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## Thermal Expansion of Tungsten at Low Temperatures\*

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Lattice parameters, thermal expansion coefficients, and Grüneisen parameters of tungsten are determined by an x-ray method in the temperature range of 180–40 K without the use of liquid gases. Lattice parameters are expressed as a function of temperature. Thermal-expansion coefficients decrease with temperature and show no anomaly in contrast to a hypothesis proposed by Featherston and Neighbours. Grüneisen parameters  $\gamma$  are decreasing with temperature in accordance with the theoretical predictions.

### I. INTRODUCTION

A detailed and accurate knowledge of the temperature dependence of thermal expansion of solids at low temperatures provides a better understanding of the equation of state of solids. In the Grüneisen<sup>1</sup> theory of thermal expansion, the Grüneisen parameter  $\gamma$  is defined by  $\gamma = 3\alpha VK_T/C_V$ , where  $\alpha$  is the coefficient of linear thermal expansion,  $K_T$  is the isothermal bulk modulus,  $C_V$  is the heat capacity at constant volume, and  $V$  is the molar volume. The Grüneisen parameter was generally regarded as a constant, independent of temperature and lattice vibrational frequency.

This simple picture turned out to be inadequate for real solids. Theoretical works of Barron<sup>2</sup> and Blackman<sup>3</sup> have shown that variations in the values of  $\gamma$  are to be expected at temperatures below  $0.2\theta$ , where  $\theta$  is the Debye characteristic temperature. With the intention to check these expectations, we have determined the lattice parameters, thermal-expansion coefficients, and Grüneisen parameters of tungsten below the  $\theta$  temperature (375 K)<sup>4</sup> within the temperature range 180–40 K.

### II. EXPERIMENTAL METHOD AND APPARATUS

Lattice-expansion measurements were made in a symmetrical back-reflection x-ray diffraction camera, cooling the powder sample with a Joule-Thompson pump without the use of liquid gases. The principles of the method and a detailed description of the apparatus will appear elsewhere.<sup>5</sup> The use of x-ray diffraction technique in the measurements of lattice expansion has unique advantages, as it determines the size of the crystal unit cell directly, needing no reference standards or bulk samples. This makes the method and the measurements quite absolute.

The tungsten powder of 99.9% purity was obtained from Koch Light Labs., Colnbrook, England. The average particle size of the powder was found to be  $10 \mu$ . No mechanical or heat treatments were necessary since the powder produced a very distinct diffraction pattern.

### III. MEASUREMENTS AND CALCULATIONS

Using cobalt radiation ( $K_{\alpha(1),\lambda} = 1.78892 \text{ \AA}$ ), this

tungsten powder gave sharp and uniform lines from the (222) plane at the Bragg angle of  $\sim 78^\circ$ . The standard deviation of the lattice parameters  $a$  (from 18 experimental measurements) was  $\pm 1.2 \times 10^{-5} \text{ \AA}$ . These calculations were programmed on an IBM 360/50 computer to obtain the best least-squares fit and to develop Eq. (1) relating the lattice parameter  $a(\text{\AA})$ , as a function of temperature  $T$  (K) in the temperature range 198–40 K:

$$a(\text{\AA}) = 3.16277 - 3.8045 \times 10^{-6}T + 7.07788 \times 10^{-8}T^2 - 1.02188 \times 10^{-10}T^3 \dots \quad (1)$$

The lattice parameter at 298 K was taken from Parrish,<sup>6</sup>  $a_{298 \text{ K}} = 3.16522 \text{ \AA}$ . The results of the lattice parameter calculations<sup>7</sup> (from three experimental runs) are shown in Fig. 1.

