Thermal-expansion and elastic constants of dilute Cr-Al alloys

H. L. Alberts and J. A. J. Lourens
Department of Physics, Rand Afrikaans University, P.O. Box 524, Johannesburg 2000, Republic of South Africa
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The thermal-expansion and elastic constants of dilute Cr-Al alloys containing 0.67, 1.06, 1.70, 2.19, 2.74, and 3.22 at. % Al are reported in the temperature range $4<T<400$ K. Magnetic anomalies were found at T_N in both the thermal-expansion coefficients (α) and bulk moduli (B) of all the alloys. At the Néel temperature these anomalies display a λ-type behavior for $c<2$ at. % Al and a jumplike behavior for $c>2$ at. % Al. The magnetic contribution to B at 0 K reaches a large peak value of about 1.1×10^{11} N m^{-2} for alloys near 2 at. % Al. It is concluded that antiferromagnetism is not suppressed for $2<c<4$ at. % Al in contrast with suggestions based on previous resistivity and susceptibility measurements. The magnetovolume, $\Delta\omega$, of all the alloys as well as of pure chromium is found to vary as $\Delta\omega(T)=a+bT^2+cT^4$ at $T<T_N$. This type of relation is predicted by itinerant-electron theories which, however, fail to produce the correct temperature variation of the magnetic contribution (ΔB) to B. The measured values of $\Delta\omega$ and ΔB are used to calculate the concentration dependence of dT_N/dp. It decreases rapidly with Al concentration in the range $2<c<4$ at. % Al where the absence of resistivity anomalies at T_N previously precluded the direct determination of dT_N/dp.

I. INTRODUCTION

Magnetoelasticity in transition metals and their alloys has received much attention[1] during recent years. Of these materials, Cr and its alloys are of particular interest due to their unique type of itinerant-electron antiferromagnetism. In Cr the antiferromagnetic ordering is associated with transverse spin-density waves below the Néel temperature T_N and with longitudinal spin-density waves below the spin-flip temperature T_{sf}. The presence of the spin-density wave (SDW) state in Cr is distinctively manifested by anomalies that appear at T_N and at T_{sf} in the thermal-expansion coefficient[2] and elastic constants[3–5] of the material.

We[6] recently used this fact in a brief report on the anomalous behavior of the longitudinal sound velocity to elucidate the complex antiferromagnetic behavior of Cr-Al alloys. It was found that antiferromagnetism in Cr-Al alloys persists in the concentration range $2<c<4$ at. % Al, as distinct from suggestions based on previous resistivity measurements.[7,8] Contrary to the disappearance of the magnetic anomaly in the resistivity between 2 and 4 at. % Al, we found the magnetic contribution to the velocity of sound, extrapolated to 0 K, to reach a maximum value in this concentration range. This, together with the sharp increase[7] in $|dT_N/dp|$ just below 2 at. % Al, indicates the existence of correspondingly low magnetovolume effects in the range $2<c<4$ at. % Al.

In this paper we report a detailed study of the magnetovolume and magnetoelastic effects in a series of Cr-Al alloys in the concentration range $0<c<4$ at. % Al. The magnetoelastic effect is found to become large near 2.2 at. % Al, while the magnetovolume effect is smallest at that concentration. For $T<T_N$ the magnetovolume $\Delta\omega$ can be well fitted to an expression of the form

$$\Delta\omega=a+bT^2+cT^4\ ,$$

as predicted by existing itinerant-electron theories. By using these theories as well as Walker's[9] phenomenological theory, dT_N/dp is calculated from the observed magnetic contributions to the volume and bulk moduli for each alloy. This gives new information above 2 at. % Al, where dT_N/dp could not be obtained directly from resistivity measurements[7] due to the disappearance of the resistivity anomaly at T_N.

II. EXPERIMENTAL PROCEDURE

Dilute Cr-Al alloys containing 0.67, 1.06, 1.70, 2.19, 2.74, and 3.22 at. % Al were prepared as previously described.[7] Cylinders were spark-cut from the ingots, and each was prepared for sound-velocity measurements with a pair of flat and parallel faces by spark planing.

The experimental techniques employed to measure the bulk modulus (B), shear modulus (G), and thermal expansion of each alloy are described elsewhere.[10] The same samples were used for all measurements. In calculating B and G from the measured ultrasonic wave velocities, we used densities for the alloys as determined from their lattice parameters.[11] The experimental error in the absolute values of B and G is estimated to be about 1%, while changes in B and G of the order of 1 in 10^4 with temperature could be detected easily. The errors in $\Delta L/L$ and α are estimated to be $\pm5\%$ and $\pm10\%$. All measurements were made in the temperature range 4—400 K.

