Dynamics of charge density waves (CDWs) has been of interest for decades [1]. In particular, the effect of a magnetic field to CDW dynamics remains unsolved. Phase of a CDW determines the initial position of the density wave, and collective motion of the CDW is described in terms of delocalization of quantum interference of CDWs extending over the $c$ plane, namely, the chain axis of the crystal. A twofold symmetry axis, namely, the chain axis of the crystal. A twofold symmetry was found in the magnetoresistance. The magnetoresistance amplitude exhibited maxima when the field was parallel to the $a$ axis, whereas it vanished to the $b$ axis. The observed anisotropy may come from a difference in interchain coupling of adjacent CDWs along the $a$ and $b$ axes. Comparison of the anisotropy to the scanning tunneling microscope image of CDWs allows us to provide a simple picture to explain the magnetoresistance in terms of delocalization of quantum interference of CDWs extending over the $b$-$c$ plane. Our observation is an unexpected phenomenon, which will provide an important key to understand the AB oscillations of the CDWs.

Single crystals of $\alpha$-TaS$_3$ were grown using a standard chemical vapor transportation method. A pure tantalum sheet and sulfur powder were placed in a quartz tube. The quartz tube was evacuated to $1 \times 10^{-6}$ Torr and heated in a furnace at 530 °C for two weeks. The grown crystals were ribbonlike whiskers. The chain direction of $\alpha$-TaS$_3$ is along the $c$ axis, and the flat surface of the ribbon is reported to be a $b$-$c$ plane, perpendicular to the $a$ axis [7–9]. The crystal orientation of each sample was determined and confirmed to be consistent with the previous reports by the electron backscattering diffraction (EBSD) technique (OIM TSL). The electrodes were made using 50-µm-diameter silver wires glued with silver paint. Gold thin film was deposited on the crystal before the silver wires were attached to reduce the contact resistance to $1 \Omega$ at room temperature.

The resistance of the sample was measured with a standard four-probe technique. As described in a previous study [10], a high-impedance digital voltmeter (Keithley 6512; $Z_m > 200 \Omega$) was employed. All measurements were performed with constant currents generated by a current source (Keithley 220). A magnetic field was applied with a couple of superconducting coils. The sample holder was rotated along an axis perpendicular to the magnetic line. In this experiment the axis of rotation was aligned with the chain axis of the sample.
The sample was glued to the sample holder with the ribbon surface facing the holder. Since Joule heat induced by eddy current might have caused the temperature to increase when the holder was rotated, each measurement was performed after the temperature was stabilized in less than 4 mK rise.

Figure 1 shows the typical temperature dependence of resistance ($R-T$). The sample cross section is $15 \times 0.5 \, \mu\text{m}^2$. The room temperature resistivity of the sample is $2.8 \times 10^{-6} \, \Omega\text{m}$, which is consistent with previous reports ($\sim 3 \times 10^{-6} \, \Omega\text{m}$) [11,12]. By lowering the temperature, the system undergoes a Peierls transition at 220 K, below which the electrons at the Fermi surface condense into a charge density wave state. In the 100 to 200 K temperature range, the resistance obeys an Arrhenius law with an activation energy of 860 K (broken line in the inset of Fig. 1). Discrepancy from the Arrhenius law is found below 100 K. In the 40 to 100 K temperature range, a smaller activation energy ($\sim 200$ K; solid line) is applicable, and it becomes higher ($\sim 400$ K; dotted line) at temperatures below 30 K. Such behavior is reproducible and also consistent with previous reports [11,13].

