Electron correlation and two dimensionality in the spin-density-wave phase of (TMTTF)$_2$Br under pressure

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The incommensurate spin-density-wave (SDW) phase in (TMTTF)$_2$Br was investigated through transport measurements under pressure and magnetic fields parallel to the $c^*$ axis. For the incommensurate SDW phase of (TMTTF)$_2$Br stabilized above 0.5 GPa, the SDW transition temperature $T_{SDW}$ increases with the applied magnetic field. The field dependence of $T_{SDW}$ is described by a quadratic behavior and the coefficient of the quadratic term increases with increasing pressure. These results are consistent with the prediction of the mean-field theory based on the suppression of the SDW transition by two-dimensionality. From the relation between the coefficient of the quadratic term and $T_{SDW}$ at zero magnetic field, we determined the role of electron correlation and two dimensionality in the SDW phase of (TMTTF)$_2$Br under pressure and found that the SDW transition in (TMTTF)$_2$Br can be well explained within the mean field theory by taking into account the reduction of the coupling constant $N(0)I$ by pressure.

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The family of organic salts (TMTCF)$_2X$ ($C = $ Se or S and $X = $ PF$_6$, AsF$_6$, ClO$_4$, Br, etc.) show quasi-one-dimensional (Q1D) electric properties and have rich ground states, spin-Peierls, antiferromagnetism (AF), spin density wave (SDW), superconductivity, etc. The ground state in these salts is influenced by the different donors and anions that constitute the compounds. A universal phase diagram for the TMTCF salts as a function of pressure has been proposed by Jérome. The differences of the donors and the anions change the distance between molecules, transfer integral, etc. and this means that these act as a chemical pressure in the universal phase diagram. With increasing pressure, the electron transfer along the interchain direction increases, and the two dimensionality of the system increases. In addition to the role of the two dimensionality, it is also well known that the electron correlation must be strong to cause such a rich phase diagram in the Q1D compounds.

The ground state of the sulfur-based salt (TMTTF)$_2$Br is antiferromagnetism (AF) at ambient pressure. In the temperature range above this AF phase, the system shows a broad minimum of resistivity $\rho_{\text{min}}$ at about $T_{\rho} = 100$ K. Between this temperature and the AF transition temperature, the system has a charge-localized (CL) state, in which the resistance increases with decreasing temperature. On applying the pressure, the ground state changes from the AF state to the incommensurate SDW state above about 0.5 GPa and $\rho_{\text{min}}$ vanishes. It is known that this incommensurate SDW state in (TMTTF)$_2$Br is essentially the same as that in (TMTSF)$_2$X, which is stabilized by the nesting of the Fermi surface in the Q1D electron band.

When the magnetic field is applied to the SDW state suppressed by the imperfect nesting of Fermi surface, it has been predicted that $T_{SDW}$ increases nearly quadratically with the field in low magnetic fields and shows a saturation behavior to the transition temperature for the perfect nesting case $T_{SDW_0}$ in high magnetic fields. In fact, the quadratic magnetic field dependence and the saturation behavior have been confirmed by the experiments for (TMTSF)$_2$PF$_6$ and quenched (TMTSF)$_2$ClO$_4$. In a previous paper, however, we have estimated $T_{SDW_0} = 16$ K for (TMTSF)$_2$PF$_6$ under low pressure. This value of $T_{SDW_0}$ is slightly small to explain the universal phase diagram using the simplest model of the mean-field (MF) theory.

In this paper, we present the results of resistivity measurements for the incommensurate SDW phase in (TMTTF)$_2$Br under pressure and magnetic field, and discuss the validity of the universal phase diagram for the TMTCF salts through the role of electron correlation and two dimensionality in the SDW phase of (TMTTF)$_2$Br under pressure.

Single crystals of (TMTTF)$_2$Br were synthesized by the electrochemical method. The resistance measurements were performed by a dc four-wire method along the highly conducting $a$ axis. Electric leads of 10 $\mu$m gold wire were attached with silver paint onto gold evaporated contacts. The current contacts covered the whole areas of both ends of the sample for a uniform current. The typical sample size was $0.4 \times 0.2 \times 0.02$ mm$^3$, where the length along the $a$-axis direction is that between voltage contacts. The temperature was measured using a Cernox CX-1050-SD resistance thermometer calibrated by a capacitance sensor in magnetic fields. The pressure was applied using the beryllium-copper clamp cell up to 2.1 GPa. We used Daphne 7373 oil as a pressure medium. It is known that the pressure decreases at low temperature...
pressures. At ambient pressure, the resistance shows the broad minimum at about 100 K, corresponds to the crossover from the metallic to the CL state. Then, the resistance increases with decreasing temperature and shows a kink at 18 K. From the NMR measurements, it is well known that (TMTTF)$_2$Br exhibits an AF transition at 14 K. However, the kink temperature is 4 K higher than this and $d(\ln R)/d(1/T)$ below 18 K is almost flat. The resistance shows no characteristic structure at 14 K. This indicates that the AF ordering does not change the behavior of conductivity. The kink structure at 18 K in the resistance at ambient pressure may be caused by charge disproportionation. The temperature for the resistance minimum $T_{\text{min}}$ rapidly decreases with increasing pressure up to 0.3 GPa and vanishes at about 0.4 GPa. These results are consistent with a previous report. As shown in Fig. 1 the kink at 18 K in the resistance at ambient pressure changes to the broad peak structure in the derivative of the logarithm of the resistance with 1/T. The temperature of this peak in $d(\ln R)/d(1/T)$ increases with increasing pressure and shows a maximum of 23 K at 0.3 GPa. Above 0.3 GPa, the peak temperature decreases from 23 K at 0.3 GPa to 19.5 K at 0.5 GPa. A detailed discussion of this phase diagram below 0.5 GPa is contained in a previous paper.

