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A high-pressure ultrasonic investigation of the Néel transition in a Cr + 0.07 at.% Ir alloy single crystal

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Abstract. Measurements of the ultrasonic wave velocity, ultrasonic attenuation, thermal expansion and electrical resistivity of a Cr + 0.07 at.% Ir alloy single crystal are reported as functions of temperature through the Néel point. Ultrasonic wave velocities were also measured as a function of temperature at different constant applied pressures and as a function of pressure at different constant temperatures. The pressure derivative of the Néel temperature corresponds closely to that observed for pure Cr and for several other dilute Cr alloys exhibiting a phase transition from an incommensurate spin-density wave to a paramagnetic phase at the Néel point. Hysteresis effects were observed in the temperature and pressure dependences of the ultrasonic wave velocity below the Néel temperature. These effects probably originate from a difference in the antiferromagnetic domain pattern existing in the alloy on cooling or heating through the Néel transition. No firm conclusions could be reached on whether the addition of 0.07 at.% Ir to Cr is enough to drive the first-order Néel transition of Cr to a second-order one.

1. Introduction

The Néel transition at the Néel point (T_N) of Cr is a transition from an incommensurate (I) spin-density wave (SDW) to paramagnetic (P) phase [1]. This transition is weakly first order, showing a small discontinuity in the thermal expansion and neutron diffraction intensity at T_N [2, 3]. The first-order nature of the Néel transition in Cr is driven to a second-order one by the addition of small quantities (of the order of 0.1 at.% or less) of impurities like for instance V or Mn. The problem of the effects of impurities on the order of the Néel transition of Cr was recently addressed theoretically [4]. The first-order nature of the transition is caused by a charge-density wave (CDW) that is induced by the Coulomb interactions between two hole Fermi surface sheets of Cr. Scattering by impurities suppresses the CDW and drives the transition to second order. It was shown [4] that as the first-order transition in Cr is weak only a very small impurity concentration is needed to accomplish this. For V impurities the critical concentration was calculated to be $x_t = 0.15$ at.% V, which corresponds well with experimental results [4]. In the case of Mn impurities x_t seems experimentally to be below 0.05 at.% Mn [5].

The magnetic behaviour of Cr alloys with group 8 non-magnetic transition metals (Ru, Os, Rh, Ir and Pt) is currently receiving attention in the literature [6–10], particularly as regards their elasticity at atmospheric pressure and effects of applied high pressure on their elastic constants and magnetic transition temperatures. The magnetic phase diagrams of these alloys contain a triple point where the ISDW, commensurate (C) SDW and P phases coexist. For alloy concentrations (x) below the triple-point concentration (x_p) the alloys

remain in the ISDW phase for all $T < T_N$, while alloys with $x > x_p$ exhibit both ISDW and CSDW phases for $T < T_N$. Cr alloys with group 8 non-magnetic transition metals show the most interesting magnetoelastic behaviour that is brought about by the interaction of the SDW with the lattice [11]. The pressure effects on the magnetoelasticity of this class of material were only recently studied for the first time, namely on a Cr + 0.3 at.% Ru alloy single crystal [6]. Remarkable anharmonic effects were found in this crystal and the studies clearly demonstrate the usefulness of ultrasonic wave velocity measurements under high applied pressure as regards obtaining more information on the order of the magnetic phase transitions in this class of alloys. The first reason for this is that magnetoelastic effects in these alloys are very large, giving very sharp and huge anomalies of magnetic origin, much larger than for other physical properties, in the temperature dependence of the elastic constants and therefore also in the ultrasonic wave velocities, particularly the longitudinal wave velocities, at the magnetic phase transitions. The second reason is that velocity of sound in materials can be measured easily to a sensitivity of 1 in 10^5 , making velocity of sound measurements as a function of pressure a very sensitive tool for probing for hysteresis effects under pressure near the magnetic phase transition or for following the magnetic transition temperature as a function of applied pressure. Pressure effects on the magnetoelasticity of Cr–Ir alloys of the above group of Cr alloys have not yet been studied. We succeeded in growing a good quality Cr + 0.07 at.% Ir alloy single crystal and report here on high-pressure longitudinal ultrasonic wave velocity studies on this crystal through the Néel transition. Measurements of the thermal expansion and electrical resistivity of the crystal are also reported. The Ir concentration of this crystal is below the triple-point concentration $x_p \approx 0.15$ at.% Ir, so the crystal remains in the ISDW phase for all $T < T_N$. One can therefore investigate whether 0.07 at.% Ir in Cr is enough to drive the first-order Néel transition to a second-order one. We also obtain the pressure–temperature magnetic phase diagram of this alloy crystal.

