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Pressure dependence of the elastic constants of Cr + 0.3 at.% Ru near the commensurate–incommensurate spin density wave transition

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Abstract. The hydrostatic pressure dependences of the elastic stiffnesses c_{11} and $c_L = (c_{11}+c_{12}+2c_{44})/2$ for a Cr + 0.3 at.% Ru alloy have been determined in the pressure range up to 1.5 kbar and at several fixed temperatures between 248 K and 298 K. This temperature region spans the antiferromagnetic incommensurate (I)–commensurate (C) spin density wave (SDW) transition. At room temperature both c_{11} and c_L soften under pressure in a similar manner to that seen in the ferromagnetic Invar material $Fe_{72}Pt_{28}$. By contrast, for a Cr + 5 at.% V alloy, which remains paramagnetic at all temperatures, the elastic constants are found to increase slightly with pressure. Below room temperature, $(\partial c_{11}/\partial p)$ and $(\partial c_L/\partial p)$ for the Cr + 0.3 at.% Ru alloy exhibit rapid increases over a narrow pressure range together with hysteresis behaviour. Such features are characteristic of a first-order CSDW–ISDW transition at T_{IC}. The observations suggest that Cr + 0.3 at.% Ru is an antiferromagnetic Invar material.

1. Introduction

Many dilute Cr alloy systems exhibit a triple point in their magnetic phase diagrams at a concentration where the commensurate (C) spin density wave (SDW), the incommensurate (I) SDW and the paramagnetic (P) phases coexist. The CSDW–P or ISDW–P transitions at the Néel point (T_N) of these systems are usually characterized by large anomalies in the temperature dependences of their physical properties. Anomalies are found in the electrical resistivity (ρ), magnetic susceptibility, thermal expansion and elastic constants. To date, the anomalies at T_N, and their concentration and pressure dependences, have been extensively studied in several Cr alloy systems. In particular, the anomalies in the ρ–T curves at T_N for different applied pressures (p) were used to determine dT_N/dp. Resistivity measurements are very suitable for this purpose as it is a relatively easy parameter to measure as a function of both temperature and pressure. By contrast, studies of the ISDW–CSDW transition at the temperature T_{IC} are less extensive than at T_N. This is especially so for the pressure dependence of T_{IC}. One reason for this is that in most cases the resistivity shows no anomaly at T_{IC}. The exceptions to this are Cr–Ge [1], Cr–Ga [2] and Cr–Mo [3] alloys.

Recently [4,5], it was found that both polycrystalline and single-crystal elastic constants of dilute Cr–Ru alloys show well defined anomalies at T_{IC}. This system is presently the only Cr alloy system known for which all the single-crystal elastic constants were found to give well defined anomalies near T_{IC}. However, unpublished data by Boshoff *et al* [6] show that similar behaviour also occurs in Cr + 0.5 at.% Re. As the resistivity measurements show no anomalies near T_{IC} for Cr–Ru [7], measurements of the pressure dependence of the elastic constants for this system should be ideally suited to determine dT_{IC}/dp. Furthermore, such a study should give information on the pressure dependence of the magnetic contributions to the elastic constants in both the ISDW and CSDW phases. This is necessary for developing a fundamental microscropic theory for the magnetoelasticity of SDW systems. Such a theory is presently still lacking.

We report here an initial study of the pressure dependence of the elastic constants of a Cr + 0.3 at.% Ru single crystal at different constant temperatures near T_{IC}. This concentration is ideally suited for such a study, as T_{IC} at atmospheric pressure is around 255 K and is therefore readily accessible. The pressure dependence of the elastic constants of a Cr + 5 at.% V single crystal are also reported. This remains paramagnetic at all temperatures [8] and serves to describe the non-magnetic behaviour of the Cr + 0.3 at.% Ru sample.

As Cr + 0.3 at.% Ru shows antiferromagnetic Invar type properties [4], the measurements are complementary to a recent study by Mãnosa *et al* [9] on the pressure dependence of the elastic constants of ferromagnetic Invar alloys. In addition the results also allow the experimental determination of the sign of dT_{IC}/dp. This sign is expected [10] to be positive as both ΔB and $\Delta\beta$ (the magnetic contributions to the bulk modulus and coefficient of volume thermal expansion at T_{IC}) are of opposite sign. As far as we are aware, this is the first study of the pressure dependence of the elastic constants of SDW Cr alloy single crystals.

