

## Low-temperature thermal expansion of single- $\mathbf{Q}$ chromium and of dilute antiferromagnetic $\text{CrV}$ alloys

A. B. Kaiser,\* G. K. White, and E. Fawcett†

*Commonwealth Scientific and Industrial Research Organization, Division of Applied Physics,  
P.O. Box 218, Lindfield, Sydney 2070, Australia*

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Measurements of the uniaxial thermal expansion of a Cr single crystal having a single spin-density-wave vector  $\mathbf{Q}$  show that the large negative term linear in temperature in the expansivity at low temperatures has an anisotropy of about 50%. This linear term is found to *increase* in magnitude in multidomain antiferromagnetic  $\text{CrV}$  alloys as the impurity concentration increases, although the Néel temperature *decreases*. The corresponding positive magnetovolume measured by comparison with paramagnetic  $\text{Cr}_{95}\text{V}_5$  is found to scale with the square of the Néel temperature, as predicted if this magnetovolume arises from expansion of the lattice in the magnetic state to minimize the kinetic-energy cost of ordering.

### I. INTRODUCTION

The antiferromagnetic properties of chromium have been known to be strongly volume dependent since Bridgman<sup>1</sup> first observed the effect of pressure on the resistivity anomaly which we now know to be associated with the Néel transition. Following Overhauser's recognition<sup>2</sup> that chromium is an example of a spin-density-wave (SDW) system, Lomer<sup>3</sup> pointed out that the incommensurate wave vector  $\mathbf{Q}$  of the SDW corresponds to the nesting vector connecting similar roughly octahedrally shaped electron and hole sheets of the Fermi surface.

McWhan and Rice<sup>4</sup> ascribed the large volume dependence of the Néel temperature  $T_N$  to the effect of dilation,  $\omega = \Delta V/V$ , on the area of nesting between the electron and hole surfaces. This provides a mechanism relating  $d \ln T_N / d\omega$  to the negative thermal expansivity, linear in temperature, seen in chromium at low temperatures. When combined with the linear term in the electronic specific heat, the thermal expansion yields a large negative Grüneisen parameter<sup>5</sup> which, in the model of McWhan and Rice, is consistent with the strong positive volume dependence of  $T_N$  that they observed.

We note, however, that large negative terms proportional to temperature are observed at low temperatures in the thermal expansivity of many magnetic metals, including the antiferromagnet  $\alpha$ -Mn and Fe-Ni-Cr alloys.<sup>6</sup> The resultant large negative Grüneisen parameter seems to be a general characteristic of weak itinerant magnets, which, like the strong pressure dependence of the ordering temperature and the large forced magnetostriction,<sup>7</sup> is a manifestation of strong magnetoelastic coupling. Therefore, it seems unlikely, except by coincidence, that nesting of the Fermi surface, which is peculiar to chromium, should be the explanation of its large negative thermal expansion at low temperature.

Fletcher and Osborne<sup>8</sup> pointed out, furthermore, that a band-structure calculation for paramagnetic chromium shows the area of nesting to be essentially independent of volume strain, apart from the hydrostatic scaling of di-

mensions in reciprocal space. The band structures of the homologous metals Mo and W are similar to that of paramagnetic Cr, and, in particular, all three metals have similar nesting electron and hole surfaces. Thus experimental studies<sup>9,10</sup> of the strain dependence of the Fermi surfaces of both Mo and W, which showed that the nesting between the electron and hole octahedra changes little with volume strain, provide indirect experimental confirmation of the results of Fletcher and Osborne. Fermi-surface studies of antiferromagnetic chromium itself under pressure<sup>11</sup> show that the wave vector  $\mathbf{Q}$  of the SDW changes little with volume so that one would expect the nesting area also to change little.

