Electrical and thermal transport properties of Cr-Si alloy single crystals

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Abstract. Electrical resistivity (\( \rho \)), Seebeck coefficient (\( S \)) and thermal conductivity (\( \kappa \)) measurements on Cr\(_{1-x}\)Si\(_x\) alloy single crystals with \( x = 0.005, 0.012 \) and \( 0.016 \) reveal novel features not previously detected in polycrystalline alloys. Our results prove thermal transport to be an experimental probe that is especially well disposed towards exposing spin-density-wave ordering in an itinerant system such as Cr-Si.

1. Introduction
The magnetic phase diagram of the dilute Cr\(_{1-x}\)Si\(_x\) alloy system [1] contains a commensurate (C) spin-density-wave (SDW), an incommensurate (I) SDW and a paramagnetic (P) phase that co-exist at a triple point \( x_L \approx 0.01 \) and \( T = 230 \) K, with a re-entrant CSDW phase predicted [1] below 150 K in the range \( 0.01 < x < 0.015 \). The Cr-Si system, together with Cr-Fe, are remarkable as they show pronounced first-order CSDW-P Néel transitions for \( x > x_L \) [1]. The phase diagram also contains a phase line separating transverse (T) and longitudinal (L) SDW phase regions.

The physical parameters electrical resistivity (\( \rho \)), thermopower (Seebeck coefficient, \( S \)) and thermal conductivity (\( \kappa \)) are all related in simple models to a common factor, the density of states at the Fermi level (\( E_F \)). The latter is critically affected in Cr and its alloys by nesting of the electron and hole Fermi surface (FS) sheets, resulting in annihilation of the nesting portions on SDW formation and the appearance of an energy gap in the band structure, with a concomitant loss of current carrier density. The above mentioned three parameters are therefore ideal, particularly \( S \) which depends on the energy derivative of the conductivity at \( E_F \), to probe the role and signatures of Fermi surface effects on SDW type magnetic phase transitions in Cr alloys. \( \rho, S \) and \( \kappa \) of the Cr-Si alloy system have previously [1, 2, 3] been reported mainly on polycrystalline samples, with the only exception, that we are aware of, being \( \rho(T) \) measurements [4] on the same three Cr-Si alloy single crystals used in this study. The \( \rho(T) \) measurements are revisited here, extending the measurements to below 77 K. Measurements on polycrystalline material have however limited value, particularly in investigations of the role of the SDW Q-domain state, single Q-domain or multi Q-domain, on the above mentioned properties of Cr and its alloys. Here Q denotes the SDW wave vector. The availability of good quality Cr\(_{1-x}\)Si\(_x\) single crystals should be useful for novel investigations of SDW energy gap effects on the thermopower and searches for signatures of the re-entrant CSDW phase and spin-flip transition in the transport properties of this alloy system. To this effect, detailed measurements of the temperature and magnetic field dependence of \( \rho, S \) and \( \kappa \) on single crystalline Cr\(_{1-x}\)Si\(_x\) alloys, having \( x = 0.005, 0.012 \) and \( 0.016 \), are reported here. These three concentrations are representative examples to cover all known magnetic
phase transitions that are allowed by the magnetic phase diagram. Applied magnetic fields up to 90 kOe were used to test for possible influences of the Q-domain state on these properties.

2. Experimental

The Cr-Si single crystals used in the present study are the same crystals previously used for electrical resistivity, elastic constants and thermal expansion measurements [4, 5, 6].

Measurements of $\rho$, $S$ and $\kappa$ were done in a temperature range 2 K $\leq T \leq$ 380 K using a Quantum Design (QD) Physical Properties Measurement System (PPMS), incorporating QD resistivity and thermal transport options. The electrical current or thermal gradient for the measurements were directed along the cubic [100] direction for all the samples. A typical thermal gradient of 3% of the actual temperature was used for the thermal transport measurements.

Previous measurements [7] on a pure Cr single crystal show enhancement of about 50% in the size of the magnetic anomaly induced in $S(T)$ on cooling through the ISDW-P Néel transition if the crystal is prepared in a single Q-state, obtained by field cooling in a strong magnetic field ($H=60$ kOe), rather than in a multi Q-state. For this reason the present resistivity measurements on the Cr-Si crystals were done on cooling down from 370 K to 2 K, both in zero and in $H=90$ kOe applied fields. The field was applied along [011], perpendicular to the [100] direction, for the cases of Cr$_{0.995}$Si$_{0.005}$ and Cr$_{0.989}$Si$_{0.016}$. For the Cr$_{0.986}$Si$_{0.012}$ crystal measurements were taken for two cases; with the field applied either along [011] or parallel to the [100] direction. Thermal transport measurements ($S$ and $\kappa$) were conducted for all the crystals in zero field, as well as in $H=90$ kOe or 80 kOe aligned along [100].