Above 0.5 GPa the peak temperature still decreases with pressure, but the rate of decrease becomes smaller. It has been confirmed by the NMR measurements that the SDW with the incommensurate wave vector is stabilized above 0.5 GPa. With decreasing temperature, the resistance shows a steep increase associated with the SDW transition and thermally activated behavior at low temperature as shown in Fig. 1. The SDW transition temperatures in zero field $T_{\text{SDW}}(0)$ determined from the peak of the derivative of the logarithm of the resistance with 1/T $d(\ln R)/d(1/T)$ are plotted against field in Fig. 2. With increasing pressure, $T_{\text{SDW}}(0)$ decreases from 19.5 K at 0.5 GPa to 12.5 K at 2.1 GPa. This pressure dependence of $T_{\text{SDW}}(0)$ above 0.5 GPa is qualitatively consistent with the prediction of the MF theory based on the imperfect nesting of the Fermi surface.

Figure 3 shows the temperature dependence of the resistance of (TMTTF)$_2$Br along the a-axis under various magnetic fields at 2.1 GPa. In each field, the resistance shows the increase with decreasing temperature associated with the SDW gap. The SDW transition temperature $T_{\text{SDW}}$ in a magnetic field, determined from $d(\ln R)/d(1/T)$ (the bottom of Fig. 3), is shown in Fig. 4. We find that, with increasing magnetic field along the c*-axis, $T_{\text{SDW}}$ increases at various pressures. The magnetic-field dependence of $T_{\text{SDW}}$ is described by the quadratic function: $T_{\text{SDW}}(B) = T_{\text{SDW}}(0) + CB^2$, where C is constant, for each pressure. The coefficient of the quadratic term C increases with increasing pressure. All the results between 0.75 GPa and 2.1 GPa, are well fitted by the quadratic function without any saturation behavior even at 24 T. Such a behavior is also found in TMTSF salts and is consistent with the prediction of the MF theory.
As a result, we conclude that the SDW transition in (TMTTF)$_2$Br is an ordinary SDW transition based on the nesting of the Fermi surface as for (TMTSF)$_2$X.

Figure 3 shows the relation between the coefficient of the quadratic term $C$ and $T_{SDW}$ at zero magnetic field $T_{SDW}(0)$ for (TMTTF)$_2$Br. The coefficient $C$ is mainly determined by the two dimensionality of the electron band in the MF theory, however, $C$ also depends on other parameters, e.g., Fermi velocity $v_F$. In the figure, the result for (TMTSF)$_2$PF$_6$ is also shown for comparison. The results of (TMTTF)$_2$Br under high pressure seem to smoothly connected to those of (TMTSF)$_2$PF$_6$, and the SDW-superconductivity phase diagram in (TMTTF)$_2$Br above 2.1 GPa is quite similar to that in (TMTSF)$_2$PF$_6$ under pressure. Therefore, we expect that the parameters in (TMTTF)$_2$Br at 2.1 GPa to be close to those in (TMTSF)$_2$PF$_6$ at ambient pressure. From the observed curvature of the line for the relation between $T_{SDW}(0)$ and $C$ for (TMTSF)$_2$PF$_6$, we had determined several parameters for the SDW transition in the MF theory, assuming that the SDW transition temperature for the perfect nesting case $T'_{SDW}$ is independent of pressure as in a previous paper. The dashed line in Fig. 3 is the fit by this theory to the experimental data of (TMTSF)$_2$PF$_6$ and the agreement is good. The fit gives $T'_{SDW} = 16$ K and $v_F = 1.03 \times 10^5$ m/s for (TMTSF)$_2$PF$_6$, with a lattice parameter along the $b'$ axis of $7.7 \times 10^{-10}$ m. However, the data for (TMTTF)$_2$Br in the region where $T_{SDW}(0)$ is higher than 13 K, corresponding to 1.7 GPa, are slightly above the calculated dashed curve. This fact indicates that the data for (TMTTF)$_2$Br and (TMTSF)$_2$PF$_6$ cannot be described by the constant value of $T_{SDW}$ with the pressure. Even if we consider only the results for (TMTTF)$_2$Br, the almost linear relation between $T_{SDW}(0)$ and $C$ in a log-log scale, shown in Fig. 3, is also hardly explained by the MF theory with constant $T_{SDW}$. In such a simple model, it is expected that $T_{SDW}(0)$ is almost constant for the imperfect nesting parameter $\epsilon_0/\Delta_0$ in the region where $\epsilon_0/\Delta_0$ is small and $T_{SDW}(0)$ is close to $T_{SDW}$, in contrast to the observed behavior. As a result, the observed slighty steep and straight dependence of $T_{SDW}$ suggests that $T_{SDW}$ varies with the pressure.

