Possible charge-density wave transition in $PrPt_2Si_2$

M. Kumar,[1] V. K. Anand,[2] C. Geibel,[1] M. Nicklas,[1,*] and Z. Hossain[2]
[1]*Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, 01187 Dresden, Germany*
[2]*Department of Physics, Indian Institute of Technology, Kanpur 208016, India*
(Received 22 September 2009; revised manuscript received 17 February 2010; published 8 March 2010)

The physical properties of the ternary intermetallic compound $PrPt_2Si_2$ have been investigated by specific-heat and electrical-resistivity measurements under pressure up to 2.85 GPa. Two phase transitions at T_0 =88 K and T_x=54 K are inferred from anomalies in the electrical resistivity. Only the transition at T_0 is visible in the specific-heat data. A strong thermal hysteresis suggests a first-order character of both transitions. The resistivity anomaly at T_0 has the resemblance to a charge-density wave transition. Furthermore, low-temperature powder x-ray diffraction data do not show any signature of a structural phase transition. The nonmagnetic character of both transitions is confirmed by the absence of a signature in magnetic susceptibility and a magnetic field independent transition temperature. Application of pressure suppresses $T_0(p)$ but has a very weak effect on $T_x(p)$.

DOI: 10.1103/PhysRevB.81.125107 PACS number(s): 71.45.Lr, 72.15.−v, 62.50.−p, 71.20.−b

I. INTRODUCTION

Both charge-density wave (CDW) and spin-density wave (SDW) transitions correspond to an instability of the Fermi sea induced by nesting of the Fermi surface.[1] The theory of CDWs was addressed by Fröhlich in 1954 (Ref. 2) and by Peierls in 1955 (Ref. 3) and later in 1962 Overhauser introduced the concept of the SDW.[4] While the CDW ground state exhibits a periodic variation in the charge density and of the lattice distortion, the SDW ground state is characterized by a spatial variation in spin density and is magnetic in character. In contrast to the SDWs, CDWs usually require a strong anisotropy of the material, i.e., the presence of a quasi-two-dimensional (2D) or quasi-one-dimensional electronic system. However, recently CDWs have also been reported in compounds such as $Lu_2Ir_3Si_5$ and $Lu_5Ir_4Si_{10}$, which *a priori* are rather three-dimensional metals.[5,6] The appearance of an energy gap in the density of states, which is a consequence of the nesting of the Fermi surface, causes many CDW systems to behave like a semimetal and sometimes can lead to a metal-to-insulator transition. Special interest in CDWs exists when they appear together with other ordering phenomena such as superconductivity or magnetism.

The simultaneous observation of superconductivity and CDW order in, e.g., $Bi_2Rh_3Se_2$, $Lu_2Ir_3Si_5$, and $Lu_5Ir_4Si_{10}$ has brought a new perspective to the research in this field.[5–7] The CDW transition temperatures of the quasi-2D rare-earth tritelluride compounds RTe_3 (R=La-Nd, Sm, and Gd-Tm) display striking systematics across the rare-earth series.[8–12] In case of tritellurides a second CDW transition is observed for the heavier rare-earths Dy-Tm.[11,12] More interestingly, a reduction in lattice parameters by application of chemical pressure (chemical substitution) or external pressure suppresses the upper CDW transition temperature while it enhances the lower CDW transition temperature.[11,13] The optical reflectivity by Sacchetti *et al.* indicates that the CDW gap belonging to the upper transition of $CeTe_3$ closes under increasing external pressure and evidences that external and chemical pressure have qualitatively a similar effect.[10,13] Moreover, pressure induced superconductivity has been observed in $TbTe_3$ (Ref. 14) and $ZrTe_3$.[15] The appearance of re-entrant superconductivity evidences the complicated interplay of the different ordering phenomena. On increasing pressure the CDW ordering temperature, T_0, decreases while the superconducting transition temperature, T_c increases.[14]

