Successive superconducting transitions and Anderson localization effect in Ta₂S₂C

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A complex carbide Ta₂S₂C consists of van der Waals (vdw)-bonded layers with a stacking sequence ... C-Ta-S-vdw-S-Ta-C ... along the c axis. The magnetic properties of this compound have been studied from DC and AC magnetic susceptibility. Ta₂S₂C undergoes successive superconducting transitions of a hierarchical nature at \( T_{cl} = 3.61 \pm 0.01 \) K \( |H_{cl}^{(0)} (0) = 28 \pm 2 \) Oe and \( H_{cl}^{(0)} (0) = 7.7 \pm 0.2 \) kOe \( T_{cu} = 9.0 \pm 0.2 \) kOe \( |H_{cu}^{(0)} (0) = 6.0 \pm 0.3 \) kOe]. The intermediate phase between \( T_{cu} \) and \( T_{cl} \), where \( \delta X (\chi_{F,C} - \chi_{F,C}) \approx 0 \), is an intra-grain superconductive state occurring in the Ta-C layers in Ta₂S₂C. The low temperature phase below \( T_{cl} \), where \( \delta X \) clearly appears, is an inter-grain superconductive state. The magnetic susceptibility \( H_{c} \) well above \( H_{cl}^{(0)} (0) \) is described by a sum of a diamagnetic susceptibility and a Curie-like behavior. The latter is due to the localized magnetic moments of conduction electrons associated with the Anderson localization effect, occurring in the 1T-Ta₂S₂ type structure in Ta₂S₂C.

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I. INTRODUCTION

Ta₂S₂C has a unique layered structure, where a sandwiched structure of C-Ta-S-vdw-S-Ta-C is periodically stacked along the c axis. The magnetic properties of this compound have been studied from DC and AC magnetic susceptibility. Ta₂S₂C undergoes successive superconducting transitions of a hierarchical nature at \( T_{cl} = 3.61 \pm 0.01 \) K \( |H_{cl}^{(0)} (0) = 28 \pm 2 \) Oe and \( H_{cl}^{(0)} (0) = 7.7 \pm 0.2 \) kOe \( T_{cu} = 9.0 \pm 0.2 \) kOe \( |H_{cu}^{(0)} (0) = 6.0 \pm 0.3 \) kOe]. The intermediate phase between \( T_{cu} \) and \( T_{cl} \), where \( \delta X (\chi_{F,C} - \chi_{F,C}) \approx 0 \), is an intra-grain superconductive state occurring in the Ta-C layers in Ta₂S₂C. The low temperature phase below \( T_{cl} \), where \( \delta X \) clearly appears, is an inter-grain superconductive state. The magnetic susceptibility \( H_{c} \) well above \( H_{cl}^{(0)} (0) \) is described by a sum of a diamagnetic susceptibility and a Curie-like behavior. The latter is due to the localized magnetic moments of conduction electrons associated with the Anderson localization effect, occurring in the 1T-Ta₂S₂ type structure in Ta₂S₂C.

The pristine TaC\(_{1-x}\) is a superconductor with a fairly high \( T_c \) \( (9.7 \) K) for \( x \approx 1 \), but for \( x \approx 0.8 \) no superconductivity is found.\(^{12,13}\) The small change of the Fermi surface due to the nonstoichiometry of the carbides considerably reduces the bulk phonon anomaly (dip) resulting in the reduction of \( T_c \).\(^{14}\) In the pristine 1T-Ta₂S₂ the charge density wave (CDW) becomes commensurate with an undistorted host lattice in a first order transition below 200 K.\(^{15}\) The electrical resistivity increases about 10-fold at the 200 K transition and below 2 K the resistivity diverges following the relation \( \rho = \rho_0 \exp[(T_0/T)^n] \) with \( n = 1/3 \), where \( \rho_0 \) is a constant resistivity and \( T_0 \) is a characteristic temperature.\(^{16}\) This is characteristic of the Anderson localization of the conduction electrons due to a random potential. The susceptibility shows a Curie-like behavior due to the localized magnetic moments of conduction electrons.\(^{17,18}\)
II. EXPERIMENTAL PROCEDURE

Powdered samples of Ta$_2$S$_2$C were prepared by Pablo Wally. The detail of the synthesis and structure is described by Wally and Ueki. X-ray powder diffraction pattern shows that Ta$_2$S$_2$C sample consists of a 3R phase as a majority phase and a 1T phase as a minority phase. The sample characterization was also carried out by scanning tunneling microscopy and X-ray photoelectron spectroscopy. The measurements of DC and AC magnetic susceptibility were carried out using a SQUID magnetometer (Quantum Design MPMS XL-5). A polycrystalline powdered sample (mass 253.2 mg) was used in the present work. Before setting up a sample at 298 K, a remnant magnetic field was reduced to less than 3 mOe using an ultra-low field capability option. For convenience, hereafter this remnant field is noted as the state $H = 0$. The detail of the measurements of DC and AC magnetic susceptibility is described in Sec. III.

III. RESULT

A. $\chi'$ and $\chi''$

Figure 1 shows the $T$ dependence of the AC magnetic susceptibility [(a) the dispersion $\chi'$ and (b) the absorption $\chi''$], where the AC frequency $f$ (= 1 Hz) and the magnitude of the AC field ($h = 0.5$ Oe) are used. The $T$ dependence of $\chi'$ and $\chi''$ is strongly dependent on $H$. The sign of $\chi'$ is negative at least for $1.9 < T < 5$ K, while the sign of $\chi''$ is positive. For $H = 0$ (see the inset of Fig. 1(a)), $\chi'$ increases with increasing $T$. It shows a kink at a critical temperature $T_{cl}$ (= 3.61 K) where the derivative $d\chi'/dT$ undergoes a discontinuous jump. The system undergoes a superconducting transition at $T_{cl}$. The dispersion $\chi'$ increases with further increasing $T$ and tends to zero around $T_{cu}$ (= 9.0 K). Similar kink behavior is observed at higher $H$, although the kink is hardly seen in Fig. 1(a) because of very small magnitude of $\chi'$ at $T_{cl}(H)$. The critical temperature $T_{cl}(H)$ decreases with increasing $H$, forming the $H$-$T$ phase diagram (see Sec. III.B). The absorption $\chi''$ at $H = 0$ shows a drastic decrease around $T_{cl}$. It shows a tail above $T_{cl}$ and is reduced to zero around 5 K. The temperature at which the tangential line $\chi''$ vs $T$ with the steepest slope intersects the $\chi'' = 0$ axis coincides with $T_{cl}$.