2. Experimental method

The Cr + 0.3 at.% Ru single crystal was grown by a floating-zone technique using RF heating. The nominal concentration is 0.3 at.% Ru. Electron microproble analysis at different positions on the crystal shows that the crystal is fairly homogeneous with an actual concentration of (0.28 ± 0.02) at.% Ru. The Cr + 5 at.% V crystal was the same as that used in a previous study [8]. Both crystals were prepared with parallel (100) and (110) faces.

Ultrasonic (10 MHz) pulse transit times were measured using the pulse–echo overlap technique which has a sensitivity to changes of 1 part in 10^6. The experimental details are described by Brassington and Saunders [11]. Transit times were measured as a function of hydrostatic pressure up to 1.5 kbar at different fixed temperatures in the range 248 K–298 K. The pressure dependence of the longitudinal wave velocities associated with $c_L = \frac{1}{2}(c_{11} + c_{12} + 2c_{44})$ and c_{11} for the two crystals is given here.

3. Results and discussion

From the data we calculate the pressure dependence of the relative change in natural velocity, $\Delta W/W_0$, defined [12] by $\Delta W/W_0 = W_p/W_0 - 1$, where W_0 is the velocity at $p = 0$ and W_p that at pressure p. This is shown in figure 1 for Cr + 5 at.% V

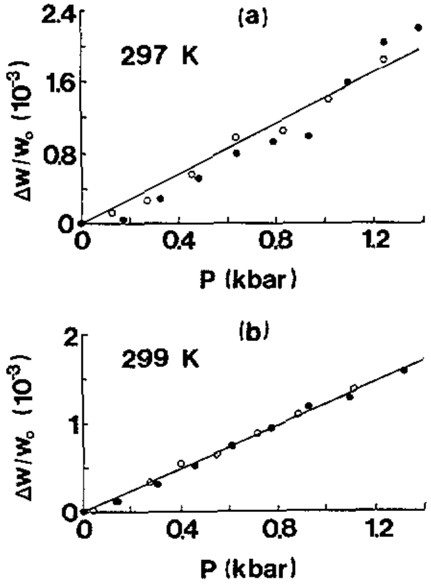

Figure 1. Relative change in natural velocity $\Delta W/W_0$ with pressure for longitudinal waves propagating along (a) [110] and (b) [100] in Cr + 5 at.% V. The modes are associated respectively with $c_L = \frac{1}{2}(c_{11} + c_{12} + 2c_{44})$ and c_{11}. The straight lines are least squares fits to the data. Increasing pressure: ●, decreasing pressure: ○.

at room temperature (around 297 K) for both modes of propagation. For Cr + 0.3 at.% Ru it is shown in figures 2 and 3 at several different fixed temperatures.

The pressure dependence of $\Delta W/W_0$, and therefore also of c_L and c_{11}, for Cr + 5 at.% V behaves normally in that both c_L and c_{11} increase linearly with applied pressure. These increases are of the order of 0.32 % kbar^{-1} and 0.24 % kbar^{-1}, respectively, for c_L and c_{11}. Normal behaviour of Cr + 5 at.% V is expected as the crystal is paramagnetic at all temperatures [8]. In the case of Cr + 0.3 at.% Ru, however, the behaviour of both longitudinal modes is unusual as shown in figures 2 and 3. At room temperature both c_{11} and c_L decrease linearly and quite rapidly with increasing pressure. The decreases are respectively of the order of 1.6% kbar^{-1} and 2.1% kbar^{-1} for c_L and c_{11}, which is some 5 to 10 times larger than the increases observed for Cr + 5 at.% V.

Below room temperature, both c_{11} and c_L for the Cr + 0.3 at.% Ru alloy show further anomalous behaviour, characterized by a sharp step in these quantities at a certain critical pressure. This is also shown in figures 2 and 3. As the temperature dependences of c_L and c_{11} for the Cr + 0.3 at.% Ru alloy show a step at T_{IC} = (257 ± 5) K for zero applied pressure [5], the steps in the curves of figures 2 and 3 are attributed to a transformation from the CSDW to the ISDW phase with increasing pressure. The results also show large hysteresis effects with increasing and decreasing pressure suggesting a first-order CSDW–ISDW transition. Recently, Boshoff *et al* [6], using neutron-diffraction measurements on a Cr + 0.3 at.% Ru crystal cut from the same boule as the present crystal, found hysteresis effects in the temperature dependence of the neutron-diffraction intensities near T_{IC}. This confirms the suggestion of a first-order CSDW–ISDW phase transition on application

