Interplay between charge density wave order and superconductivity in $LaAuSb_2$ under pressure

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We report superconductivity below $T_c = 0.64$ K in the charge density wave (CDW) ordered material $LaAuSb_2$, from measurements of the electrical resistivity, specific heat, and ac magnetic susceptibility. To investigate the interplay between superconductivity and CDW order in $LaAuSb_2$, we measured the resistivity under pressures up to 2.0 GPa and constructed the temperature-pressure phase diagram. With the application of pressure, T_c increases gradually before exhibiting a sudden jump at around 0.64 GPa, while the CDW order is suppressed to lower temperatures before abruptly vanishing at the same pressure. We suggest that the jump of T_c may be due to the enhancement of the density of states with the closure of the CDW energy gap when CDW order is suppressed. On the other hand, the normalized upper critical field H_{c2} changes little with pressure, suggesting that orbital limiting is the dominant pair-breaking mechanism in $LaAuSb_2$.

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I. INTRODUCTION

The nature of the complex interplay between different coexisting electronic states in correlated systems remains an open topic in condensed matter physics. A prime example is the competition or coexistence between magnetic order and superconductivity (SC) in heavy fermions and iron pnictides [1–4]. In these systems, quantum critical points (QCPs) surrounded by a superconducting dome are revealed when the magnetic order is suppressed to zero temperature by nonthermal parameters such as pressure, magnetic fields, and doping. The interplay between charge-density-wave (CDW) order and SC has also been of great interest [5–12] since their coexistence in some systems leads to similar phase diagrams to those of heavy fermion and iron pnictide systems with magnetism and SC.

CDW order corresponds to a periodic modulation of the conduction electron density, which was initially found in low-dimensional materials [5,6]. The coexistence of CDW and SC has been reported in some systems, where SC is typically enhanced upon the suppression of CDW order [5,6], which can be well understood from Bardeen-Cooper-Schrieffer (BCS) superconducting theory since the suppression of CDW order leads to a closure of the the CDW gap on the Fermi surface, and hence enhances the density of states at the Fermi level $N(E_F)$. However, in some other cases, the interplay between CDW and SC is more complicated [7–12]. For instance, a CDW QCP is reported in $Lu(Pt_{1-x}Pd_x)_2In$ [9], where critical fluctuations are considered to play an important role in the enhancement of T_c. A clear difference between magnetic QCPs and CDW QCPs is that non-Fermi liquid behavior is usually absent in the case of the latter [9,10]. However, whether the superconducting dome is associated with the CDW QCP still remains controversial. For instance, the superconducting dome was found to be separate from the CDW QCP in pressurized 1T-$TiSe_2$ [11]. Therefore, whether critical quantum fluctuations can promote SC in the vicinity of a CDW QCP still remains unresolved.

$LaAuSb_2$ belongs to the $LaTSb_2$ (T = Ag, Au, Cu) family crystallizing in the $ZrCuSi_2$-type structure [13]. $LaAgSb_2$ exhibits two CDW transitions at $T_{CDW1} = 207$ K and $T_{CDW2} = 186$ K, while SC is absent down to 0.3 K [14,15]. Upon applying pressure, T_{CDW1} is suppressed to 120 K at around 2.12 GPa, without the emergence of SC [16]. On the other hand, no signature of CDW order is detected in $LaCuSb_2$, and there is a superconducting transition at $T_c = 0.9$ K [17]. Recently, a CDW transition at $T_{CDW} \approx 88$ K was reported in $LaAuSb_2$, together with the absence of SC down to 2 K [13]. However, whether there is a superconducting transition in $LaAuSb_2$ at lower temperatures still remains unknown. In addition, the lattice volume of $LaAuSb_2$ is smaller than $LaAgSb_2$, but larger than $LaCuSb_2$. These results suggest that pressure may significantly affect the charge density wave order and SC in $LaTSb_2$. In this work, we report the discovery of superconductivity with $T_c = 0.64$ K in $LaAuSb_2$ at ambient pressure from measurements of the electrical resistivity, specific heat, and ac magnetic susceptibility. Based on resistivity measurements under pressure up to 2.0 GPa, we construct the temperature-pressure phase diagram of $LaAuSb_2$.