B. $M$-$H$ loop

Figure 2(a) shows the hysteresis loop of the magnetization $M$ at $T = 1.9$ K. After the sample was quenched from 298 to 1.9 K at $H = 0$, the measurement was carried out with varying $H$ from 0 to 1 kOe at $T$, from $H = 1$ to -1 kOe, and from $H = -1$ to 1 kOe. The $M$-$H$ curve at 1.9 K shows a large hysteresis and a remnant magnetization. Structural imperfections or defect in the sample may play a role of flux pinning, resulting in an inhomogeneous type-II superconductor. Figure 2(b) shows typical data of zero-field cooled (ZFC) magnetization $M_{ZFC}$ vs $H$ at various $T$. Before each measurement, the sample was kept at 20 K at $H = 0$ for 20 minutes and then it was quenched from 20 K to $T$ ($< 4$ K). The magnetization $M_{ZFC}$ at $T$ was measured with increasing $H$ ($0 \leq H \leq 120$ Oe). The magnetization $M_{ZFC}$ exhibits a single local minimum at a characteristic field for $T < T_{cl}$, shifting to the low-$H$ side with increasing $T$. The lower critical field $H_{c1}(T)$ is defined not as the first minimum point of the $M_{ZFC}$ vs $H$, but as the first deviation point from the linear portion due to

FIG. 1: $T$ dependence of (a) $\chi'$ and (b) $\chi''$ with and without $H$ for Ta$_2$S$_2$C. $h = 0.5$ Oe. $f = 1$ Hz. The inset shows the detail of $\chi'$ at $H = 0$ around $T_{cl}$.
The penetration of magnetic flux into the sample: typically, $H_{cl}^{(i)}(T = 1.9\text{K}) = 20\text{ Oe}$ and $H_{cl}^{(i)}(T = 2.7\text{K}) = 12\text{ Oe}$. The least-squares fit of the data of $H_{cl}^{(i)}$ vs $T$ to a conventional relation $H_{cl}^{(i)}(T) = H_{cl}^{(i)}(0)[1 - (T/T_{cl})^2]$ yields $H_{cl}^{(i)}(0) = 28 \pm 2\text{ Oe}$.

**C. $\chi_{ZFC}$ and $\chi_{FC}$**

The measurements of $\chi_{ZFC}$ ($= M_{ZFC}/H$) and $\chi_{FC}$ ($= M_{FC}/H$) were carried out as follows, where $M_{FC}$ is the field-cooled (FC) magnetization. After annealing at 50 K for 1200 sec in the absence of $H$, the sample was quenched from 50 to 1.9 K. The magnetic field $H$ was applied at 1.9 K and then $\chi_{ZFC}$ was measured with increasing $T$. The sample was again heated up and annealed at 50 K for 1200 sec in the presence of $H$. Then $\chi_{FC}$ was measured with decreasing $T$. Figures 2(a) and (b) show the $T$ dependence of $\chi_{ZFC}$, $\chi_{FC}$, and $\delta \chi$ at $H = 1\text{ Oe}$, where $\delta \chi = \chi_{FC} - \chi_{ZFC}$. The scale of the susceptibility is enlarged in Fig. 2(b). The susceptibility $\chi_{ZFC}$ ($\chi_{FC}$) exhibits a kink at $T_{cl} (= 3.61\text{ K})$, where $d\chi_{ZFC}/dT$ ($d\chi_{FC}/dT$) undergoes a drastic decrease. The deviation of $\chi_{ZFC}$ from $\chi_{FC}$ is clearly seen below $T_{cl} (= 3.61\text{ K})$, indicating that the extra magnetic flux is trapped during the FC process. Between $T_{cl}$ and $T_{cu}$ ($= 9.0\text{ K}$), $\delta \chi$ is still positive but nearly equal to zero. The sign of $\chi_{ZFC}$ ($\chi_{FC}$) changes from negative to positive at $T_{cu}$. Similar behavior is also observed at $H = 20\text{ Oe}$ as shown in Figs. 2(c) and (d). Figure 2 shows the $T$ dependence of $\delta \chi$ at various $H$. The value of $\delta \chi$ at fixed $T$ decreases with increasing $H$. The difference $\delta \chi$ undergoes a drastic decrease at $T_{cl}(H)$.

The minimum value of $\chi_{ZFC}$ at $H = 1\text{ Oe}$ is $-1.7 \times 10^{-3}\text{ emu/g}$ at 1.9 K, while the minimum value of $\chi'$ at $H = 0$ is $-1.8 \times 10^{-3}\text{ emu/g}$ at 1.9 K. Using the value of $\chi'$ at 1.9 K ($\approx -1.8 \times 10^{-3}\text{ emu/g}$) and the calculated density $\rho_{cal} = 9.23\text{ g/cm}^3$ for 3R-Ta$_2$S$_2$C, the fraction of flux expulsion relative to complete diamagnetism ($\chi_0 = -1/4\pi = -0.0796\text{ emu/cm}^3$) is estimated as 21 %, suggesting that the system consists of small grains. This is in contrast to 38 % of the diamagnetic volume fraction reported for Nb$_2$S$_2$C by Sakamaki et al.