2. Experimental method

The Cr + 0.07 at.% Ir single crystal was grown by a floating-zone technique using RF heating in a pure argon atmosphere as previously described [12]. The starting materials were 99.996% pure Cr and 99.9% pure Ir. Ir is soluble in Cr up to about 12 at.% at 1680 °C [13]. The actual concentration of 0.07 ± 0.02 at.% Ir was determined through electron microprobe analyses at different points on the crystal. The crystal was prepared with a pair of flat and parallel (100) faces. The distance between these faces is about 8 mm. A standard pulse-echo overlap (10 MHz) technique [14] was used to measure the temperature and pressure dependence of the longitudinal ultrasonic wave velocity propagated along [100]. Hydrostatic pressure in the pressure range 0 to 0.16 GPa was generated by a high-pressure gas system using nitrogen gas as a pressure medium. Measurements were made as a function of pressure at different constant fixed temperatures as well as as a function of temperature at constant fixed pressures. The temperature could be kept constant to within 0.2 K or better during the pressure runs and pressure to better than 10^{-3} GPa during the temperature runs. A waiting time long enough for thermal equilibrium to set in was allowed for each data point. Velocity data were corrected for transducer diffraction effects by using methods developed by Kittinger [15]. The error in the absolute values of the sound velocity is about 0.5% while the sensitivity of the measuring technique is 1 part in 10^5 . Thermal expansion was measured using a strain gauge technique, and electrical resistivity by a standard four-probe method. These measurements were made to complement the ultrasonic measurements.

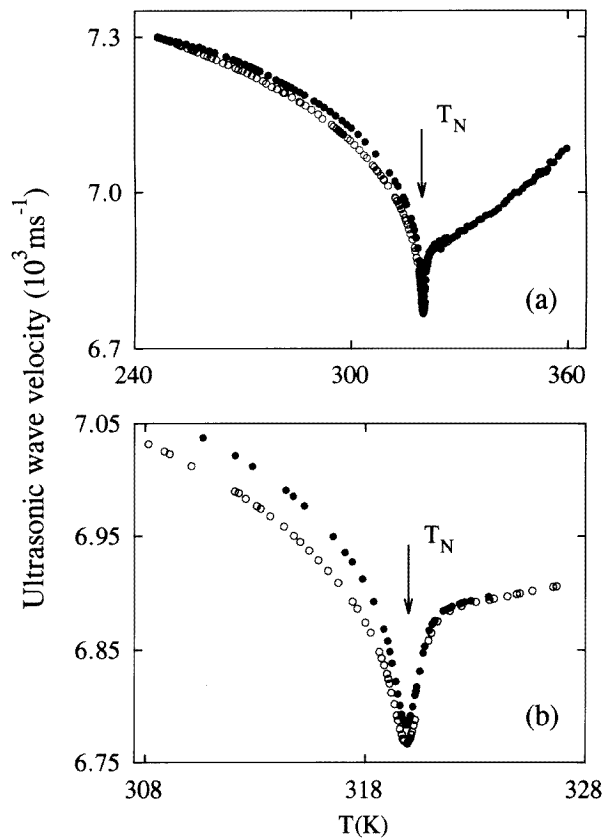


Figure 1. The temperature dependence of the longitudinal ultrasonic wave velocity of a Cr + 0.07 at.% Ir alloy single crystal for [100] wave propagation at atmospheric pressure. (a) shows the behaviour between 240 K and 360 K and (b) that close to the Néel point T_N . Closed circles show the results for a heating run and the open circles those for a cooling run.

