quality potassium bromide were prepared. To register the multiplets of the Dy^{3+} ion in a wide spectral region, MCT and InSb detectors and a silicon diode were used. Temeperature-dependent transmission spectra were measured using Cryomech PT403 closed-cycle optical helium cryostat.

3. Results and discussion

Figure 1 shows the transmission spectra of Dy_2BaNiO_5 over a wide range of frequencies at temperatures of 4.2 K and 265 K. Groups of relatively narrow lines are multiplets of the Dy^{3+} ion, indicated by their names. At helium temperature, the spectral lines are intense and narrow. At temperatures close to room temperature, the lines broaden significantly. In high-temperature spectra, additional lines are visible on the low-frequency side of each multiplet, corresponding to transitions from excited CF levels of the ground multiplet, the population of which increases with increasing temperature. These lines provide information on the energy structure of the ground multiplet.

Figure 2 shows a diagram of the energy levels of the Dy^{3+} Kramers ion. The CF of C_{2v} symmetry splits the multiplets of the free Dy^{3+} ion into $(2J + 1)/2$ Kramers doublets (J is the total moment). In the absence of a magnetic field, absorption is observed due to optical transitions from the CF levels of the ground $^6H_{15/2}$ multiplet of the Dy^{3+} ion, designated by the Latin numerals I, II, etc. to the CF levels of the excited multiplets $^{2S+1}L_J$, denoted by capital Latin letters A, B, etc.). Figure 2 shows the transition IA from level I to level A. In the AFM phase, the internal magnetic field B_{eff} splits the Kramers doublets. In the general case, the spectral line splits into four components.

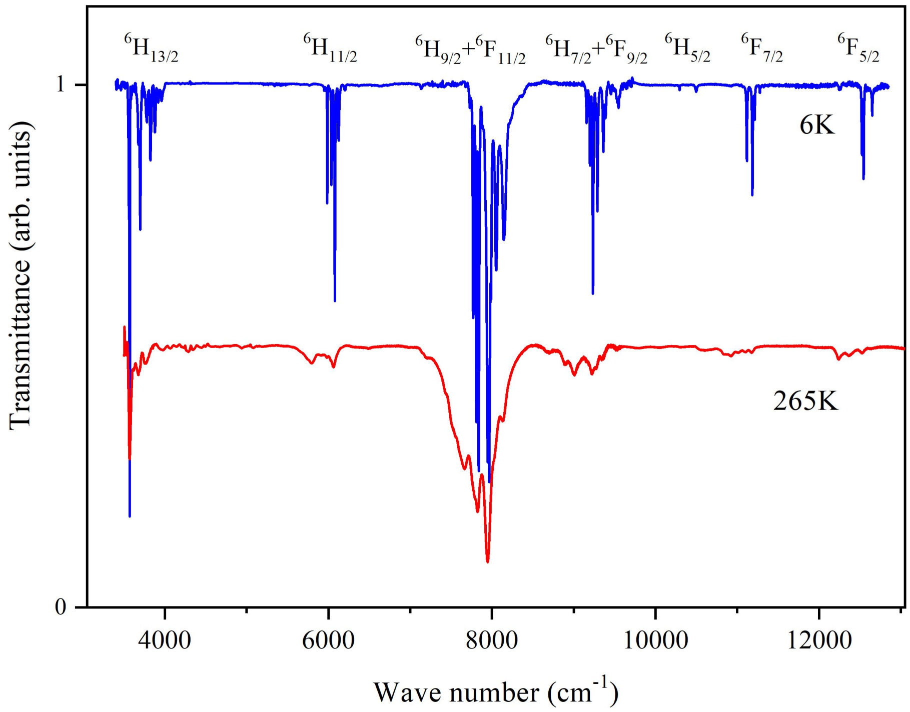

Figure 1. Transmission spectra of Dy_2BaNiO_5 at temperatures of 4.2 K and 265 K (the transmission spectrum at 265 K is shifted along the ordinate axis for clarity). The symbols for the energy multiplets of the Dy^{3+} ion are shown at the top.

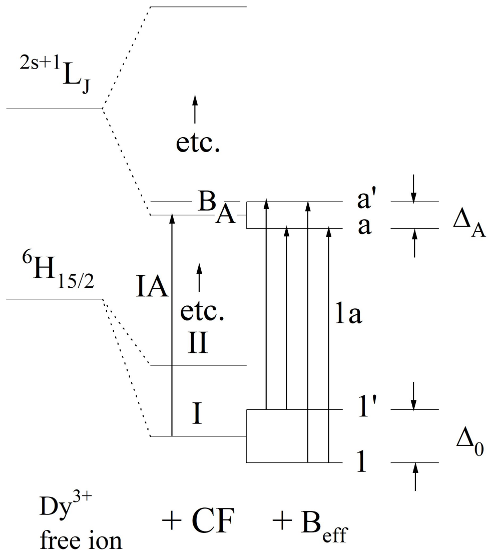

Figure 2. The energy levels scheme of the Dy^{3+} Kramers ion in paramagnetic and AFM states.