On the other hand, Fletcher and Osborne<sup>8</sup> found in their calculations that tetragonal strain distorts the Fermi surface of Cr as one might expect and strongly affects the nesting. This is evident also in the experimental studies of Mo and W. This suggested that perhaps the negative thermal expansivity of a single SDW domain of Cr, i.e., a monocrystal having a single wave vector  $\mathbf{Q}$ , which had been assumed to be essentially isotropic, might in fact be highly anisotropic. The mechanism responsible for such spontaneous tetragonal distortion might be that the consequent reduction in magnetic order, and hence the increase in entropy, would minimize the free energy.

However, experimental studies<sup>12</sup> had hinted at only a weak anisotropy of the low-temperature thermal expansivity  $\alpha$  of single- $\mathbf{Q}$  Cr, with  $\alpha_{\parallel} = dc/dT$  being more negative than  $\alpha_{\perp} = da/dT$ , i.e.,  $|\alpha_{\parallel}| > |\alpha_{\perp}|$  with both negative ( $a$  and  $c$  are the lattice parameters perpendicular and parallel to  $\mathbf{Q}$ , respectively). We have measured a single- $\mathbf{Q}$  sample of Cr with considerably higher accuracy and we have confirmed this tentative result.

This work is described in Sec. II and the interpretation in Sec. III leads to an analysis (see Sec. IV) of measurements of the thermal expansion of Cr and  $\text{CrV}$  alloys reported by White, Roberts, and Fawcett.<sup>13</sup> The emphasis of the present paper is on the fact that both the negative term linear in temperature in the low-temperature thermal expansion of Cr and the antiferromagnetic  $\text{CrV}$  alloys,

and the strong strain dependence of their Néel temperatures, are essentially volume effects and not uniaxial strain effects. The implications of this finding for our understanding of the close parallelism between the effects of alloying and of hydrostatic pressure upon the Néel temperature will be discussed elsewhere.<sup>14</sup>

## II. EXPERIMENT: SINGLE-Q CHROMIUM

A sample of chromium, an approximately 5-mm cube, was spark cut and planed with edges parallel to the cube axes from a single crystal supplied by the Materials Preparation Center, Ames, Iowa. After lapping, the thermal expansion was measured in a three-terminal capacitance dilatometer<sup>15</sup> from 2 to 35 K. The sequence of measurements was as follows: (a) The cube was field-cooled through the Néel temperature,  $T_N=311$  K, in a magnetic field of 10 T parallel to the [001] axis to produce a single-Q domain. The thermal expansion was then measured parallel to this axis ( $\alpha_{||}$ ). (b) The cube was then rotated through 90° and the thermal expansion measured perpendicular to the Q direction ( $\alpha_{\perp}$ ). (c) The cube was then warmed above  $T_N$ , cooled in zero field, and the thermal expansion measured parallel to [001] and (d) normal to [001], i.e., along [010]. (e) The cube was again field cooled, parallel to the [010] axis, and remeasured parallel to Q. (f) The cube was rotated a few degrees and remeasured parallel to Q.

The small length of the sample relative to the 50-mm copper dilatometer magnifies the influence of hysteresis due to intersurface movements and of calibration errors including end effects. To allow for these latter effects we made a calibration run with a silicon rod 5.8 mm long and applied appropriate corrections to our chromium data with results shown in Fig. 1. Note that irreproducibility due to hysteresis near 30 K is significant [e.g., between (e) and (f)] compared with the thermal expansion and prevents us from obtaining meaningful data for the lattice contribution ( $T^3$  term) to  $\alpha$ . However, below 10 K, the "electronic" ( $T$  term) is large compared with the copper-dilatometer expansion and values for  $\alpha$  are reproducible and meaningful at the  $\pm 5\%$  level.

Data sets (a), (e), and (f) give values for  $\alpha_{||}=[-(5.2\pm 0.3)\times 10^{-9}]T$  K<sup>-1</sup> ( $T < 10$  K), while (b) gives  $\alpha_{\perp}=[-(3.3\pm 0.3)\times 10^{-9}]T$  K<sup>-1</sup>. This yields an average  $\alpha=(2\alpha_{\perp}+\alpha_{||})/3=-3.9\times 10^{-9}T$  K<sup>-1</sup>, which may be compared with data on a 41.6-mm polycrystalline rod<sup>13</sup> for which  $\alpha=-3.6\times 10^{-9}T$  K<sup>-1</sup>. Sets (c) and (d) on the 5-mm crystal in the multidomain state give, respectively,  $(3.0\pm 0.3)$  and  $(3.6\pm 0.3)\times 10^{-9}T$  K<sup>-1</sup>.