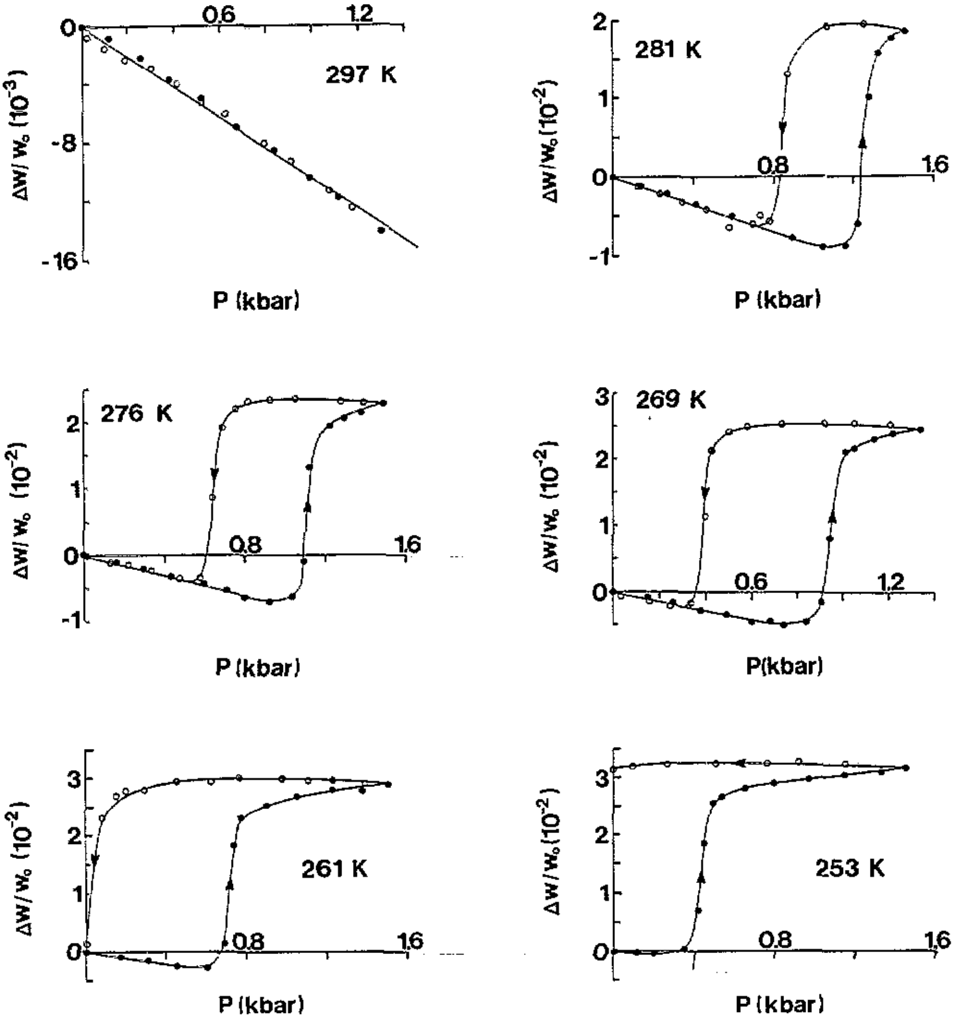

Figure 2. Relative change in natural velocity $\Delta W/W_0$ with pressure for longitudinal waves propagating along [100] at different constant temperatures in Cr + 0.3 at.% Ru. The mode is associated with c_{11}. Increasing pressure: ●, decreasing pressure: ○. The straight line is a least squares fit to the data. Note the different pressure scales in the figures.

of pressure. The steps in c_L and c_{11} at the critical pressure for the CSDW–ISDW transitions in figures 2 and 3 amount to a change of about 5 to 6% in both cases. This is similar to the steps observed [5,6] at T_{IC}. The widths of the hysteresis loops of figures 2 and 3 are fairly large, roughly 0.5 kbar, and show a tendency to increase slightly on decreasing the temperature towards T_{IC} ($p = 0$). At 253 K, the curves for both the c_L and c_{11} elastic modes show an additional important feature. Starting from $p = 0$, the crystal transforms from the CSDW to the ISDW phase at a critical pressure of between 0.3 and 0.4 kbar. When the pressure is reduced from above 1 kbar, the sample remains in the ISDW phase down to $p = 0$. This suggests that the crystal can be in either the CSDW or ISDW phase at temperatures near T_{IC}, depending