3.1. CF levels structure of the ground $^6H_{15/2}$ multiplet of Dy^{3+} in Dy_2BaNiO_5

Figure 3 shows the transmission spectra in the low-frequency region of the $^6H_{15/2} \rightarrow ^6F_{5/2}$ transition in the Dy^{3+} ion at temperatures above and below T_N. The spectral line splits in the AFM phase due to the lifting of the Kramers degeneracy by the effective magnetic field B_{eff}. With heating above T_N, new lines appear on the low-frequency side of the IA line. As was mentioned above, they are due to transitions from excited levels of the ground multiplet. Two broad structured bands are observed. One of them, separated by about 150 cm^{-1} from the IA line becomes noticeable at temperatures of about 50 K. The other one is about 250–300 cm^{-1}

apart from IA line and becomes observable at temperatures above 80 K. At further increase in temperature no new lines appear, but the ratio of intensities changes. Apparently, each of the two broad bands is a combination of several lines. They include the whole set of lines IIA – VIIIA, which is shown in Fig. 3. For other multiplets, a similar pattern is observed, namely, two broad structured bands on the high-frequency side of the IA line grow as the temperature increases.

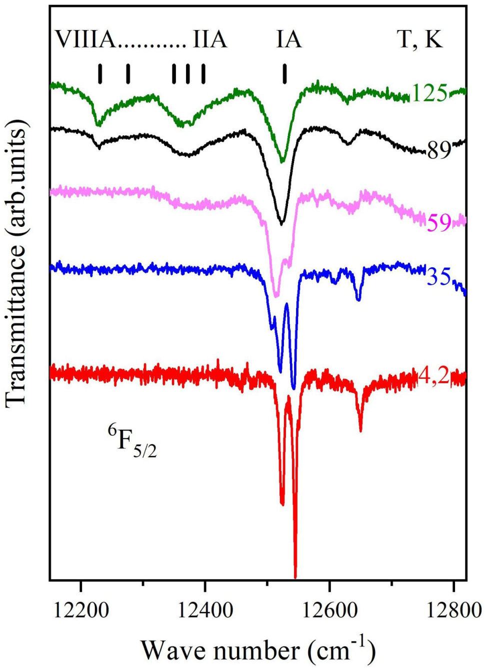

Figure 3. Transmission spectra of Dy$_2$BaNiO$_5$ in the low-frequency region of the ^{6}H$_{15/2}$ →^{6}F$_{5/2}$ transition at different temperatures.

To further elucidate the energy structure of the ^{6}H$_{15/2}$ ground multiplet of the Dy^{3+} ion, we studied the behavior of the temperature dependences of the integral intensities of several spectral bands, see Fig. 4. These intensities should be proportional to the populations of the initial levels of the corresponding spectral transitions. The populations, in turn, follow the Boltzmann distribution. The ^{6}H$_{15/2}$ ground multiplet of the Dy^{3+} ion consists of eight Kramers doublets. Their populations n_i are described by the following relation:

$$n_i(T) = \frac{e^{-\Delta_i/k_\mathrm{B}T}}{1 + \sum_{i=2}^{8} e^{-\Delta_i/k_\mathrm{B}T}}, \tag{1}$$

where Δ_i is the energy of the i-th doublet, k is the Boltzmann constant, T is the temperature. The experimental intensities are best described using formula (1) for the following set of energy states: 0, 133, 150, 156, 178, 190, 260, 290 cm^{-1}. This set agrees well with the prediction of [1] based on the CF calculation performed by B.Z. Malkin.