Thus we see that the anisotropy  $(\alpha_{||}-\alpha_{\perp})/\alpha$  is about 50%. Alternatively, if we identify the expansion  $\alpha_{MO}$  due to magnetic order as the difference between  $\alpha$  for Cr and that for a paramagnetic Cr<sub>95</sub>V<sub>5</sub> alloy (see Fig. 2), the anisotropy  $(\alpha_{||}-\alpha_{\perp})/\alpha_{MO}$  in the low-temperature expansion due to magnetic order is about 40%. It can be seen from Fig. 1 that the difference between  $\alpha_{||}$  and  $\alpha_{\perp}$  decreases as temperature increases. This is consistent with the measurements of Steinitz *et al.*<sup>12</sup> which show that the anisotropy  $|c-a|/a$  in the lattice parameter in single-Q Cr does not exceed  $25\times 10^{-6}$  at any temperature. This aniso-

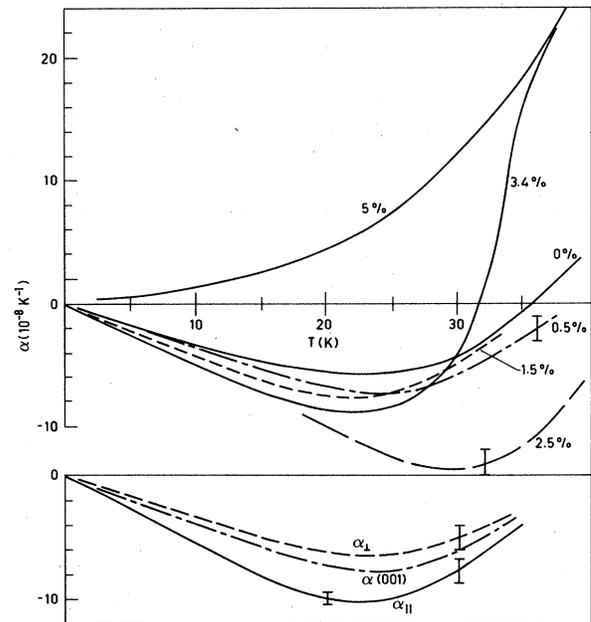


FIG. 1. Linear thermal expansivity  $\alpha$  of single-Q chromium, and of CrV alloys (from Ref. 14). Curves in the upper section are labeled with the concentration  $x$  of V in at.%, with  $x=0$  at.% corresponding to pure polycrystalline Cr. The lower section shows  $\alpha_{||}$  and  $\alpha_{\perp}$ , parallel and perpendicular, respectively, to the Q direction, and  $\alpha(001)$  parallel to the [001] direction in the multidomain state.

tropy is clearly very weak. This suggests that the nesting property of the Fermi surface of chromium, while being responsible for the incommensurate nature of the SDW, nevertheless is not the principal factor determining the thermal expansion. If it were, one might expect  $\alpha_{||}$  and  $\alpha_{\perp}$

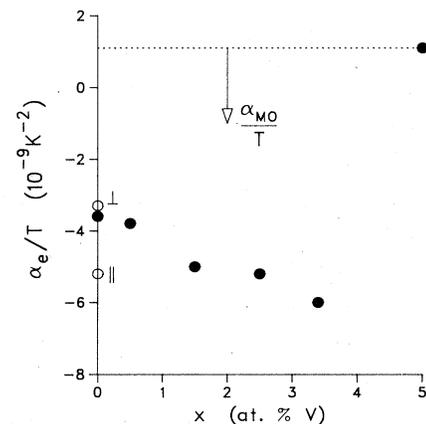


FIG. 2. Measured linear terms (solid dots) in the low-temperature expansion of Cr and dilute CrV alloys as a function of the concentration  $x$  of V (Ref. 13), showing the sudden change when the alloy remains paramagnetic down to zero temperature. The expansion term  $\alpha_{MO}$  due to SDW order is estimated by subtracting the expansion of the paramagnetic 5-at.% alloy. Also shown are the expansions for Cr parallel (||) and perpendicular ( $\perp$ ) to the Q direction (open dots).