III. RESULTS

A. Thermal expansion

In order to determine the magnetovolume

$$\Delta\omega = 3\left[\left[\frac{\Delta L}{L}\right]_{\text{latt}} - \left[\frac{\Delta L}{L}\right]_{\text{meas}}\right]$$

at different temperatures, one needs to extrapolate the measured $\Delta L/L$-vs-T curves backwards from above T_N towards lower temperatures to obtain $(\Delta L/L)_{\text{latt}}$ at each temperature. In our previous study[10] on Cr-Co alloys this was done by using the Gruneisen relation. Recent work of Roberts *et al.*,[2] however, shows that magnetovolume effects continue to exist in Cr up to temperatures as high as 500 K, and that a more reliable temperature dependence of the magnetovolume could be obtained by comparing $\Delta L/L$ of Cr with that of Cr + 5 at. % V. Since the addition of more than 4 at. % V destroys the antiferromagnetism in Cr without altering the lattice spacing or density by more than 1–2 parts in a thousand, this alloy appears to be ideally suited for the determination of the magnetic part of $\Delta L/L$ as a function of temperature, assuming that the nonmagnetic states of the alloys are identical to that of pure Cr.

Thermal expansions were measured by using the strain-gauge technique in which two identical strain gauges, one bonded to the Cr-Al sample and the other to the Cr + 5 at. % V reference sample, were placed in opposite arms of a resistance bridge. In this way the temperature dependence of

$$\left[\frac{\Delta L}{L}\right]_{Cr_{95}V_5} - \left[\frac{\Delta L}{L}\right]_{\text{Cr-Al}} = \left[\frac{\Delta L}{L}\right]_{\text{nm Cr-Al}} - \left[\frac{\Delta L}{L}\right]_{\text{Cr-Al}}$$

(where subscript nm means nonmagnetic) could be directly measured. Differentiation of the measured $\Delta L/L$-vs-T curves with respect to T yields the temperature dependence of the magnetic contribution to the thermal-expansion coefficient

$$\Delta\alpha = \alpha_{\text{nm Cr-Al}} - \alpha_{\text{Cr-Al}} \cdot \tag{1}$$

The results of Roberts *et al.*[2] for $\alpha_{Cr_{95}V_5}$ were used to calculate $\alpha_{\text{Cr-Al}}$ from Eq. (1) for each alloy. Figure 1 shows the variation of α with temperature for each of the Cr-Al alloys.