3. Experimental results for the ultrasonic measurements

The temperature dependence of the longitudinal ultrasonic wave velocity (v_L) in the temperature range 240–360 K for [100] propagation is shown in figure 1(a) for both heating and cooling runs at atmospheric pressure. Figure 1(b) shows v_L-T in a smaller temperature range around T_N . T_N is taken at the minimum in the v_L-T curve, giving $T_N = 319.9 \pm 0.3$ K for both heating and cooling runs at atmospheric pressure. Figure 2 shows v_L-T runs for heating at different constant hydrostatic pressures. Cooling runs at constant pressure give to within 0.5 K the same T_N as the heating runs in figure 2 and show hysteresis effects below T_N similarly to in figure 1. The pressure dependence of T_N obtained from the temperature runs at different constant pressures is given in figure 3. Figure 4 shows v_L as a function of pressure at different constant temperatures, taken during both increasing and decreasing pressure runs.

The attenuation coefficient, γ , for longitudinal ultrasonic waves propagating along [100], obtained for a heating run at temperatures close to T_N , is shown in figure 5.

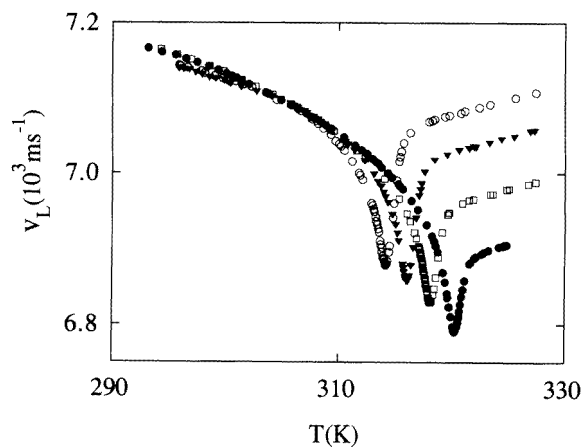


Figure 2. The temperature dependence of the longitudinal ultrasonic wave velocity (v_L) of a Cr + 0.07 at.% Ir alloy single crystal for [100] wave propagation at different constant pressures. The points are marked as follows: ●, atmospheric pressure; □, 34.5 MPa; ▼, 69 MPa; ○, 103.4 MPa. Only the data for increasing pressure runs are shown.

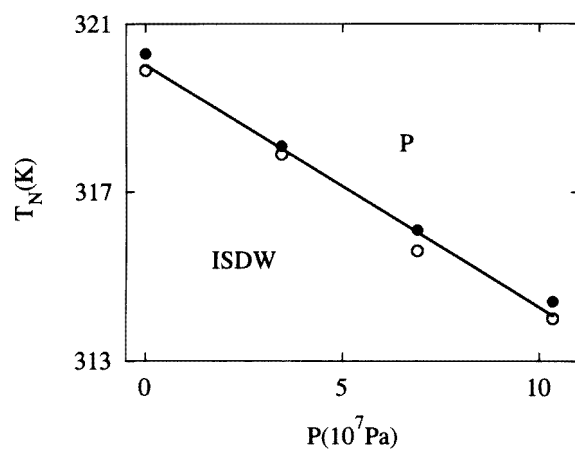


Figure 3. The pressure dependence of the Néel temperature, T_N , of a Cr + 0.07 at.% Ir alloy single crystal as determined from measurements of v_L as a function of temperature at different constant pressures. The closed circles are the results for T_N obtained for increasing temperature at a fixed pressure and the open circles those obtained during cooling at the same fixed pressure.

4. Discussion

The minimum in the v_L - T curve (figure 1) at $T = 319.9$ K corresponds closely to the temperature $T = 320.4$ K of the peak in the γ - T curve of figure 5. This in a sense justifies the identification of T_N at the minimum in the former curve and at the peak in the latter one. The value of T_N defined in this way corresponds well with $T_N \approx 318$ K obtained from the electrical resistivity data of Butylenko and Nevdacha [16] on polycrystalline Cr-Ir alloys.