to be opposite in sign, i.e., the anisotropy would be substantially greater than 100%. There must be another mechanism responsible for the overall negative low-temperature thermal expansion along all axes.

### III. THEORY

When we seek an explanation for this negative thermal expansion we first note that many magnetic metals have a positive magnetovolume, which gives rise to a negative thermal expansivity as the magnetic moment, and, therefore, the magnetovolume, decreases with increasing temperature.<sup>7,16</sup> The principal mechanism for positive magnetovolume in itinerant magnetic systems is as follows (e.g., Janak and Williams<sup>17</sup>). The spin polarization splits the electronic energy bands, resulting in a gain of exchange energy, which is counterbalanced by an increase in the kinetic energy of the electrons. This kinetic-energy increase may be reduced by increasing the density of electron states by expanding the lattice in all directions. Thus the metal establishes an effective repulsive force proportional to the magnetic energy density, i.e., to the square of the moment, which is often referred to as a positive magnetic pressure.

In a more general treatment, Holden, Heine, and Samson<sup>18</sup> derived an expression for the magnetovolume change in magnetic metals. For the case of chromium, their model leads<sup>19,20</sup> to a magnetovolume of a form similar to that of the model of Moriya and Usami<sup>21</sup> for weak ferromagnets (and that of the Stoner model<sup>7</sup>):

$$\omega_M(T) = \frac{C}{B} \langle M^2(T) \rangle. \quad (1)$$

Here  $\langle M^2(T) \rangle$  is the mean-square magnetic moment, the angular brackets signifying both a thermal and spatial average,  $C$  is the magnetoelastic coupling constant, and  $B$  is the bulk modulus.

Although the bulk modulus has a significant temperature dependence in chromium near the Néel temperature, we can take it to be constant at the low temperatures of interest here.<sup>22</sup> Thus the magnetic volume thermal expansivity is

$$\beta_M = \frac{d\omega_M}{dT} = \frac{C}{B} \frac{d}{dt} \langle M^2(T) \rangle. \quad (2)$$

The third law of thermodynamics requires  $\beta_M$  to approach zero as temperature  $T$  approaches zero. The experimental data,<sup>13</sup> as illustrated in Fig. 1, show that, in fact, the thermal expansivity of dilute CrV alloys, like that of Cr itself, is linear in temperature below about  $T \approx 10$  K; the linear term being negative in the antiferromagnetic alloys.

We may therefore assume in an expansion in ascending powers of  $T$  to describe  $\langle M^2(T) \rangle$  that the first term in  $T$  will be of order  $T^2$ ,

$$\lim_{T \rightarrow 0} \langle M^2(T) \rangle = \frac{1}{2} M_0^2 (1 - a_2 T^2). \quad (3)$$

We write  $\frac{1}{2} M_0^2$  without angular brackets to emphasize that the mean-square moment at absolute zero is obtained by taking a spatial average only and that we are considering here a sinusoidal incommensurate SDW of amplitude

$M_0$ . The negative coefficient  $-a_2$ , together with Eq. (2), is consistent with the observed negative thermal expansivity. Furthermore, the negative sign indicates a mean-square moment decreasing with increasing temperature, as we expect, for example, in the Fedders-Martin<sup>23</sup> nesting model of an itinerant antiferromagnet, in which the amplitude  $M_0$  decreases due to single-particle excitations across the energy gap. We note, however, that a quadratic decay of the mean-square moment may also result when spin-fluctuation effects are dominant as in weak itinerant ferromagnets.<sup>24</sup>

