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Low-temperature phase transition in Dy

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We give a theoretical explanation for the low-temperature phase transition in Dy first experimentally observed by Rhyne *et al.* Our results are in excellent agreement with experimental observations. We also demonstrate that the phase transition is a first-order transition, a result until now not reported in the literature.

I. INTRODUCTION

In order to study the magnetoelastic properties of a rare-earth metal, several theoretical models have been developed.[1–3] However, none of these models have been worked out in sufficient detail in order to explain the phase transition at low temperature occurring in Dy. We believe that many interesting results may yet be obtained with the use of the models already known. The principal problem is that until now they have not been studied with great generality in order to obtain numerical results to compare with experiment. In order to do so, we have developed a very complete computer program.

In the present report we are interested in the theoretical explanation of the phase transition at low temperature in Dy. As reported by Rhyne *et al.*[4] and more recently by Liebermann and Graham,[5] this phase transition at a temperature of 4.2 K occurs with an external magnetic field of 74 kOe applied in the hard direction of magnetization of Dy crystals (*c* axis) Fig. 1. As can be observed in this figure, if we increase the magnetic field beyond the value of 70 kOe, we obtain a nonlinear increase of the *z* component of the reduced mag-

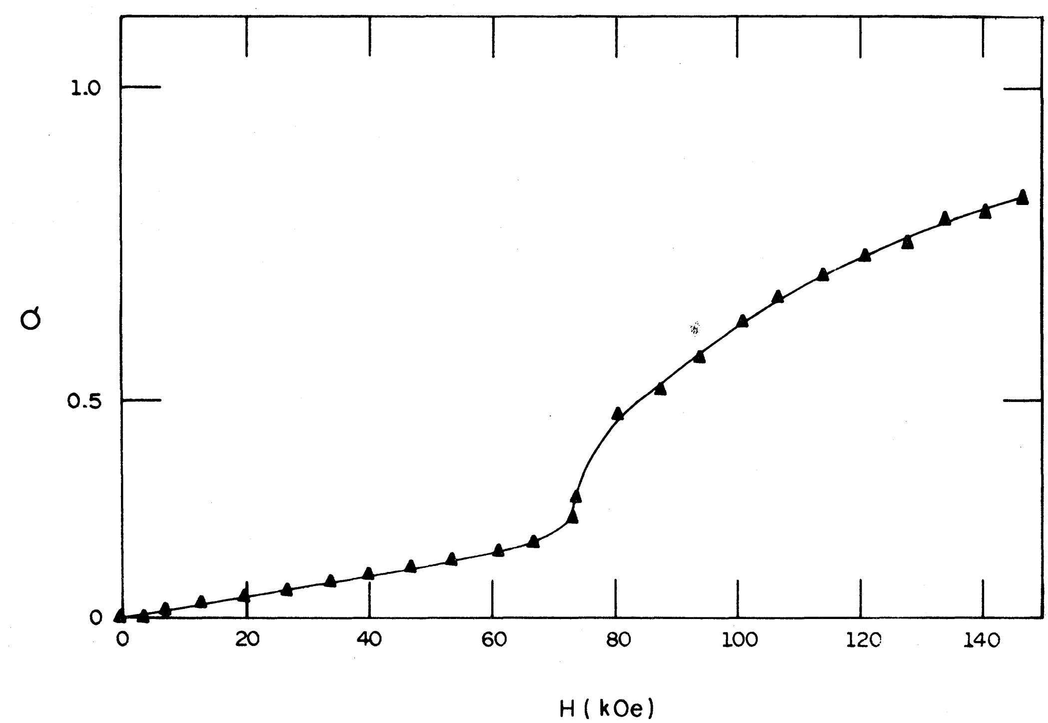

FIG. 1. Experimental determination of low-temperature phase transition in Dy (Ref. 4).

netization. The magnetic properties of the crystal are then permanently changed. Rhyne *et al.* observed that magnetic-moment measurements with higher fields along the *c* axis indicate a considerable reduction in axial anisotropy. Measurements in the pulsed field up to 380 kOe along the *c* axis actually produced a reversal in sign of the observed anisotropy. Such behavior, according to Rhyne *et al.*, may be consistent with the development of large internal strains and lattice changes, possibly of magnetostrictive origin.

Liebermann and Graham have reported recently that plastic deformation occurs in Dy at 4.2 K with applied magnetic field varying between 70 to 90 kOe. For Liebermann and Graham the mechanism is a mechanical twinning on the $\langle 10\bar{1}2 \rangle$ and $\langle 10\bar{1}1 \rangle$ system and the most likely driving force for the deformation seems to be the decrease in magnetic energy of the twins, which are oriented with an easy $\langle 10\bar{1}0 \rangle$ direction almost parallel to the field. Another possibility is that the magnetostrictive strain simply exceeds the yield strain. The experiments and their interpretation are made difficult by the large magnetic torque acting on a sample that is not perfectly aligned in the field.