A feature that was not previously reported for the physical properties of Cr-Ir alloys is the hysteresis effect observed in the v_L - T curve from just above T_N , starting at about

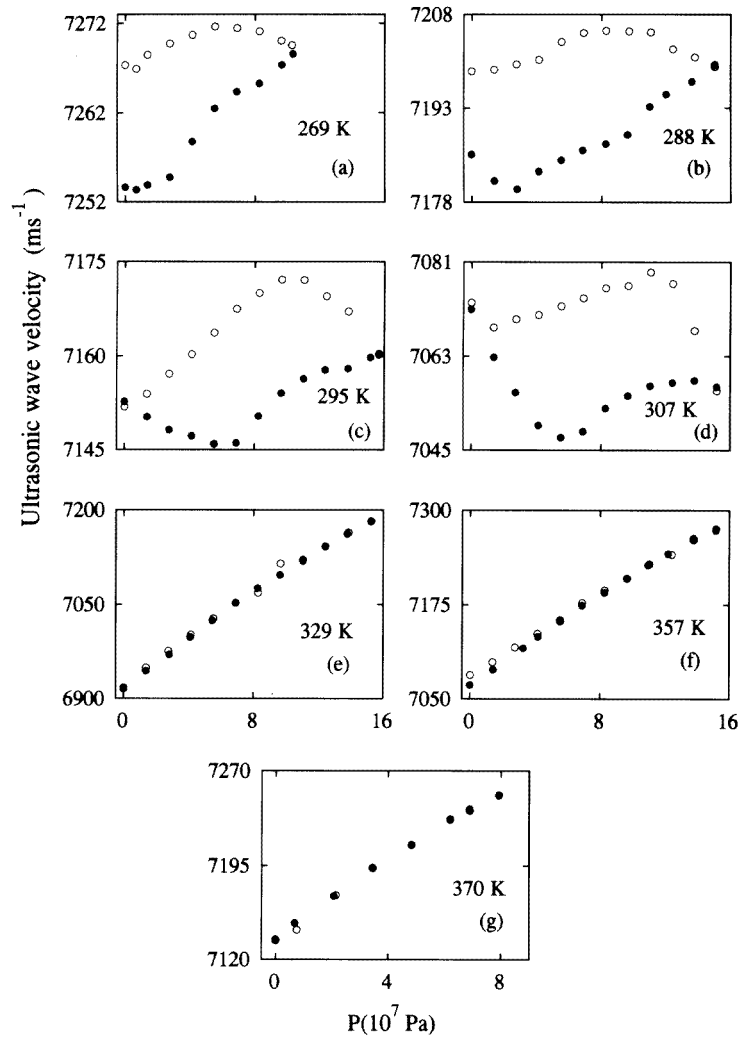


Figure 4. The ultrasonic wave velocity for longitudinal wave propagation along [100] as a function of pressure at different constant temperatures: (a) 269 K; (b) 288 K; (c) 295 K; (d) 307 K; (e) 329 K; (f) 357 K; (g) 370 K. The closed circles represent the results obtained for increasing pressure runs and the open circles those for decreasing pressure runs. Note the different pressure range in (g).

321 K, down to about 240 K (figure 1). Heating and cooling runs give about the same T_N , to within 0.5 K, but the velocity of sound for each $240 < T < 321$ K is smaller during cooling runs than during heating runs. Above 321 K, v_L is the same for both heating and cooling runs. A similar type of effect was previously [6] also observed for a Cr + 0.3 at.% Ru alloy single crystal, but in this case near the ISDW–CSDW magnetic phase transition temperature T_{IC} . For this crystal, hysteresis effects persist to well below (to about 40 K below) T_{IC} where v_L for cooling is also smaller than for heating runs. For $T > T_{IC}$, v_L on heating and cooling is the same. For the latter crystal T_{IC} differs for heating and cooling runs by as much as 20 K.