The magnetovolume

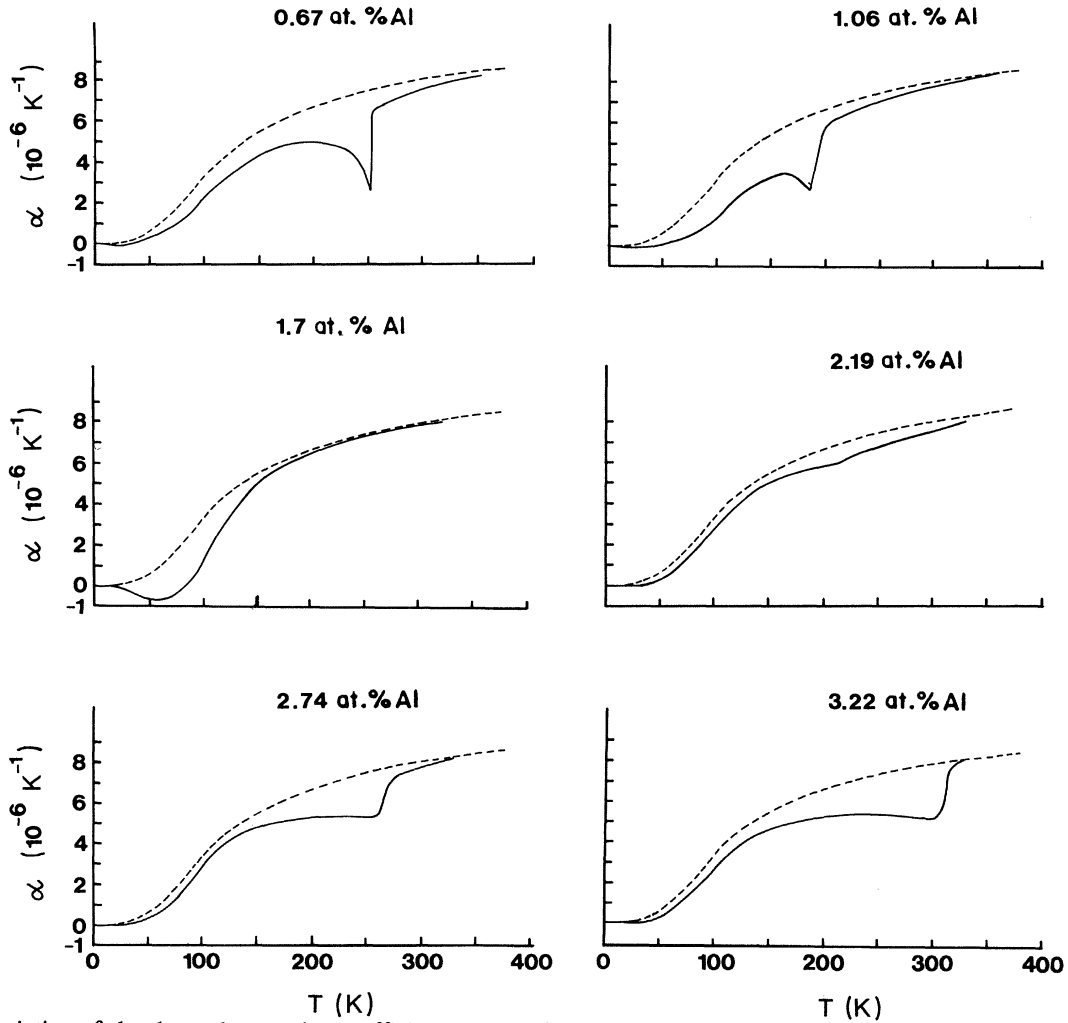

FIG. 1. Variation of the thermal-expansion coefficient (α) as a function of temperature. The solid lines represent our results for the Cr-Al alloys and the dotted lines those of Roberts *et al.* (Ref. 2) for $Cr_{95}V_5$. The measurements for the Cr-Al alloys were made relative to the thermal expansion of $Cr_{95}V_5$, which was used to represent the nonmagnetic state of the Cr-Al alloys.

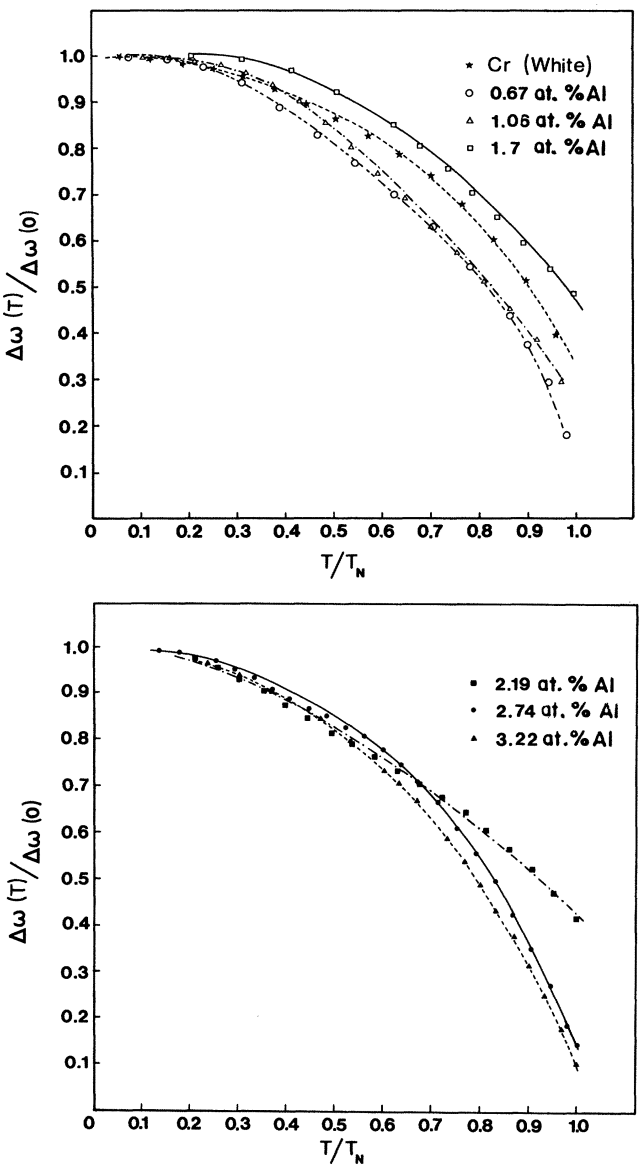

FIG. 2. Ratio of the magnetovolume at temperature T to that at 0 K as a function of T/T_N. The solid or dashed curves through the data points for each alloy are best fits of $\Delta\omega(T)/\Delta\omega(0)=A_0+A_1(T/T_N)^2+A_2(T/T_N)^4$.