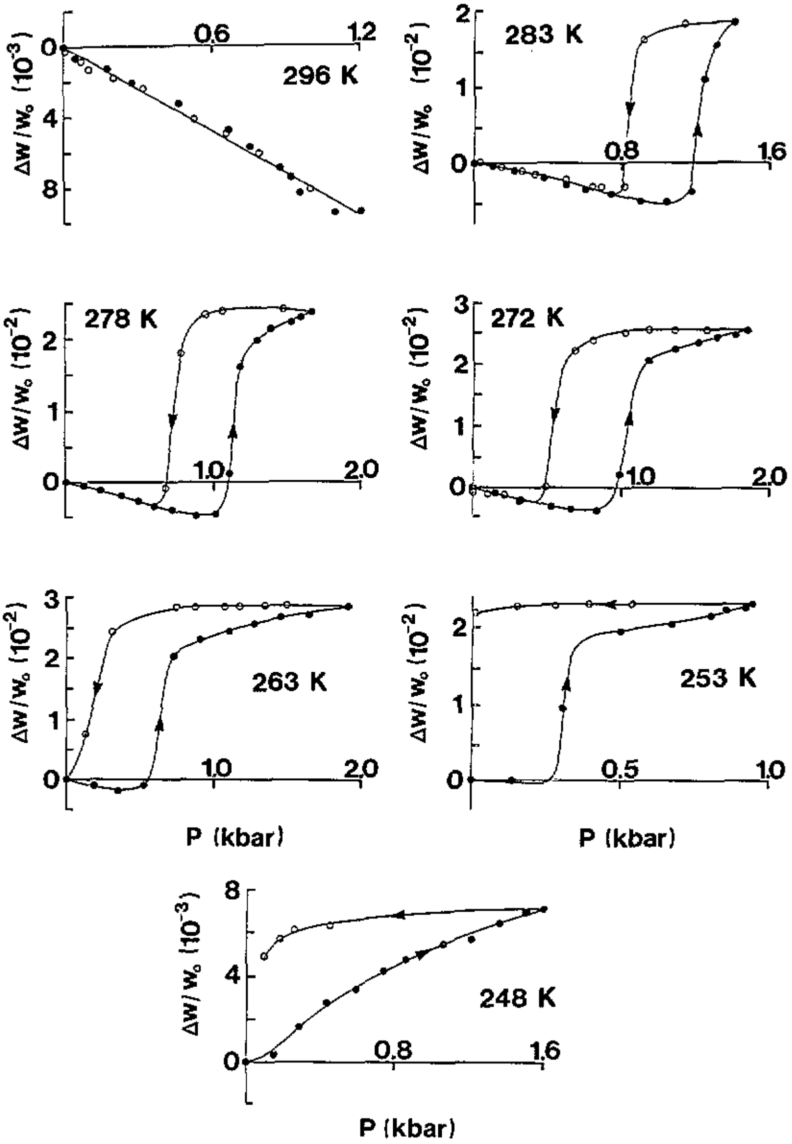

Figure 3. Relative change in natural velocity $\Delta W/W_0$ with pressure for longitudinal waves propagating along [110] at different constant temperatures in Cr + 0.3 at.% Ru. The mode is associated with c_L. Increasing pressure: ●, decreasing pressure: ○. The straight line is a least squares fit to the data. Note the different pressure scales in the figures.

on whether it had been previously pressurized or not. In the case of figure 3 at $T = 248$ K ($< T_{IC}$), the natural velocity, W, at $p = 0$ was observed to be almost the same (to within 0.3%) as that in the ISDW phase in which the sample remains at 253 K of figure 3 after decreasing the pressure to zero. On increasing the pressure at $T = 248$ K with the sample in the ISDW phase, the velocity increases continuously but still shows hysteresis effects on decreasing the pressure. The continuous increase of c_L with pressure in the ISDW phase is in agreement with the results of Katahara *et al* [13] on pure Cr. This is in the ISDW phase from 312 K to 0 K. Katahara *et al* [13] observed a continuous increase in both c_{11} and c_L with increasing pressure for the ISDW state of Cr up to about 1 kbar, after which a transformation to the paramagnetic state is induced by applied pressure. The origin of the hysteresis effects in the Cr + 0.3 at.% Ru crystal at $T = 248$ K ($< T_{IC}$), where the sample is supposed to be completely in the ISDW phase, is presently unknown and requires further investigation.