3. Results and discussion

Figure 1 shows the $\rho(T)$ for the Cr$_{0.988}$Si$_{0.012}$ crystal in zero field. Three distinct $\rho(T)$ anomalies are observed in this figure. The first is in the form of a large, nearly discontinuous, jump in $\rho(T)$ at the Néel temperature, $T_N = (234\pm2)$ K on cooling and $(242\pm2)$ K on heating. The second anomaly, in the form of a relatively weaker, but also closely discontinuous jump, appears at $(140.5\pm0.8)$ K on cooling and at $(163.7\pm0.8)$ K on heating. This anomaly was not observed in previous [4] measurements on the same crystal down to 77 K. It is probably attributed to the particular temperature cycle (320 to 77 to 320 to 77 K) used in the previous measurements, resulting in a broadening of the low temperature hysteretic anomaly (at around 100 K), to such an extent that it overlaps the temperature range of the ISDW-CSDW transition. The temperature where the second anomaly in figure 1 occurs (on heating), corresponds reasonably well with the ISDW-CSDW phase transition temperature $T_{IC} = 150$ K (on heating), obtained from velocity of sound measurements on the same crystal [6]. A third relatively large hysteretic anomaly is also observed in figure 1 in the temperature range 60 < $T$ < 110 K, approximately similar in shape, but much narrower in temperature range, than that previously observed [4, 6] in $\rho$ and thermal expansion measurements on the same crystal. It is now evident from the new measurements that the third $\rho(T)$ anomaly of figure 1 was mistakenly interpreted in the previous resistivity and thermal expansion measurements as due to an ISDW-CSDW phase transition. Although this anomaly occurs at the approximate temperature expected for the spin-flip transition of this alloy, it should be noted that this transition in Cr-alloys usually has no significant influence on $\rho(T)$. A further complication is the possibility of an unstable mixed magnetic state [4] in which CSDW, LISDW and TISDW phases co-exist in this crystal at low temperatures, as $x = 0.012$ of this crystal is close to the point ($x = 0.015$, $T = 50$ K) where the phase lines separating these three phases merge on the magnetic phase diagram [1]. It is therefore difficult to speculate on the origin of the low temperature $\rho$-anomaly of the Cr$_{0.988}$Si$_{0.012}$ crystal. Present measurements of $\rho(T)$ on the Cr$_{0.995}$Si$_{0.005}$ and Cr$_{0.984}$Si$_{0.016}$ crystals down to 2 K, show magnetic anomalies, non-hysteretic and hysteretic, respectively, only near one temperature, namely $T_N$, in accordance with the magnetic phase diagram [1] and previous measurements [4].

$S(T)$ and $\kappa(T)$ of the ISDW Cr$_{0.995}$Si$_{0.005}$ crystal (measured as part of this study, but not shown here) both display only one pronounced non-hysteretic anomaly of SDW origin at $T_N = (275.3\pm0.1)$ K, quite
The electrical resistivity ($\rho$) as a function of temperature for the Cr$_{0.988}$Si$_{0.012}$ crystal with $H = 0$. The solid curve was measured on heating and the broken curve on cooling.

Similar in shape to that previously [2] observed for $S(T)$ of ISDW Cr$_{0.996}$Si$_{0.004}$ and for $\kappa(T)$ of Cr$_{0.991}$Si$_{0.009}$ [3] polycrystalline alloys. In these regards single and polycrystalline alloys behave similarly.

$S$-$T$ and $\kappa$-$T$ curves for the Cr$_{0.988}$Si$_{0.012}$ and Cr$_{0.984}$Si$_{0.016}$ crystals are shown in figure 2 (a) and (b) and figure 3 (a) and (b), respectively. Both these crystals show relatively sharp step-like CSDW-P Néel transitions in both $S(T)$ and $\kappa(T)$, without the presence of thermal hysteresis effects. The absence of hysteresis for these two crystals is attributed to masking thereof due to the smallness of the temperature width of the Néel transition (~8 K from $\rho(T)$), compared to the temperature gradient (amounting to ~7 K at 235 K) used during the measurements.

Figure 3(a) for Cr$_{0.984}$Si$_{0.016}$ shows a small but prominent new $S(T)$ minimum at $T_N = (249 \pm 2)$ K, not previously observed in polycrystalline Cr-Si alloys [2]. Signatures of such a minimum is absent in $\rho(T)$ and $\kappa(T)$ for this crystal. Measurements during heating and cooling runs for the Cr$_{0.988}$Si$_{0.012}$ crystal in the temperature range of the two lower $\rho$-anomalies of figure 1, reveal no signatures of the expected $S(T)$ and $\kappa(T)$ anomalies of figures 2(a) and (b). The absence of the I-C transition in $S$ and $\kappa$ can probably be ascribed to the smallness of the effect and its smearing out due to the temperature gradient used in the measurements. The reason for the absence of signatures of the prominent lower temperature anomaly, is however unknown.

