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Analysis on an abnormal behavior of magnetization in neodymium trifluoride at low temperatures

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ABSTRACT

An abnormal phenomenon is experimentally found that the saturation magnetization in NdF_3 drops with decreasing temperatures in high magnetic fields by Guillot. In this paper, by studying the magnetic and magneto-optical properties, a thorough explanation is presented by a two-sublattice model. Our theory points out that the abnormal magnetic behavior mainly originates from the vibration of the exchange interaction between two Nd sublattices in NdF_3. And, the saturation characteristic of magnetization is theoretically explored at different temperatures under extremely high magnetic fields. Meanwhile, the magnetic moments of the two sublattices A and B are discussed. Based on the conclusions of the magneto-optical experiments, it further implies that light irradiation can affect the magnetic properties in NdF_3. And, the corresponding physical mechanism is revealed.

1. Introduction

In recent years, much attention has been paid to the theoretical and experimental studies on physical properties of magnetic materials under extreme conditions, for some novel magnetic properties have been brought out, especially at low temperatures and in high magnetic fields [1–3]. However, due to the complicated physical mechanism of the occurrence of these new phenomena, most of the experiments in some certain materials have not been explained by appropriate theory [4]. Furthermore, up to now, the essence of some properties has been debated in the literatures [5–7]. Now, the main aim of this paper is, using a two-sublattice model, to theoretically analyze an abnormal behavior of magnetization in NdF_3 experimentally found by Guillot [8].

As is known, Guillot et al. have systematically investigated the magnetic and magneto-optical (MO) behaviors in paramagnetic neodymium trifluoride by experiments, and revealed some special magnetic and MO properties in the low temperature range, for example, the abnormal temperature dependence of the saturated magnetization M_S, and the nonlinear dependence of the Faraday rotation θ on the applied field H_e [8,9]. In our previous paper, we have successfully interpreted the MO properties of NdF_3 under high magnetic fields by quantum theory where the nonlinear and reciprocal properties of MO effects in paramagnetic media under high magnetic fields are pointed out [10]. Also, using one-ion model, Zhang et al. present a theoretical analysis of magnetic and MO characteristics, where it is found that the theoretical calculations of magnetization cannot excellently agree with the experiments under high magnetic fields [11]. Our point of view is that the existence of the difference between the theoretical and experimental curves mainly originates from the neglect of the exchange interaction among Nd^{3+} ions under high magnetic fields.

2. Outline of theoretical model

As to paramagnetic media, we have ever pointed out that the contribution of the exchange interaction cannot be ignored in the calculation of magnetization under high magnetic fields [12]. Also, as to the calculation of the magnetic properties in paramagnetic $Nd_3Ga_5O_{12}$, a two-sublattice model has been presented [13]. In this paper, based on two-sublattice model, a deep and thorough analysis on the above unusual properties in NdF_3 is carried out.

It is known that, rare earth trifluorides, such as PrF_3 and NdF_3, are antiferromagnetic, consisting of two magnetic sublattices at low temperatures, and will convert into paramagnetism at above Néel temperature. According to Ref. [6], the magnetic susceptibility χ_M of NdF_3 follows a Curie–Weiss law for $T > 30$ K. Then, suppose that the two magnetic sublattices, marked as A and B, exist in NdF_3, and, M_A and M_B are their magnetizations, respectively. According to Langevin theory of paramagnetism, M_A and M_B can be written as

$$M_i = M_{si}B(y_i) = N_i J g_J \mu_B B(y_i) \quad (i = \text{A or B}), \tag{1}$$

where g_J, J and μ_B stand for the Lander factor, the total quantum number and the Bohr magneton, respectively; N_i represent the number of atoms contributing to magnetization per unit volume in A and B sublattices, respectively; $B(y_i)$ are the Brillouin

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functions, and $y_i = Jg_J\mu_B H_i/(K_B T)$, where H_i are the total effective fields in A and B sublattices, respectively.Taking account of the exchange interactions between magnetic atoms in the neighboring as well as the same sublattices, we have [14]

$$H_A = H_e + \nu_{AB} M_B + \nu_{AA} M_A, \quad (2)$$
$$H_B = H_e + \nu_{BA} M_A + \nu_{BB} M_B, \quad (3)$$

where H_e is the applied magnetic field, ν_{AA} and ν_{BB} signify the exchange interaction coefficients of magnetic atoms within the same sublattices, and ν_{AB} and ν_{BA} represent those between magnetic atoms in neighboring sublattices. Assume χ_A and χ_B as the magnetic susceptibilities of A and B sublattices, respectively, hence $M_A = \chi_A H_e$, $M_B = \chi_B H_e$. Then y_A and y_B can be rewritten as

$$y_A = Jg_J\mu_B(1+\alpha_A)H_e/(K_B T), \quad (4)$$
$$y_B = Jg_J\mu_B(1+\alpha_B)H_e/(K_B T), \quad (5)$$

where $\alpha_A = \nu_{AB}\chi_B + \nu_{AA}\chi_A$, $\alpha_B = \nu_{BA}\chi_A + \nu_{BB}\chi_B$. Therefore, the total magnetization M of NdF_3 can be defined as the sum of M_A and M_B.

3. Results and discussion

Based on the above theoretical model, we made a theoretical fitting of the experimental $M - H_e$ curves of NdF_3. Due to the antiferromagnetic property, it can be supposed that $M_{sA} = M_{sB}$. Table 1 gives the fitting parameters of the α_A and α_B, where it shown that the values of α_A and α_B change with the increase of the temperatures. Meanwhile, it is worthy to note that the values of α_A and α_B become unequal with the increase of temperatures, while implies the difference of the magnetization in A and B magnetic sublattices. Different with the results in Ref. [11], the theoretical results, seen from Fig. 1, are in good agreements with the experimental data, which demonstrates that the exchange interaction has important influence on the magnetization characteristic of NdF_3 in the range of low temperatures.