Our calculation does not include plastic deformation effects in the Dy crystal. We have used a Hamiltonian which includes all terms already considered by Callen and Callen[1] and Southern and Goodings.[3] We have developed a very extensive computer program and tried to include as much information as possible in order to have as much detail as possible about the magnetostriction properties of all rare-earth metals which interest us. In the present report we have specialized our computer program in order to study the low-temperature phase transition of Dy reported by Rhyne *et al.*

II. THEORETICAL DETERMINATION OF THE PHASE TRANSITION

In the preceding paper we have considered some details of the general Hamiltonian used in our calculations. After the diagonalization of this Hamiltonian it is possible, among other things, to calculate numerically the Helmholtz free energy (divided by the Boltzmann constant *k*)

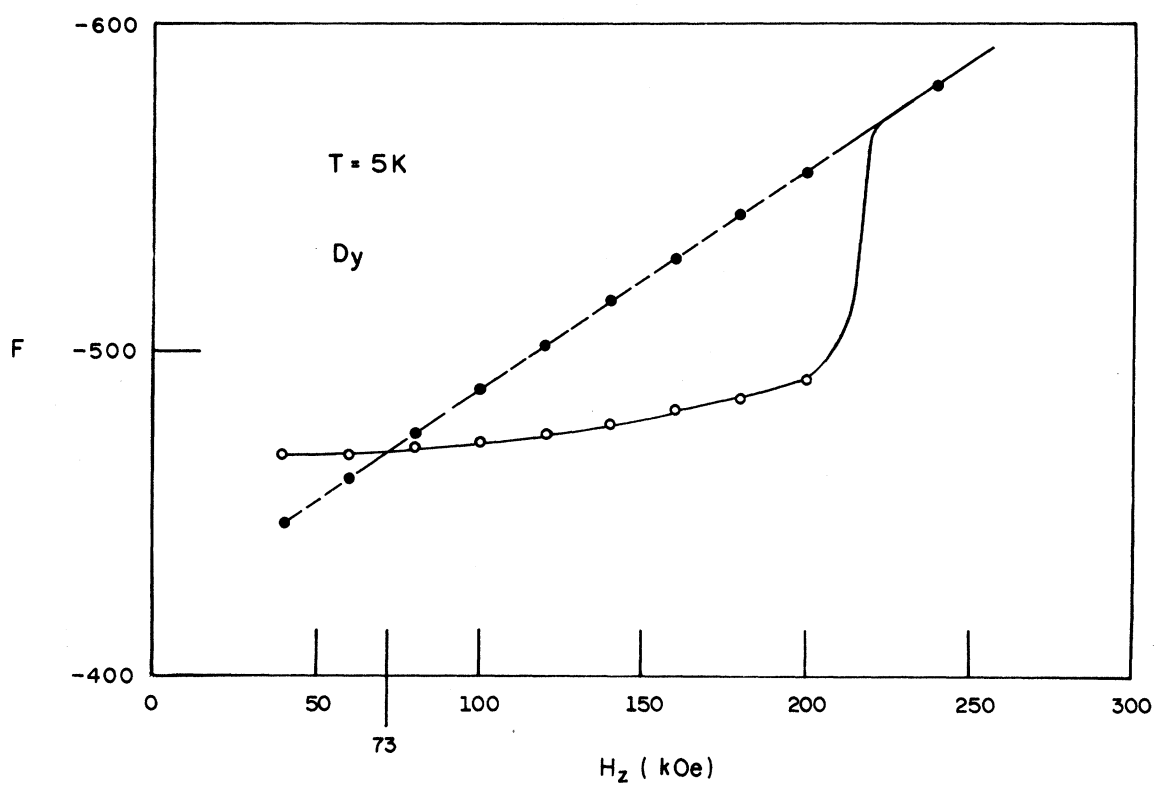

FIG. 2. Variation of Helmholtz free energy with external magnetic field applied in the Dy hard direction of magnetization. At high fields there appears a sort of saturation, the paths with increasing and decreasing field are different and they meet at 73 kOe.

