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dc Anisotropic Magnetization at 20.4 K and the Ideal Crystal-Field-Only Behavior of TmSb

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The anisotropic magnetization of TmSb at 20.4 K has been measured in applied dc fields up to 144 kOe. The experimental results are in excellent agreement with isothermal crystal-field-only theory for parameters deduced from earlier pulsed-field experiments at 1.5 K. This resolves an ambiguity posed by earlier pulsed-field magnetization experiments at 20.4 K, the results of which fell between the adiabatic and isothermal crystal-field-only theoretical curves. High-resolution dc measurements of the magnetization at 1.5 K have also been made up to 52 kOe and are in good agreement with the earlier pulsed-field data.

I. INTRODUCTION

Analysis of the low-field susceptibility as a function of temperature and of the high-field anisotropic magnetization at 1.5 K for TmSb led to the conclusion that TmSb serves as a model crystal-field-only paramagnet.[1,2] This conclusion was subsequently verified by inelastic-neutron-scattering experiments.[3] The fact that TmSb acts as an ideal crystal-field-only material is of interest for two reasons. First, the magnetization and neutron experiments provide information on crystal field parameters that any *ab initio* theory of the crystal field must explain. Second, anisotropic-magnetization experiments serve as a valuable tool in the study of exchange effects[4,5] as one approaches the critical value of exchange for magnetic ordering in singlet ground-state systems[6]; the behavior of TmSb, for which there is no detectable exchange,[2,3] serves as a basis of comparison in discussing exchange effects in other systems such as[4,5] $Tb_zY_{1-z}Sb$.

Only one discrepancy occurred[1] in the comparison of the magnetic data for TmSb with those predicted on the basis of crystal-field-only behavior. The results of pulsed-field experiments at 20.4 K did not agree with the predictions of isothermal crystal-field-only theory for the same parameters that described the behavior of the low-field susceptibility as a function of temperature and the high-field anisotropic magnetization at 1.5 K. In Ref. 1, it was suggested that this discrepancy could arise if the spin system is unable to remain in equilibrium with the bath at 20.4 K during the pulse. If that were so, one would expect the pulsed-field magnetization data at 20.4 K to lie between the isothermal and adiabatic crystal-field-only theoretical values as observed in Ref. 1. (This question does not arise at 1.5 K. There is effectively no distinction between the adiabatic and isothermal magnetizations at 1.5 K, since essentially all the population is in the ground state at all fields experimentally attained.) It was also pointed out that the comparison of the experimental anisotropic magnetization of TmSb at 20.4 K with the prediction of crystal-field-only theory could be made unambiguously by performing the magnetic-moment measurements in a dc field, so that isothermal conditions would strictly pertain. The results of such dc experiments are reported here—the agreement with the crystal-field-only theoretical isothermal magnetization for the parameters indicated by the earlier susceptibility, the 1.5-K magnetic data,[1,2] and neutron-scattering experiments[3] is excellent. We conclude that the description of TmSb as a model crystal-field-only paramagnet is justified.

II. EXPERIMENTAL METHODS

The dc magnetization measurements were made on two oriented TmSb single crystals; one was employed for the [100] data and one for the [111] data. No chemical analysis was made on these crystals; however, they were chosen from the same source materials as those used for the earlier reported data.[1,2]

The magnetic moment was measured at high fields in water-cooled Bitter solenoids with a low-frequency vibrating-sample magnetometer (VSM).[7] The samples were immersed in liquid H_2 for measurements from 14 to 20.4 K. Only the 20.4-K data are presented here because these are directly compared with the earlier pulsed-field measurements. Because we were interested in clarifying relatively small differences, considerable care was used in calibration. The high-field data were normalized with much-higher-resolution measurements in a conventional[8] VSM adapted to a superconducting solenoid. These VSM data extended to 60 kG and to $T \simeq 30$ K. The samples were in contact with He gas and the temperature was measured with thermocouples to ± 0.1 K. The accuracy of the magnetic moment vs field of the high-field data (normalized to the lower-field data) is well within 2%. The data presented below show the applied field vs magnetization. Depolarization corrections are estimated to be less than 2%.

