

## Dislocation Scattering in Teflon at Low Temperatures\*

T. Scott

*Department of Physics and Astronomy, University of Wyoming, Laramie, Wyoming 82070*

and

M. Giles

*Department of Mathematics and Physics, Missouri Western College, St. Joseph, Missouri 64501*

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The thermal conductivity of a sample of Teflon was measured in the temperature range 0.17 to 4.0°K. A good fit to these data was obtained by using dislocation scattering of phonons and a Debye  $\omega^2$  frequency distribution.

Previous measurements<sup>1-3</sup> of the thermal conductivity of polymers have indicated that boundary scattering from crystallite boundaries and structure scattering in amorphous regions are the dominant low-temperature phonon scattering mechanisms in such semicrystalline polymers as polyethylene and nylon. Our measurements suggest that at temperatures between 0.17 and 4°K, the dislocation scattering of phonons plays a more significant role in determining the thermal conductivity of Teflon (polytetrafluoroethylene) than either of the above-mentioned scattering processes.

In the present work, measurements of the thermal conductivity of a commercial sample of Teflon were taken between 0.17 and 4°K. The sample dimensions and experimental procedure were similar to those described by Giles and Terry.<sup>3</sup> Between 0.17 and 1.2°K, a He<sup>3</sup>-He<sup>4</sup> dilution refrigerator was used to cool the sample. Metal-encased germanium resistance thermometers were used in this temperature range, their calibration being obtained from the magnetic susceptibility of cerium magnesium nitrate and the vapor pressure of He<sup>3</sup>. Above 1.2°K, a standard He<sup>4</sup> refrigerator was used,  $\frac{1}{8}$ -W, 200- $\Omega$  Allen-Bradley carbon resistors being used as thermometers. These thermometers were calibrated against the vapor pressure of He<sup>4</sup>. The results of the thermal conductivity measurements are shown in Fig. 1.

In addition, specific-heat measurements were taken on Teflon, using a standard calorimetric technique in temperature range 0.3 to 1.3°K. These measurements showed a Debye  $T^3$  temperature dependence over the entire temperature range, the data being represented by  $C = (350 \pm 50)T^3$  erg/°K g. The work of other investigators<sup>4</sup> indicates that this expression is probably valid up to 4°K, justifying our later use of a Debye  $\omega^2$  phonon frequency distribution.

Perhaps the most significant feature of the thermal conductivity data is their near-perfect  $T^2$  temperature dependence below 1.2°K. This behavior suggests the scattering of phonons by dislocations, and we utilize such a mechanism to obtain an expression for the low-temperature thermal conductivity of Teflon. According to Klemens<sup>5</sup> there are two contributions to the scattering of phonons from dislocations. First, the central core of the dislocation produces scattering with mean free path  $l_c = Aq^{-3}$ , and secondly the strain field surrounding the dislocation pro-

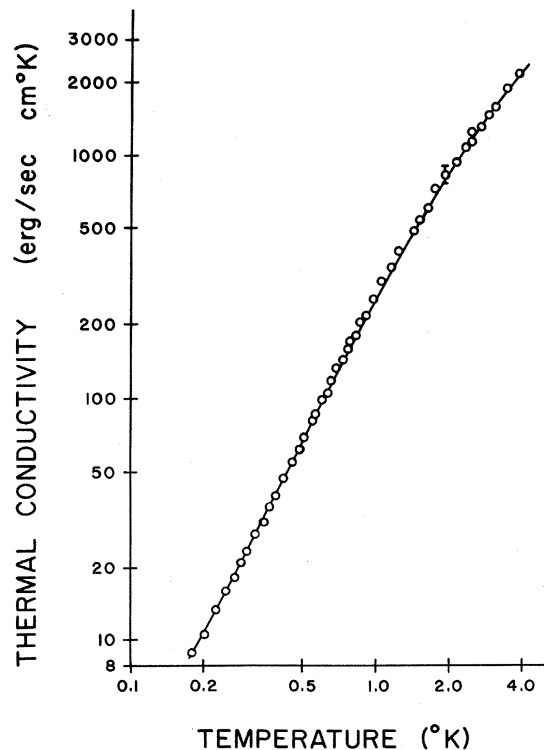


FIG. 1. Thermal conductivity of Teflon as a function of temperature. The solid line represents the fit obtained using Eq. (1).