$$F = -T \ln \sum_n \exp\left[-\frac{E_n}{T}\right], \qquad (2.1)$$

where E_n are the total-energy eigenvalues. In our calculation we have considered the following values for the anisotropy constants (in K/ion): $P_2^0 = 0.5$, $P_4^0 = -2\times 10^{-3}$, $P_6^0 = 4.0\times 10^{-7}$, and $P_6^6 = -9.77 \times 10^{-9}$. For one-ion magnetostriction coefficients we considered the values (in K/ion) $B_1^\alpha = -15$, $B_2^\alpha = -4.19$, $B^\gamma = 10.176$, and $B^\epsilon = 8.5$ and for the two-ion magnetostrictive coefficients (also in K/ion) $G_1^{\alpha,0} = 1.02$, $G_2^{\alpha,0} = 10.6$, $G_1^{\alpha,2} = 6.24$, $G_2^{\alpha,2} = 6.24$, $G^\gamma = -14.5$, and $G^\epsilon = 0$. These values fit the experimental results better. As we shall see later, the theoretical results we obtain here are in very good agreement with the experimental results. To our knowledge this is the first theoretical explanation for the low-temperature phase transition in Dy, whose experimental results were reported many years ago by Rhyne *et al.*

Our theoretical procedure to determine the phase transition is the following. We fix the temperature. Then, as in the laboratory, we apply increasing magnetic fields in the hard magnetization direction (*c* axis) of Dy crystal. For each value of the external magnetic field we calculate, through the computer program among other results of interest, the value of the Helmholtz free energy. We then plot the values of the free energy versus external magnetic fields (Fig. 2). As we see in this figure, the behavior of free energy versus magnetic field is different if the magnetic field increases or decreases. In Fig. 2 the low curve corresponds to the increasing magnetic fields while the upper curve corresponds to decreasing magnetic fields. At low fields there is only one crossover point. This point corresponds to a minimum of free energy of the two processes when increasing and decreasing the magnetic field.

In order to obtain the crossover point at the critical field stated in literature, we have varied some coefficients appearing in the Hamiltonian. We have observed that if we change in a large range the numerical values of these coefficients, although fixing the negative sign of the uniaxial anisotropy coefficient P_4^0, we obtain two different curves as in Fig. 2. The important change observed using the varied set of values is in the numerical value of the

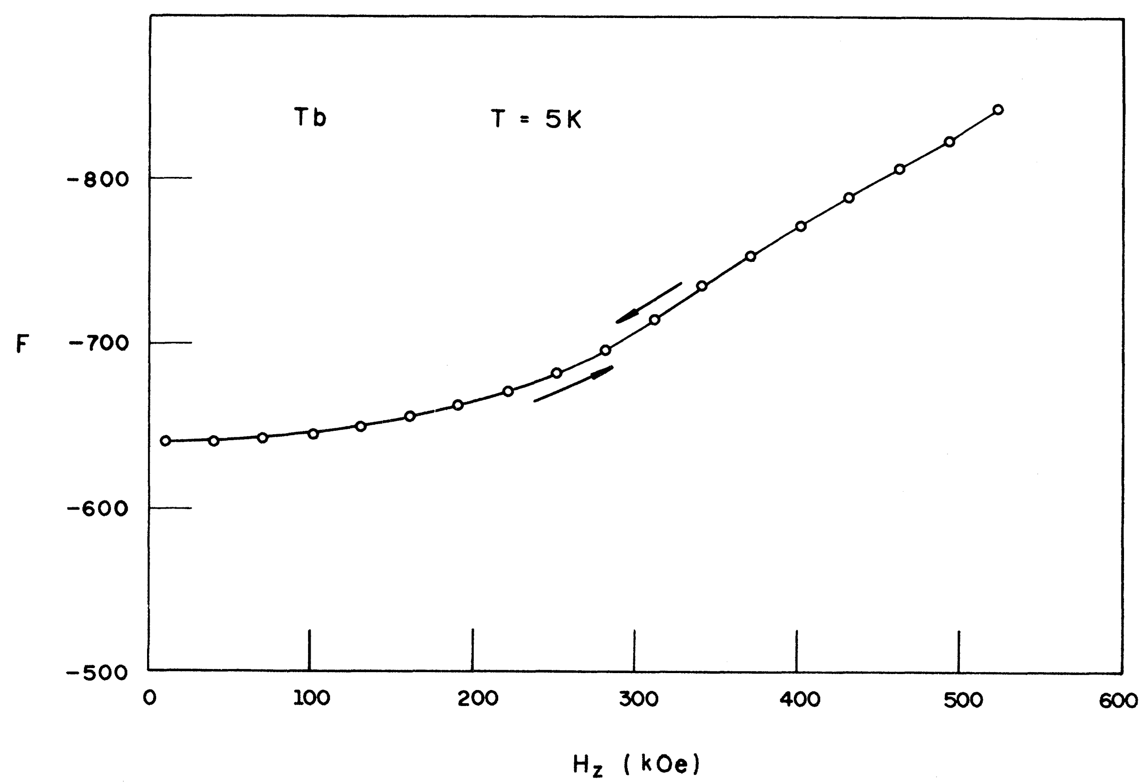

FIG. 3. Same situation as in Fig. 2 for Tb. The values of free energy at increasing and decreasing magnetic field are equal. There is no phase transition for Tb.