III. RESULTS AND DISCUSSION

In Fig. 1, the present dc magnetization values are superimposed on Fig. 2 of Ref. 1. Figure 2 of Ref. 1 gives the pulsed-field magnetization curves for TmSb at 20.4 K and the theoretical isothermal and adiabatic magnetization curves for fourth-order-only crystal field anisotropy with the W parameter giving the crystal field splitting chosen to match the low-field susceptibility in the limit of $T = 0$ K. (In the present Fig. 1, the theoretical isothermal curves have been extended to somewhat higher field than in Fig. 2 of Ref. 1.)

The present dc data, shown in Fig. 1, are distinctly different from the earlier pulsed-field data, and are in close agreement with the isothermal crystal-field-only theory. Presumably this difference indicates that the spin system is in thermal equilibrium with the bath only for the dc measurements.

The crystal field Hamiltonian for a rare-earth ion in an octahedral crystal field has the form

$$\mathcal{H}_{CF} = B_4(O_4^0 + 5O_4^4) + B_6(O_6^0 - 21O_6^4) \ . \quad (1)$$

Here O_4^0, O_4^4, O_6^0, and O_6^4 are specified operators for a given J ($J = 6$ for Tm^{3+}), and the axis of quantization has been chosen parallel to a crystal axis. The operators O_4^0 and O_4^4 are fourth order in the components of $\vec{J}$, while O_6^0 and O_6^4 are sixth order in $\vec{J}$. Thus the crystal field Hamiltonian is completely determined by symmetry considerations except for the constants B_4 and B_6. Rather than deal with B_4 and B_6, it is often more convenient to treat two other parameters,[9] x and W. The ratio of fourth- to sixth-order anisotropy is given by x and the absolute scaling of the crystal field energy levels is given by W:

$$\frac{B_4}{B_6} = \frac{x}{1 - |x|} \frac{F(6)}{F(4)} \ , \quad (2)$$

$$B_4 F(4) = Wx \ . \quad (3)$$

Here $F(4)$ and $F(6)$ are numerical factors known for a given J.

The crystal field parameters used in Fig. 1 are $x = -1$ (fourth-order-only anisotropy) and $W = -0.887$ K. These were the parameters obtained in Ref. 1 by taking $x = -1$ and choosing W to match the low-field susceptibility (χ) in the limit of zero temperature. Actually, the susceptibility data by themselves are not sufficient to give a good determination of x and W independently, but only provide a linear relationship between them. This is because the $1/\chi$-vs-T behavior depends strongly only on the crystal field splitting from the Γ_1 ground state to the Γ_4 first excited state. The $1/\chi$ value as $T \to 0$ requires that this splitting be 26.6 K. This in turn implies the relationship

$$(22x - 8)W = 26.6 \text{ K} \ . \quad (4)$$

Now in Ref. 2 it was shown that the anisotropy of the high-field magnetization at 1.5 K served to restrict x to the predominantly fourth-order range from $x = -0.6$ to $x = -1$, where for any x, W is de-

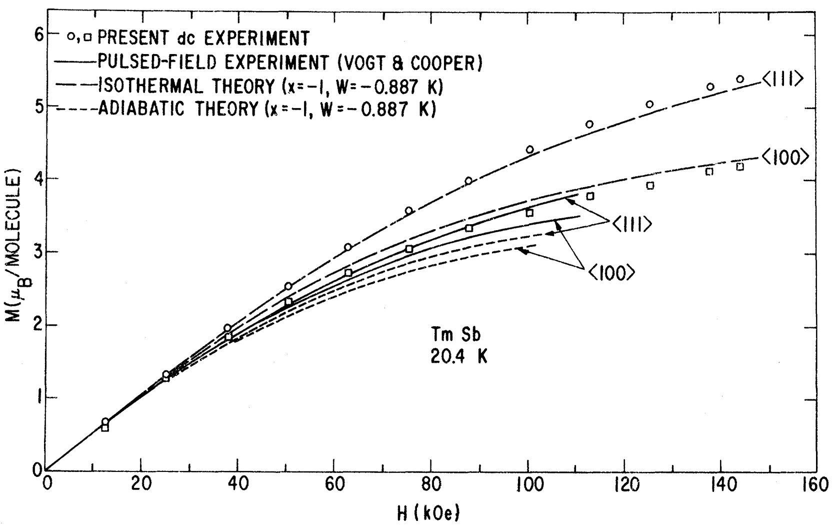

FIG. 1. Magnetization of TmSb at 20.4 K. The pulsed-field data of Fig. 2 of Vogt and Cooper (Ref. 1) are reproduced along with the isothermal crystal-field-only theoretical curves extended to higher field and the present dc data.