Recently, we started working on Pr-based compounds and investigated several ternary compounds of 122 composition that included $PrRh_2Si_2$, $PrPd_2Si_2$, and $PrPt_2Si_2$. $PrRh_2Si_2$ and $PrPd_2Si_2$ were found to order antiferromagnetically below 68 K and 3 K, respectively.[16,17] However, no magnetic order was observed in $PrPt_2Si_2$ down to 2 K (Ref. 17) in agreement with the paramagnetic behavior reported previously in literature.[18] $PrPt_2Si_2$ crystallizes in the $CaBe_2Ge_2$-type primitive tetragonal structure. We found two broad anomalies with large hysteresis in electrical-resistivity measurement runs performed on cooling and heating cycles between 1.8 and 300 K. We will discuss that these anomalies are connected with the formation of a CDW. Further, we will show that application of hydrostatic pressure suppresses the CDW state. While the structure of $PrPt_2Si_2$ can formally be described as a stacking of Pr and Pt_2Si_2 layers, the electronic and magnetic properties of this RT_2Si_2 type of compounds (R=rare-earth element and T=transition-metal element), such as, e.g., band structure or spin-wave excitations, are usually clearly three dimensional,[19,20] in contrast to the pronounced two-dimensional character of the archetypical CDW systems based on selenides or tellurides. Thus $PrPt_2Si_2$ is likely a new example for a CDW in a compound with a three-dimensional Fermi surface.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $PrPt_2Si_2$ were prepared by standard arc melting on a water-cooled copper hearth under an inert argon atmosphere starting with high-purity (99.99% and above) elements in a stoichiometric ratio. The samples were annealed at 1000 °C for 1 week to improve the quality (for details see Ref. 17). The polycrystalline samples look dense and shiny but are a little bit brittle. The samples were

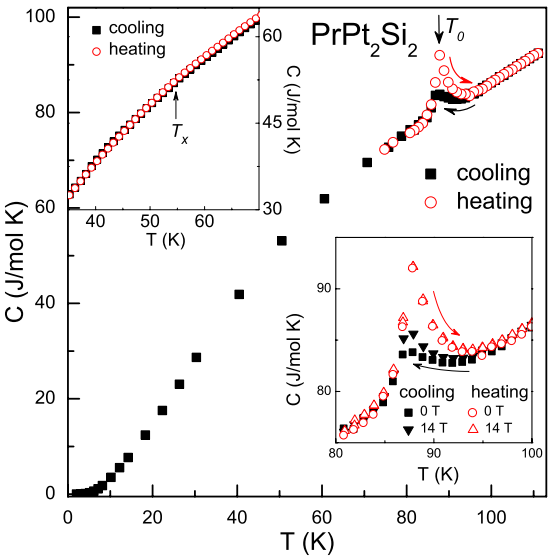

FIG. 1. (Color online) Specific-heat data, $C(T)$, of $PrPt_2Si_2$ taken on cooling and heating cycles, respectively. The upper inset shows $C(T)$ between 35 and 70 K. The position of the T_x anomaly observed in the electrical resistivity is indicated by an arrow. The lower inset displays an expanded view of the region near T_0.

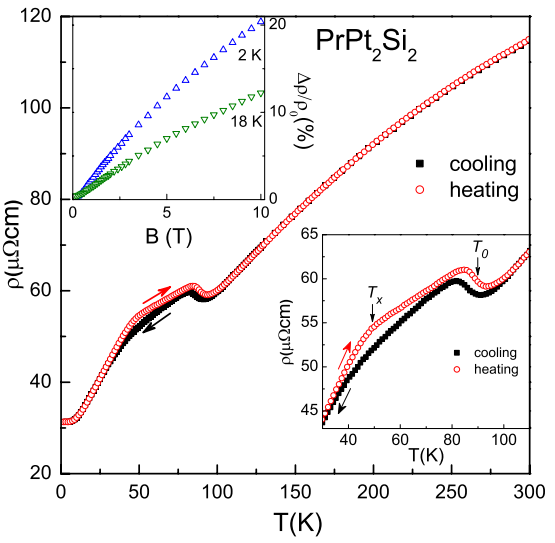

FIG. 2. (Color online) Temperature dependence of electrical resistivity, $\rho(T)$, of $PrPt_2Si_2$. The upper inset shows the magnetoresistance $\Delta\rho/\rho_0$ up to 10 T. The lower inset presents the anomalies in the resistivity in detail. The arrows denote the phase transitions at T_0 and T_x.

characterized using powder x-ray diffraction and scanning electron microscopy equipped with energy-dispersive x-ray analysis. Specific heat was measured by a relaxation method and electrical resistivity by a standard four-probe ac technique. Magnetic field up to 14 T and temperatures down to 1.8 K were achieved in a physical properties measurement system (PPMS, Quantum Design). The reproducibility of the results has been checked by performing the measurements on a second batch of samples too. For the high-pressure electrical-resistivity experiments a double-layer piston-cylinder-type pressure cell with silicone fluid as pressure-transmitting medium was utilized. Pressure up to 3 GPa could be generated. The shift of the superconducting transition temperature of Pb with pressure served as a pressure gauge. The superconducting transition remained sharp at all pressures indicating a pressure gradient less than 1–2 % of the applied pressure along the sample.

