

higher-order coupling by Axe *et al.*²⁰ Neither of these treatments includes damping effects which are certainly extremely important in discussing the dynamical as opposed to the limiting static behavior of BaTiO₃, so that a quantitative comparison is not possible at the present time.

The present observations on tetragonal BaTiO₃ have raised further interesting questions concerning the cubic phase as well. For example, the extreme anisotropy of the soft phonon branch must persist into the cubic phase if this is the origin of the diffuse x-ray scattering. Due to larger sample size, it is also possible in the cubic phase to reach more

definite conclusions concerning the central question of the frequency distribution of the diffuse scattering, particularly with regard to the possibility of a large static component. These topics form the basis of a forthcoming paper.¹⁹

ACKNOWLEDGMENTS

We are grateful to V. Belruss for growing the excellent single crystal, and to M. DiDomenico for kindly carrying out the poling to produce the single-domain sample. We also wish to thank W. Cochran, M. DiDomenico, and Y. Yamada for many stimulating discussions.

†Work performed under the auspices of the U. S. Atomic Energy Commission and Advanced Research Project Agency.

¹W. Cochran, *Advan. Phys.* **9**, 387 (1960); **10**, 401 (1961).

²P. A. Fleury and J. M. Worlock, *Phys. Rev.* **174**, 613 (1968).

³R. A. Cowley, *Phys. Rev.* **134**, A981 (1964).

⁴Y. Yamada and G. Shirane, *J. Phys. Soc. Japan* **26**, 396 (1969).

⁵G. Shirane, R. Nathans, and V. J. Minkiewicz, *Phys. Rev.* **157**, 396 (1967).

⁶G. Shirane, J. D. Axe, J. Harada, and J. P. Remeika, *Phys. Rev. B* **2**, 155 (1970).

⁷A. S. Barker, Jr., *Phys. Rev.* **145**, 391 (1966).

⁸Y. Yamada, G. Shirane, and A. Linz, *Phys. Rev.* **177**, 848 (1969).

⁹J. Harada and G. Honjo, *J. Phys. Soc. Japan* **22**, 45 (1967); G. Honjo, S. Kodera, and N. Kitamura, *ibid.* **19**, 351 (1964).

¹⁰R. Comes, M. Lambert, and A. Guinier, *Solid State Commun.* **6**, 715 (1968); **7**, 305 (1969).

¹¹A. Hüller, *Z. Physik* **220**, 145 (1969).

¹²A. Linz, V. Belruss, and C. S. Naiman, *J. Electrochem. Soc.* **112**, 60C (1965).

¹³M. DiDomenico, Jr., S. H. Wemple, S. P. S. Porto, and R. P. Bauman, *Phys. Rev.* **174**, 522 (1968).

¹⁴S. H. Wemple, M. DiDomenico, Jr., and I. Camlibel, *J. Phys. Chem. Solids* **29**, 1797 (1968).

¹⁵A. Pinczuk, W. Taylor, E. Burstein, and I. Lefkowitz, *Solid State Commun.* **5**, 429 (1967).

¹⁶L. Rimai, J. L. Parsons, J. T. Hickmott, and T. Nakamura, *Phys. Rev.* **168**, 623 (1968).

¹⁷D. Berlincourt and H. Jaffe, *Phys. Rev.* **111**, 143 (1958).

¹⁸M. J. Cooper and R. Nathans, *Acta Cryst.* **23**, 357 (1967).

¹⁹J. Harada, J. D. Axe, and G. Shirane (unpublished).

²⁰J. D. Axe, J. Harada, and G. Shirane, *Phys. Rev. B* **1**, 1227 (1970).

²¹A. Pinczuk, E. Burstein, and S. Ushioda, *Solid State Commun.* **7**, 139 (1969).

²²A. R. Hutson and D. L. White, *J. Appl. Phys.* **33**, 40 (1962).

²³V. Dvorak, *Phys. Rev.* **167**, 525 (1968).

²⁴M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Clarendon, Oxford, England, 1962), Chap. V; P. B. Miller and J. D. Axe, *Phys. Rev.* **163**, 924 (1967).

²⁵W. Cochran, *Advan. Phys.* **18**, 157 (1969).

Spin-Wave Spectrum and Magnon Contribution to the Specific Heat of EuTe

Fred Masset and J. Callaway

Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803

(Received 13 July 1970)

Measurements of the specific heat of EuTe at low temperatures are analyzed in conjunction with results of antiferromagnetic resonance experiments to determine first- and second-neighbor exchange parameters $J_1 = (0.07 \pm 0.02)^\circ\text{K}$ and $J_2 = -(0.21 \pm 0.02)^\circ\text{K}$.

