Pressure effect on competition between charge density wave and superconductivity in $ZrTe_3$: Appearance of pressure-induced reentrant superconductivity

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We report on an intricate competition between charge density wave (CDW) formation and superconductivity under pressure up to 11 GPa in the low-dimensional conductor $ZrTe_3$. As pressure increases, the CDW transition temperature T_{CDW} initially increases, then begins to decrease at 2 GPa and abruptly disappears near 5 GPa. On the other hand, while the superconducting transition temperature T_C falls to below 1.2 K at ~0.5 GPa and is not observed at up to 5 GPa above 2.5 K, a superconducting transition emerges beginning at ~5 GPa and T_C increases steeply up to 11 GPa. This is an observation of pressure-induced reentrant superconductivity. The results are discussed in terms of the change in the reduced area of the Fermi surface due to CDW formation.

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The transition metal trichalcogenide $ZrTe_3$ undergoes a phase transition at 63 K due to charge density wave (CDW) formation.[1–3] The crystal structure of $ZrTe_3$ consists of infinite chains formed by stacking $ZrTe_3$ prisms along the b direction.[4] The electrical resistivity in the metallic phase is anisotropic with ($\rho_a:\rho_b:\rho_c=1:1:10$) and only ρ_a and ρ_c exhibit a hump anomaly in resistivity due to CDW formation. This is in contrast to the case in isostructural $NbSe_3$, which is a well-known quasi-one-dimensional (1D) CDW compound that exhibits a resistivity anomaly along the chain, i.e., in the b direction.[5,6] Direct measurements of superlattice spots due to electron and x-ray diffraction have shown that the CDW nearly commensurate with the underlying lattice and have revealed the CDW q vector in the a^*-c^* plane.[7,8] Surprisingly, no b^* component is observed in the q vector. The q vector determined experimentally agrees well with the nesting vector deduced from the calculated Fermi surface (FS).[9,10] These results show that CDW formation for $ZrTe_3$ is driven by a nesting of FS. At a low temperature near 2 K, $ZrTe_3$ becomes superconductive,[1,3] i.e., the remnant FS after CDW formation is responsible for the superconductivity. This is consistent with the prediction based on a band-structure calculation in which the CDW and the superconductivity coexist on different portions of the FS.[9,10]

The pressure dependencies of the CDW transition temperature T_{CDW} and the superconducting transition temperature T_C are unusual, at least up to 1.1 GPa.[11–13] CDW formation is enhanced by pressure and T_{CDW} reaches ~105 K at 1.1 GPa,[13] while superconductivity is suppressed and T_C decreases to 1.2 K at 0.5 GPa. This is in contrast to the general case of competition between superconductivity and CDW formation under pressure, where CDW formation is suppressed by an increase in three-dimensionality due to the application of pressure and T_C is enhanced by the restoration of the density of states at the Fermi level E_F.[14–17] A greater pressure is expected to raise the three-dimensionality in $ZrTe_3$. Therefore, it would be very interesting to investigate the unusual pressure dependencies of T_{CDW} and T_C at high pressure. In this study, we investigated the pressure dependencies of T_{CDW} and T_C over a wide pressure range up to 11 GPa using a cubic anvil pressure cell to examine the possible existence of competition between superconductivity and CDW formation.

The single crystal used in the present study was prepared by the iodine transport method.[1] Typical crystal dimensions were on the order of $0.7\times0.1\times0.05$ mm^3, where the long axis is the crystallographic a axis. For each sample, the resistance was measured along the a axis using a standard four-probe configuration. Electrical contacts were prepared by evaporating indium (In) in a vacuum. Electrical leads, 20 μm Au wires, were glued to the evaporated In contacts with a conducting silver (Ag) paste. All measurements were carried out under a current-controlled bias. Resistance was measured in the temperature range of 2.5–300 K. External pressure from 2 to 11 GPa was generated using a cubic anvil pressure cell.[18] Nearly hydrostatic pressure was produced in a Teflon cell filled with a fluid pressure-transmitting medium consisting of a 1:1 mixture of Fluorinert FC70 and FC77.