II. EXPERIMENTAL METHODS

Single crystals of $LaAuSb_2$ were synthesized using the self-flux method described in Ref. [13]. The heat capacity down to 0.35 K was measured using a Quantum Design

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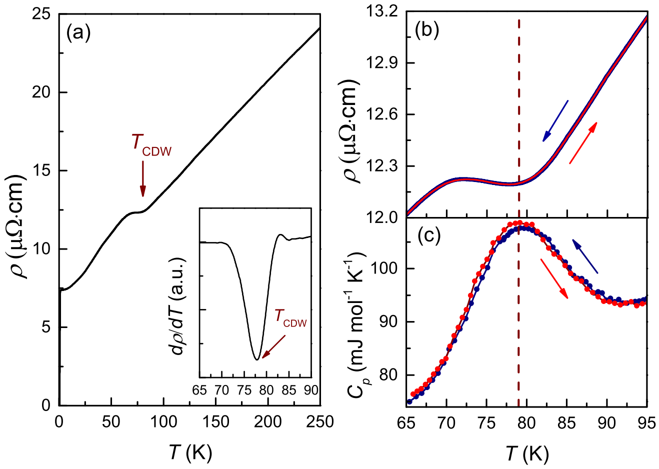

FIG. 1. (a) Resistivity of $LaAuSb_2$ at ambient pressure as a function of temperature $\rho(T)$ from 250 K down to 0.3 K. (inset) $d\rho/dT$, which was used to define T_{CDW}. Temperature dependence of the (b) resistivity and (c) heat capacity C_p, both measured upon warming (red lines) and cooling (navy lines) which show the presence of a CDW transition at around 78 K, as marked by the vertical dashed line.

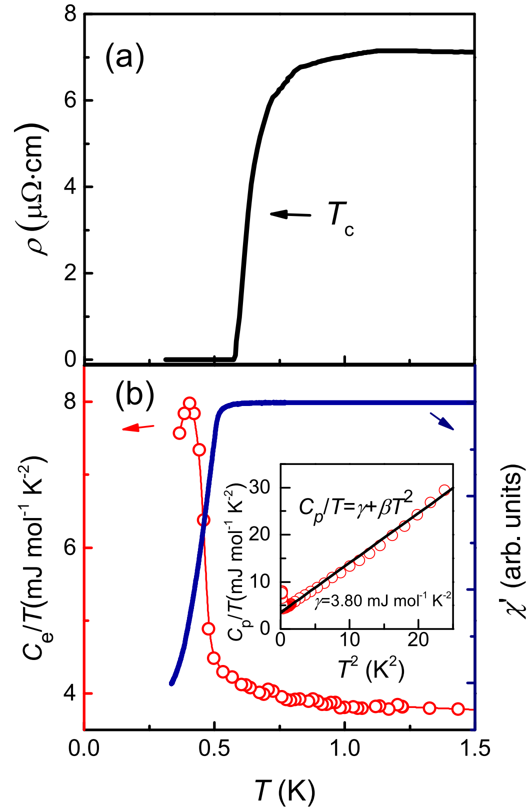

FIG. 2. (a) Low temperature $\rho(T)$ of $LaAuSb_2$, which shows the presence of a superconducting transition. The arrow marks the position of T_c, corresponding to the midpoint of the resistivity drop. (b) Electronic contribution to the heat capacity as C_e/T and the ac susceptibility $\chi_{ac}(T)$ at low temperature, where the inset shows $C_p(T)/T$ versus T^2 and the corresponding fitting with $C_p/T = \gamma_n + \beta T^2$.

Physical Property Measurement System (PPMS) with a ^3He insert, using a standard pulse relaxation method. The ac susceptibility and resistivity measurements were performed in a Teslatron-PT system with an Oxford ^3He refrigerator, with a temperature range of 0.3 to 300 K and a maximum applied magnetic field of 8 T. Resistivity measurements using the standard four-probe method under pressure were carried out using a piston-cylinder-type pressure cell up to 2.0 GPa, and Daphne 7373 was used as the pressure-transmitting medium to obtain good hydrostaticity. The applied pressure was determined by the shift in T_c of a high-quality Pb single crystal.