termined from Eq. (4). Figure 2 reproduces part of the results of Ref. 2. As shown in Fig. 2, for the range of x from -0.6 to -1, the variation of anisotropy of the 1.5-K magnetization as x varied [subject to Eq. (4)], was small—trying to select a particular x was unjustified with the existing experimental accuracy. On the other hand, as discussed thoroughly in connection with Fig. 4 in Ref. 2, one could use the 1.5-K anisotropic-magnetization data to eliminate x values outside the predominantly fourth-order range.

High-resolution magnetization measurements have been carried out between 4.2 and 1.5 K in order to search for any systematic deviation between the pulsed-field experiments and the various theoretical curves in Fig. 2. Magnetization data at 1.5 K and up to 52 kOe are in excellent agreement with the earlier pulsed-field data and the $x=-1$, $W=-0.887$ K curves. At 42 kOe and 1.5 K, $M[100]=3.18$ μ_B/molecule and $M[111]=3.96$ μ_B/molecule. (The pulsed-field data are $M[100]=3.14$ μ_B/molecule and $M[111]=3.91$ μ_B/molecule.) The change in M at 52 kOe is $\leq 0.5\%$ from 4.2 to 1.5 K. (The calculated change of M vs temperature is

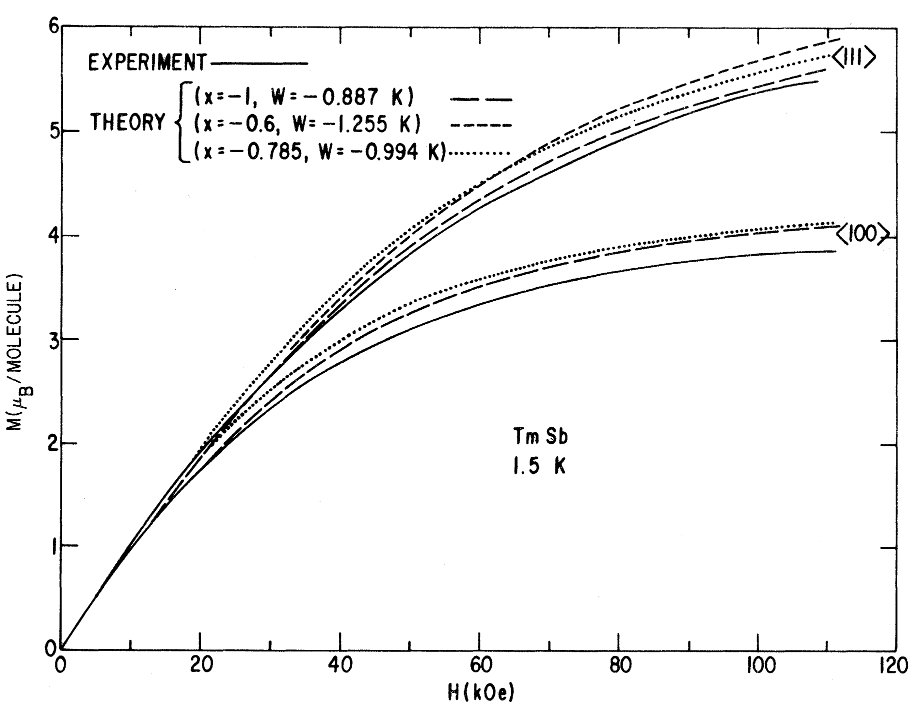

FIG. 2. Magnetization of TmSb at 1.5 K. The experimental results are the pulsed-field data of Vogt and Cooper (Ref. 1). The $x=-1$ and -0.6 crystal-field-only theoretical curves have been given previously by Cooper and Vogt (Ref. 2) and the $x=-0.785$ curves are for parameters from the neutron-scattering experiments of Birgeneau *et al.* (Ref. 3). (The ⟨100⟩ curve for $x=-0.6$ coincides with the $x=-1$ curve.)