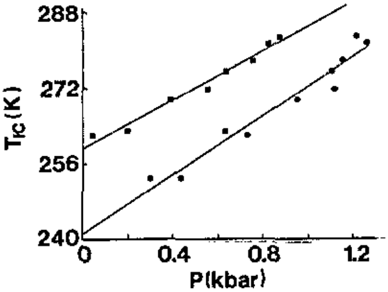

Figure 4. Pressure dependence of T_{IC} for Cr + 0.3 at.% Ru. The results for both the c_L and c_{11} modes are shown by the lower straight line (●) for increasing pressure and by the upper straight line (■) for decreasing pressure. The straight lines are least squares fits to the data. The experimental error in determining the pressures at T_{IC} is ± 0.05 kbar.

Figure 4 shows the variation of T_{IC} for Cr + 0.3 at.% Ru with pressure, for increasing and decreasing pressures, for the measurements of both the c_L and c_{11} modes. The pressure chosen for the ISDW–CSDW transition was taken as the midpoint of the sharp increase on the curves of figures 2 and 3. The error in this pressure is estimated to be ± 0.05 kbar. The average values of dT_{IC}/dp for increasing and decreasing pressures are $+(31 \pm 3)$ K kbar^{-1} and $+(26 \pm 2)$ K kbar^{-1}, respectively. dT_{IC}/dp is positive, as has also previously been found for Cr–Ge [1,14] and Cr–Mn [15]. It is, however, of the opposite sign to that found in Cr–Ga [16,17], Cr–Fe [15] and Cr–Co [15]. However, the mean absolute value obtained for dT_{IC}/dp for Cr + 0.3 at.% Ru is of the same order of magnitude as that for all of the alloys mentioned above. The positive sign of dT_{IC}/dp for Cr + 0.3 at.% Ru was recently predicted by Fawcett and Alberts [10] from the fact that the anomalies in the coefficient of thermal expansion and bulk modulus at T_{IC}, $\Delta\beta$ and ΔB, respectively, are of opposite signs. Fawcett and Alberts [10] estimated from $\Delta\beta$ and ΔB measurements, assuming the transition at T_{IC} to be continuous as was observed in polycrystalline Cr + 0.3 at.% Ru, that $d \ln T_{IC}/dw = -5$, where w is a volume strain. This gives $dT_{IC}/dp = +0.8$ K kbar^{-1} which is much smaller than the above-mentioned values of $+(31 \pm 3)$ K kbar^{-1} and $+(26 \pm 2)$ K kbar^{-1} that we obtained by direct measurement. This large difference between the calculated and directly observed values of dT_{IC}/dp can be attributed to the fact that the value of Fawcett and Alberts [10] was calculated from the Ehrenfest equation that is only valid for a second-order transition at T_{IC}. The present measurements on the single crystal show that there is in fact a first-order transition taking place. The sign of the calculated [10] and directly observed values of dT_{IC}/dp is nevertheless the same.

The non-magnetic components of c_{11} and c_L for Cr + 0.3 at.% Ru are taken [8] to be the same as that for the Cr + 5 at.% V alloy. The magnetic contribution to

c_{11} and c_L of Cr + 0.3 at.% Ru at any temperature below T_N is then given by

$$\Delta c(T) = c_{\mathrm{Cr-V}}(T) - c_{\mathrm{Cr-Ru}}(T)$$

where c refers to c_{11} or c_L. Therefore

$$\frac{\mathrm{d}\Delta c(T)}{\mathrm{d}p} = \frac{\mathrm{d}c_{\mathrm{Cr-V}}(T)}{\mathrm{d}p} - \frac{\mathrm{d}c_{\mathrm{Cr-Ru}}(T)}{\mathrm{d}p}.$$

From the last equation and figures 1 to 3 it follows that $\mathrm{d}\Delta c(T)/\mathrm{d}p > 0$ at room temperature, which corresponds to an increase of the magnetic contributions to c_{11} and c_L on application of pressure.

Recently Mãnosa *et al* [9] have studied the longitudinal acoustic mode softening and Invar behaviour of ferromagnetic $Fe_{72}Pt_{28}$. They found the first experimental evidence for softening of the longitudinal modes in this material under pressure and argued that this behaviour is central to understanding the source of the negative thermal expansion of ferromagnetic Invar alloys. The present measurements reveal that the same kind of mode softening occurs in Cr + 0.3 at.% Ru. This fact together with an earlier observation [4] that the coefficient of thermal expansion is zero near 400 K strongly suggest that Cr + 0.3 at.% Ru show antiferromagnetic Invar type behaviour; this will be the subject of further investigation.

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