We can make further progress in studying this negative thermal expansion and gain a more quantitative understanding by considering the behavior of antiferromagnetic alloys of Cr with other transition metals. Koehler *et al.*<sup>25</sup> found that the amplitude  $M_0(x)$  of the SDW was approximately proportional to the Néel temperature  $T_N(x)$  when dilute concentrations  $x$  of various impurities (e.g., V, Mn, Mo, Ru, W, and Re) were added to Cr. Such behavior is not unexpected for an itinerant antiferromagnet like Cr, for which, at least in the Fedders-Martin model,<sup>23</sup> both  $M_0(x)$  and  $T_N(x)$  are proportional to the size of the energy gap induced by the SDW.

In light of Eq. (1) this result gives the ground-state magnetovolume  $\omega_0(x)$  at zero temperature (if we assume that  $C$  and  $B$  remain unchanged in the dilute alloys):

$$\frac{\omega_0(x)}{\omega_0} = \frac{M_0^2(x)}{M_0^2} = \frac{T_N^2(x)}{T_N^2}, \quad (4)$$

in terms of  $T_N(x)$  and the magnetovolume  $\omega_0$  and Néel temperature  $T_N$  of pure Cr. In the case of an impurity like V having fewer electrons per atom than Cr both  $T_N(x)$  and  $\omega_0(x)$  decrease with increasing  $x$ .

Furthermore, if the shape of the curve describing the decrease of  $\langle M^2(T, x) \rangle$  as a function of  $T/T_N(x)$  is the same for different alloys, as we might expect if the same model describes them all, and keeping in mind that the leading term in the expansion of  $\langle M^2(T, x) \rangle$  must be quadratic in  $T$ , we may write

$$\frac{\langle M^2(T, x) \rangle}{M_0^2(x)} = f \left[ \frac{T^2}{T_N^2(x)} \right], \quad (5)$$

where the universal function  $f$  is the same for different impurities. This means, when we retain only the leading term in  $T^2$  as in Eq. (3), that  $a_2(x)$  will be proportional to  $T_N^2(x)$ , so that we obtain

$$\lim_{T \rightarrow 0} \langle M^2(T, x) \rangle = \frac{1}{2} M_0^2(x) \left[ 1 - a \frac{T^2}{T_N^2(x)} \right]. \quad (6)$$

Equations (1) and (2) then give

$$\beta_M(x) = - \frac{aC}{B} \frac{M_0^2(x)}{T_N^2(x)} T, \quad (7)$$

and with Eq. (4) we obtain

$$\beta_M(x) = \beta_0 = - \frac{aC}{B} \frac{M_0^2}{T_N^2} T. \quad (8)$$

Thus the magnetic volume thermal expansivity of dilute

TABLE I. Low-temperature thermal expansivity (linear term  $\alpha_e$  and cubic term  $\alpha_l$ ) and "electronic" Grüneisen parameter  $\gamma_e$  of CrV alloys.  $\omega_0$  is the ground-state magnetovolume relative to the  $x=5$  at. % alloy, the values of the linear specific-heat coefficient  $C_e/T$  are from Heiniger *et al.* (Ref. 33) and Takeuchi *et al.* (Ref. 34).

$x$ (at. %)	0	0.5	1.5	2.5	3.4	5
$T_N$ (K)	311	263	150	105	28	0
$10^9 \frac{\alpha_e}{T}$ (K <sup>-2</sup> )	-3.6	-3.8	-5.0	-5.2	-6.0	1.1
$10^{12} \frac{\alpha_l}{T^3}$ (K <sup>-4</sup> )	2.4	1.9				2.7
$10^6 \omega_0(x)$	1430	920		165	10	
$10^8 \frac{\omega_0(x)}{T_N^2(x)}$ (K <sup>-2</sup> )	1.47	1.32		1.50	1.27	
$\frac{C_e}{T}$ (mJ mol <sup>-1</sup> K <sup>-1</sup> )	1.45	1.5	1.6	1.8	2.1	2.3
$\gamma_e$	-10.5	-11	-13	-12	-12	2.0