Figures 2(a) and (b) show the $T$ dependence of $\chi_{ZFC}$ at various $H$, where the measurement was carried out between 1.9 and 11 K with increasing $T$. There is a drastic increase in the diamagnetic contribution in $\chi_{ZFC}$ associated with the superconducting transition at $T_{cl}(H)$. Nevertheless, a diamagnetic contribution in $\chi_{ZFC}$ still remains above $T_{cl}(H)$, increases with further increasing $T$, and reduces to a zero at a upper critical temperature $T_{cu}(H)$. For $H = 150\text{ Oe}$, for example, $\chi_{ZFC}$ exhibits a kink at $T_{cl}(H)$. The sign of $\chi_{ZFC}$ changes from negative to positive around 9 K with increasing $T$. At $H = 5\text{ kOe}$, $\chi_{ZFC}$ is positive at least between 1.9 and 6 K, showing a broad peak at 2.65 K. At $H = 10\text{ kOe}$, $\chi_{ZFC}$ decreases with increasing $T$, showing a Curie-like behavior. Figure 2(a) shows the $T$ dependence of $\delta \chi$ at various $H$. The magnitude of $\delta \chi$ between $T_{cu}(H)$ and $T_{cl}(H)$ is very small compared to that below $T_{cl}(H)$, but still show the irreversible effect of magnetization. The difference $\delta \chi$ at fixed $H$ decreases with increasing $T$ and reduces to zero at $T_{cu}(H)$. Note that $\delta \chi$ at fixed $T$ (for example 5 K) increases increasing $H$, showing a maximum around $H = 500\text{ Oe}$, and decreases with further increasing $H$. This feature is in contrast to the $H$ dependence of $\delta \chi$ below $T_{cl}(H)$, which decreases with increasing $H$. Figure 2(b) shows the $T$ dependence of the derivative $d\chi_{ZFC}/dT$ for $5\text{ Oe} \leq H \leq 10\text{ kOe}$. Clearly $d\chi_{ZFC}/dT$ at low $H$ undergoes two step-like changes around $T_{cl}(H)$ and $T_{cu}(H)$. The critical temperatures $T_{cl}(H)$ and $T_{cu}(H)$ decreases with increasing $H$, forming the $H$-$T$ phase diagram (see Sec. III E).
FIG. 3: $T$ dependence of $\chi_{ZFC}$, $\chi_{FC}$, and $\delta \chi = \chi_{FC} - \chi_{ZFC}$ for $H = 1$ Oe. (a) and (b) for $H = 1$ Oe. (c) and (d) for $H = 20$ Oe. $T$ dependence of $\chi_{ZFC}$ ($5 \leq T \leq 10$ K) is also shown for comparison. The definition of $\chi_{ZFC}$ is given in Sec. III D.

D. $\chi_{ZFC}$ in a quasi-equilibrium state

Here we present our peculiar results on the $T$ dependence of $\chi_{ZFC}$. The method of the measurement for $\chi_{ZFC}$ is a little different from that for the conventional $\chi_{ZFC}$. First, the sample was annealed at 50 K for 1200 sec in the absence of $H$ and then it was quenched to 1.9 K. After the sample was kept at 1.9 K for 100 sec in the presence of fixed $H$, it was quickly heated up to $T_0$ between $T_{cl}$ and $T_{cu}$. Then $\chi_{ZFC}$ was measured with increasing $T$ from $T_0$ to 11 K. Figures 4(a)-(d) show the $T$ dependence of $\chi_{ZFC}$, $\chi_{ZFC}$, and $\chi_{FC}$ at various $H$ ($5 \leq H \leq 100$ Oe), where $T_0 = 5$ K. The $T$ dependence of $\chi_{ZFC}$ at the same $H$ is independent of $T_0$ when $T_0$ is at least between 4 and 8 K. The data of $\chi_{ZFC}$ vs $T$ at $H = 1$ and 20 Oe are shown in Figs. 4(b) and (d), respectively. The susceptibility $\chi_{ZFC}$ increases with increasing $T$ and reduces to zero at $T_{cu}(H)$. The magnitudes of $\chi_{ZFC}$, $\chi_{ZFC}^*$, and $\chi_{FC}$ at the same $H$ are strongly dependent on $H$: $\chi_{ZFC} > \chi_{FC} > \chi_{ZFC}$ for $H = 1 - 30$ Oe, $\chi_{FC} > \chi_{ZFC} > \chi_{ZFC}$ for $H = 50$ Oe, and $\chi_{FC} > \chi_{ZFC} = \chi_{ZFC}$ for $H = 100$ and 150 Oe. Note that $\chi_{ZFC}$ at $H = 1$ Oe (see Fig. 3(b)), whose sign is positive, decreases with increasing $T$ and reduces to zero at $T_{cu}$. Between $T_{cl}$ and $T_{cu}$, the space of states is divided into at least three states, the ZFC state, $ZFC^*$ state, and FC state. The system lies in the FC states under the cooling from 11 K, the ZFC state under the slow heating from 1.9 K, and the $ZFC^*$ state under the rapid heating from 1.9 K. The value of $H (= 30$ Oe) is a little higher than the lower critical field $H_{c1}(T = 1.9$K) = 20 Oe. The susceptibility of each state provides a measure for the corresponding induced magnetic flux density $B$ which is trapped in the superconducting grains: $B = H + 4\pi M$. The inequality $\chi_{ZFC}^* > \chi_{FC}$ indicates that the induced magnetic flux density (or the number of fluxoids over the system) in the $ZFC^*$ state is higher than that in the FC state for $H < 30$ Oe. Such a relatively high flux density in the $ZFC^*$ state may be due to a flux compression as a result of the rapid redistribution of the grain-pinned vortices which occurs during a change of $T$ from 1.9 K to $T_0$. This effect exists only at low $H$. In this sense, the...
The present effect is similar to the paramagnetic Meissner effect observed in $\chi_{FC}$ of the pristine Nb$^{20,23}$: $\chi_{FC}$ at low $H$ becomes positive below the superconducting transition temperature. According to Koshelev and Larkin$^{22}$ the surface supercurrent inhomogeneously trap the magnetic flux in the sample interior, as a vortex. In Sec. IV A we discuss a possible distribution of vortices around the grains in the ZFC and FC states below $T_{cl}$ and between $T_{cl}$ and $T_{cu}$.