$$\Delta\omega(T)=3\int_0^{T_f}\Delta\alpha\,dT-3\int_0^{T}\Delta\alpha\,dT\ ,$$

where T_f is the maximum temperature at which measurements were made, was determined from the curves in Fig. 1 by numerical integration. The variation of $\Delta\omega(T)/\Delta\omega(0)$ with T/T_N for each alloy is shown in Fig. 2. The variation for pure Cr was obtained from the results of Roberts *et al.*[2]

B. Elastic moduli

The temperature dependence of the bulk modulus B for each sample is shown in Fig. 3. All the samples display large magnetic effects in B when cooled from high temperatures, whereas the shear modulus G increases smoothly with decreasing temperature without any trace of such

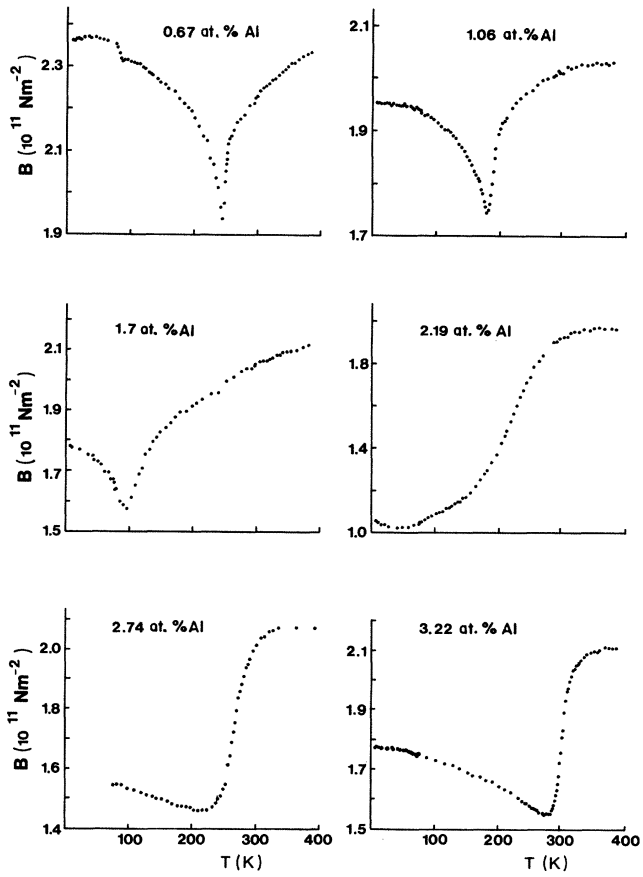

FIG. 3. Variation of the bulk modulus of each alloy as a function of temperature.

effects. In the case of Cr + 0.67 at. % Al, we observed two distinct anomalies in the B-vs-T curve. The one at the higher temperature was associated with the antiferromagnetic-paramagnetic transition and the smaller one at 88 K with the spin-flip transition, i.e., $T_{sf}=88$ K. The spin-flip transition was not seen in the other alloys down to 4 K.

C. Néel temperatures

As mentioned earlier, the electrical-resistivity—versus—temperature curves cannot be used to determine T_N in the intermediate concentration range $2<c<4$ at. % Al, where the anomaly in the resistivity disappears. However, since anomalies in the B-vs-T and α-vs-T curves are very clearly detected at all concentrations, T_N can be readily determined from these results in the intermediate concentration range. In the 0.67-, 1.06-, and 1.70-at. % Al samples, the λ-type point in the B-vs-T curves was used to define T_N, while the inflection point at the sharp rise of B vs T was used for the other samples. These Néel temperatures are shown in Fig. 4, together with the Néel temperatures that were determined from the α-vs-T curves. T_N in these curves was taken to occur at the temperature where

$$\Delta\alpha=\alpha_{Cr_{95}V_5}-\alpha_{Cr\text{-}Al}$$

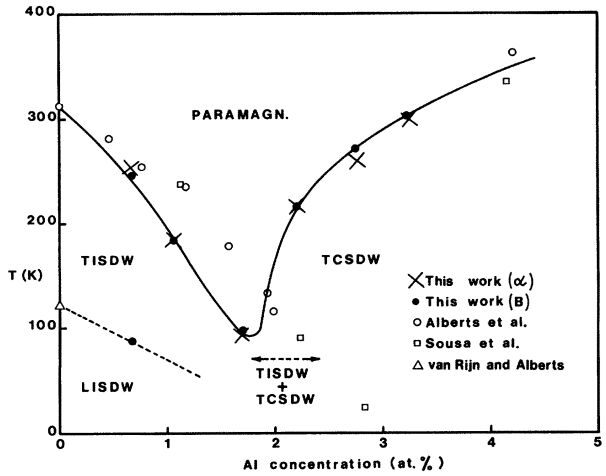

FIG. 4. Magnetic phase diagram of dilute Cr-Al alloys. Néel points are determined from the thermal-expansion coefficient (Fig. 1), the bulk modulus (Fig. 3), electrical resistivity, and susceptibility (Refs. 7 and 8) data. Spin-flip temperatures are determined from Fig. 3 for the 0.67-at.% Al sample and from Ref. 3 for pure Cr.