alloys of Cr having incommensurate SDW's like Cr itself is predicted to be roughly constant and equal to its value  $\beta_0$  in pure Cr. The essential point is that although the magnetovolume  $\omega_0(x)$  in the case of Cr +  $x$  at. % V alloys decreases as  $x$  increases, so does the temperature range in which this occurs, in accordance with Eq. (4), whose quadratic form yields a constant negative expansivity. The thermal expansivity at low temperatures should abruptly change sign when  $x$  is increased to the point where the alloy remains paramagnetic down to zero temperature.

In the Fedders-Martin<sup>23</sup> model of an itinerant antiferromagnet, comprising perfectly nesting electron and hole spheres, the temperature dependence of the mean-square moment  $\langle M^2(T) \rangle$  is given by the Bardeen-Cooper-Schrieffer function. In this case the coefficient  $a$  in Eq. (8) giving the temperature dependence for  $T \ll T_N$  is very small,  $a \ll 1$ . In the case of the Stoner model<sup>26</sup> of an itinerant ferromagnet with parabolic energy bands, Eq. (6) describes the temperature dependence over the whole range up to the Curie temperature with  $a=1$ .

In the case of chromium a satisfactory model for the temperature dependence of  $\langle M^2(T) \rangle$  has not been given even in the low-temperature limit. Since in the limit of zero temperature the Fedders-Martin model corresponds to an antiferromagnetic insulator, we might expect the value of  $a$  for chromium to be closer to that for the Stoner model, i.e.,  $a \leq 1$ . The model of Holden, Heine, and Samson<sup>18</sup> for the thermal expansion of magnetic transition metals gives<sup>20</sup> a value  $C/B=2.2\%/ \mu_B^2$  for the ratio of the magnetoelastic coupling constant of Cr to its bulk modulus,  $B=195$  GPa.<sup>22</sup> The model of Kübler<sup>27</sup> based on a calculation in the local-spin-density approximation gives a value  $C/B=0.84\%/ \mu_B^2$ , as discussed by Kaiser and Haines.<sup>20</sup> With the root-mean-square moment,  $M_0/\sqrt{2}=0.43\mu_B$ , the Néel temperature,  $T_N=311$  K, and  $a \leq 1$ , we obtain, for Cr, using Eq. (8),

$$\frac{\beta_0}{T} \lesssim -3.2 \times 10^{-8} \text{ to } -8.4 \times 10^{-8} \text{ K}^{-2}. \quad (9)$$

These limits are large enough to account for the observed

value,  $3\alpha_{MO}/T = -1.7 \times 10^{-8} \text{ K}^{-2}$ , of the volume thermal expansivity of pure Cr at low temperature measured relative to the paramagnetic alloy Cr<sub>95</sub>V<sub>5</sub>, as illustrated in Fig. 2 and tabulated in Table I [ $\alpha_{MO} = \alpha_e(0\%) - \alpha_e(5\%)$ ].

#### IV. EXPERIMENT: CrV ALLOYS

We have explored the predictions of Eq. (4), that  $\omega_0(x)/T_N^2(x)$  is constant and equal to  $\omega_c/T_N^2$  for pure Cr, and of Eq. (8), that  $\beta_M(x)$  is constant and equal to  $\beta_0$  for pure Cr, by using the experimental data of White, Roberts, and Fawcett<sup>13</sup> for the linear thermal expansion of Cr and CrV alloys. We can assume throughout that the expansion is isotropic and therefore  $\beta=3\alpha$ , since we use the data for pure polycrystalline Cr, and the CrV samples are all polycrystalline with the sole exception of the Cr + 1.5 at. % V, which would, however, be multidomain and therefore also isotropic. The results are given in Figs. 1 and 2 and in Table I, in which  $\alpha_e$  and  $\alpha_l$  are the low-temperature "electronic" and "lattice" thermal expansivities, linear and cubic in temperature, respectively.