3.2. Estimation of the magnetic g factor of the Dy^{3+} ion

For the whole set of observed splittings of spectral lines, we analyzed the temperature dependence of lines splittings. As a result, we deduced the dependence $\Delta_0(T)$, the splitting of the ground Kramers doublet of the Dy^{3+} ion in the Dy$_2$BaNiO$_5$, see Fig. 5. The derivative of $\Delta_0(T)$ has a minimum at the Néel temperature $T_\mathrm{N} = 61 \pm 1$ K. As can be seen, the splitting is not equal to zero even for temperatures much higher than T_N, which confirms the presence of magnetic

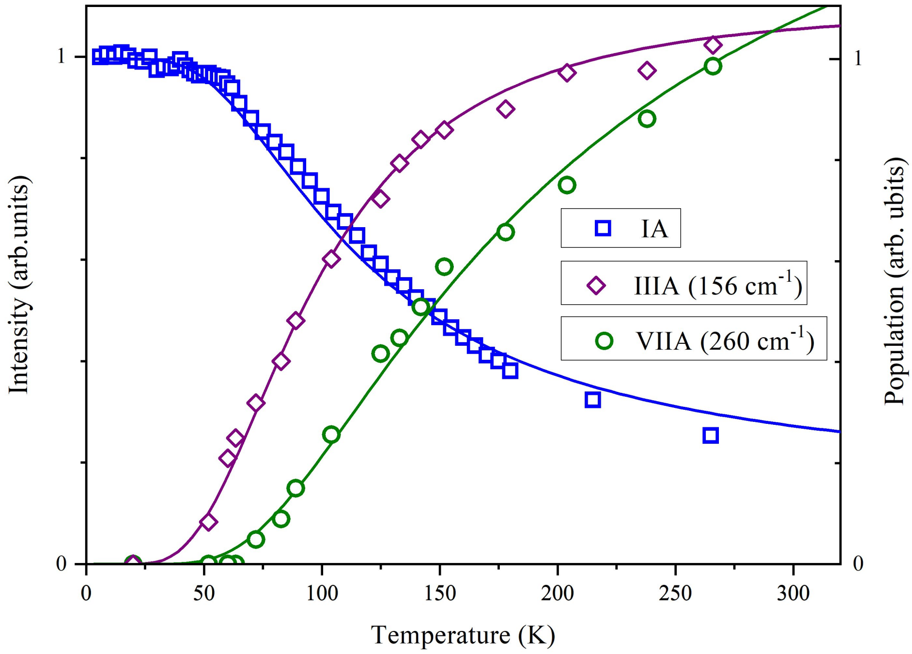

Figure 4. Intensities of the IA, IIIA, and VIIA spectral bands and calculated populations of the Stark levels of the $^6H_{15/2}$ ground multiplet of the Dy^{3+} ion in the Dy_2BaNiO_5 crystal.

correlations in Ni^{2+} magnetic chains.

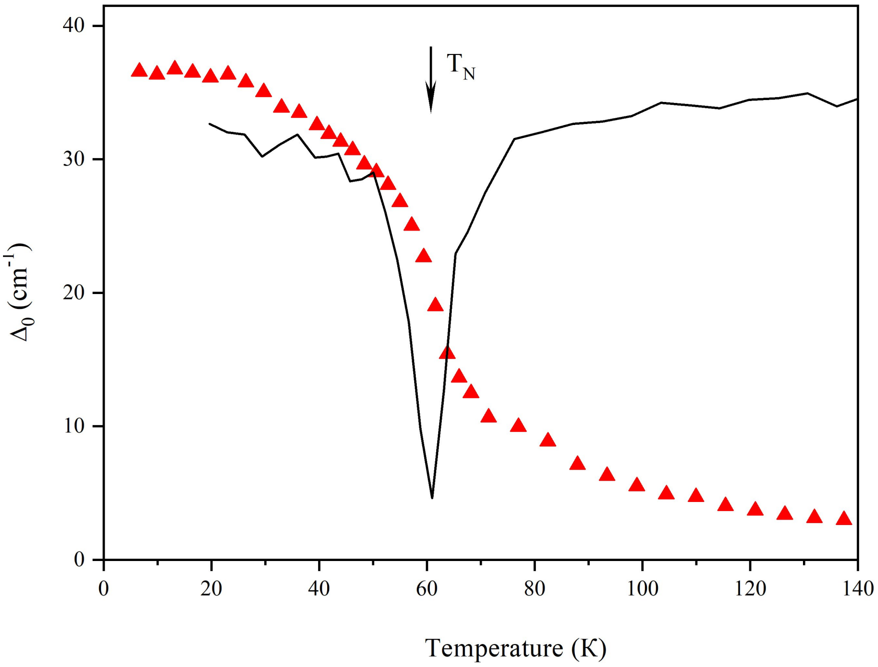

Figure 5. Splitting $\Delta_0(T)$ of the ground Kramers doublet of the Dy^{3+} ion as a function of temperature (symbols) and the derivative of $\Delta_0(T)$ (line).