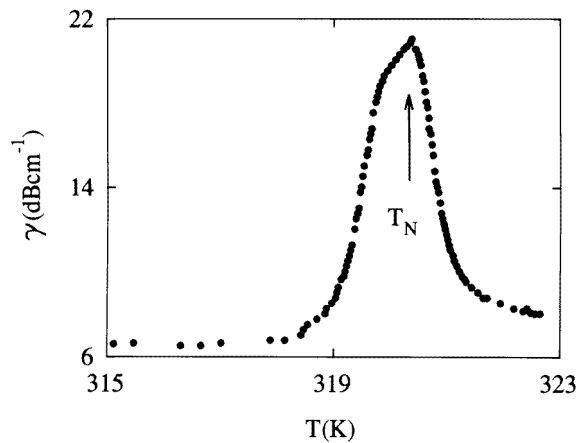


Figure 5. The ultrasonic wave attenuation coefficient, γ , of a Cr + 0.07 at.% Ir alloy single crystal for [100] wave propagation as a function of temperature near the Néel point.

The v_L - T curve of the Cr+0.07 at.% Ir crystal is very similar in shape to that previously [12, 17] reported for pure Cr. The overall width of the Néel transition in Cr + 0.07 at.% Ir seems however to be roughly two times smaller than that observed for a pure Cr crystal grown by the same technique as described above [12].

The pressure derivative $dT_N/dP = -58 \pm 4 \text{ K GPa}^{-1}$ obtained from figure 3 is very close to the value $dT_N/dP = -57 \text{ K GPa}^{-1}$ observed [18] for pure Cr which also shows an ISDW-P transition at T_N . This value also corresponds with that observed for other dilute Cr alloys at an ISDW-P transition: -50 K GPa^{-1} for Cr-Mn, -50 K GPa^{-1} for Cr-Ru, -60 K GPa^{-1} for Cr-Ge, but differs for some other dilute Cr alloys at their ISDW-P transitions: -100 to -200 K GPa^{-1} for Cr-Fe, -100 K GPa^{-1} for Cr-Si (see table VIII of [11]). It was found [19] for Cr-V alloys that $|dT_N/dP|$ decreases markedly as the electron concentration, e_A , in Cr is decreased by alloying Cr with up to 2.62 at.% V. Ir in Cr should increase the electron concentration, but due to the very low Ir content of the Cr + 0.07 at.% Ir crystal, we are unable to draw any conclusions concerning the effect of increasing e_A on $|dT_N/dP|$ for this case.

The hysteresis effects observed in figure 1 for v_L below T_N are more clearly displayed in the v_L - P curves at different constant temperatures of figure 4. The v_L - P curves of figure 4 show well defined hysteresis behaviour at temperatures below $T_N = 320 \text{ K}$ which are absent above this temperature. For $T > T_N$, v_L increases with increasing pressure without hysteresis as is expected for a paramagnetic material. The increase is however not exactly linear as usually observed for normal materials, but shows a slight curvature towards the pressure axis. Similar behaviour was also observed [20] for pure Cr, about 60 K above T_N , and is probably due to the presence of spin-fluctuation effects that persist in the crystal to temperatures well above T_N . The high-pressure studies [20] on pure Cr were only done for increasing pressures and therefore do not give information on hysteresis effects below T_N .

It is of interest to note the relatively small pressure effect on v_L for $T < T_N$ compared to that for $T > T_N$ in figure 4. The peculiar behaviour of v_L - P at $T < T_N$ in figure 4 probably results from the presence of the SDW and the hysteresis effects observed in figure 1 below T_N . One is inclined to ascribe the hysteresis effects observed near T_N as a manifestation of