Figure 2 shows the current-voltage ($I-V$) characteristics of the sample at 4.2 K with flowing current $I = 2$ nA. At this temperature, the ohmic resistance exceeds $10^9 \, \Omega$, as deduced from an extrapolation of the $R-T$ curve. However, the slope of the $I-V$ curve corresponds to $1.3 \times 10^9 \, \Omega$, which comes from a tiny current accompanied by relaxation. No nonlinear conduction threshold was observed in the $I-V$ characteristics at this temperature where there were almost no thermally activated quasiparticles. A slight hysteresis was also observed in the $I-V$ curve in the neighborhood of $I = 0$. This phenomenon can be interpreted as a rearrangement of the CDW dislocations, which hold electric charges, as reported for blue bronze [14,15] and TaS$_3$ [16]. In a higher current range $|I| > 1 \times 10^{-9}$ A, the hysteresis became insignificant. The following experiments were performed in this current range.

Figure 3 shows the magnetoresistance of the sample observed at 4.2 K. The magnetic field was applied perpendicular to the current flow (c axis). $\theta = 0^\circ$ means that the field is directed along the $b$ axis, and at $\theta = 90^\circ$ the field is along the $a$ axis, as shown in the inset. The current is 2 nA, which stays in the nonlinear regime of the $I-V$ characteristics. The error bar represents $\pm 2\sigma$, where $\sigma$ is the standard deviation of the data. Solid lines are the guide to the eyes.
observed magnetoresistance is independent of the current direction. Moreover, the ratio \( V(B)/V(0) \) was constant over the nonlinear regime of the \( I-V \) characteristics in the field of \( B = 5.2 \, \text{T} \). As shown by the error bars in Fig. 3, which correspond to \( \pm 2\sigma \) where \( \sigma \) is a standard deviation of raw data, the data seem to be noisy, probably because of influence of the CDW’s collective motion, e.g., narrow band noise.

Magnetoresistance of quasi-one-dimensional conductors has been intensively studied [17]. A magnetic field changes electron motion on the Fermi surface. This provides an increase of resistivity, namely, *positive magnetoresistance* with anisotropic angle dependence according to the shape and topology of the Fermi surface. On the contrary, the sign of the magnetoresistance of \( \alpha \)-TaS\(_3\) was negative (Fig. 3). Moreover, no normal carrier in \( \alpha \)-TaS\(_3\) is left at the Fermi surface in the CDW state, and thermally activated quasiparticles are negligible at 4.2 K. Hence our observation should not be understood in terms of the conventional magnetoresistance. A comparison with the magnetoresistance of the NbSe\(_3\) case [6] is also noteworthy. NbSe\(_3\) has two CDW transitions at \( T_1 = 145 \, \text{K} \) and \( T_2 = 59 \, \text{K} \). Even below \( T_2 \) there remain normal carriers on the Fermi surface. A large positive magnetoresistance was observed below \( T_2 \), accompanied by an increase of the number of CDW carriers. Magnetic response to the dynamics of a CDW was hindered in the previous experiment. Our result also excludes the possibility of a spin-related phenomenon being a major contributor to the observed magnetoresistance, as with the negative magnetoresistance of TaS\(_2\) [18], which is essentially isotropic.

Figure 4 shows the angle dependence of the magnetoresistance, which reveals the twofold symmetry of the magnetoresistance. As the first approximation for twofold symmetry, we tried to fit the angle dependence of the observed magnetoresistance with the formula: \( \Delta R = -A \sin^2(\theta - \theta_0) \), where \( A \) is the amplitude of magnetoresistance and \( \theta_0 \) is the offset angle. The parameters were determined by nonlinear least square fitting to be \( A = 5.78 \times 10^6 \, \Omega \) and \( \theta_0 = -7^\circ \). The solid line in Fig. 4 shows the result of the fit. The residual error of the fit was roughly the same as the distribution of the observed data, which is shown with the error bars of 2\( \sigma \) in Fig. 4. The magnetoresistance amplitude exhibited maxima when the field was applied along the \( a \) axis, whereas it vanished when the field was along the \( b \) axis. The offset angle \( \theta_0 = -7^\circ \) represents a slight misalignment of the crystal axes to the magnetic field direction. This angle coincides with the direction of the \( b \) axis of the sample determined with the EBSD technique.