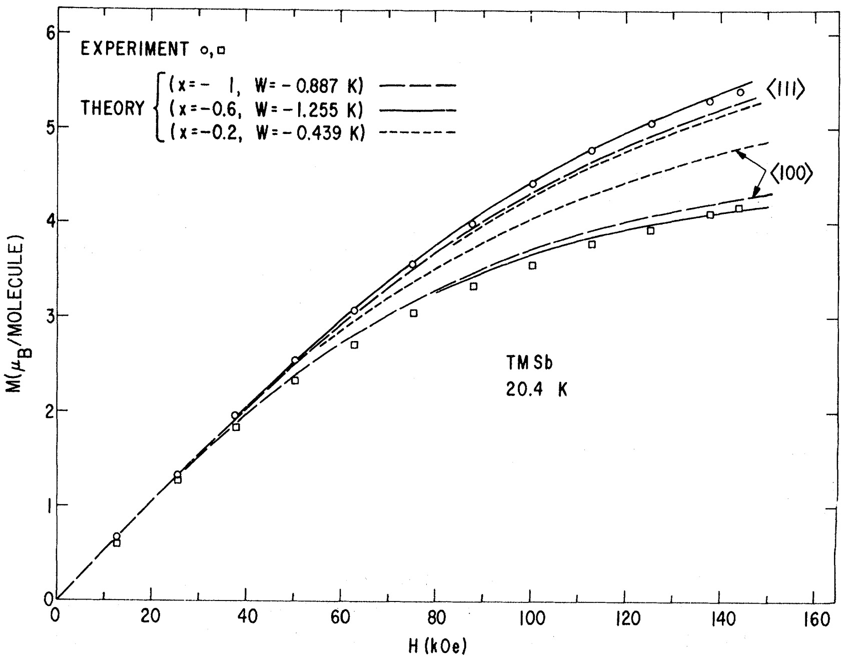

FIG. 3. Comparison of experimental results of dc magnetization of TmSb at 20.4 K with crystal-field-only isothermal theory for several values of x and W.

~ 0.1% for [111] and < 10^{-4} for [100] for the crystal field chosen above.) Furthermore, the field dependence of the magnetization is essentially the same for both 1.5 and 4.2 K for each orientation. For each orientation the susceptibility at low field ($H \leq 3$ kOe) is independent of temperature (within ± 2%) between 4.2 and 1.5 K and slightly (< 10%) lower than that in Ref. 1. [A corresponding larger splitting would be in order in Eq. (4); the calculated results would not be very different from those presented here.]

In Fig. 3, we compare the present dc experimental values of magnetization at 20.4 K to the crystal-field-only isothermal magnetization for $x = -0.6$ and -1, with W given by Eq. (4). This then shows the full range of crystal-field-only anisotropic-magnetization behavior at 20.4 K that is consistent with both the susceptibility as $T \to 0$ and the anisotropic magnetization at 1.5 K. The ⟨111⟩ dc data fall between the $x = -0.6$ and $x = -1$ curves, while the ⟨100⟩ data fall slightly below the $x = -0.6$ and $x = -1$ curves. Thus the 20.4-K dc data are completely consistent with the behavior expected for the range of x values selected by the 1.5-K data of Refs. 1 and 2. Moreover, this is distinctly different from the behavior expected for other x values (see the $x = -0.2$ curves in Fig. 3).

The possibility of using the present 20.4-K data to restrict x to some particular value or very narrow range of values within the present $x = -0.6$ to -1 range is attractive. It would be interesting to see the closeness of agreement with the values $x = -0.785$ and $W = -0.994$ K found in the neutron-