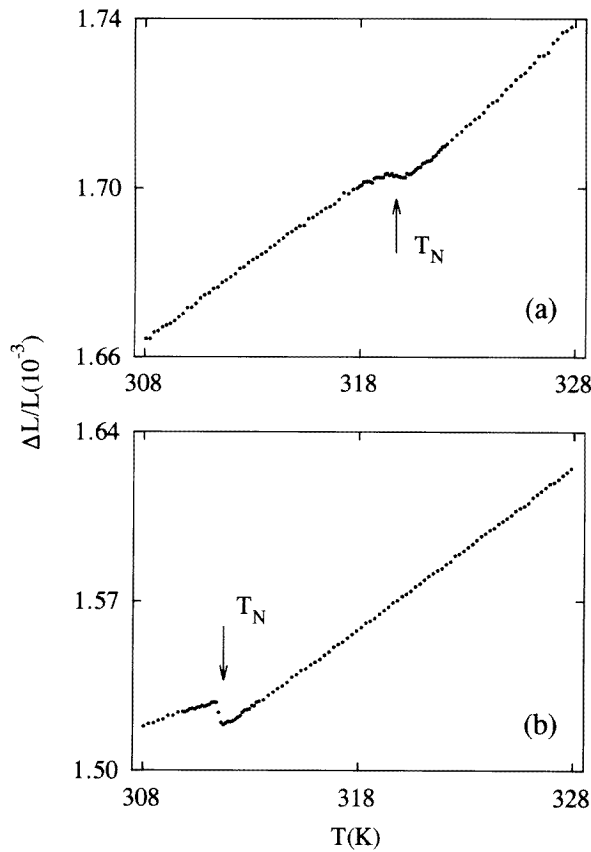


Figure 6. The temperature dependence of the thermal expansion, $\Delta L/L$, near the Néel point for (a) a Cr + 0.07 at.% Ir alloy single crystal and (b) a pure Cr crystal. (b) is after [5]. Measurements of $\Delta L/L$ relative to the length at 77 K are shown.

the possibility that the Néel transition in the alloy may be a first-order one. Measurements of the thermal expansion and electrical resistivity of the crystal through T_N , however, suggest that this is not the case. Figure 6 shows the thermal expansion ($\Delta L/L$) as a function of temperature measured on the Cr + 0.07 at.% Ir crystal near T_N for a heating run. Heating and cooling runs give within experimental error the same results. Also shown in figure 6 is the $\Delta L/L-T$ curve for pure Cr measured in our laboratory [5]. Pure Cr displays a discontinuous-like transition of width 0.3 K (figure 6) in $\Delta L/L-T$ at T_N , which reflects its first-order Néel transition. For the Cr + 0.07 at.% Ir crystal the transition is however clearly continuous (figure 6).

Figure 7 shows the temperature dependence of the electrical resistivity (ρ) measured on the Cr + 0.07 at.% Ir crystal. No hysteresis effects were observed within the experimental error for $\rho-T$ measured during heating and cooling runs. Clearly the Néel transition of the Cr + 0.07 at.% Ir crystal appears to be continuous from the $\rho-T$ and $\Delta L/L$ measurements. For the case of pure Cr in which the Néel transition is first order, $d\rho/dT$ changes nearly discontinuously at T_N [21] which is not the case for Cr + 0.07 at.% Ir in figure 7. One must however keep in mind that concentration inhomogeneities in the Cr + 0.07 at.% Ir alloy will smear a first-order transition out, making it appear continuous and second-order

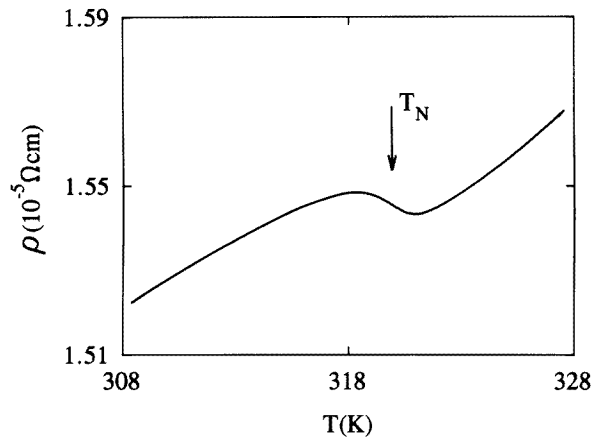


Figure 7. The temperature dependence of the electrical resistivity of a Cr + 0.07 at.% Ir alloy single crystal near the Néel transition temperature T_N . The smooth curve represents the best curve through about 1200 data points. These points do not scatter by more than about 0.7% around this smooth curve.

