Giant magnetostriction of terbium titanates

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A giant magnetostriction has been observed in the terbium titanates $Tb_2Ti_2O_7$ and Tb_2TiO_2 in the paramagnetic state. The experimental results can be explained qualitatively and quantitatively by the crystal-field model.

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In this letter we are reporting measurements of the magnetostriction, magnetic susceptibility, and magnetization of the polycrystalline terbium titanates $Tb_2Ti_2O_7$ and Tb_2TiO_5.

These compounds have different crystal structures. Terbium dititanate, $Tb_2Ti_2O_7$, is a cubic compound with the pyrochlore structure; its unit cell has Tb^{3+} ions at trigonal-symmetry positions. The compound Tb_2TiO_5 exists in two polymorphic modifications: a low-temperature orthorhombic structure (the α phase) and a high-temperature hexagonal structure (the β phase). The crystal structure of these compounds and the techniques for preparing samples, by codeposition followed by high-temperature annealing and quenching are described in Refs. 1 and 2.

Figure 1 shows the experimental results on the magnetostriction $\Delta l/l$, plotted as a function of the magnetic field H, for $Tb_2Ti_2O_7$ and for the α and β phases of Tb_2TiO_5 at $T=4.2$ K. We see that the external magnetic field causes a pronounced deformation of these compounds, which reaches a value $\Delta l/l \approx 5 \times 10^{-5}$ in fields $\sim$50 kOe; such deformations are called "giant" deformations in the literature.[3,4] The highest magnetostriction is exhibited by the compound $Tb_2Ti_2O_7$, with the cubic structure, although its concentration of magnetic ions is lower than that in Tb_2TiO_5. Shown for comparison in Fig. 1 is the corresponding curve for a magneti-

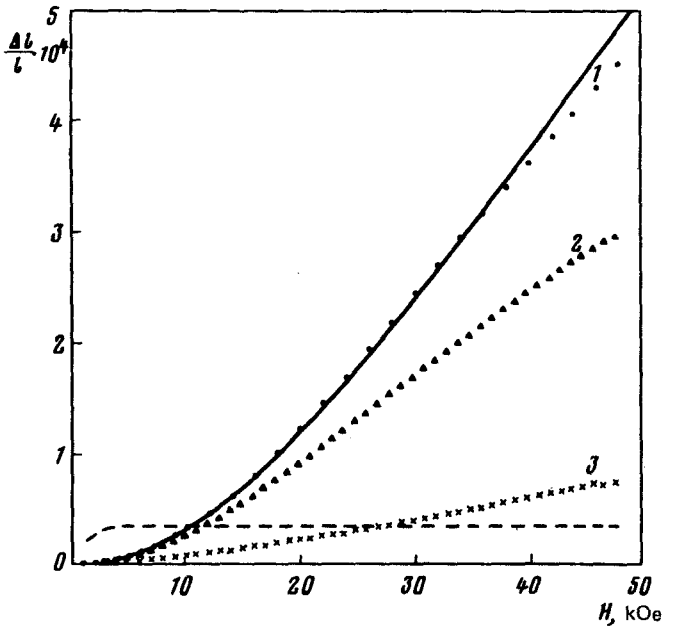

FIG. 1. Dependence of the magnetostriction on the external magnetic field. •–Experimental data for $Tb_2Ti_2O_7$; ▲–experimental data for α-Tb_2TiO_5; x–experimental data for β-Tb_2TiO_5; solid curve–calculation for $Tb_2Ti_2O_7$.

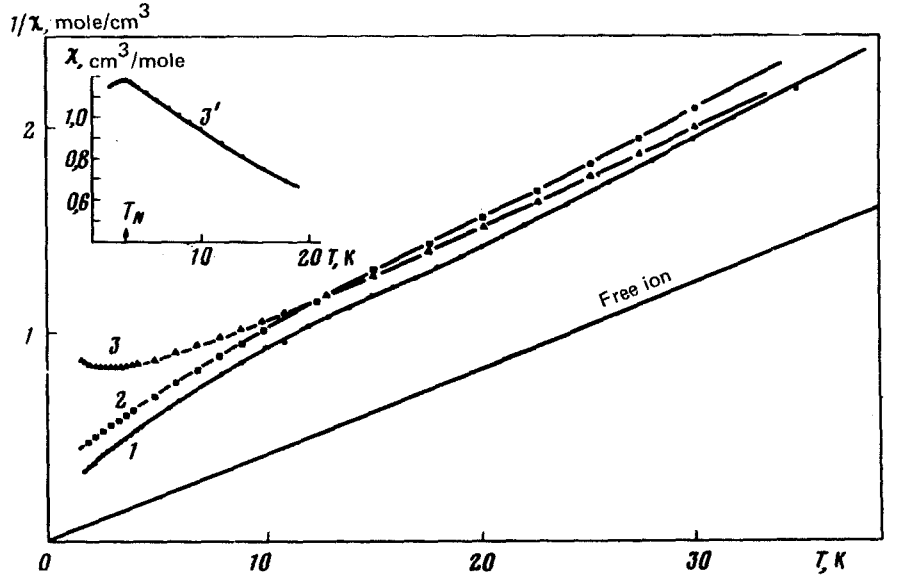

FIG. 2. Temperature dependence of χ^{-1}. 1–$Tb_2Ti_2O_7$; 2–β-Tb_2TiO_5; 3–α-Tb_2TiO_5; 3'–temperature dependence of the susceptibility for α-Tb_2TiO_5.