I. INTRODUCTION

Europium telluride is a fcc (rocksalt structure) antiferromagnetic insulator at low temperatures ($T_n = 9.81^\circ\text{K}$). The magnetic structure is of type II.¹ Antiferromagnetic resonance experiments have

shown that the spins are confined to the (111) planes with a weak preference for $(11\bar{2})$ -type directions.² Some lattice distortion occurs below T_n ; however, the amount has not been determined and is probably small. Exchange parameters have been estimated; J_1 (nearest neighbor) being approximately 0.03°K

and J_2 (second neighbor) being in the range from -0.12 to -0.17°K .³

The arguments which have lead to estimates of the exchange parameters appear to have been based mainly on molecular-field theory. The objective of the present paper is to obtain values for these parameters by a different method. We calculate a spin-wave spectrum for EuTe from a Hamiltonian containing unknown exchange constants. The low-temperature spin-wave specific heat of EuTe is calculated from the spectrum, and compared to the experimental specific-heat measurements of Passenheim and McCollum,⁴ supplemented by the antiferromagnetic resonance results of Battles and Everett.² A similar process has previously been employed to obtain exchange parameters in ferromagnetic EuS.⁵

II. SPIN-WAVE SPECTRUM

The calculation of the spin-wave spectrum follows that of Lines and Jones,⁶ except that we neglect the effects of possible lattice distortion in the antiferromagnetic state. Specifically, our Hamiltonian is

$$H = 2 \sum_{(i,j)} J_{ij} \vec{S}_i \cdot \vec{S}_j + \sum_i (D_1 S_{ix}^2 + D_2 S_{iy}^2). \quad (1)$$

The only nonzero values of the exchange parameters included here are those pertaining to the first neighbors (J_1) and second neighbors (J_2). The summation is over different pairs. The z axis, which is that of the spin alignment, is in a (111) plane, while the x and y axes are, respectively, perpendicular and parallel to a (111) plane.

The spin-wave spectrum for this Hamiltonian was calculated by Lines and Jones.⁶ Subsequently, a paper by Reissland and Begum⁷ appeared which treated a similar Hamiltonian, but obtained a different result. We have calculated the spin-wave energies using the above Hamiltonian by the method of Reissland and Begum, but have obtained a result in agreement with Lines and Jones, and therefore conclude that the spectrum of Reissland and Begum is erroneous.

The spin-wave spectrum contains two branches. The energies are as follows:

$$E_1(k) = 2S [(a_k - b_k - c) (a_k + b_k + c)]^{1/2}, \quad (2)$$

$$E_2(k) = 2S [(a_k - b_k + c) (a_k + b_k - c)]^{1/2}, \quad (3)$$

where

$$a_k = J_1 \gamma_1(k) + \frac{1}{2} Z J_2 + \frac{1}{2} (D_1 + D_2), \quad (4)$$

$$b_k = J_1 \gamma_2(k) + J_2 \gamma_3(k), \quad (5)$$

$$c = \frac{1}{2} (D_1 - D_2), \quad (6)$$

$$\gamma_1(k) = 2\{\cos[(k_x - k_y)a/2] + \cos[(k_y + k_z)a/2] + \cos[(k_x - k_z)a/2]\}, \quad (7)$$

$$\gamma_2(k) = 2\{\cos[(k_x + k_y)a/2] + \cos[(k_y + k_z)a/2] + \cos[(k_x + k_z)a/2]\}, \quad (8)$$

$$\gamma_3(k) = 2(\cos k_x a + \cos k_y a + \cos k_z a). \quad (9)$$

Here Z is the number of nearest neighbors (12), and a is the lattice constant. These results agree with those of Lines and Jones. It must be noted, however, that our definition of the Hamiltonian gives exchange parameters smaller than those in the paper of Lines and Jones by a factor of 2. (This usage is consistent with most previous definitions of the exchange parameters in rare-earth chalcogenides.)

Our expressions for the spin-wave energies also agree with those computed by Collins⁸ provided that the constants D_1 and D_2 of (1) are related to the phenomenological anisotropy constants K_1 , K_2 , K_3 of Keffer and O'Sullivan⁹ by

$$D_1 = -3K_1/2NS^2, \quad (10)$$

$$D_2 = -(9K_2 - K_3)/NS^2, \quad (11)$$

where N is the concentration of Eu ions. The anisotropy portion of the Hamiltonian used by Collins⁸ is the same as that employed by Battles and Everett² in their discussion of antiferromagnetic resonance. The spin-wave spectrum presented above may therefore be expected to give a correct description of the low-lying excited states of the coupled spins in EuTe.