The temperature dependence of resistance was measured under various pressures between 2 and 11 GPa for three samples. Figure 1 shows the typical high-temperature resistance measured for sample B of $ZrTe_3$ crystal. Under 3 GPa, the resistance increases sharply at 114 K due to CDW formation. This resistance anomaly is quite similar to those observed under pressures below 1.1 GPa[11–13] and it becomes smeared with increasing pressure. T_{CDW} is defined here as the temperature at which the temperature derivative of the resistance exhibits a minimum. T_{CDW} decreases monotonously with increasing pressure up to 5 GPa, but abruptly decreases above 5 GPa. At $P=5.5$ GPa, no resistance anomaly due to CDW formation is observed down to 4 K, but a gradual drop in resistance is observed at temperatures below 3.5 K.

Figure 2(a) shows the typical low-temperature resistance measured for sample B of $ZrTe_3$ crystal. No drop in resis-

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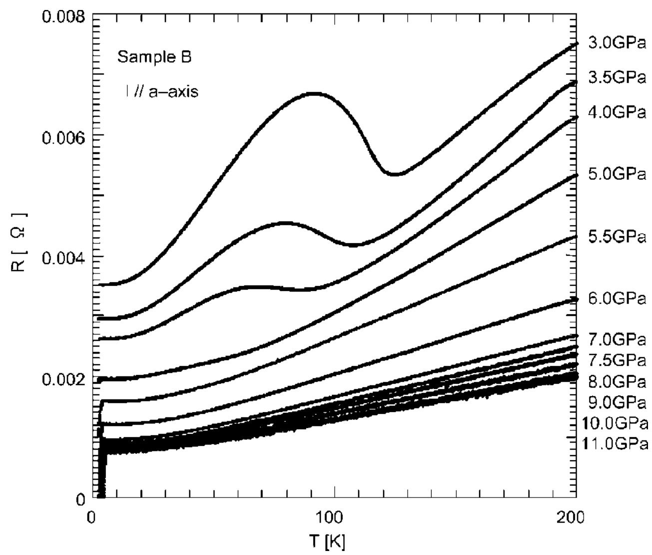

FIG. 1. High-temperature resistance for sample B of $ZrTe_3$ measured under various pressures.

tance was observed above 2.5 K in the pressure range between 3 and 5 GPa. As the pressure was increased over 5.5 GPa, the resistance drop became large and sharp, and zero resistance was clearly observed above $P=7$ GPa. Figure 2(b) shows the zero-resistance transition at various magnetic fields H applied parallel to the c^* axis. Here, a finite resistance remained below 3 K is due a strong current dependence of the resistance as shown in the inset of Fig. 2(b). The zero-resistance transition is lowered with increasing H. At $H=2.5$ T the resistance above 3 K almost restores to the value of the normal resistance. From these findings it can be safely concluded that $ZrTe_3$ again undergoes a superconducting phase transition above 5.5 GPa. The detail superconducting properties will be published elsewhere.[19] The superconducting transition temperature T_C is defined as half of the resistance in the metallic phase at low temperature. T_C increases with increasing pressure and T_C at 11 GPa is more than twice that at ambient pressure. Thus the pressure-induced suppression of CDW and enhancement of superconductivity are observed above 2 GPa, which is contrary to the pressure dependence of CDW and superconductivity observed at pressures below 1.1 GPa.[11–13]

Figure 3(a) shows the pressure-dependencies of T_{CDW} and T_C obtained for three samples, together with those for samples measured earlier at low pressure. As pressure increases, T_{CDW} initially increases, then starts to decrease at ~2 GPa and abruptly disappears to below 2.5 K between 5.0 and 5.5 GPa. Thus, CDW of $ZrTe_3$ is sensitive to the application of external pressure. On the other hand, T_C decreases with increasing pressure and falls below 1.2 K at ~0.5 GPa. No superconducting transition is observed above 2.5 K in the pressure range of 1.1–5 GPa, but a superconducting transition emerges beginning at ~5 GPa. T_C increases steeply with increasing pressure up to 11 GPa. T_{onset}, which is observed at pressures below 5 GPa, continuously increases with increasing pressure. In contrast to the abrupt disappearance of T_{CDW}, no discontinuous change in T_{onset} is observed near 5 GPa. Furthermore, as shown in Fig. 1 the temperature dependence of resistance in the metallic state varies systematically for pressure from 3.5 to 11 GPa and any anomalous change is not observed around 5 GPa. In addition, resistance at room temperature continuously decreased with increasing pressure up to 11 GPa. These findings strongly suggest that the abruptly disappearance of T_{CDW} near 5 GPa is not due to a structural phase transition. Thus we can conclude that superconductivity observed above 5 GPa is induced by a change in the Fermi surface under pressure, but not by a structural change. This is an observation of pressure-induced reentrant superconductivity. Figure 3(b) shows the pressure dependence of T_C plotted on a linear scale. T_C in the pressure range of 0.5–5 GPa is shown by extrapolation from those at <0.5 GPa and >5 GPa. The minimum T_C can be estimated to be around 2 GPa, which is comparable to the pressure corresponding to the peak of T_{CDW}. Such a remarkable pressure-dependent competition between T_{CDW} and T_C has