The magnitude of $\Delta_0(T)$ determines the low-temperature dynamics of the studied magnetic material (Schottky anomaly in heat capacity, peculiarities of magnetic susceptibility and thermal conductivity, etc.) [38–42]. We use the obtained experimental dependence $\Delta_0(T)$ to calculate the contribution of dysprosium to the magnetic susceptibility of Dy_2BaNiO_5, since data on the temperature dependence of the magnetic susceptibility are presented in the literature [7,14]. In the simplest model of isolated ground doublet, we can use the following relation [41]:

$$\chi = \frac{2N_\mathrm{A} m_\mathrm{Dy}^2(0)}{3m_\mathrm{mol} k_\mathrm{B} T} \cosh^{-2} \frac{\Delta_0(T)}{2k_\mathrm{B}T}, \tag{2}$$

where N_A is Avogadro constant, m_mol is the molar mass, $m_\mathrm{Dy}(0)$ is the magnetic moment of Dy^{3+} at zero temperature.

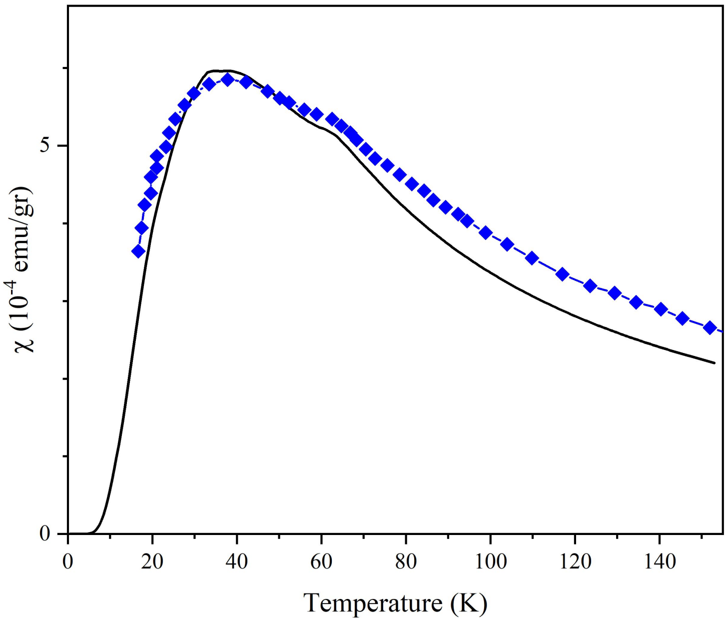

Figure 6. Temperature dependence of the magnetic susceptibility. Experimental data [14] (symbols) and calculation using spectroscopic data (line).

The solid line in Fig. 6 is the result of calculation using Eq. 2. The best fit to the experimental data was obtained for the value of the magnetic moment of dysprosium $m_\mathrm{Dy}(0) = 9\,\mu_B$. This value agrees well with the data predicted by B.Z. Malkin [1]. In general, the calculated curve qualitatively well describes the experiment. In the temperature region below 40 K, the discrepancy is most likely due to the presence of uncontrolled paramagnetic impurity in the sample, which was studied in [14]. In the temperature region above 40 K there is also a small discrepancy. In this temperature region, levels IIA and IIIA start to be populated, see Fig. 4. These levels give an additional contribution to the susceptibility, which is not taken into account in the model described by Eq. 2. Thus, the main contribution to the magnetic susceptibility of Dy$_2$BaNiO$_5$ at low temperatures is due to the splitting of the ground Kramers doublet of the Dy^{3+} ion.

3.3. Spectroscopic information on the magnetic anisotropy of the Dy^{3+} ion in Dy$_2$BaNiO$_5$

To get an additional information on the magnetic anisotropy of the Dy^{3+} ion in Dy$_2$BaNiO$_5$, we apply the erbium probe method. In this method we use impurity RE (Er) probe, thus the probe does not disturb the structure, and its spectral characteristics contain information about the properties of the studied crystals [43, 44]. In the framework of the mean-field model, the splitting of Kramers doublets of the Er^{3+} ion is described by the following formula:

$$\Delta = \mu_\mathrm{B}\sqrt{(g_x B_{\mathrm{eff},x})^2 + (g_y B_{\mathrm{eff},y})^2 + (g_z B_{\mathrm{eff},z})^2}, \tag{3}$$