E. $H$-$T$ diagram

Figure 5 shows the $H$-$T$ phase diagram, where $T_{cl}(H)$ is determined from the data of $\chi'$ vs $T$ and $\chi''$ vs $T$, and $T_{cu}(H)$ is determined from the data of $\chi_{ZFC}$ vs $T$. These lines correspond to the lines $H_{cl}^{(i)}(T)$ and $H_{cl}^{(h)}(T)$. The least squares-fit of the data of $H$ vs $T$ for the line $H_{cl}^{(i)}(T)$ to a conventional relation $H_{cl}^{(i)}(T) = H_{cl}^{(i)}(0)[1 - (T/T_{cl})^2]$ yields $H_{cl}^{(i)}(0) = (7.7 \pm 0.2)$ kOe and $T_{cl} = 3.61 \pm 0.01$ K, where the data of $\chi'$ vs $T$ and $\chi''$ vs $T$ are used. The value of $H_{cl}^{(i)}(0)$ thus obtained is comparable to that estimated using an empirical relation given by Werhammer et al.$^{24}$ $H_{cl}^{(i)}(T = 0K) = -0.69 T_{cl}(dH_{cl}^{(i)}/dT)_{T = T_{cl}}$. In fact, the value of $H_{cl}^{(i)}(0)$ is calculated as $6.3 \pm 0.2$ kOe, where we use $T_{cl} = 3.61$ K and a slope $(dH_{cl}^{(i)}/dT)_{T = T_{cl}} = -2500 \pm 189$ (Oe/K) obtained from the linear relation in $H_{cl}^{(i)}$ vs $T$ in the vicinity of $T = T_{cl}$ and $H_{cl}^{(i)} = 0$.

The coherence length $\xi$ and the magnetic penetration depth $\lambda$ are related to $H_{cl}^{(i)}(0)$ and $H_{cl}^{(i)}(0)$ through relations $H_{cl}^{(i)}(0) = \Phi_0/(2\pi \xi^2)$ and $H_{cl}^{(i)}(0) = (\Phi_0/4\pi \lambda^2)\ln(\lambda/\xi)$, where $\Phi_0 = 2.0678 \times 10^{-7}$ Gauss cm$^2$ is the fluxoid.$^{19}$ When the values of $H_{cl}^{(i)}(0) = (27.7$ Oe) and $H_{cl}^{(i)}(0) = (7730$ Oe) are used, the values of the Ginzburg-Landau parameter $\kappa = (\lambda/\xi)$, $\lambda$ and $\xi$ can be estimated as $\kappa = 20.5 \pm 0.3$, $\xi = 210 \pm 10$ A and $\lambda = 4200 \pm 100$ A. Our results of $T_{cl}$, $H_{cl}^{(i)}(0)$, $H_{cl}^{(h)}(0)$, $\kappa$, $\lambda$, and $\xi$ in Ta$_2$S$_2$C thus obtained are compared to those in Nb$_2$S$_2$C$^{14}$. $T_c = 7.6$ K, $H_{c1}(0) = 227 \pm 4$ Oe and $H_{c2}(0) = 9950 \pm 180$ Oe, $\kappa = 6.37$, $\xi = 182 \pm 2$ A, and $\lambda = 1160 \pm 20$ A. Both the pristine Nb and Ta are superconductive elements with the critical temperatures $T_c$(Nb) = $9.25 \pm 0.02$ K and $T_c$(Ta) = $4.47 \pm 0.04$ K$^{10}$ and have a body-centered cubic structure. We find that the ratio $T_c$(Nb$_2$S$_2$C)/$T_c$(Ta$_2$S$_2$C) = (2.11) is close to the ratio $T_c$(Nb)/$T_c$(Ta) = (2.06). The values of $T_c$ and $H_{c2}$
for Ta$_2$S$_2$C are comparable to those of 2$H_x$-TaS$_x$Se$_{2-x}$
(0.4 < $x$ < 1.8): $T_c$ = 3.9 K and $H_{c2} = 6.7$ kOe for $x$ = 0.8, $T_c = 3.7$ K and $H_{c2} = 9 - 11$ kOe for $x$ = 1, $T_c = 3.9$ K and $H_{c2} = 11.5 - 12.8$ kOe at $x$ = 1.2.

What kind of the superconductivity occurs at $T_{cu}(H)$? The least squares-fit of the data of $H$ vs $T$ for the line $H_{c2}^{(0)}(T)$ to a conventional relation $H_{c2}^{(0)}(T) = H_{c2}^{(0)}(0)[1 - (T/T_{cu})^2]$ yields $H_{c2}^{(0)}(0) = (6.0 \pm 0.3)$ kOe and $T_{cu} = 8.98 \pm 0.06$ K, where the data of $\chi_{ZFC}$ vs $T$ are used. The origin of the superconductivity at $T_{cu}(H)$ may be due to Ta-C layers in Ta$_2$S$_2$C. According to Giorgi et al., the critical temperature $T_c$ of the pristine TaC$_{1-x}$ increases with decreasing $x$ and is equal to 9.0 K at $x = 0.019$. Fink et al.$^{13}$ have reported that the values of $H_{c1}$ and $H_{c2}$ at $T = 1.2$ K for TaC are 220 Oe and 4.6 kOe. These values are in good agreement with the values of $T_{cu}$ and $H_{c2}^{(0)}(0)$ in Ta$_2$S$_2$C. The origin of the superconductivity at $T_{cu}$ and $T_{cl}$ will be discussed in Sec. IV A.

F. $\chi_{ZFC}$ and $\chi_{FC}$ at high $H$ and high $T$

In Fig. 6 we show the $T$ dependence of $\chi_{ZFC}$ and $\chi_{FC}$ at various $H$. The susceptibility $\chi_{FC}$ (also $\chi_{ZFC}$) at $H = 20$ Oe show a sharp peak around 9.5 K and decreases with further increasing $T$. The deviation of $\chi_{FC}$ from $\chi_{ZFC}$ is observed for $15 \leq T \leq 19$ K. The sign of $\chi_{FC}$ changes from positive to negative around 16 K. The discontinuous jump of $\chi$ observed in the vicinity of $\chi = 0$ is an artifact due to the SQUID measurement. In contrast, $\chi_{FC} (= \chi_{ZFC})$ at $H = 10$ kOe decreases with increasing $T$ from 1.9 K and merge to the curves of $\chi_{FC}$ at $H = 20$ Oe above 9.5 K.