III. RESULTS

Figure 1(a) shows the temperature dependence of the resistivity $\rho(T)$ measured from 250 K down to 0.3 K, which exhibits a phase transition near 78 K, consistent with heat capacity measurements [Fig. 1(c)]. In this context, the minimum of $d\rho/dT$ is used to define T_{CDW}, as shown in the inset of Fig. 1(a). The derived value of the CDW-ordering temperature T_{CDW} is 78 K, which is compatible with the results reported in Ref. [13]. We note that T_{CDW} in Ref. [13] is determined from the onset temperature of the transition. Both the $\rho(T)$ and heat capacity measurements were performed upon warming up and cooling down between 65 and 95 K as shown in Figs. 1(b) and 1(c). The overlap between the curves indicates the absence of thermal hysteresis, suggesting that the CDW transition may be second order or weakly first order.

Figure 2 displays $\rho(T)$, the electronic contribution to the specific heat as $C_e(T)/T$ and the ac magnetic susceptibility $\chi_{ac}(T)$ at low temperature. $LaAuSb_2$ exhibits a superconducting transition at $T_c \approx 0.64$ K, which is defined as the midpoint of the resistivity drop in Fig. 2(a). The onset temperatures of the superconducting transition in the specific heat and ac magnetic susceptibility are around 0.56 K, close to 0.59 K where the resistivity drops to zero. The slight variations of T_c between different measurements may be due to the resistivity being more sensitive to the sample surface, while the former two techniques reflect the bulk properties. The specific heat in the normal state was fitted using $C_p/T = \gamma_n + \beta T^2$ as shown in the inset of Fig. 2(b), where γ is the Sommerfeld coefficient and β represents the lattice contribution, yielding $\gamma = 3.80\,\text{mJ mol}^{-1}\ \text{K}^{-2}$ and $\beta = 1.037\ \text{mJ mol}^{-1}\ \text{K}^{-4}$. A Debye temperature $\Theta_D = 196$ K was calculated from $\Theta_D = (12\pi^4 nR/5\beta)^{1/3}$, where $n = 4$ is the number of atoms per formula unit and R is the molar gas constant. From this, we estimate an electron-phonon coupling strength $\lambda_{ep} \approx 0.44$ from the McMillan formula [18] assuming a Coulomb pseudopotential $\mu^* = 0.13$. The relatively small value of λ_{ep} indicates that $LaAuSb_2$ is a weakly coupled superconductor. The specific heat jump at T_c is determined to be $\Delta C/\gamma T_c = 1.13$, being slightly smaller than the BCS value of 1.43 which may be due to gap anisotropy or multigap SC.

Figure 3 displays the resistivity from 150 to 0.3 K under various pressures. It can be seen that the CDW transition broadens and is gradually suppressed to lower temperatures with the application of pressure up to 0.64 GPa, as shown

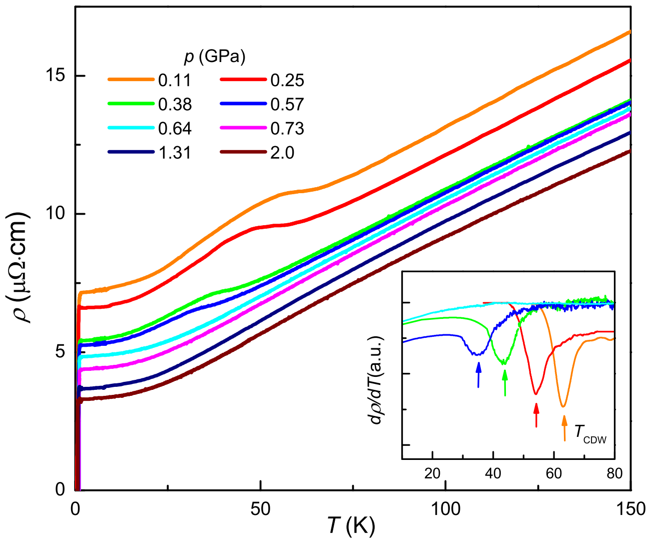

FIG. 3. Temperature dependence of the resistivity of $LaAuSb_2$ under various pressures, measured upon cooling. The inset shows the derivative $d\rho/dT$, where the arrows mark the CDW transition temperatures.

by $d\rho/dT$ in the inset. With further increasing pressure, there is no signature of the CDW transition in the resistivity, indicating that the CDW transition has been totally suppressed by pressure and $\rho(T)$ shows good metallic behavior with an absence of non-Fermi liquid behavior down to the superconducting transition. The low-temperature resistivity under various pressures is shown in Fig. 4(a). With increasing pressure, T_c increases before reaching a maximum value of $T_c^{max} \approx 1.05$ K, and then decreases to 0.65 K at the maximum measured pressure. The maximum T_c is reached close to the pressure where the CDW transition disappears.