From the BCS relation in the MF theory, $T_{SDW}$ is given as $k_B T_{SDW} = 1.14 D \exp(-1/N(0)I)$, where $D$, $N(0)$, and $I$ are the bandwidth, the density of state at Fermi level, and the on-site Coulomb energy, respectively. With increasing pressure, we can expect that (a) the on-site Coulomb energy $I$ decreases due to the increase of the screening of Coulomb potential, (b) the density of state $N(0)$ decreases due to the increase of the transfer energy, and (c) the bandwidth $D$ increases due to the increase of the transfer energy along the $a$ axis. The contribution of $N(0)I$ to $T_{SDW}$ is expected to be larger than that of $D$ because the variation of $N(0)I$ affects $T_{SDW}$ exponentially. After all, we can expect $T_{SDW}$ to decrease due to the decrease of the coupling constant $N(0)I$ with increasing pressure. Indeed, the reduction of $N(0)I$ by applying pressure was suggested from the NMR measurements in the metallic phase of (TMTCF)$_2$X under the pressure. It is natural to consider that $T_{SDW}$ decreases as the ap-

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**FIG. 3:** Top: Temperature dependence of the resistance in (TMTTF)$_2$Br for various magnetic fields at 2.1 GPa. Bottom: The derivative of the logarithm of the resistance by $1/T$ offset from zero for clarity.

**FIG. 4:** Magnetic-field dependence of the SDW transition temperature $T_{SDW}$ for various pressures. The solid lines are the best fit of the quadratic function.
stant against the pressure; it is expected that the SDW transition temperature and a small pressure dependence of $T_{SDW}$ due to the reduction of $N(0)I$ by pressure.

In the case of $(TMTSF)_2PF_6$, the pressure dependence of $T_{SDW}(0)$ is mainly determined by the imperfect nesting parameter $\epsilon_0/\Delta_0$, because $\epsilon_0/\Delta_0$ is fairly large and $T_{SDW}(0)$ shows a rapid decrease with increasing $\epsilon_0/\Delta_0$. In such a situation, the reduction of $N(0)I$ with pressure makes only a small contribution to $T_{SDW}(0)$. As a result, the simplest model with constant $T_{SDW0}$ reproduces the observed relation between $T_{SDW}$ and $\epsilon_0$ for $(TMTSF)_2PF_6$, as described in a previous paper. Accordingly, as shown in Fig. 5, the incommensurate SDW phase in both $(TMTSF)_2PF_6$ and $(TMTTF)_2Br$ is understood systematically and quantitatively in the common pressure axis, where its origin corresponding to ambient pressure is shifted for these two salts, with the MF theory based on the nesting of the Fermi surface taking into account the reduction of the coupling constant with pressure.

In summary, resistivity measurements have been performed under pressure and in magnetic fields in $(TMTTF)_2Br$. Above 0.5 GPa, $(TMTTF)_2Br$ exhibits an incommensurate SDW state at low temperatures. With increasing pressure, the SDW transition temperature $T_{SDW}$ decreases from 19.5 K at 0.5 GPa to 12.5 K at 2.1 GPa. When the magnetic field is applied to this SDW phase, substantially suppressed by the pressure, $T_{SDW}$ increases quadratically with the field and the coefficient $C$ of the quadratic term increases with increasing pressure. These results are consistent with the prediction of the MF theory based on the nesting of the Fermi surface in the Q1D electronic band. From the analysis of the relation between the coefficient $C$ and $T_{SDW}$ at zero magnetic field, the magnetic field and pressure dependence of $T_{SDW}$ in $(TMTTF)_2Br$ can be well explained by the MF theory by taking into account the reduction of the coupling constant $N(0)I$ with pressure. As a result, the incommensurate SDW phase in both $(TMTSF)_2PF_6$ and $(TMTTF)_2Br$ is well described by the MF theory taking into account electron correlation and two dimensionality of the system in the universal phase diagram.

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**FIG. 5:** The relation between the SDW transition temperature at zero magnetic field $T_{SDW}(0)$ and the coefficient $C$ of quadratic term in $T_{SDW}$ in $(TMTTF)_2Br$ and $(TMTSF)_2PF_6$. The broken line represents the theoretical curve with $T_{SDW0} = 16$ K. The solid line represents the theoretical curve taking into account the pressure dependence of $N(0)I$ shown in Fig. 6.
With the increasing pressure, we can expect the increase of the transfer energy along the $a$ axis, which works as the increase of $v_F$, and the decrease of the lattice parameter along the $a$ axis, which works as the decrease of $v_F$. Although a quantitative estimation of the pressure dependence of $v_F$ is difficult at present, a small pressure dependence does not change our conclusion.

If the value of $v_F$ increases 20% with increasing pressure from 0.75 GPa to 2.1 GPa, $T_{SDW_0}$ at 0.75 GPa increases from 20.0 K to 20.6 K.