Figure 6 shows the $T$ dependence of $\chi_{FC}$ at $H = 10$ kOe for $1.9 \leq T \leq 298$ K. The susceptibility $\chi$ consists of a diamagnetic susceptibility and a Curie-like susceptibility. The susceptibility $\chi_{FC}$ slightly decreases with increasing $T$ above 20 K and reaches a negative constant (diamagnetic susceptibility) above 150 K: $\chi_d = (-1.44 \pm 0.01) \times 10^{-7}$ emu/g at 298 K. Similar $T$ dependence of $\chi$ is observed in 1T-TaS$_2$ below 150 K.$^{17,18}$

A step-like change $\chi$ near 200 K for 1T-TaS$_2$ is due to the CDW transition between a high-temperature incommensurate phase and a low-temperature commensurate phase. As seen in Fig. 6 there is no such anomaly above 20 K in Ta$_2$S$_2$C. The inset of Fig. 6 shows the $T$ dependence of $\chi_{FC}$ at $H = 10$, 20, and 30 kOe. The susceptibility is strongly dependent on $H$ below 10 K, indicating that $M_{FC}$ is nonlinear in $H$ (see also the data of $M_{ZFC}$ vs $H$ in Fig. 7). Note that the discontinuous jump of $\chi_{FC}$ observed in the vicinity of $\chi_{FC} = 0$ is an artifact due to the SQUID measurement. Almost all the data between 10 and 16 K are removed from the figure. The least-squares fit of the data of $\chi_{FC}$ vs $T$ at $H = 10$ kOe for $4 \leq T \leq 43$ K to

$$\chi_{FC} = \chi_0 + C/(T - \Theta),$$

yields the $T$-independent susceptibility $\chi_0 = (-1.80 \pm 0.02) \times 10^{-7}$ emu/g, the Curie-Weiss constant $C = (2.91 \pm 0.04) \times 10^{-6}$ emu K/g, and the Curie-Weiss temperature $\Theta = -2.37 \pm 0.07$ K. The magnitude of $\chi_{ZC}$ is a little larger than that of $\chi_d$ determined above. The susceptibility $\chi_{FC}$ consists of a Curie-like behavior at low $T$ and a diamagnetic contribution at high $T$. We assume that the Curie-like behavior is due to the localized electron spins having the effective magnetic moment $P_{eff} = g\langle S(S + 1) \rangle^{1/2}\mu_B = \sqrt{3}\mu_B$, where the Landé $g$-factor $g = 2$ and the spin $S = 1/2$. This localized magnetic moment could be related to the Anderson localization (see Sec. IV B).

Through the repulsive Coulomb interaction between the electrons, a singly occupied state exists in the vicinity of $\epsilon_F$. Then the number of spins per gram of Ta$_2$S$_2$C ($N_g$) can be estimated as $N_g = 4.67 \times 10^{18}$/g (or 4.3 $\times$
FIG. 7: (a)-(d) \( T \) dependence of \( \chi_{ZFC}^*, \chi_{ZFC}^x, \) and \( \chi_{FC}^* \) at various low \( H \). The definition of \( \chi_{ZFC}^* \) is given in Sec. III D. The measurement of \( \chi_{ZFC}^* \) was made with increasing \( T \) from \( T_0 (= 5 \text{ K}) \) after the sample was quickly heated from 1.9 K (ZFC state) to \( T_0 \).

There may be another possibility that the Curie-Weiss behavior is due to magnetic impurities (for example, Fe\(^{2+}\)), which may be contained in original Ta (typically 15 ppm Fe in the pristine 1T-TaS\(_2\)). However, this possibility may be ruled out in the following way. If each Fe\(^{2+}\) ion has the effective magnetic moment (= 5.4\( \mu_B \)), then the number of Fe\(^{2+}\) spins per gram of Ta\(_2\)S\(_2\)C is estimated as \( N_g = 2.0 \times 10^{19} \) /g. The magnitude of Fe\(^{2+}\) impurities is estimated as 1800 ppm, which is too large compared to \( \approx 15 \text{ ppm as major metallic impurities of Ta}_2\)S\(_2\)C. Similar Curie-Weiss behavior is observed in the susceptibility of 1T-TaS\(_2\): \( \chi_0 = -1.96 \times 10^{-7} \text{ emu/g}, \) \( \Theta = 0.4 \text{ K}, \) and \( C = 0.806 \times 10^{-6} \text{ emu K/g}. \) These values of \( \chi_0, \Theta, \) and \( C \) are comparable to those derived in the present work for Ta\(_2\)S\(_2\)C. The Curie-like behavior in 1T-TaS\(_2\) is not due to magnetic impurities, but due to the localized magnetic moments related to the Anderson localization effect. The diamagnetic susceptibility is common to the 1T-polytypes showing CDW’s. In 1T-TaS\(_2\), the effect of the ordering of the commensurate CDW below 200 K is clearly observed in a discontinuous jump in the electrical resistivity. In Ta\(_2\)S\(_2\)C the \( T \) dependence of the electrical resistivity \( \rho \) has been reported by Ziebarath et al.\(^{17,18}\) using a pressed- and sintered-sample. The resistivity increases with increasing \( T \) for \( 4.2 \leq T \leq 300 \text{ K} \) (\( \rho \approx 0.8 \text{ m\( \Omega \)cm at 4.2 K and 2.2 m\( \Omega \)cm at 300 K} \), showing a metallic behavior. Unlike the resistivity of 1T-TaS\(_2\), no discontinuous change in \( \rho \) has been observed below 300 K.
FIG. 8: $H$-$T$ phase diagram, where $T_{cl}(H)$ is determined from the measurements of $\chi'$ vs $T$ ($\bullet$) and $\chi''$ vs $T$ ($O$), and $T_{cu}(H)$ is a temperature where $\chi_{ZFC}$ becomes zero ($\triangle$). Solid lines are least-squares fitting curves for the data of $T_{cl}(H)$ and $T_{cu}(H)$ to the form $H_{cl}(T) = H_{cl}(0)[1 - (T/T_{cl})^2]$. The fitting parameters are given in the text. $T_{cl} = 3.61$ K. $T_{cu} = 9.0$ K.