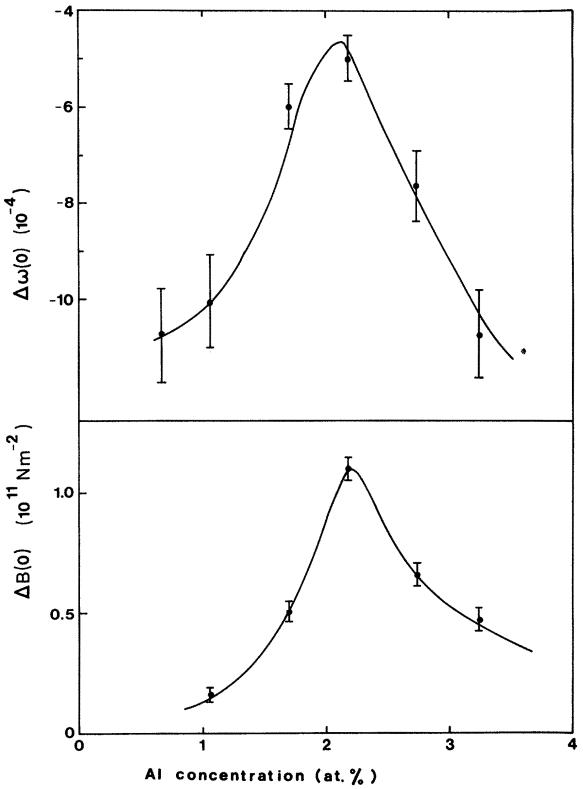

FIG. 5. Variation of the magnetic contribution to the bulk modulus (ΔB) and of the magnetovolume ($\Delta\omega$) at 0 K as function of Al concentration.

reaches its maximum value. The T_N values obtained from both types of curves agree very well (Table I). Also shown in Fig. 4 are T_N values determined from our resistivity measurements[7] as well as those of Sousa *et al.*[8]

IV. DISCUSSION

A. Magnetic phase diagram

Resistivity measurements[7] suggest that antiferromagnetism in Cr-Al alloys should disappear in the concentration range $2 < c < 4$ at.% Al where the resistivity (ρ) anomaly disappears. Further evidence of the disappearance of antiferromagnetism in this concentration region stemmed from measurements of $d\rho/dT$ and the susceptibility by Sousa *et al.*,[8] who concluded that antiferromagnetism is virtually suppressed at about 3 at.% Al. The results of the present investigation (Figs. 1 and 3) clearly show that the magnetic anomalies in the bulk modulus and the thermal-expansion coefficient do not disappear in the "critical" concentration region. In fact, the magnetic contribution to the bulk modulus at zero temperature as determined by back extrapolation from higher temperatures attains its maximum value in this region (Fig. 5). The anomaly in the thermal expansion also persists in this region, reaching an easily observable minimum around 2.19 at.% Al. These results therefore point to the continued existence of antiferromagnetism in the concentration range $2 < c < 4$ at.% Al.

Of all the alloys, only the 0.67-at.% Al one displayed a transition to a longitudinal incommensurate spin-density-wave state (LISDW) (cf. Fig. 3). Since only transverse states[12] appear at higher concentrations below the Néel temperature, a rough extrapolation of the dashed line in Fig. 4 shows that the LISDW state disappears somewhere around 2 at.% Al. Since the neutron-diffraction results of Kallel and De Bergevin[12] indicate a crossover from an

TABLE I. Néel temperatures, magnetovolume, and magnetoelastic data for Cr-Al alloys. A_0, A_1, and A_2 are the coefficients in the expansion $\Delta\omega(T)/\Delta\omega(0) = A_0 + A_1 T^2/T_N^2 + A_2 T^4/T_N^4$, $\Delta\omega(0)$ the magnetovolume at 0 K, and $\Delta B(0)$ the magnetic contribution to the bulk modulus at 0 K.