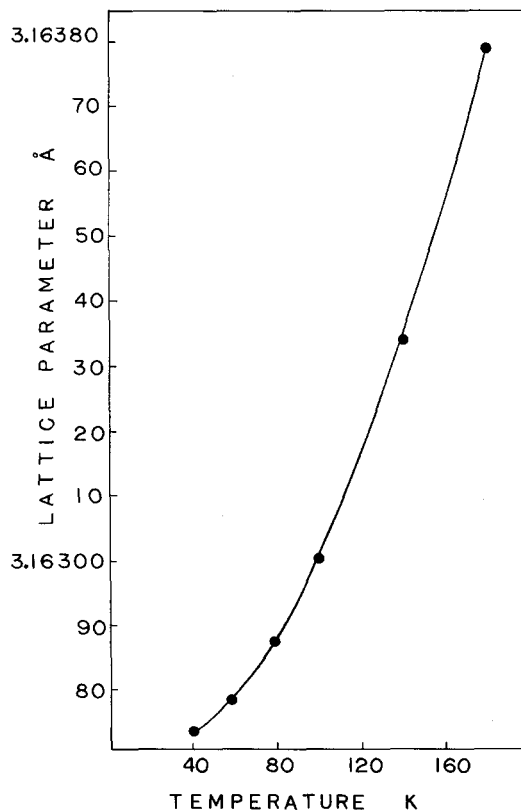


FIG. 1. Lattice parameters of tungsten as a function of temperature.

TABLE I. Lattice parameters, expansion coefficients, and Grüneisen parameters of tungsten at low temperature.

$T$ (K)	$a$ (Å)	$\alpha \times 10^6$ (K <sup>-1</sup> )	$\alpha \times 10^6$ (K <sup>-1</sup> )	$\gamma$
180	3.16379	3.7	3.89 <sup>a</sup>	1.47
140	3.16335	3.2	3.38 <sup>a</sup>	1.39
100	3.16300	2.3	2.70 <sup>a</sup>	1.26
80	3.16287	1.8	2.20 <sup>b</sup>	1.20
60	3.16277	1.1	1.50 <sup>b</sup>	1.14
40	3.16273	0.4	0.60 <sup>b</sup>	1.17

<sup>a</sup>See Ref. 8.<sup>b</sup>See Ref. 9.

Due to the lack of lattice parameter data for tungsten below 298 K, we could only test our experimental results by comparing our linear thermal-expansion coefficients with those of Nix and McNair.<sup>8</sup>

Even in this case, their measurements terminate at 120 K. Therefore, below 120 K we have compared our results with the extrapolated values of Corruccini and Gniewek,<sup>9</sup> Thermal-expansion coefficients  $\alpha = a^{-1}(\partial a / \partial T)_p$  (Table I) were obtained by graphical as well as numerical methods. The agreement between the present work and the published data cited above is poor.

To calculate the Grüneisen parameter  $\gamma$ , we have used our values of  $\alpha$ , the  $C_V$  (actually  $C_p$ , since  $C_V \sim C_p$  at low temperatures) data compiled by Corruccini and Gniewek,<sup>10</sup> and  $K_T$  data of Bolef and Klerk.<sup>11</sup> The Grüneisen parameters are tabulated in Table I together with the lattice parameters and thermal-expansion coefficients.

#### IV. DISCUSSION

The disagreement between our thermal-expansion data and the published data may be due to the fact that they have used bulk samples obtained by compacting and solid-state sintering of tungsten powder.<sup>12</sup> Such bulk samples never attain the theoretical density<sup>13</sup> in contrast to molten metals, for instance, iron.<sup>14</sup> In the present work, lattice expansion was

measured by the x-ray method which is independent of presence of microvoids.

In accordance with the theoretical predictions of Barron, our  $\gamma$  values decrease with temperature.

In their work on the bulk modulus, Featherston and Neighbours<sup>15</sup> show that bcc  $4d-5d$  metals like tantalum, tungsten, and molybdenum behave anomalously, the anomalous range of tungsten being 200–140 K. For an explanation, they propose antiferromagnetic ordering in this temperature range and further stipulate that tungsten should show an anomalous thermal-expansion behavior in the same temperature range. However, our results are quite contrary to this hypothesis; they show that the thermal-expansion coefficients  $\alpha$  of tungsten decrease with temperature without any anomaly.

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