III. SPECIFIC HEAT

We have used the spin-wave spectrum as given by (2) and (3) to calculate the low-temperature specific heat. Interactions between spin waves have been neglected. We must evaluate the expression

$$C = \frac{\Omega R}{(2\pi)^3} \sum_{i=1}^2 \int \left(\frac{E_i}{KT} \right)^2 \frac{e^{E_i/KT}}{(e^{E_i/KT} - 1)^2} d^3k. \quad (12)$$

In this integral, E_i is the energy of the i th branch given by (2) or (3), K is Boltzmann's constant, R is the gas constant, and Ω is the volume of the unit cell. The integral is to be performed over the magnetic Brillouin zone, which is different from the zone defined considering the chemical structure.

It seemed more convenient to us to use the ordinary Brillouin zone for the fcc lattice for the purposes of numerical integration even though the energy must be evaluated at more points. However, if this is done, proper account must be taken of the fact that the spin-wave energies do not have full cubic symmetry, but are instead invariant with respect to a group relevant to the magnetic structure. The symmetry properties of the functions $E_i(k)$ have been described by Daniel and Cracknell.¹⁰ A procedure for subdividing the Brillouin zone into small cubes for the purpose of performing the nu-

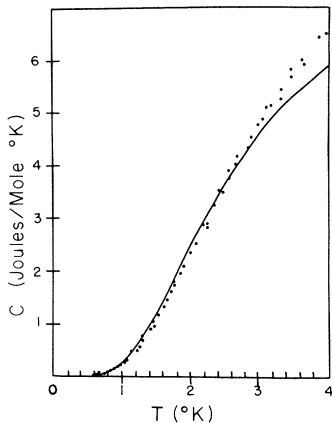


FIG. 1. Calculated specific heat for EuTe is shown (smooth curve) and compared with the experimental points of Passenheim and McCollum.

merical integration with weights assigned by reference to these symmetry considerations was developed and applied.

In order to assist in determining the parameters, we took values for E_1 and E_2 at the center of the zone from the measurement of Battles and Everett to be 2 and 68 GHz respectively. At $k=0$, Eqs. (2) and (3) become

$$E_1(0) = 2S\{D_2[D_1 + 12(J_1 + J_2)]\}^{1/2} \quad (13)$$

and

$$E_2(0) = 2S\{D_1[D_2 + 12(J_1 + J_2)]\}^{1/2}. \quad (14)$$

We were able to obtain a good fit to the specific-heat measurements for temperatures below 3°K ($\frac{1}{3}T_n$). The lattice contribution to the specific heat may safely be neglected in this temperature range. The exchange parameters found in this way are $J_1 = 0.07 \pm 0.02^\circ\text{K}$ and $J_2 = -0.21 \pm 0.02^\circ\text{K}$. The uncertainties indicate a range of values in which fits of roughly equal quality are obtained. The quantities D_1 and D_2 depend sensitively on $(J_1 + J_2)$ as indicated in Eqs. (13) and (14). For the case $J_1 + J_2 = -0.14^\circ\text{K}$, these quantities are determined to be $D_1 = -0.13^\circ\text{K}$, $D_2 = -0.00010^\circ\text{K}$. The negative signs are consistent with Eqs. (10) and (11). The calculated and measured specific heats are shown in Fig. 1. Inclusion of a temperature-dependent renormalization of the spin-wave energies would probably improve the fit around 4°K .

ACKNOWLEDGMENTS

We have benefitted from several discussions with Dr. G. E. Everett and Dr. John T. Marshall, and wish to thank Dr. Passenheim, Dr. McCollum, Dr. Battles, and Dr. Everett for furnishing their experimental results prior to publication.

¹G. Will, S. J. Pickart, H. A. Alperin, and R. Nathans, *J. Phys. Chem. Solids* **24**, 1769 (1963).

²J. W. Battles and G. E. Everett, *Phys. Rev. B* **1**, 3021 (1970).

³S. Methfessel and D. C. Mattis, in *Encyclopedia of Physics*, edited by S. Flügge (Springer, Berlin, 1968), Vol. 18, Part 1, p. 389.

⁴B. C. Passenheim and D. C. McCollum, *J. Phys. Chem. Solids* (to be published).

⁵B. C. Passenheim, D. C. McCollum, and J. Callaway, *Phys. Letters* **23**, 634 (1966).

⁶M. E. Lines and E. D. Jones, *Phys. Rev.* **139**, A1313

(1965).

⁷J. A. Reissland and N. A. Begum, *J. Phys. C* **2**, 874 (1969).

⁸M. F. Collins, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (The Institute of Physics and the Physical Society, London, 1965), p. 319.

⁹F. Keffer and W. O'Sullivan, *Phys. Rev.* **108**, 637 (1957).

¹⁰M. R. Daniel and A. P. Cracknell, *Phys. Rev.* **177**, 932 (1969).