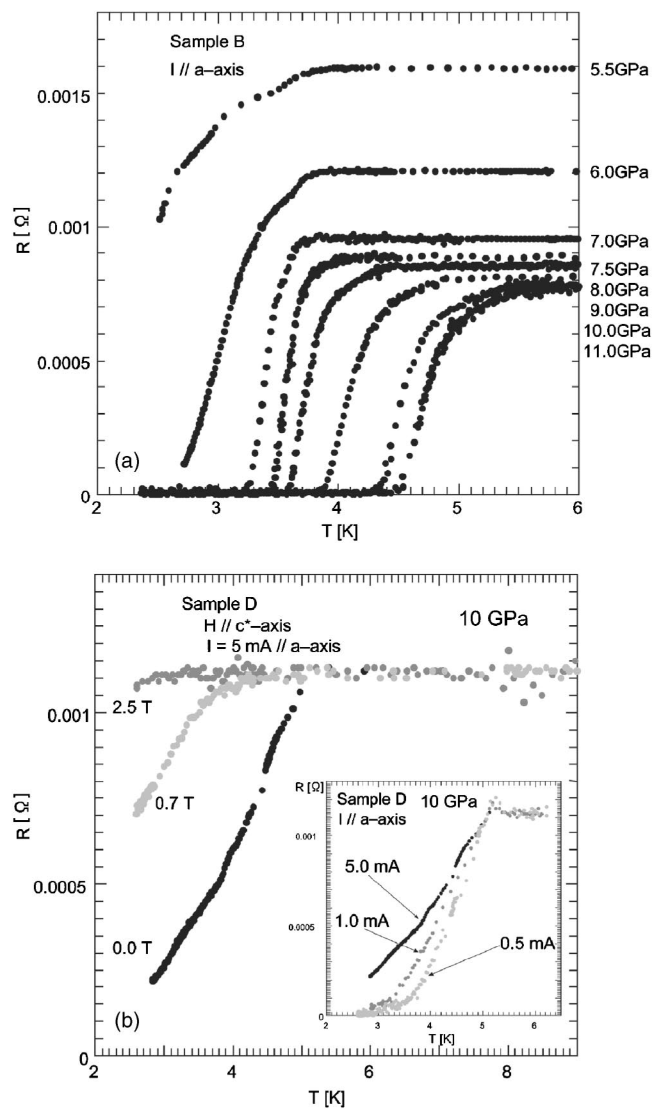

FIG. 2. Pressure dependence of zero-resistance transition for sample B (a) and magnetic field dependence of zero-resistance transition for sample D (b) with field along the c^* axis under 10 GPa. The inset in Fig. 2(b) shows current dependence of the transition at zero magnetic field.