FIG. 9: $T$ dependence of $\chi_{FC}$ at various $H$ near $T_{cu}$.

G. $M_{ZFC}$ vs $H$ at low $T$ and high $H$

Figure 11 shows the $H$ dependence of $M_{ZFC}$ at fixed $T$ below $T_{cl}$. The susceptibility $M_{ZFC}$ is negative at low $H$ because of the Meissner effect. The sign of $M_{ZFC}$ changes to positive at a zero-crossing field $H_0(T)$. The value of $H_0 (= 5.6$ kOe at $1.9$ K) coincides with that evaluated from the relation $H^{(0)}_c(T) = H^{(0)}_c(0)[1 - (T/T_{cl})^2]$. The magnetization $M_{ZFC}$ increases with further increasing $H$. It shows a broad peak, which shifts to the high-$H$ side with increasing $T$: $22.5$ kOe at $1.9$ K and $29$ kOe at $3.5$ K. This peak arises from a competition between the Curie-like behavior ($\Delta M_{ZFC}$) and the diamagnetic...
The difference \( \delta \chi \) at a fixed \( T \) below \( T_{cl} \) decreases with increasing \( H \).

The successive phase transitions at \( T_{cl} \) and \( T_{cu} \) in YBa\(_2\)Cu\(_4\)O\(_8\) can be qualitatively explained by Kawachi et al. by taking into account of the role of the superconducting grain below \( T_{cu} \) and clusters below \( T_{cl} \). According to their model, our results of Ta\(_2\)S\(_2\)C can be explained as follows. Below \( T_{cu} \) the superconductivity occurs in each grain. In the presence of \( H \) well above \( H_{cl}^{(i)} \), the fluxoids are pinned by pinning centers such as defects and vacancies within grains. A very weak irreversible effect of magnetization suggests that the density of flux-oids pinned in each grain for the FC state is slightly larger than the ZFC state. The distribution of fluxoids in the FC state is more uniform, while in the ZFC state more fluxoids are concentrated in the grain-boundary, which does not contributes to \( \chi_{ZFC} \). Below \( T_{cl} \), the superconductivity occurs in clusters formed of grains coupled through weak inter-grain Josephson couplings. In the presence of \( H \), the system is divided into relatively free regions (the cluster-boundary region) and strong-pinned regions (within the clusters). In the FC state, the distribution of the fluxoids is more uniform inside the clusters. In the ZFC state, more fluxoids are concentrated in the cluster-boundary regions which may have little contribution to \( \chi_{ZFC} \). In the presence of \( H < H_{cl}^{(i)} \) applied to the system in the ZFC state, the fluxoids will enter only the cluster-boundary regions around the clusters. The fluxoids in the inter-cluster region can have a path through the system without being caught by clusters. When \( H > H_{cl}^{(i)} \), some free fluxoids are caught by the clusters and pinned strongly, which contributes to \( \chi_{ZFC} \). Because of such an increase in \( \chi_{ZFC} \), \( \delta \chi \) decreases with increasing \( H \).

The possible existence of mesoscopic grains in the Ta-C layers of Ta\(_2\)S\(_2\)C would be essential to the successive transitions having a hierarchical nature. The superconductive ordering proceeds in two steps from the intraplanar Josephson couplings between grains in the same Ta-C layers to the interplanar Josephson interaction between grains in adjacent Ta-C layers separated by Ta\(_2\)S\(_2\) type structure. In the intermediate phase between \( T_{cu} \) and \( T_{cl} \), each grain in Ta-C layers becomes a superconductor. Through the intraplanar Josephson coupling between grains, the region of the superconducting grains becomes larger as \( T \) decreases below \( T_{cu} \), forming a 2D superconducting phase. Thermal fluctuations overcome the interplanar Josephson coupling. Just below \( T_{cl} \), the effective interplanar Josephson coupling becomes strong enough to give rise to a 3D superconducting phase. The 2D superconducting systems are coupled to each other through a weak interplanar Josephson coupling between adjacent Ta-C layers.

We note that the magnetic analogy to the successive phase transitions of such a hierarchical nature is seen in a stage-2 CoCl\(_2\) graphite intercalation compound (GIC)\(^{20}\) \( T_{cu}^{(2)} \) (= 8.9 K) and \( T_{cl} \) (= 6.8 – 7.2 K). The existence of islands is essential to the successive phase transitions.

FIG. 12: Difference \( \Delta M_{ZFC} (= M_{ZFC} - \chi_d H) \) as a function of \( H^*/T \) where \( \chi_d = -1.44 \times 10^{-7} \) emu/g, \( H = H^* \times 10^4 \) (\( H \) and \( H^* \) are in the units of Oe and Tesla, respectively), and the data of Fig. 11 for \( M_{ZFC} \) are used. The solid lines denote Brillouin functions given by \( N_g \mu_B \tanh(0.6717H^*/T) \) with \( N_g = (4.7, 2.1, 1.3, 0.91) \times 10^{18} \)/g.

\[ \Delta M_{ZFC}(10^3 \text{emu/g}) \]

| \( H^*/(\text{Tesla/K}) \) | 1.9 K | 2.1 | 2.5 | 3.0 | 3.5 |
|-----------------|-----|-----|-----|-----|-----|
| \(-5\)          | 1.3 | 2.1 | 1.9 | 0.91|
| \(0\)           | 1.3 | 2.1 | 1.9 | 0.91|
| \(1\)           | 1.3 | 2.1 | 1.9 | 0.91|
| \(1.5\)         | 1.3 | 2.1 | 1.9 | 0.91|
| \(2\)           | 1.3 | 2.1 | 1.9 | 0.91|
| \(2.5\)         | 1.3 | 2.1 | 1.9 | 0.91|
| \(3\)           | 1.3 | 2.1 | 1.9 | 0.91|