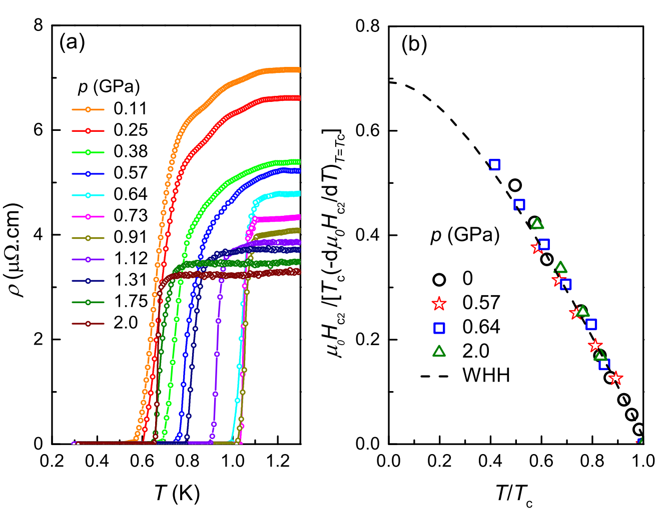

FIG. 4. (a) Low-temperature resistivity of $LaAuSb_2$ under various pressures, showing the evolution of the superconducting transition with pressure. (b) The upper critical field (H_{c2}) of $LaAuSb_2$ as a function of T/T_c at various pressures, normalized by the product of T_c and the slope of H_{c2} near T_c. The calculated curve from the WHH model (dashed line) is also displayed.

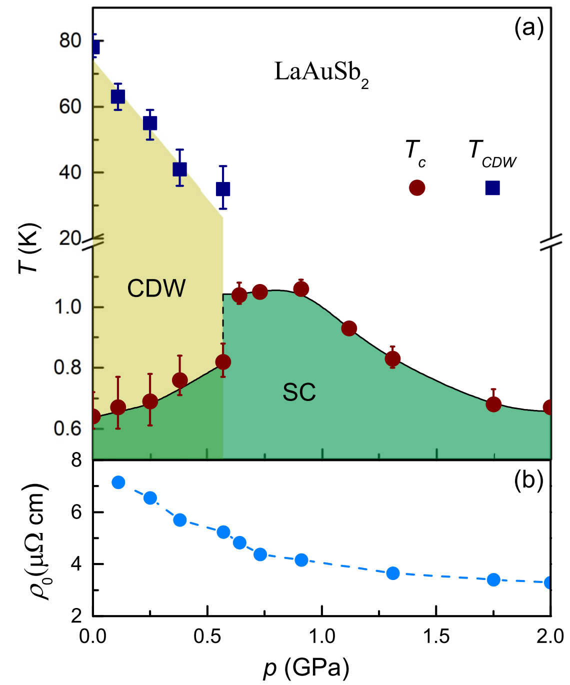

FIG. 5. (a) Temperature-pressure phase diagram of $LaAuSb_2$. T_{CDW} is determined from the derivative of the resistivity, where the error bars in T_{CDW} correspond to the full width at half minimum of the feature at the transition in $d\rho/dT$. T_c from $\rho(T)$ corresponds to the temperature where there is a drop to 50% of the normal-state value. The error bars in the T_c of the resistivity represents where there is a 10% and 90% drop of the resistivity. The error bars for the pressure are all less than 0.015 GPa, and are, therefore, smaller than the symbol size. (b) Pressure dependence of the residual resistivity ρ_0.

Figure 4(b) shows the values of upper critical field H_{c2} plotted as a function of T/T_c at 0, 0.57, 0.64, and 2.0 GPa from resistivity measurement under applied magnetic fields, and they are normalized by the product of T_c and the derivative of H_{c2} near T_c. It can be seen that the normalized H_{c2} vs T/T_c overlap for the different pressures and are fitted well with the Werthamer-Helfand-Hohenberg (WHH) model shown by the dashed line [19]. This suggests that orbital limiting is the dominant pair breaking mechanism in $LaAuSb_2$ at all pressures.