Application of a magnetic field has no influence on any of the measurements, except for the resistivity of the ISDW $x = 0.005$ sample for which the field reduces $\rho$ at $T < 100$ K significantly.

Figures 1 and 2 show nearly identical magnetic anomalies $\Delta \rho/\rho \approx - \Delta \kappa/\kappa \approx 10\%$ in $\rho$ and $\kappa$, except for the sign, on cooling the Cr$_{0.988}$Si$_{0.012}$ crystal through $T_N$. This is expected as the magnetic contributions to these parameters have the same origin, namely a reduction in the carrier density due to a gapping of the FS on SDW formation. The above equivalence is also in line with the Wiedemann-Franz law, $\kappa \rho = L_0 T$, where $\kappa$ and $\rho$ are the electronic heat conductivity and resistivity, respectively, both at temperature $T$ and $L_0=2.44 \times 10^8$ $\text{V}^2\text{K}^{-2}$ is the free electron Lorentz number.

**Figure 2** The temperature dependence of (a) the Seebeck coefficient ($S$) and (b) the thermal conductivity ($\kappa$) for the Cr$_{0.988}$Si$_{0.012}$ single crystal. The regions around the Néel temperature are shown in the insets. Symbols $\bigcirc$ and $\triangle$ represent heating and cooling runs, respectively.
Figure 3 The temperature dependence of (a) the Seebeck coefficient ($S$) and (b) the thermal conductivity ($\kappa$) of the Cr$_{0.984}$Si$_{0.016}$ single crystal. The region around the Néel temperature is shown in the insets. Symbols $\bigcirc$ and $\bigtriangleup$ represent heating and cooling runs, respectively. $\bigtriangleup$ show measurements on heating after field cooling in $H = 80$ kOe.

Figure 4 Temperature dependence of the Lorentz number ($L$) of Cr$_{1-x}$Si$_x$ with $x = 0.005(\bigcirc)$, $x = 0.012(\bigtriangleup)$ and $x = 0.016(\square)$. The Sommerfeld free electron value $L_0 = 2.44 \times 10^{-8}$ V$^2$K$^{-2}$ is chosen as the initial value on the vertical axis.

Figure 4 shows the temperature dependence of $L = \kappa \rho / T$ for the Cr$_{1-x}$Si$_x$ crystals. $L$ is defined in terms of the measured $\kappa$, which includes both phonon and electronic contribution. The $L$ values in figure 4 are considerably larger than $L_0$ and its relatively high values in the P phases of the alloys are linked [1, 3], as previously done for a Cr$_{0.991}$Si$_{0.009}$ alloy, to nesting effects of the Fermi surface sheets. The peaks around 50 K in figure 4, prominent in the case of $x = 0.012$ and 0.016, but relatively weak for $x = 0.005$, was previously also observed in a polycrystalline Cr$_{0.999}$Si$_{0.009}$ alloy [3] and ascribed [1, 3] to phonon contributions to $\kappa$.

4. Conclusion
Measurements of $\rho(T)$, $S(T)$ and $\alpha(T)$ on Cr$_{1-x}$Si$_x$ alloy single crystals, containing $x = 0.005$, 0.012 and 0.016, reveals aspects not previously observed in polycrystalline material. Of interest is the absence of the relative large hysteretic low temperature ($60 < T < 110$ K) $\rho$-anomaly in $S(T)$ and $\alpha(T)$ measurements on the Cr$_{0.998}$Si$_{0.002}$ crystal and the fact that cooling ISDW or CSDW Cr-Si alloy single crystals through $T_N$ in an applied field of up to 90 kOe, has no significant effect on the transport properties. No evidence could be found in the present single crystal measurements for the re-entrant CSDW phase predicted [1] on the Cr-Si magnetic phase diagram. Neutron diffraction studies are proposed to clarify the unique low temperature $\rho(T)$ anomaly observed for the Cr$_{0.998}$Si$_{0.002}$ crystal.

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References
[1] Fawcett E, Alberts HL, Galkin V Yu, Noakes DR and Yakhmi JV 1994 Rev. Mod. Phys. 66 25
[2] Arajs S, Anderson EE and Ebert EE 1971 Il Nuovo Cimento 4 40
[3] Johannesson Ch, Aström HU and Rao KV 1982 Physica Scripta 25 751
[4] Prinsloo ARE, Alberts HL and Smit P 1998 J. Phys.: Condens. Matter 10 2715
[5] Prinsloo ARE, Alberts HL and Smit P 1997 Phys. Rev. B 56 11777
[6] Anderson RA, Alberts HL and Smit P 1993 J. Phys.: Condens. Matter 5 1733
[7] Muir WB and Ström-Olsen JO 1971 Phys. Rev. B 4 988