III. EXPERIMENTAL RESULTS

Figure 1 displays the specific heat of $PrPt_2Si_2$. A sharp anomaly at $T_0=88$ K indicates a phase transition. A clear thermal hysteresis points to the first-order nature of the transition at T_0. The application of magnetic field up to $B=14$ T does not show any significant effect on T_0 supporting that the transition is not associated with any magnetic ordering (see lower inset of Fig. 1). Moreover, the absence of any signature of this transition in the magnetic susceptibility also evidences the nonmagnetic origin of this transition.[17]

To further study the nature of the transition at T_0 we carried out electrical resistivity, $\rho(T)$, measurements. Figure 2 shows $\rho(T)$ taken during cooling and heating cycles between 1.8 and 300 K. $\rho(T)$ exhibits metallic behavior with a very broad curvature above 100 K. This curvature can be attributed to crystal-field effects. On decreasing temperature $\rho(T)$ exhibits a sharp upturn between 95 and 80 K in the vicinity of T_0. This behavior is typical for the opening of a gap and hints, in absence of any magnetic signature, at the opening of a CDW gap in the density of states at the Fermi surface. A second broad anomaly is observed around $T_x=55$ K. The specific-heat data do not show any feature or anomaly at T_x (upper inset of Fig. 1). However, a broad curvature is apparently inferred between 40 and 70 K. Such a broad curvature might be associated with a transition from an incommensurate to a commensurate CDW as in $Er_5Ir_4Si_{10}$. Below T_x, the resistivity decreases strongly down to 1.8 K. We find a residual resistivity ratio of $\rho_{300\ \mathrm{K}}/\rho_{1.8\ \mathrm{K}}\approx 8$.

The electrical resistivity shows a large thermal hysteresis connected with the phase transitions. During the cooling cycle the upturn of $\rho(T)$ related to the CDW transition at T_0 starts at about 94 K and attains a maximum around 82 K while in the heating cycle the resistivity shows the maximum at ≈85 K then decreases sharply and exhibits a minimum around 99 K (lower inset of Fig. 2). The anomaly at $T_x=55$ K only shows a slight change in slope on cooling while on heating a distinct kink approximately at the same temperature hints at the presence of a second phase transition. With this transition, as with the T_0 transition, a large hysteresis is associated. The broad resistivity anomaly having a typical width of about 10–15 K and the large thermal hysteresis observed at both transitions suggest a first-order character of both phase transitions.

The magnetoresistance, $\Delta\rho/\rho_0=[\rho(B)-\rho(0)]/\rho(0)$, of $PrPt_2Si_2$ at 2 K as well as at 18 K is positive up to 10 T (upper inset of Fig. 2). The almost linear in B behavior with a slight negative curvature suggests a combination of the positive magnetoresistance of a normal metal with a negative spin-flip scattering at higher magnetic field due to the mixing of higher crystal electric field levels with increasing B.

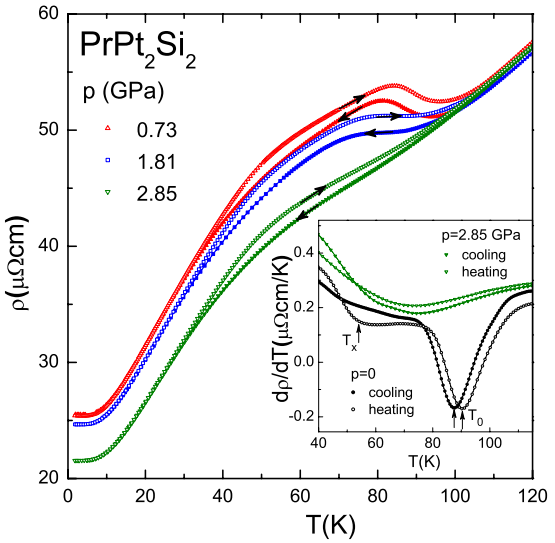

FIG. 3. (Color online) $\rho(T)$ of $PrPt_2Si_2$ at different applied pressures measured on cooling (closed symbols) and on heating cycles (open symbols), respectively. The arrows indicate the direction of temperature change. Inset: $d\rho(T)/dT$ at $p=0$ and 2.85 GPa. The arrows in the inset mark the transitions at T_0 and T_x. At 2.85 GPa T_0 and T_x cannot be defined unambiguously anymore.