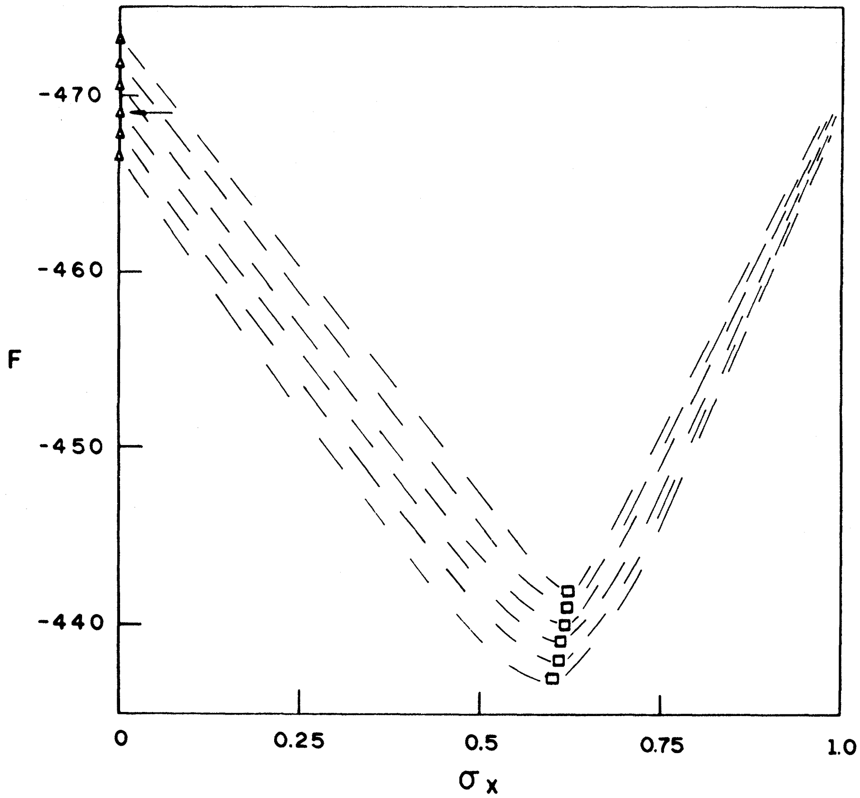

FIG. 4. Type determination of phase transition for Dy. We observe the lowering of free energy with the magnetization in the hard direction between the fields 72 and 74 kOe.

critical field (the crossover point).

We know that for Tb, the value of the P_4^0 coefficient is positive.[6] As a test for the stability of our computer program and in order to confirm the importance we have given the results of Fig. 2, which are a consequence of the negative sign of P_4^0 for Dy; we have used an analogous calculation for Tb. As can be seen in Fig. 3, if we vary the magnetic field up to 520 kOe, we obtain a completely reversible path for free energy as we increase or decrease the magnetic field.

We have determined theoretically the type of the phase transition occurring in Dy at low temperature. For this we have studied the behavior of the free energy for some input value of the x component σ_x of the reduced magnetization, when several values for the external magnetic field are applied in the c direction of Dy. For each value of σ_x and for each value of the external magnetic field, we obtain through our computer program a self-consistent value for σ_z and a value for the free energy. If we set $\sigma_x = 0$, we obtain quite naturally $\sigma_z = 1$. In this case, all values obtained for the free energy corresponding to each value of the external magnetic field are represented in Fig. 4 on the F axis. For the input values of σ_x equal to 0.5 and 1.0, we have other solutions indicated in Fig. 4. The dotted lines in this figure are not formed by solutions. We use these lines only in order to connect the three solutions for the same magnetic field. We started with a field of 68 kOe and arrived at a field of 78 kOe, varying the field in steps of 2 kOe. We see in Fig. 4 that there is a value of F between the fields of 72 and 74 kOe for which F is minimum for $\sigma_z = 1$. In general, we have two minima for the free energy corresponding to solutions with $\sigma_z = 1$ and $\sigma_x = 1$. When the magnetic field reaches some value, in our present case 73 kOe, these minima have the same depth, indicating that we have a first-order phase transition. (I am thankful to M. E. Foglio for calling my attention to this question.)

CONCLUSION

We have presented some results obtained through a detailed calculation that applied a theory of magnetostriction in many respects already known in literature. Our computer program is very extensive and with many operational possibilities. Some results presented in this paper are in excellent agreement with available experiments. We have also predicted the behavior of the critical magnetic field with temperature which is awaiting experimental confirmation.

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