We compare each alloy with paramagnetic Cr<sub>95</sub>V<sub>5</sub> in order to obtain the ground-state magnetovolume due to magnetic order,<sup>20</sup>

$$\omega_0(x) = 3 \int_{T_x}^0 [\alpha(x) - \alpha(P)] dT, \quad (10)$$

where  $\alpha(P)$  is the thermal expansivity of Cr<sub>95</sub>V<sub>5</sub> and the limit of integration  $T_x$  is the temperature where  $\alpha(x) = \alpha(P)$ .  $T_x$  may be as much as 100 K above the Néel temperature  $T_N(x)$ , as seen<sup>28</sup> for  $x=0.5$  at. % V. Above  $T_x$  the magnetic states of the CrV alloys are essentially identical<sup>20</sup> (but not necessarily nonmagnetic since Moriya-type spin fluctuations<sup>29</sup> may be present). Thus the Cr<sub>95</sub>V<sub>5</sub> expansion provides a reference for estimating the ground-state magnetovolumes  $\omega_0(x)$  relative to the  $x=5$  at. % alloy, i.e., due to magnetic order, even if large spin fluctuations are present at high temperatures in all the alloys. These ground-state magnetovolumes may be associated with the ordered ground-state moments  $M_0(x)$  provided any difference in zero-point fluctuations in the

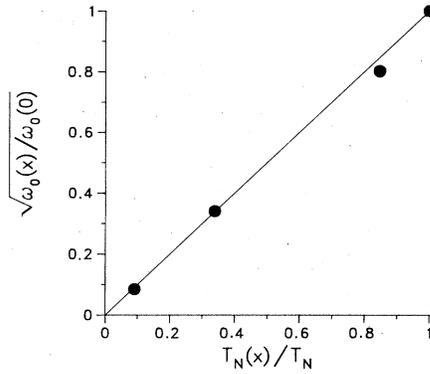


FIG. 3. Relation between ground-state magnetovolume  $\omega_0(x)$  and Néel temperature  $T_N(x)$  as impurity concentration  $x$  is varied: Data for ordered CrV alloys (dots) are compared to the prediction of Eq. (4) (solid line).

alloys is small.

We see from Table I and Fig. 3 that the ratio of magnetovolume  $\omega_0(x)$  to  $T_N^2(x)$  is roughly constant, in accordance with Eq. (4), for a range of variation of each over 2 orders of magnitude. In conjunction with the observation by Koehler *et al.*<sup>25</sup> that  $M_0(x)/T_N(x)$  is roughly constant, this provides experimental evidence for a relation

$$\omega_0(x) = \text{const} \times M_0^2(x) \quad (11)$$

which provides support for the theoretical result of Eq. (1).

The data in Table I confirm that the change from a large negative expansion term  $\alpha_e$  in Cr to a small positive expansion in paramagnetic Cr<sub>95</sub>V<sub>5</sub> is a sudden change near the critical concentration for SDW ordering (about 4 at. %V), rather than a gradual effect as  $T_N(x)$  decreases. Rather than staying constant for the ordered alloys, however, the linear expansion term  $\alpha_{MO}$  relative to Cr<sub>95</sub>V<sub>5</sub> ( $=\beta_{MO}/3$  for polycrystalline and/or multidomain samples, where  $\beta_{MO}$  is the change in volume expansion due to magnetic order) actually increases by about 50% between pure Cr and  $x=3.4$  at. % V (see Fig. 2). This increase indicates that the parameter  $a$  in Eq. (7) and (8) is not constant, but increases with V concentration. The reason for this is that in Cr there is a very strong decrease in magnetovolume<sup>20,28</sup> as the temperature increases through  $T_N$ , possibly associated with critical fluctuations above and below  $T_N$ , so that the magnetovolume contraction at low temperatures is relatively smaller. As the V concentration  $x$  increases in the CrV alloys, there is a decrease in the contraction occurring near  $T_N(x)$  relative to that at lower temperatures,<sup>14</sup> the total contraction in each case being proportional to  $T_N^2(x)$ . The change with  $x$  of the shape of the curve describing the temperature variation of  $\langle M^2(T, x) \rangle$  as a function of  $T/T_N(x)$  means that, in fact, no universal function exists to describe the behavior of all the CrV alloys as implied by Eq. (5).