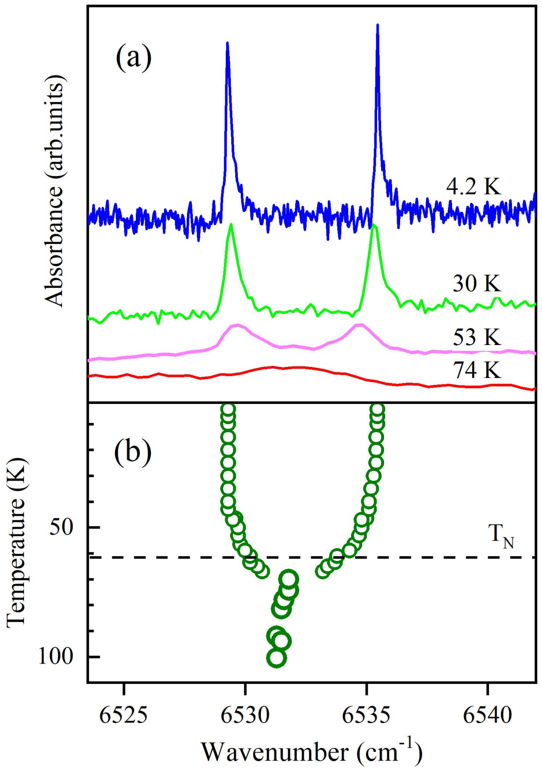

Figure 7. (a) The absorption spectra in the region of the lowest-frequency spectral line of the $^4I_{15/2} \rightarrow {}^4I_{13/2}$ transition in the Er^{3+} ion in a Dy_2BaNiO_5:Er (1 at.%) crystal at different temperatures. (b) Temperature dependence of the position of the 6532 cm^{-1} spectral line in the paramagnetic phase and the positions of the split components in the magnetically ordered state.

where g_i and $B_{\mathrm{eff},i}$ are the components of the g factor and of the internal effective field, respectively. If the components of the g factor of the probe are very different, the splitting will be sensitive to the direction of the internal magnetic field. For example, if $g_y = g_z = 0$, the splitting will occur only for the $B_{\mathrm{eff}} \parallel x$ field. This is exactly the situation for the Er^{3+} ion in Er_2BaNiO_5 (see Table 1).

Table 1. Components of the g factor of the ground doublet of the Er^{3+} ion in Er_2BaNiO_5 obtained by spectroscopic study [45] and by neutron scattering [14].

	g_x	g_y	g_z
Optical spectroscopy	15.53	0.26	0.04
Neutron scattering	14.46	0	0.64

The results of the spectroscopic study of Dy_2BaNiO_5:Er (1 at.%) are presented in Fig. 7. At the Néel temperature $T_N = 61$ K, a magnetic phase transition occurs and the effective magnetic field B_{eff} removes the Kramers degeneracy of Er^{3+} levels in Dy_2BaNiO_5:Er (1 at.%), resulting in a splitting of erbium spectral lines. The low-frequency component of the split line does not disappear with decreasing temperature. Instead, its intensity increases, as well as the intensity of the high-frequency component. This suggests that the line splitting is caused by the splitting of the upper level of the transition. At the same time, the splitting of the ground doublet is zero, which is confirmed by the absence of other components in the spectra.

The large component g_x makes the erbium probe very sensitive to the magnetic field in this direction, while small values of g_y and g_z are unlikely to distinguish the field along these directions.

The absence of ground state splitting in the erbium probe allows us to conclude that the magnetic moments in dysprosium nickelate are oriented perpendicularly to the x axis, which is consistent with the prediction of B.Z. Malkin, as well as with the conclusions of [14] that the magnetic moments of dysprosium and nickel in dysprosium nickelate are oriented along the z axis

4. Conclusions

The infrared transmission spectra of chain nickelates Dy_2BaNiO_5 and Dy_2BaNiO_5:Er (1at.%) were investigated in a wide range of frequencies and temperatures. The energies of CF levels of the $^6H_{15/2}$ ground multiplet of the Dy^{3+} ion were determined to be, most likely, 0, 133, 150, 156, 178, 190, 260, and 290 cm^{-1}. By the splitting of spectral lines corresponding to the *f-f* transitions in Dy^{3+} and Er^{3+} ions, the magnetic ordering of Dy_2BaNiO_5 at $T_N = 61 \pm 1\,\mathrm{K}$ was registered. Using the spectroscopic data on the splitting $\Delta_0(T)$ of the ground Kramers doublet of the Dy^{3+} ion, we calculated the contribution of dysprosium into the magnetic susceptibility of Dy_2BaNiO_5. There is good agreement with experiment provided that the magnetic moment of dysprosium equals to $9\,\mu_B$. The study of temperature-dependent *f-f* transitions in the Er^{3+} ion in Dy_2BaNiO_5:Er (1at.%) leads us to the conclusion that the g_x component of the magnetic g factor of the Dy^{3+} ion in Dy_2BaNiO_5 is zero. All these results coincide with the theoretical predictions of Boris Malkin [1].

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