At.% Al	T_N from $\alpha(T)$ (K)	T_N from $B(T)$ (K)	A_0	A_1	A_2	$\Delta\omega(0)$ ($\times 10^{-4}$)	$\Delta B(0)$ (10^{11} N m^{-2})
0[a]	311	311	1.00	−0.41	−0.25	−15.0	0.20
0.67	255	246	1.00	−0.75	−0.05	−10.7	
1.06	185	184	1.03	−0.81	+0.03	−10.1	0.16
1.70	95	98	1.01	−0.43	−0.15	−6.0	0.50
2.19	215	216	1.00	−0.72	+0.16	−5.0	1.10
2.74	260	272	1.00	−0.48	−0.36	−7.6	0.66
3.20	299	303	1.00	−0.63	−0.26	−10.7	0.47

[a]Values determined for Cr from the work of Refs. 2 and 4.

incommensurate (ISDW) to a commensurate (CSDW) spin-density-wave state somewhere between 1 and 4 at. % Al, one may expect a mixture of ISDW and CSDW states between 1 and 4 at. % Al. Since T_N is usually higher for the CSDW state than for the ISDW state, such a mixture is probably responsible for the broad transitions seen both in B and α near 2 at. % Al. Our measurements therefore do not provide conclusive evidence of the existence of a mixture of states near 2 at. % Al as indicated in the magnetic phase diagram (Fig. 4).

The minimum in Fig. 4 occurs at $(c,T)=(\sim 2$ at. % Al, ~ 100 K), differing significantly from the result of Arajs *et al.*[13] (1.2 at. % Al, 200 K) based on resistivity work and also from Pop *et al.*,[14] who observed two minima in their susceptibility measurements.

B. Comparison with existing theories

The last few years have seen much theoretical activity on magnetoelastic effects in itinerant-electron metallic materials (see Ref. 1 and references cited therein). In these studies the emphasis was mainly concentrated on itinerant ferromagnetic metals and alloys, and although they have succeeded in explaining many of the observed magnetoelastic properties of these materials, controversies still exist about the relative importance of the Stoner single-particle excitations and the spin fluctuations to their magnetoelastic behavior. As was pointed out by Wohlfarth,[15] magnetovolume effects seem to be useful in assessing this relative importance.

Theoretical work on itinerant-electron antiferromagnets such as Cr is scarce. Recently Shimizu *et al.*[16] and Yamada[17] discussed magnetoelastic effects in itinerant-electron antiferromagnets. Shimizu *et al.* calculated the contributions of spin fluctuations to the magnetovolume in paramagnetic Cr, while Yamada studied the anomalous temperature dependence of the elastic constant c_{11} in antiferromagnetic metals. Some authors[18–20] have suggested that the itinerant-electron theories for ferromagnets should also be valid for itinerant-electron antiferromagnets. For the antiferromagnetic case one only needs to replace the ferromagnetic magnetization with the sublattice magnetization. We therefore discuss our results in terms of the existing itinerant electron theories, for both ferromagnets and antiferromagnets, as well as in terms of Walker's[9] phenomenological theory of the SDW state in Cr.

In the framework of the Stoner-Wohlfarth theory (SW theory) of itinerant-electron magnetism the magnetic anomaly in the spontaneous volume magnetostriction is given by[18]

$$\Delta\omega(T)=\left[\frac{4\phi_m}{B}\right]\frac{\partial \ln T_N}{\partial\omega}\left[1-\frac{T^2}{T_N^2}\right]$$
$$=CM^2(0)\left[1-\frac{T^2}{T_N^2}\right], \tag{2}$$

and the magnetic anomaly in the bulk modulus for the case of Cr and Cr alloys is given approximately by[10,18]

$$\Delta B=-4\phi_m\left[\frac{\partial \ln T_N}{\partial\omega}\right]^2\left[3-\frac{T^2}{T_N^2}\right]. \tag{3}$$

Here ϕ_m is an excitation potential and $M(0)$ the sublattice magnetization for the antiferromagnetic case at 0 K. The anomalies in ω and B, i.e., $\Delta\omega$ and ΔB, are defined by

$$\Delta X(T)=X^{\text{high}}(T)-X^{\text{low}}(T),$$

where $X^{\text{high}}(T)$ is the extrapolated value of X from above the transition temperature T_N and $X^{\text{low}}(T)$ is the observed value at any temperature below T_N.

In deducing Eqs. (2) and (3) it was assumed that the magnetization varies with temperature according to[21]

$$M^2(T)=M^2(0)\left[1-\frac{T^2}{T_N^2}+\cdots\right],$$

in which terms up to T^2 only were retained. In analyzing our magnetovolume results of Fig. 2, it was found that the data could be well fitted to an equation of the form

$$\frac{\Delta\omega(T)}{\Delta\omega(0)}=A_0+A_1\left[\frac{T}{T_N}\right]^2+A_2\left[\frac{T}{T_N}\right]^4.$$

The numerical values of A_0, A_1, and A_2 determined from these fits are given in Table I for each alloy. The best fits through the experimental points are shown in Fig. 2.