The Grüneisen parameter  $\gamma_e$  defined in the usual way by the expression

$$\gamma_e = \frac{3\alpha_e B_s V_0}{C_e} \quad (12)$$

varies considerably less than  $\alpha_e$ ; in Eq. (12)  $C_e$  is the electronic term in the specific heat, linear in temperature at low temperatures, while  $V_0=7.2$  cm<sup>3</sup> is the molar volume and  $B_s=195$  BPa is the adiabatic bulk modulus for pure Cr at low temperatures.  $V_0$  and  $B_s$  (except near  $T_N$ ) do not change significantly with small additions of vanadium.

## V. DISCUSSION

Our measurements, together with those of Steinitz *et al.*,<sup>12</sup> indicate only a weak spontaneous tetragonal strain in Cr. Furthermore, other experimental observations militate strongly against the theory of Fletcher and Osborne<sup>8</sup> that the tetragonal strain dependence of the nesting area of the Fermi surface is the dominant factor determining the volume dependence of the Néel temperature  $T_N$ . Uniaxial stress measurements have failed to show any significant anisotropy of the stress dependence of  $T_N$  in pure Cr.<sup>30,10</sup> In the single-crystal measurements of Fawcett *et al.*<sup>10</sup> the values of the stress dependence of  $T_N$  for compressive stress  $\sigma$  along [110] was—to within experimental accuracy—the same as for stress along [100], with an average value of  $dT_N/d\sigma$  in agreement with that obtained from the pressure dependence if  $dT_N/d\sigma$  is assumed to be isotropic. It should be noted also that the isotropy of the stress dependence of  $T_N$  is consistent with the fact that the anisotropy of the elastic constants is observed<sup>31</sup> to be relatively weak in single-Q Cr. These results show that the proposed explanation<sup>8</sup> of Fletcher and Osborne for the large pressure dependence of  $T_N$  must be wrong.

Thus the present and previous results indicate that, although there is some weak anisotropy in expansion, the magnetovolume in Cr is mainly due to expansion to take advantage of the reduced kinetic-energy cost of forming the SDW. The net energy difference between the ordered and paramagnetic states in Cr is very small, as shown by the very small heat capacity associated with magnetic order.<sup>32</sup> It is not surprising, therefore, that the application of pressure (which increases the kinetic-energy cost of ordering) should have a considerable effect on the energy balance in Cr, leading to a strong pressure dependence of  $T_N$ . Our conclusions do not contradict the mechanism of McWhan and Rice,<sup>4</sup> in which the fraction of the Fermi surface exhibiting the SDW gap varies with volume, nor their relation between the large pressure dependence of  $T_N$  and low-temperature expansion, but suggest that the origin of each of these effects lies more in the strongly-volume-dependent energy balance in Cr than in changes in Fermi-surface—nesting topology.<sup>14</sup>

In conclusion, we summarize our experimental findings and their implications for understanding antiferromagnetism in chromium and dilute Cr alloys: (1) the anisotropy of the magnetic low-temperature thermal expansion in single-Q chromium is only weak and therefore Fermi-surface nesting appears to play a smaller part in determining the large negative Grüneisen parameter than previously suggested; (2) the zero-temperature magnetovolume scales with the square of the Néel temperature, which provides support for a general theory ascribing magnetovo-

lume to the expansion of the lattice to minimize the kinetic-energy cost of ordering the electron spins.

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\*Permanent address: Physics Department, Victoria University of Wellington, Private Bag, Wellington, New Zealand.

†Permanent address: Department of Physics, University of Toronto, Toronto, Canada M5S 1A7.

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