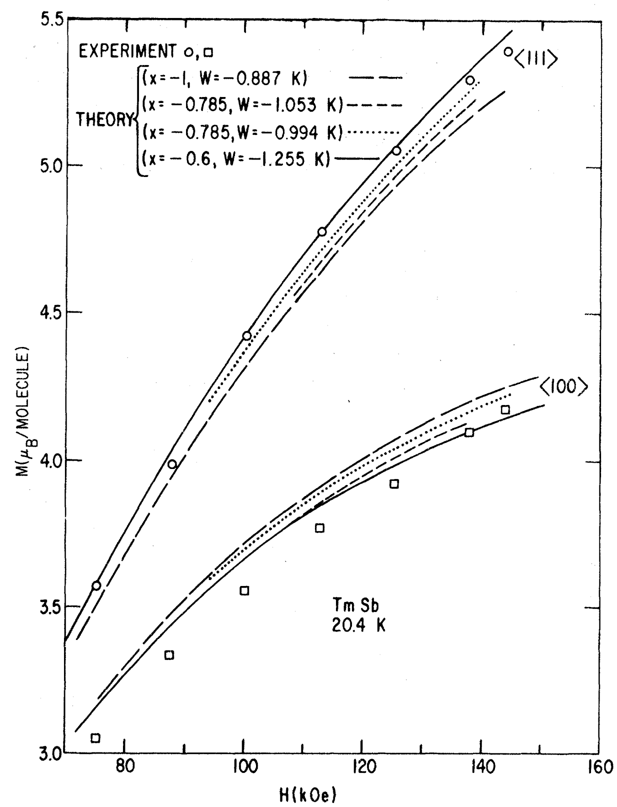

FIG. 4. Comparison of dc magnetization data of TmSb (above 70 kG) with crystal-field-only theory for several values of x and W including the $x = -0.785$ value obtained from the neutron-scattering experiments of Birgeneau *et al.* (Ref. 3).

scattering work of Birgeneau *et al.*[3] This x value is almost exactly in the middle of our allowed range, while the W value is about 6% smaller than the W given for that x by Eq. (4).

In Fig. 2, we have included the theoretical magnetization curves at 1.5 K for the x and W of Birgeneau *et al.*[3] There is no reason for choosing or rejecting their x and W compared to the $x=-0.6$ and $x=-1$ already discussed. Figure 4 includes the magnetization curves at 20.4 K for the Birgeneau *et al.*[3] x and W, a curve for $x=-0.785$, but $W=-1.053$ K as given for that x by Eq. (4), as well as the $x=-0.6$ and -1 curves already shown in Fig. 3. We note that the $x=-0.785$ curve with W chosen by Eq. (4) is much further from the $x=-1$ curve for $\langle 100\rangle$ than for $\langle 111\rangle$. The differences between the various curves in Fig. 4 are such as to preclude selecting a more specific x value within the accuracy of the present data.

While the accuracy of the magnetization experiments could be improved to choose an x more narrowly defined within the -0.6 to -1 range, the effort involved in doing this does not seem warranted since the neutron-scattering experiments[3] have selected a very definite x value. For that x value, $x=-0.785$, the low-temperature susceptibility behavior indicates a W value about 6% greater than that given by the neutron experiments. This difference in W values is within the combined experimental uncertainties.

We can then say that the susceptibility-vs-temperature and high-field anisotropic-magnetization data at 1.5 K (pulsed-field data of Refs. 1 and 2) and 20.4 K (present dc data) are all consistent with one another, indicating an x between -0.6 and -1 and a W given by Eq. (4) above, and that this result is consistent with the x and W found in the neutron-scattering experiments.

In conclusion, the present dc magnetization data at 20.4 K eliminate the apparent discrepancy with the crystal-field-only picture, and thus lend further support to the conclusion that TmSb acts as a model crystal-field-only paramagnet.

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Dynamic Structure of Vortices in Superconductors. III. Graphs for $B \approx H_{c2}$*

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Graphs are presented illustrating the backflow generated by vortices moving in a superconductor as driven by a transport current.

In a previous paper[1] we have studied the dynamic structure of vortices in superconductors carrying a transport current. It was found that backflow will generally accompany a moving vortex unless a new dynamic screening length ξ for the electric field is equal to the static screening length λ of the magnetic field. An explicit expression was given for the change in the local field $\delta\vec{B}_b$ generated by the backflow current $\vec{j}_b$ when the static applied field is near the upper critical field H_{c2}. The purpose of this addendum is to present graphs of this function so that the reader can more easily visualize the phenomenon. The set of graphs we present below is contours of equal magnitudes of $\delta\vec{B}_b(x,y)$