Since the experimental results point to the existence of a significant T^4 term, the sublattice magnetization is expanded as follows:

$$M^2(T)=M^2(0)\left[1-\alpha\left[\frac{T}{T_N}\right]^2+\beta\left[\frac{T}{T_N}\right]^4\right], \tag{4}$$

which, of course, gives the required temperature variation of $\Delta\omega(T)=CM^2(T)$. Another effect[22] which can also give rise to a fourth-degree term in $\Delta\omega(T)$ is through a temperature-dependent magnetoelastic coupling constant

$$C(T)=C(0)\left[1-\xi\left[\frac{T}{T_N}\right]^2\right]. \tag{5}$$

By using Eqs. (4) and (5), the expression for the magnetovolume reduces to

$$\Delta\omega(T)=C(0)M^2(0)\left[1-(\xi+\alpha)\left[\frac{T}{T_N}\right]^2+(\beta+\alpha\xi)\left[\frac{T}{T_N}\right]^4\right]. \tag{6}$$

With $\alpha=1$ and $\beta=0$, the sum of the coefficients of the second- and fourth-degree terms in Eq. (6) equals -1, whereas experimentally A_1+A_2 tends to a maximum value of ~ -0.5 around 2 at. % Al, diminishing to ~ -0.9 at 3.22 at. % Al. Thus the temperature variation of $\Delta\omega(T)$ cannot be explained by the second effect [Eq. (5)] alone. Both the above effects as well as the inclusion of the effects of spin fluctuations are probably necessary for such an explanation.

We briefly discuss the relevance of spin fluctuations to our work. Evidence favoring the existence of fluctuating moments in Cr above T_N was found by Fincher and

Shirane[23] using inelastic neutron scattering techniques. With the aid of Eqs. (4) and (5), the spin-fluctuation theory of Moriya and Usami[24] produces the same temperature dependence of $\Delta\omega$ as the SW theory below the critical temperature T_c. However, for $T \geq T_c$, $\Delta\omega_{SW}=0$ while the spin-fluctuation theory predicts $\Delta\omega \neq 0$ and $\alpha_{mag}=$constant for $T \gg T_c$. The thermal-expansion coefficient of Cr-Al alloys (Fig. 1) and of pure Cr indeed contains a magnetic contribution above the magnetic transition point, but since T_N cannot uniquely be determined from our $\alpha(T)$ and $B(T)$ curves, we were not able to extract quantitatively the spin-fluctuation contribution from the results in Fig. 1.

Although the above-mentioned theories are successful in predicting the observed temperature dependence of the magnetovolume effect, one has problems in applying it to the behavior of the bulk moduli of the Cr alloys. Equation (3) gives the correct order of magnitude for ΔB at low temperatures,[10] but fails to predict its observed temperature behavior. This is partly due to an additional stress-induced contribution which is estimated[10] to be of the same order of magnitude as that calculated from Eq. (3). The stress-induced contribution is smallest at 0 K and its magnitude increases with increasing temperature.[25]

From Eqs. (2) and (3) it follows that at 0 K

$$\Delta B(0) = -3B \left[\frac{\partial \ln T_N}{\partial \omega} \right] \Delta\omega(0) , \tag{7}$$

which may be used to calculate dT_N/dp for each alloy from the observed $\Delta B(0)$ and $\Delta\omega(0)$ values. $\Delta B(0)$ was determined as before.[10] The observed variation of $\Delta B(0)$ and $\Delta\omega(0)$ with Al content is shown in Fig. 5 and the values for each alloy in Table I. The experimental value of $|\Delta B(0)|$ reaches a maximum value at 2.2 at.% Al, while $|\Delta\omega(0)|$ reaches a minimum value there. The $|dT_N/dp|$ values calculated from the above equation are shown in Fig. 6 together with values that we determined directly from electrical resistivity measurements[7] in the range $0 < c < 2$ at.% Al. For concentrations in this range these values of $|dT_N/dp|$ correspond fairly well. The presence of a stress-induced term at 0 K would result in smaller values of $|dT_N/dp|$. The fair agreement between the calculated and measured $|dT_N/dp|$ for $0 < c < 2$ at.% Al is probably indicative of a negligibly small stress-induced contribution at 0 K.