IV. DISCUSSION

A. Origin of successive phase transitions at \( T_{cl} \) and \( T_{cu} \)

We find that Ta\(_2\)S\(_2\)C undergoes successive superconducting transitions at \( T_{cl} \) and \( T_{cu} \). The dymagnetic volume fraction (21 %) indicates that our system is formed of many small grains. The \( T \) and \( H \) dependence of \( \chi_{ZFC} \) and \( \chi_{FC} \) below \( T_{cl} \) is very different from that between \( T_{cl} \) and \( T_{cu} \): \( \delta \chi > 0 \) for \( T < T_{cl} \) and \( \delta \chi \approx 0 \) (\( \delta \chi > 0 \)) for \( T_{cl} < T < T_{cu} \). The difference \( \delta \chi \) at a fixed \( T \) below \( T_{cl} \) decreases with increasing \( H \). In contrast, \( \delta \chi \) at a fixed \( T \) between \( T_{cl} \) and \( T_{cu} \) increases with increasing \( H \), showing a maximum around \( H = 500 \) Oe, and decreases with further increasing \( H \). We note that similar successive transitions have been reported in a ceramic superconductor YBa\(_2\)Cu\(_4\)O\(_8\) (\( T_{cl} = 37 \) K and \( T_{cu} = 80 \) K)\(^{25,26}\) which consists of small grains. Below \( T_{cu} \), \( \chi_{ZFC} (= \chi_{FC}) \) becomes negative. The difference \( \delta \chi \) appears below \( T_{cl} \). The contribution \( \chi_d H \). In Fig. 12 we show the plot of a magnetization \( \Delta M_{ZFC} \) as a function of \( H^*/T \), where \( \Delta M_{ZFC} = M_{ZFC} - \chi_d H \) with \( \chi_d = -1.44 \times 10^{-7} \) emu/g, and \( H = 10^3 H^* \) (\( H \) and \( H^* \) are in the units of Oe and Tesla, respectively). The curve of \( \Delta M_{ZFC} \) vs \( H^*/T \) which are slightly different for different \( T \), increases with increasing \( H^*/T \). The function form of \( \Delta M_{ZFC} \) vs \( H^*/T \) will be discussed in Sec. IV.]
The nearest neighbor spins inside islands are ferromagnetically coupled with intraplanar exchange interactions. On approaching $T_{cu}$ from the high-$T$ side, spins come to order ferromagnetically inside islands. At $T_{cu}$ these ferromagnetic islands continue to order over the same layer through interisland interactions (mainly ferromagnetic), forming a 2D ferromagnetic long-range order. Below $T_{cu}$ a reentrant spin-glass-like phase order is established through effective antiferromagnetic interplanar interactions between spins in adjacent intercalate layers, where the antiferromagnetic phase and the spin glass phase co-exist.

B. Anderson localization effect

We have shown that in Ta$_2$S$_2$C, the susceptibility $\chi_{FC}$ at low $T$ and high $H$ obeys the Curie law, which is a direct evidence of the appearance of local magnetic moments of unpaired electrons due to the Anderson localization effect. A theory for the magnetic susceptibility in systems with both localization and electron-electron interactions is presented by Kobayashi et al. and Aoki. It is assumed that there are localized states with energies very close to the Fermi level $\epsilon_F$. Singly occupied states are energetically favorable by virtue of intrastate Coulomb interaction ($U > 0$) between spin-up and spin-down electrons in the same localized state. In the presence of $H$, the magnetization contribution $m_i$ of the $i$-th localized state with energy $\epsilon_i$, relative to $\epsilon_F$, can be written as

$$m_i = \mu_B [e^{-\beta(\epsilon_i - \mu_B H)} - e^{-\beta(\epsilon_i + \mu_B H)}]/Z$$

(2)

where $\beta = 1/(k_B T)$, $\mu_B$ is the Bohr magneton, and $Z$ is the partition function given by

$$Z = 1 + e^{-\beta(\epsilon_i - \mu_B H)} + e^{-\beta(\epsilon_i + \mu_B H)} + e^{-\beta(2\epsilon_i + U)},$$

(3)

corresponding to an empty state, a state occupied by a spin-up electron with the energy $\epsilon_i - \mu_B H$, a state occupied by a spin-down electron with the energy $\epsilon_i + \mu_B H$, and a state occupied by spin-up and spin-down electrons with $2 \epsilon_i + U$. Then the total magnetization $M_e$ is obtained as

$$M_e = \frac{N(0) \mu_B \sinh(\beta \mu_B H)}{\beta(\cosh^2(\beta \mu_B H) - e^{-\beta U})^{1/2}} \ln \frac{\cosh(\beta \mu_B H) + [\cosh^2(\beta \mu_B H) - e^{-\beta U}]^{1/2}}{\cosh(\beta \mu_B H) - [\cosh^2(\beta \mu_B H) - e^{-\beta U}]^{1/2}}$$

(4)

where $N(0)$ is the density of states at $\epsilon_F$. For $U = 0$, $M$ is equal to the Pauli paramagnetism $M_P = 2 N(0) \mu_B^2 H$. Here we define $\Delta M^*$ as $\Delta M^* = (M_e - M_P)/(k_B N(0) \mu_B)$, where $H$ is in the unit of Oe and $U$ is in the unit of K; $U = 27$ K for the pristine 1T Ta$_2$S$_2$. Figure 13 shows the plot of $\Delta M^*$ as a function of $H^*/T$ for $0 \leq H^* \leq 10$ Tesla and $T = 1.9$ and 3.5 K as $U$ is changed as a parameter, where $H^*$ is in the units of Tesla. For comparison, the Brillouin function given by $M_B^0 = U \tanh(\beta \mu_B H)$ is also plotted for each $U$. Although $\Delta M^*$ depends on both $H^*/T$ and $U/T$ for each $U$, the curve of $\Delta M^*$ vs $H^*/T$ for each $U$ fits well with $M_B^*$ with the same $U$. Note that $M_B^*$ is a little larger than $\Delta M^*$ at the same $H^*/T$ for $0 \leq H^*/T \leq 3$. The magnetization $\Delta M^*$ at $H^*/T = 5$ is equal to the saturation magnetization ($= U$) of $M_B^*$. This implies that $\Delta M^*$ with $U$ coincides with the magnetization of free electron spins whose number is given by $N(0)/U$.