From the resistivity measurements under pressure, we constructed the temperature-pressure phase diagram for $LaAuSb_2$, which is displayed in Fig. 5(a). The CDW order is gradually suppressed almost linearly with the application of hydrostatic pressure, similar to that observed in $LaAgSb_2$ [16,20]. Above around 0.64 GPa, the CDW transition is no longer observed. Meanwhile, the residual resistivity ρ_0 continuously decreases across the whole pressure range. On the other hand, T_c initially increases with pressure before reaching a maximum value at around the pressure where CDW order disappears, then slowly decreases up to the maximum measured pressure. Upon the suppression of the CDW transition, T_c exhibits an abrupt increase, which is similar to other examples of systems with SC competing with CDW order [6,21,22].

IV. DISCUSSION AND SUMMARY

For pressures lower than 0.64 GPa, the CDW transition temperature is suppressed linearly with pressure, similar to $LaAgSb_2$ [20], while the value of dT_{CDW}/dP is around −0.82 K/GPa, which is about two times larger. This indicates that the CDW order in $LaAuSb_2$ is more sensitive to the application of pressure. Furthermore, it was found that the charge carrier density of $LaAuSb_2$ at ambient pressure shows a drop of about 22% across T_{CDW} [13], and therefore it is expected that once CDW order is suppressed by pressure, there will be an enhancement of the density of states. The residual resistivity decreases more rapidly with pressure in the CDW state than it does once CDW order is suppressed, which could be related to the closing of the CDW gap upon suppressing the CDW order. T_c exhibits an abrupt jump at 0.64 GPa upon the disappearance of CDW order, which may be due to the sudden closure of the CDW gap, leading to an enhancement of $N(E_F)$. Upon further increasing the pressure, T_c decreases slowly which has also been reported in other materials such as $LaPt_2Si_2$ [23]. This may be due to band broadening which reduces the density of states upon compressing the lattice. However, further band-structure calculations are required to determine the origin of this behavior.

In $LaTSb_2$ (T = Ag, Au, Cu), the differences between the CDW transition temperatures of these three compounds has been attributed to different dimensionalities of the electronic structures [16,24,25]. From band-structure calculations, the Fermi surfaces of $LaAgSb_2$ show a strongly two-dimensional character and it exhibits the highest CDW transition temperature, while $LaCuSb_2$ is more three-dimensional and has no CDW transition [24]. In $LaAg_{1-x}T_xSb_2$ (T = Au, Cu), the CDW transition temperatures are suppressed upon decreasing c/a [25]. Furthermore, the results from applying pressure to $LaAgSb_2$ are very similar to those of the doping study of $LaAg_{1-x}Cu_xSb_2$ [16,25]. These results suggest that the dimensionality might be the possible reason for the suppression of T_{CDW} in the compounds $LaTSb_2$. Note that it was proposed that a reduction of the distance between the Sb planes around the transition metals may cause the suppression of T_{CDW} in these systems [25]. However, further bulk modulus experiments and pressure experiments on doped materials are required to determine the mechanism behind these behaviors. On the other hand, the absence of SC in $LaAgSb_2$ above 0.3 K might arise from the relatively low $N(E_F)$. Although the Debye temperature of $LaAgSb_2$ is 249 K [17], which is a bit higher than in $LaAuSb_2$, the γ of $LaAgSb_2$ is around 2.62 mJ mol^{-1} K^{-2} [17] which is considerably lower. It is possible that the superconducting transition temperature of $LaAgSb_2$ is well below 0.3 K and the application of pressure may enhance the superconductivity. To further address the interplay of superconductivity and CDW in $LaTSb_2$, it is desirable to study $LaAgSb_2$ under pressure at lower temperatures. Finally, further studies of the dynamics of the CDW order in $LaAuSb_2$ is also highly desirable to gain further comprehension of the relationship between CDW and SC in $LaAuSb_2$.

In summary, we reported the observation of superconductivity at $T_c = 0.64$ K in the CDW-ordered material $LaAuSb_2$ and determined its temperature-pressure phase diagram. With the application of hydrostatic pressure, T_c increases gradually before showing an abrupt jump at around 0.64 GPa, while the CDW order is suppressed to lower temperatures before vanishing above the same pressure. We suggest that the jump of T_c is related to the closure of the energy gap, when the CDW order is fully suppressed by pressure. Further doping and pressure experiments would be useful to gain a deeper understanding of the interplay between CDW order and superconductivity in $LaTSb_2$.

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