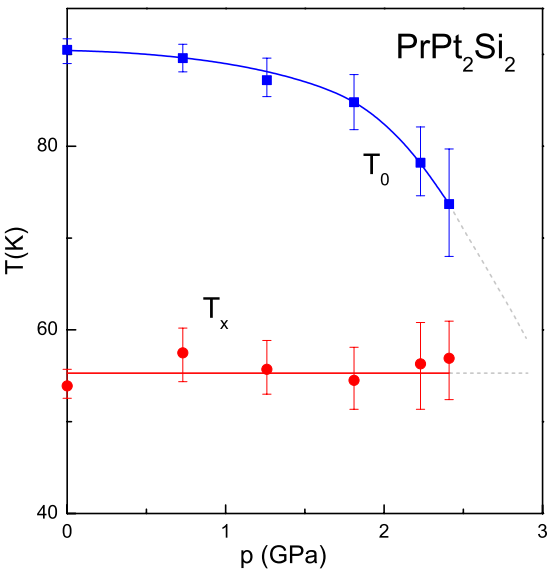

FIG. 4. (Color online) T-p phase diagram of $PrPt_2Si_2$: the transition temperatures $T_0(p)$ and $T_x(p)$ determined from the resistivity data on heating are presented. The lines are guide to the eyes.

Since CDW order is predominantly driven by Fermi-surface nesting, it is especially sensitive to pressure-induced changes in the electronic structure. Therefore, to further elucidate the nature of the transition, we performed electrical-resistivity experiments under hydrostatic pressure up to 2.85 GPa. Figure 3 shows $\rho(T)$ for selected pressures. Application of pressure rapidly shifts $T_0(p)$ to lower temperatures. We have determined T_0 by the position of minimum in the $d\rho(T)/dT$, which at ambient pressure agrees well with the position of the anomaly in the specific-heat data, and T_x as the temperature, where $d\rho(T)/dT$ shows the sharp kink below the minimum. $T_x(p)$ only shows a weak pressure dependence and remains almost unchanged up to 2.41 GPa. It is also worth noting that the hysteresis associated with the T_0 transition becomes smaller on increasing pressure, i.e., the difference between T_0 determined on heating and cooling decreases slightly. However, the hysteresis associated with $T_x(p)$ does not show any significant change and further confirms that $T_x(p)$ is only weakly depending on pressure.

We compile the results of our pressure study in the T-p phase diagram displayed in Fig. 4. For clarity we only show $T_0(p)$ and $T_x(p)$ determined from the resistivity data taken on the heating cycle. At first, $T_0(p)$ decreases almost linearly up to $p=1.81$ GPa with an initial slope of $dT_0(p)/dp|_{p=0} \approx -3$ K/GPa and then drops more rapidly on further increasing pressure. Above 2.41 GPa the anomalies at transitions at T_0 and T_x broaden more and more and move close to each other, too close and too broad to still determine the transition temperatures unambiguously. A simple extrapolation of $T_0(p)$ and $T_x(p)$ to higher pressures indicates that the CDW transition at T_0 approaches the second transition at T_x around $p \approx 3$ GPa.

IV. DISCUSSION

We first note that an anomaly quite similar to the upper one we observed at $T_0=88$ K in $PrPt_2Si_2$ was reported by Ragel *et al.*[21] in the resistivity of $LaPt_2Si_2$ at $T_0=111$ K. It is very likely that both are related to identical phenomena. Substitution of La by Ce, having a smaller ionic radius, in the alloy $La_{1-x}Ce_xPt_2Si_2$ leads to a shift of T_0 to lower temperatures and to a weakening of the associated anomaly, which was no longer observable for $x>0.7$.[21] Extrapolating the linear decrease in $T_0(x)$ to $x=1$ results in an hypothetical $T_0=60$ K in pure $CePt_2Si_2$. This is well below $T_0=88$ K we observed in $PrPt_2Si_2$, although both lattice parameters a and c (as well as the unit-cell volume) are still larger in the Ce-based than in the Pr-based compound. Thus, while the decrease in $T_0(x)$ in $La_{1-x}Ce_xPt_2Si_2$ is in general agreement with the decrease in $T_0(p)$ in $PrPt_2Si_2$ (and with the general response of such transitions to pressure, see below), the effect in the former alloy is much larger, which is likely related to the strong Kondo effect in $CePt_2Si_2$. In this compound inelastic neutron-scattering experiments indicate a rather large Kondo temperature on the order of 250 K,[22] which should strongly affect the electronic states which we suggest to be responsible for this CDW-like transition. From the other point of view, the very anomalous T dependence of the quasielastic line observed in these neutron-scattering experiments, i.e., an unexpected, pronounced increase below 50 K,[22] as well as further anomalous properties of $CePt_2Si_2$, might be a consequence of this electronic instability still being present, but too weak to lead to a real phase transition.