To further explore the saturation characteristic of magnetization at different temperatures, the external magnetic field is applied up to 3000 kOe in our calculations. And then, three complete $M - H_e$ curves are obtained. Fig. 2 shows that for T = 1.6, 4.2 and 40 K, only after H_e strides over 50, 500 and 2500 kOe does the magnetization of NdF_3 approach saturation and finally achieve a saturated one of 1.05, 2.10 and 2.90 μ_B/Mole, respectively. According to paramagnetic theory, if not considering the exchange interaction in NdF_3, it can be calculated that the applied magnetic field is up to 23.8, 62.5 and 596 kOe when the magnetization tends to saturation at 1.6, 4.2 and 40 K, respectively. However, from the above analyses, the actual values are apparently bigger than those calculated from paramagnetic theory, which further implies that the exchange interaction cannot be neglected in the calculation.

As mentioned above, the increase of the temperatures will result in the difference of the magnetization of A and B sublattices. Fig. 3 gives the ratio of M_A and M_B at different temperatures when the magnetic field is up to 3000 kOe. At 1.6 K, the magnetic moments of A and B sublattices are equal. However, at 4.2 and 40 K, it is obvious that M_A is bigger than M_B under lower magnetic fields, which means that the rotation of the magnetic moments of A sublattice along to magnetic fields is easier than that of B sublattice. Moreover, the ratios of M_A and M_B decrease with the increase of the magnetic fields. Finally, in higher magnetic fields, the values of M_A and M_B tend to be equal, that is, the magnetic moments will tend to be parallel to H_e under much stronger magnetic fields. In addition, the insert of Fig. 3 presents the values of the magnetization of A sublattice. Here, it is found that, when H_e > 45 kOe, the values of M_A is obviously bigger at 4.2 K than those at 1.6 K, hence, the corresponding whole magnetization will be larger at 4.2 K as shown in Fig. 1.

Additionally, a large saturation magnetization drop can be observed with the decrease of temperatures. From our calculations, we suggest that this abnormal phenomenon of the magnetization in NdF_3 maybe originate from the variation of the exchange interaction at low temperatures and under high magnetic fields. As is known, the thermal energy will gradually drop with the decrease of temperatures. Under high magnetic fields, the orientation of magnetic moments is inclined to the direction of the applied fields. And, the competition of the ferromagnetic and antiferromagnetic exchange interaction becomes notable. Thus, relative to the situation of paramagnetic state at high temperatures, the rotation of

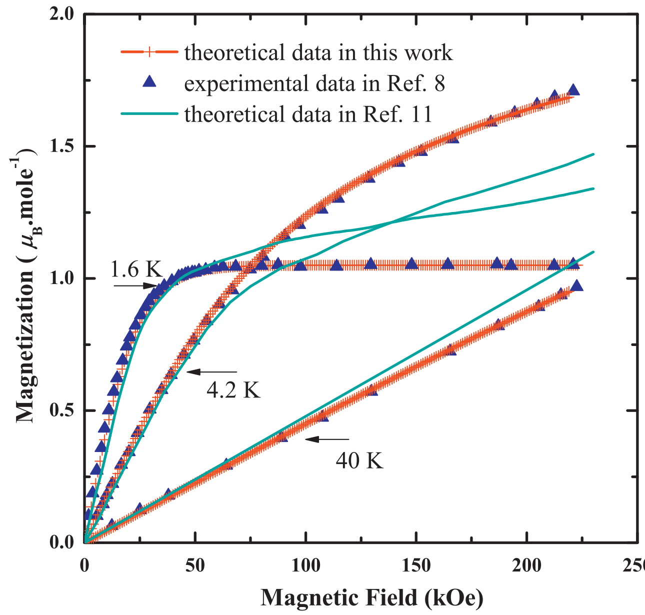

Fig. 1. Magnetization vs. the external magnetic field in NdF_3 at low temperatures.

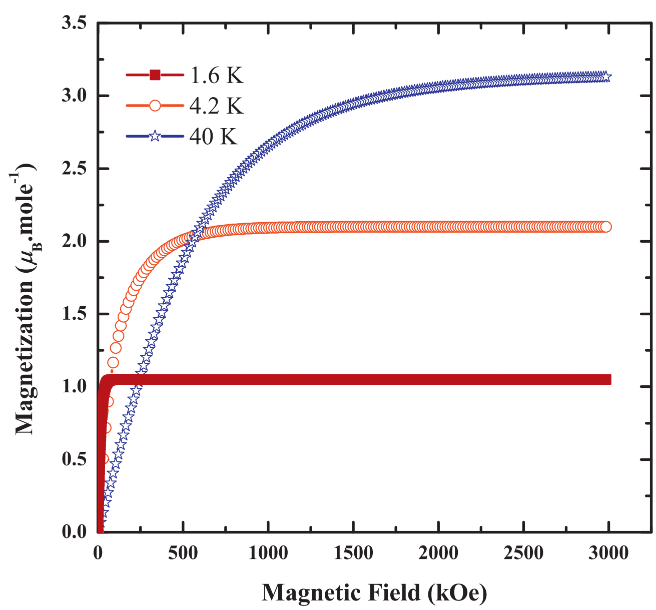

Fig. 2. Field dependence of magnetization in NdF_3 at 1.6, 4.2 and 40 K.

Table 1
The values of the parameters α_A and α_B.

	1.6 K	4.2 K	40 K
α_A	−0.02	−0.4	−0.4
α_B	−0.02	−0.85	−0.62