like. The fact that the width of the minimum in the v_L - T curve for the alloy is comparable with or smaller than that observed for pure Cr can probably be taken as evidence of fairly good homogeneity in the alloy. Unfortunately it is very difficult to make any decisive conclusions on the basis of the electron microprobe analyses at different points on the sample, concerning the homogeneity. This is due to the very low Ir concentration in the alloy, making the experimental error in the microprobe analyses relatively large. In this regard it may be mentioned that the width of the Néel transition in the $\Delta L/L$ - T curve of figure 6 is about 1 K compared to 0.3 K [5] for pure Cr. As the Néel temperature of Cr is changed by 9 K on adding 0.07 at.% Ir, a variation of about 10% in the Ir concentration throughout the sample is needed to smear a discontinuous transition out to a continuous one of width 1 K. A concentration change of 10% from point to point on 0.07 at.% Ir is difficult to detect by electron microprobe analyses.

The hysteresis effects observed in v_L - T (figures 1 and 4) may possibly be explained by the presence of antiferromagnetic domains below T_N . It is known [22, 23] that the longitudinal mode elastic constants, and therefore also the longitudinal mode sound velocity, v_L , of single- S single- Q Cr depend on the direction of the wave propagation direction relative to the direction of the SDW Q -vector and to the spin direction S . This can lead to differences of up to more than 1% between the longitudinal mode elastic constants measured for different allowed Q - and S -orientations in a single- Q single- S Cr crystal (see figure 43 of [1]). When Cr is cooled through T_N in the absence of an applied magnetic field, a multi- Q -domain phase will result consisting of single- Q domains in which the SDW Q -vector amongst them is oriented at random along the equivalent [100] directions of the bcc lattice. The S -vectors in the multi- Q phase will then also be oriented at random along the equivalent [100] directions, but perpendicular to the Q -vector of a specific Q -domain of the transverse SDW phase of Cr. It is suggested that the temperature hysteresis observed in the velocity of sound of the Cr + 0.07 at.% Ir crystal is due to the possibility that the fractional volumes occupied at each temperature, $T < T_N$, by each of the six types of antiferromagnetic Q -domain, are different on cooling or heating through T_N . A similar argument may be used to explain the observed pressure hysteresis. This type of

argument was previously [24] also used to explain the temperature hysteresis observed in the integrated neutron diffraction intensities of pure Cr below T_N in the multi- Q -domain state. The neutron diffraction experiments [24] clearly indicate that the fractional volumes occupied by each of the six types of antiferromagnetic Q -domain in Cr below T_N depend on the temperature and on the past thermal and magnetic history of the sample. The present study on Cr + 0.07 at.% Ir demonstrates that the fractional volumes are also dependent on the pressure history of the sample.

5. Conclusion

Temperature and pressure runs show the presence of hysteresis effects in the velocity of sound of a Cr + 0.07 at.% Ir alloy single crystal below the Néel temperature, effects not previously observed in other Cr alloys near the ISDW–P transition. These effects are thought to originate from a difference in the antiferromagnetic Q -domain pattern in the alloy on heating or cooling through T_N . Hysteresis effects were not observed within experimental error in the temperature dependence of the thermal expansion and electrical resistivity of the crystal below T_N . The pressure derivative of T_N of the Cr + 0.07 at.% Ir crystal is nearly the same as that observed for the ISDW–P transition of pure Cr and of low-concentration Cr–Ru alloys that are also members of the series of Cr alloys with group 8 non-magnetic transition metals. No firm conclusion could be reached concerning the order of the Néel transition in the Cr + 0.07 at.% Ir crystal. The measurements point to a continuous second-order transition, but a smearing out of the transition, by sample inhomogeneities cannot be definitely ruled out.

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