Ragel *et al.*[21] mentioned either a classical structural transition or a CDW as possible scenario for the anomaly at T_0 in $LaPt_2Si_2$, but did not analyze this transition further, because of a lack of sufficient data. Our more detailed investigations strongly support the CDW scenario in $PrPt_2Si_2$. On the one hand our results reveal a striking analogy of the resistivity of

$PrPt_2Si_2$ to that of $Er_5Ir_4Si_{10}$, which also presents a steplike increase (with decreasing temperature) at $T_0=155$ K and a kink in $\rho(T)$ connected with an increase in $d\rho(T)/dT$ below $T_x=55$ K.[23,24] In $Er_5Ir_4Si_{10}$ x-ray investigations on single crystals proved a transition toward an incommensurate CDW at T_0 followed by a lock-in transition into a commensurate CDW at T_x.[23] On the other hand classical structural transitions are in some cases also connected with a similar steplike increase in $\rho(T)$, as, e.g., in the compounds $R\mathrm{InAu}_2$.[25] A characteristic difference is the size of the structural distortion. Classical structural transitions, as, e.g., in $R\mathrm{InAu}_2$ or $R\mathrm{Cu}_6$, can be recognized in powder x-ray diffraction patterns (PXRD), since the changes in the structure are quite sizeable and associated with a lattice distortion easily observable even in powder data through the splitting of some Bragg peaks.[25,26] However, CDWs lead only to the formation of weak superstructure peaks, which are too small to be seen in PXRD and require high sensitive measurements on single crystals. Therefore, we performed temperature-dependent PXRD studies to look for a signature of both transitions. These measurements on the available polycrystals did not resolve any splitting or appearance of additional Bragg peaks, proving that the deformations of the structure at T_0 and T_x are very small, in accordance with a CDW scenario. We furthermore note that $PrPt_2Si_2$ behaves very similar to $Er_5Ir_4Si_{10}$ not only in the resistivity and in the PXRD but also in the specific heat. The shape and the size (normalized to the number of atoms per formula unit) of the anomaly at T_0 are almost identical while for both compounds no anomalies could be resolved at T_x.

There is no typical dependence of the CDW transition temperature(s) on pressure reported, e.g., Refs. 14 and 27. The different pressure response of the two CDW transitions in $PrPt_2Si_2$ observed in our experiments is most likely to be associated with critical changes in the band structure. The pressure dependence of $T_0(p)$ can be described on the basis of pressure promoted band broadening due to which the CDW state becomes unstable hence the CDW transition temperature becomes smaller.[27]

V. SUMMARY

To summarize, $PrPt_2Si_2$ exhibits sharp and well-defined anomalies in electrical-resistivity and specific-heat data at $T_0=88$ K associated with a first-order phase transition. Since no corresponding anomaly was observed in the magnetic susceptibility, the transition is most likely not related to a magnetic phase transition. Further, the low temperature x-ray diffraction data clearly show that the specific-heat and resistivity anomalies are also not related to a large, classical structural phase transition. We, therefore, argue that the anomaly found in resistivity and specific heat is connected to the appearance of a gap in the density of states at Fermi surface resulting from a CDW transition in $PrPt_2Si_2$. We surmise that the anomaly at T_x, only observable in the electrical resistivity is related to the lock-in on the CDW, as observed at ambient pressure for $Er_5Ir_4Si_{10}$.[12] However, further experiments are needed to clarify the origin of the kink in $\rho(T)$ at T_x. Application of pressure enhances the bandwidth and, therefore, reduces the density of states which results in a weakening of the CDW. The distinct pressure dependence of the two transitions is likely related to complex changes in the topology of the Fermi surface. $PrPt_2Si_2$ appears to be an interesting system to study the mechanism behind the density-wave state by excluding the complexities related to the magnetism and superconductivity and thus deserves further investigations.

ACKNOWLEDGMENTS

We thank H. Borrmann for conducting the low-temperature x-ray diffraction measurements. Z.H. and V.K.A. acknowledge the UGC-DAE CSR, Indore for providing access to the low-temperature measurement facility using PPMS.

*nicklas@cpfs.mpg.de

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