Here we discuss our result shown in Fig. 12 based on the above model. As shown Fig. 12 all the data of $\Delta M_{ZFC}$ vs $H^*/T$ do not fall on a single-valued function of $H^*/T$. This is consistent with the expression given by Eq. 4. Negative values of $\Delta M_{ZFC}$ for $H^*/T < 0.3$ is due to the Meissner effect. In
Fig. 12 a solid line denotes a Brillouin function given by \( N_\mu B \tan(h(0.6717H^*/T)) \) with \( N_\mu B = 0.019 \text{ emu/g} \) or \( N_\mu B = 2.1 \times 10^{18} \text{ emu/g} \), where \( N_\mu = N(0)U \). Our data of \( \Delta M_{ZFC} \) vs \( H^*/T \) greatly deviates downward from this Brillouin function for \( H^*/T > 1 \). In Fig. 12 the magnetization \( \Delta M_{ZFC} \) reaches 0.012 emu/g at \( H^*/T = 2.5 \). Since the saturation magnetization is \( N_\mu B \), the value of \( N_\mu \) can be estimated as \( N_\mu = 1.3 \times 10^{18} \text{ emu/g} \). We also note \( N_\mu = 4.7 \times 10^{18} \text{ emu/g} \) (\( N_\mu B = 0.044 \text{ emu/g} \)) in the limit of \( H^*/T \to 0 \), which is evaluated from the Curie-Weiss constant (see Sec. IIII). Alternative method to determine \( N_\mu \) is as follows. We assume that \( M_{ZFC} = \chi H + N_\mu B \tan(\beta \mu B H) \). The magnetization \( M_{ZFC} \) at fixed \( T_1 \) has a local maximum at \( H^* = H_{1}^* \): \( N_\mu = -1.6053 \times 10^{24} T \chi_H \cosh^2(0.6717 T_1 H_{1}^*/T_1) \). Using the values of \( H_{1}^* \) and \( T_1 \) determined from the inset of Fig. 11 and \( \chi_H = -1.44 \times 10^{-7} \text{ emu/g} \), \( N_\mu \) is calculated as \( N_\mu = (0.91 \pm 0.05) \times 10^{18} \text{ emu/g} \). This value of \( N_\mu \) is close to that for 1T-TaS\(_2\) \( (N_\mu = 1.3 \times 10^{18} /g) \) which is calculated from the Curie-Weiss constant \( (C_\mu = 0.806 \times 10^{-6} \text{ emu/g}) \) obtained by DiSalvo and Waszczak.\(^\text{12} \) For comparison, in Fig. 12 we also show the Brillouin function with these values of \( N_\mu \), as a function of \( H^*/T \). We do not find any reasonable value of \( N_\mu \), which leads to good agreement between the result and the Brillouin function over the whole range of \( H^*/T \). Similar behavior has been also observed in 1T-TaS\(_2\): the downward deviation of the observed magnetization from the Brillouin function (corresponding to the case of \( N_\mu = 2.1 \times 10^{18} \) emu/g in Fig. 12) occurs for \( H^*/T \geq 0.25 \). One of the reason for such a difference between the theory and experiment is that \( U \) is assumed to be constant in the above model. The intra-state Coulomb energy \( U_i = U(\epsilon_i) \) is considered to decrease with increasing \( \epsilon_i \). The total energy \( 2\epsilon_i + U_i \) is needed for the state \( i \) to be occupied by the spin-up and spin-down electrons. If \( 2\epsilon_i + U_i > 0 \), the \( i \)-th state is occupied by a single electron which behaves as a free spin. If \( 2\epsilon_i + U_i < 0 \), the \( i \)-th state is occupied by paired electrons.\(^\text{5} \) The number of free spins is given by \( N(0)\Delta U \), where \( \Delta U \) is the region where free spins can situate on the localized states. The replacement of \( U \) by \( \Delta U \) leads to a decrease in the saturation magnetization. Thus the downward deviation of the magnetization from the Brillouin function with \( U \) in Ta\(_2\)S\(_2\)C and 1T-TaS\(_2\) can be qualitatively explained in terms of this replacement.

V. CONCLUSION

Two phenomena (the superconductivity and the Anderson localization effect) are observed in Ta\(_2\)S\(_2\)C. The structure of Ta\(_2\)S\(_2\)C can be viewed as a sum of Ta-C layers and TaS\(_2\)-type structure. The superconductivity is mainly due to the Ta-C layers, while the Anderson localization effect is due to TaS\(_2\)-type structure. Ta\(_2\)S\(_2\)C undergoes successive superconducting transitions of a hierarchical nature at \( T_{cl} = 3.61 \pm 0.01 \text{ K} \) and \( T_{cu} = 9.0 \pm 0.2 \text{ K} \). The intermediate phase between \( T_{cu} \) and \( T_{cl} \) is an intra-grain superconductive state occurring in the Ta-C layers in Ta\(_2\)S\(_2\)C, while the low temperature phase below \( T_{cl} \) is an inter-grain superconductive state. The \( T \) dependence of magnetic susceptibility at \( H \) well above \( H_{1/2}(0) \) and the \( H \) dependence of \( M_{ZFC} \) at low \( T \) and high \( H \) are described by a sum of a diamagnetic susceptibility and a Curie-like behavior. The latter shows that the Anderson localization effect occurs in the 1T-TaS\(_2\)-type structure in Ta\(_2\)S\(_2\)C, leading to the localized magnetic moments due to unpaired electrons just below \( \epsilon_F \).

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