duces scattering with mean free path  $l_s = Bq^{-1}$ , where  $A$  and  $B$  are constants and  $q$  is the phonon propagation vector. The second of these processes would dominate at long wavelengths, and hence would lead to the observed  $T^2$  low-temperature behavior of the data. If these mean free paths are utilized along with an  $\omega^2$  phonon frequency distribution, one obtains for the low-temperature thermal conductivity of Teflon due to the three phonon polarizations

$$\kappa = \frac{vkA}{2\pi^2} \int \frac{x^4 e^x}{(e^x - 1)^2} \left( x^3 + \frac{A}{B} \frac{v^2 \hbar^2}{k^2 T^2} x \right)^{-1} dx, \quad (1)$$

where  $v$  is the average phonon velocity for all three polarizations and  $k$  is Boltzmann's constant. The solid line in Fig. 1 represents a fit of this expression to the present data. The constant used in this fit were  $A = 9.1 \times 10^{15} \text{ cm}^{-2}$ ,  $B = 38$ , and  $v = 1.2 \times 10^5 \text{ cm/sec}$ , the velocity value being obtained from our specific-heat results.

The expressions for  $A$  and  $B$  given by Klemens<sup>5</sup> are  $A = 1.82N_d^{-1}a^{-4}$  and  $B = 30.3N_d^{-1}b^{-2}\gamma^{-2}$ , where  $\gamma$  is the Grüneisen parameter,  $b$  is the Burgers vector of the dislocation involved,  $N_d$  is the dislocation density, and  $a^3$  is the volume occupied by each vibrating unit in the sample. Solving these expressions simultaneously, using  $\gamma = 4$  as suggested by Barker,<sup>6</sup> and  $a = 5.2 \text{ \AA}$  as obtained from the density of Teflon, assuming  $\text{CF}_2$  groups to be the appropriate vibrating units, we obtain  $b = 4.4 \text{ \AA}$  and  $N_d = 2.5 \times 10^{13} \text{ dislocations/cm}^2$ .

The value obtained for  $b$  is certainly consistent with a Burgers vector of approximately 1 interatomic spacing. While the present data are probably insufficient to permit a detailed description of the types of dislocations involved, the rather large dislocation density value obtained might indicate that the dislocations are to be associated with the intercrystalline regions of the sample, in support of the crystal-defect model of polymer structure as described by Lindenmeyer.<sup>7</sup> Other possible dislocation types might be polymer-chain helix reversals in the crystalline regions as proposed by Clark,<sup>8</sup> or the rotational slippage

of one polymer-chain helix with respect to its neighbors as mentioned by Vainshtein.<sup>9</sup>

It is interesting to note the seeming absence of crystallite boundary scattering in the present data. A calculation of the mean free path for strain-field dislocation scattering using a dominant-phonon argument shows  $l_s$  equal to approximately  $2 \mu\text{m}$  at  $0.17^\circ\text{K}$ . Reese and Tucker<sup>1</sup> indicate that the average crystallite diameter in Teflon might be as large as  $1.5 \mu\text{m}$ . Hence it is possible that boundary scattering might manifest itself at temperatures lower than those covered in the present work.

While it is certainly possible that amorphous-structure scattering with mean free path proportional to  $q^{-2}$  could be playing a role in Teflon, various trial fits to the present thermal conductivity data have shown that structure scattering alone cannot account for the curvature of the data above  $1.2^\circ\text{K}$ , and that the inclusion of the more rapid  $q$  variation of central-core dislocation scattering is necessary to reproduce the data accurately.

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