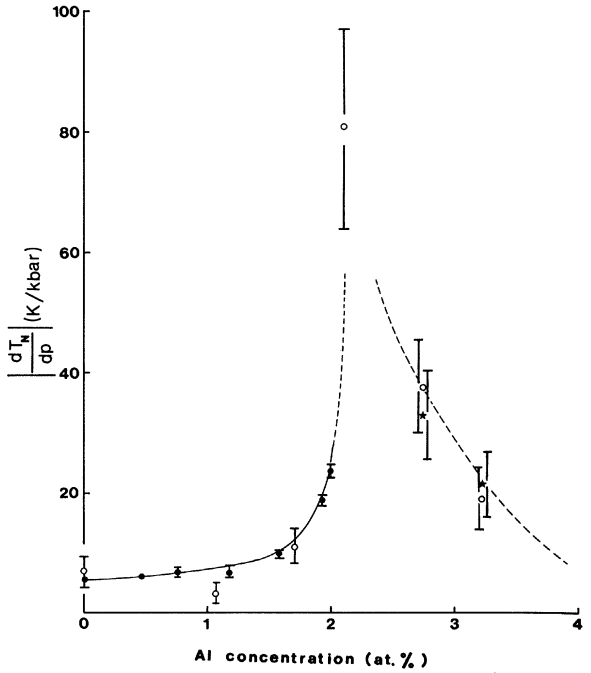

FIG. 6. Variation of $|dT_N/dp|$ as function of Al concentration. Closed circles are the results of direct measurements (Ref. 7). Open circles and stars follow from Eqs. (7) and (8). For the 2.74- and 3.22-at.% Al samples, the error bars to the left are for the open circles and those to the right for the stars.

From Walker's[9] phenomenological theory of the SDW state of Cr follows an Ehrenfest-type relation[10]

$$\Delta\beta \frac{dT_N}{dp} = \Delta\kappa . \tag{8}$$

Here $\Delta\beta$ and $\Delta\kappa$ are, respectively, the discontinuities in the volume thermal-expansion coefficient and the compressibility at T_N. In deriving Eq. (8) Walker assumed the transition at T_N to be second order. The transition is, however, observed in pure strain-free Cr to be first order and is known[26] to change to a second-order transition upon alloying with more than about 2 at.% Al. This is also clearly displayed in the behavior of the B-vs-T and α-vs-T curves of Figs. 1 and 3. The low-concentration alloys display a λ-type behavior which gradually changes to a jumplike behavior at higher concentrations. We used Eq. (8) to calculate dT_N/dp from the experimental $\Delta\beta$ and $\Delta\kappa$ values for the samples in which both B and α show a jumplike behavior at T_N, i.e., for the 2.74- and 3.22-at.% Al samples. These results are also shown in Fig. 6. Owing to the disappearance of the resistivity anomaly between 2 and 4 at.% Al, we[7] were previously unable to measure dT_N/dp directly for $c > 2$ at.% Al. The results in Fig. 6 suggest, however, that $|dT_N/dp|$ reaches a maximum value at about 2 at.% Al, whereafter it decreases sharply with increasing concentration. A similar behavior, i.e., a maximum in $|dT_N/dp|$ vs c, was also found[27] for Cr-Co alloys.

Concerning the behavior of the elastic moduli, Yamada's[17] calculations of the elastic constant c_{11} in itinerant-electron antiferromagnets show that for bcc antiferromagnets it becomes discontinuous at T_N and decreases as the sample is cooled through T_N. The 2.74- and 3.22-at.% Al samples (Fig. 3) do in fact display such such a behavior. Unfortunately, as mentioned by Yamada, his present theory cannot be applied directly to make quantitative calculations in the case of Cr, because he used a single-band model to describe the nonmagnetic state of Cr. The absence of anomalies in the polycrystalline shear moduli for the Cr-Al alloys relates to Walker's theory[9] as was also the case for dilute Cr-Co alloys.

V. CONCLUSION

It was previously concluded from the disappearance of the resistivity anomaly that antiferromagnetism is suppressed in Cr-Al alloys for $2 < c < 4$ at.% Al. In the

present study, however, we found an anomaly near T_N in the bulk moduli (B) as well as in the thermal-expansion coefficients (α) for all Cr-Al alloys studied with $c<4$ at. % Al. The magnetic anomaly in B was found to reach a maximum near 2 at. % Al, while the anomaly in α, although easily observable, reaches a minimum value there. We conclude that antiferromagnetism is not suppressed for $2<c<4$ at. % Al. The observed temperature dependence of the magnetovolume agrees well with existing theories, although these theories fail to predict the observed temperature dependence of B. From the observed magnetovolume and magnetoelastic anomalies we also found that $|dT_N/dp|$ reaches a maximum value near 2 at. % Al and that it decreases sharply for $2<c<4$ at. Al. Owing to the disappearance of the resistivity anomaly, it was previously impossible to determine dT_N/dp directly in this concentration range.

ACKNOWLEDGMENTS

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