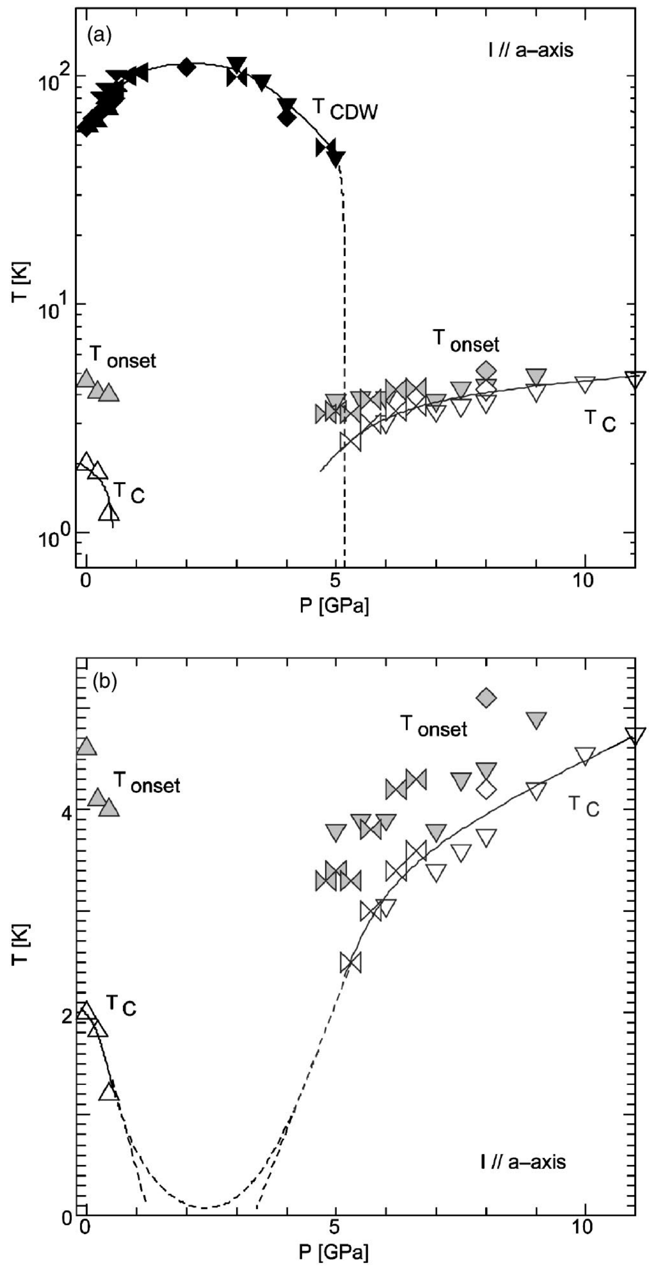

FIG. 3. Pressure dependence of the CDW transition temperature T_{CDW} (closed symbols) and the superconducting transition temperature T_{C} (open symbols) and the onset temperature of superconducting transition T_{onset} (gray symbols) plotted on a logarithmic scale (a) and the pressure dependence of T_{C} and T_{onset} plotted on a linear scale (b) for three samples of $ZrTe_3$. Previous results are also shown at pressure below 1.1 GPa. The solid and dashed lines are guides for the eye.

not been reported for other systems of low-dimensional conductors.

At a temperature where the CDW gap is sufficiently developed, the size of the resistance anomaly reflects the reduction in the density of states at E_F due to CDW formation. The resistance anomaly at T_{CDW} becomes smeared with increasing pressure. Therefore, we can focus on the size of the resistance anomaly to examine the effect of pressure on CDW

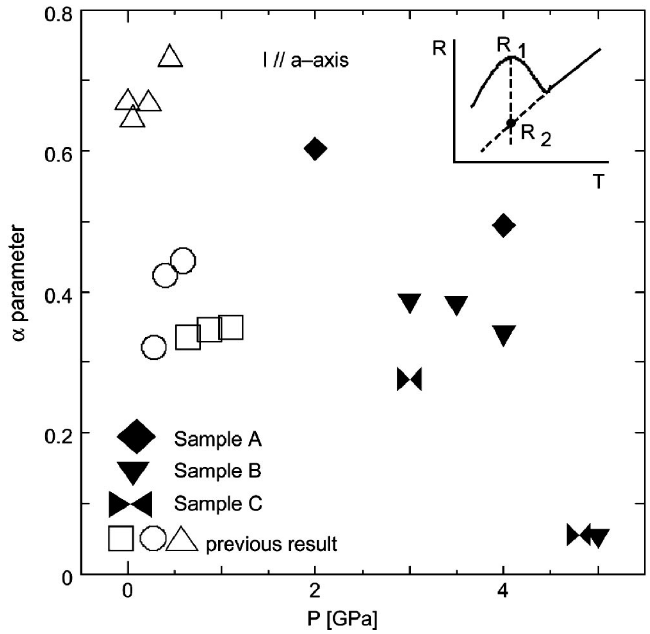

FIG. 4. Pressure dependence of the α parameter for three samples of $ZrTe_3$. Previous results are shown using open symbols.