cally ordered sample of pure nickel. In weak fields ($H \lesssim 10$ kOe) the magnetostriction is proportional to H^2, as expected.[4]

These values of the magnetostriction of terbium titanate cannot be explained on the basis of a proximity to a transition to a magnetically ordered state. From Fig. 2, which shows the measured magnetic susceptibilities of these compounds, it can be seen that the samples are paramagnetic at $T = 4.2$ K. The $Tb_2Ti_2O_7$ and the β-Tb_2TiO_5 remain paramagnetic down to 1.7 K; only for the α-Tb_2TiO_5 do we observe an anomaly on the $\chi(T)$ curve, at $T = 2.8$ K, which may mean a transition to an ordered state. As mentioned earlier, however, the magnetostriction of α-Tb_2TiO_5 is smaller than that of cubic $Tb_2Ti_2O_7$.

According to the single-ion model of magnetostriction,[5] it may be suggested that the observed effect results from a change in the equilibrium positions of the anions which constitute the nearest neighborhood of the magnetic ion and which form its crystal field.

In the absence of direct spectroscopic data on the properties of the crystal fields in these compounds, it is difficult to offer a quantitative analysis of these results. We have obtained some information on the positions of the energy levels of the Tb^{3+} ion from the low-temperature part of the $\chi(1/T)$ curve and from the field dependence of the magnetic moment of the cubic compound $Tb_2Ti_2O_7$.

The experimental curves of $M(H)$, shown by the points in Fig. 3, do not agree with calculations based on the standard representation of a two-singlet Tb^{3+} ground state with a maximum angular-momentum projection $J_z = \pm 6$ and with a slight admixture of the $|0\rangle$ state. This is the picture observed for most other compounds having Tb^{3+} ions in trigonal crystal fields.[6,7]

The general shape of the magnetization curves corresponds better to a doublet ground state, and at least one of the excited levels should be quite low. At extremely low temperatures (~2 K), at which it seems quite safe to assume that only the ground-state doublet is populated, the dependence $\chi(1/T)$ is described by

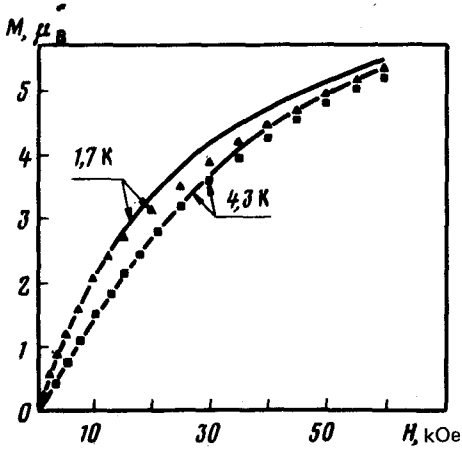

FIG. 3. Dependence of the magnetization on the magnetic field for the dititanate $Tb_2Ti_2O_7$ at 1.7 and 4.3 K. Points–Experimental; curves–theoretical.

$$\chi = \frac{C}{T} + \alpha, \qquad (1)$$

where $C = (3.9 \pm 0.1)\mu_B^2$ (deg) and $\alpha = 1.77\ \mu_B^2$ (per Tb^{3+} ion). The value of C gives us a more specific description of the structure of the gound-state doublet, while the value of α tells us the relationship between the energies of the excited states (E_n). The other expressions, which we need for E_n, can be found from a corresponding analysis of that part of the susceptibility curve on which the susceptibility begins to deviate from function (1), because the first excited state is being filled. According to our estimates, this first excited level should be at 15–20 K.

Rough calculations of the field dependence of the magnetization and the magnetostriction have been carried out on the basis of the crystal-field model and the energy-level scheme as constructed as above. The results agree well with the experimental data (Figs. 1 and 3). This agreement shows that all the measured magnetic characteristics, including the giant magnetostriction, are determined by that system of Tb^{3+} energy levels which exists in the crystal fields of these compounds. In particular, the presence of the low-lying first excited level predetermines all the magnetic properties to a large extent.

Admittedly, an analysis based solely on the low-temperature part of the susceptibility curve cannot unambiguously give us the system of quantum states. We hope that an analysis of experimental data on the susceptibility over a broad temperature range will make it possible to find the energy levels unambiguously, to carry out rigorous calculations of the functions $M(H)$ and $\Delta l/l(H)$ incorporating all the higher-lying states, and to thus eliminate the discrepancy between the theoretical and experimental curves at strong fields.

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