formation. This is referred to as the α parameter and α is usually defined as

$$\alpha = (R_1 - R_2)/R_1 = (\sigma_2 - \sigma_1)/\sigma_2, \tag{1}$$

where R_1 (σ_1) is the peak resistance (conductivity) in the resistance anomaly and R_2 (σ_2) is the resistance (conductivity) that is expected in the absence of CDW formation.[16,20] A schematic definition of R_1 and R_2 is shown in the inset of Fig. 4. Using the relation of metallic conductivity presented in detail by Kawabata,[20] α can be rewritten as

$$\alpha = \Delta N/N_0, \tag{2}$$

where N_0 is the density of states at E_F in the absence of CDW formation and ΔN denotes a reduction of N_0 due to CDW formation. Thus the α parameter represents the ratio of the reduction of the density of states at E_F due to CDW formation to that in the absence of CDW formation. Since ΔN is proportional to the reduced area of the FS due to CDW formation, α is expected to provide information on the size of the nesting area of the FS.

Figure 4 shows the pressure dependence of α obtained for three samples measured at above 2 GPa, together with those for samples measured earlier at low pressure.[11–13] Although the value of α varies from sample to sample, α initially increases with increasing pressure but then begins to decrease at 2 GPa and finally disappears at ~5 GPa. The pressure dependence of α is quite similar to that of T_{CDW}. The similarity between the pressure dependencies of T_{CDW} and α shows that the remarkable change in T_{CDW} with the application of pressure originates from the change in the size of the nesting area of the FS. On the other hand, superconductivity is expected to form on remnant FS's after CDW formation. The density of states at E_F after CDW formation N_S is given by

$$N_S = N_0 - \Delta N = N_0(1-\alpha). \quad (3)$$

Equation (3) shows that N_S/N_0 can be represented using $(1-\alpha)$. As found easily from Fig. 4, $(1-\alpha)$ decreases below 1.1 GPa and begins to increase at 2 GPa; i.e., the pressure-dependence of $(1-\alpha)$ shows a pattern similar to that of T_C. This similarity between the pressure dependencies of T_C and $(1-\alpha)$ suggests that the remarkable change in T_C under pressure originates from the change in the size of the remnant FS's after CDW formation. The present result confirms that the strong pressure dependence of T_C results from that of N_S, which is caused by the remarkable change in the CDW state due to the application of pressure. This conclusion is a typical example of competition between CDW formation and superconductivity; i.e., T_C is suppressed under conditions in which CDW formation is enhanced and vice versa.

Band-structure calculations predict that the FS of $ZrTe_3$ consists of quasi-1D electron like FS sheets and a 3D hole like FS.[9,10] The former FS sheets have dominant Te 5*px* character originating in the Te(2)-Te(3) chain,[4] while the latter FS has dominant Zr 4*d* character in the center of $ZrTe_3$ prisms. Due to the coexisting quasi-1D and 3D FS's, a van Hove singularity (vHs) is formed in the *k*-space where FS's of differing dimensionality overlap. Thus, a notable feature of the FS of $ZrTe_3$ is the differing dimensionality of quasi-1D, dual quasi-1D+3D and 3D, which was confirmed by a recent study using angle-resolved photoemission spectroscopy (ARPES).[21] The observation of large specific-heat anomaly at the CDW transition also supports existance of the quasi-1D FS.[22] It has been shown theoretically and experimentally that the 1D sheets are responsible for CDW formation, while the vHs with dual quasi-1D+3D character is responsible for superconductivity. Therefore, the competition between CDW formation and superconductivity established in the present study suggests the strong hybridization of Te 5*px* and Zr 4*d* bands.

It is known that pressure affects CDW by modifying the shape of FS. When pressure increases three-dimensionality, the area of the planar portions of FS will decrease with increasing pressure. In this case, CDW formation is suppressed and α decreases. The situation above 2 GPa in $ZrTe_3$ is expected to correspond to this case, where α decreases. In the unconventional case where pressures increases the quasi one dimensionality of the FS, which is observed in some low-dimensional organic conductors,[23] CDW formation will be induced and/or enhanced and α will increase. The situation below 2 GPa in $ZrTe_3$ is considered to correspond to this case, where α increases. However, it is unclear why the one dimensionality of the FS is enhanced easily and significantly with the application of low pressure below 2 GPa. Studies on the effects of pressure on lattice parameters and the anisotropy of resistance are underway.

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