It is necessary to look carefully at the difference between the \( a \) and \( b \) axes. The STM image [9] demonstrates that maxima of CDW intensity on the \( b\)-plane are canted and form an angle of 86° with the chain axis. This angle corresponds to arctan(\( 8b_0/4c_0 \)) \( \approx 84^\circ \), where \( b_0 \) and \( c_0 \) are the lattice constants of \( \alpha \)-TaS\(_3\). This implies the wave function of CDW extends over the \( b\)-plane, and it is not firmly bound to a particular chain. The STM image is consistent with the CDW vector of \( \alpha \)-TaS\(_3\), \( \mathbf{q}_{\text{CDW}} = (0.5,0.125,0.25) \), formerly determined with diffraction studies [7,19]. CDWs of adjacent chains are out of phases along the \( a \) axis. This results from energetically favorable configuration among independent chains coupled with the Coulomb interaction [1]. Hence the response to the magnetic field may not be symmetric for the rotation upon the \( c \) axis.

The canting of CDW wave vector provides a simple picture as follows. There are two possibilities for the CDW to settle on the pristine lattice by canting left or right. Once either left or right is chosen, a certain area of CDWs are aligned to form a domain. If CDWs consist of such domains, a closed loop can exist as shown in the inset of Fig. 4. The area surrounded by the loop is represented as \( S = L^2 \sin \alpha \), where \( L \) is the length of the loop, and \( \alpha \) is the angle between CDW and the \( b \) axis. The angle \( \alpha \) is exaggerated in the inset.

![Fig. 4. Angle dependence of the magnetoresistance in the field \( B = 5.2 \, \text{T} \) observed at 4.2 K. The error bars represent \( \pm 2\sigma \) as in Fig. 3. A twofold symmetry is clearly exhibited. The solid line shows a result of a least-square fit to the formula \( \Delta R = -A \sin^2(\theta - \theta_0) \), where the offset angle \( \theta_0 = -7^\circ \). This angle coincides with the direction of \( b \) axis of the sample determined with the EBSD technique. The inset shows a schematic image of a CDW loop formed in domain structure. The thin lines represent the wave front of CDWs, and the bold line shows a loop, surrounding an area of \( L^2 \sin \alpha \), where \( L \) is the length of the loop.](Image 413x592 to 485x648)
magnetoresistance is represented as an even function of the magnetic flux, e.g., $\Delta R \propto -\Phi^2$; hence magnetoresistance would have a component of $\sin^2 \theta$. Therefore, the observed twofold symmetry in magnetoresistance (Fig. 4) can be interpreted as a natural consequence of the delocalization picture.

The coherent length $L$ was estimated both by the STM image and by x-ray diffraction. The STM image (Fig. 3 in Ref. [9]) shows CDWs in $22.5 \times 22.5 \text{ nm}^2$ area, where two domains of CDW can be distinguished. This image suggests that the size of each domain is much larger than $2 \times 10^{-8} \text{ m}$. A synchrotron x-ray study [24] exhibits the coexistence of incommensurate and commensurate CDWs in $\alpha$-TaS$_3$. Two satellite spots are separated by two pixels apart at the detector, and each spot is as narrow as one-pixel width (Fig. 1 in Ref [24]). This gives an estimation of the correlation length longer than $3 \times 10^{-7} \text{ m}$ in $c^*$ direction. If $L \sim 3 \times 10^{-7} \text{ m}$ is assumed, the corresponding area of the CDW loop becomes $1 \times 10^{-14} \text{ m}^2$, which gives the field of $0.2 \text{ T}$ for a flux quantum $h/2e$. This field can be interpreted as the minimum field for the negative magnetoresistance to occur. The observed magnetoresistance ranges above the field of $0.2 \text{ T}$, as shown in Fig. 3; hence it is consistent quantitatively with the delocalization picture.

What kinds of carrier are consistent with the delocalization picture? Quantum interference can occur for any kind of carrier as far as their phase is not lost. We have already ruled out magnetoresistance of thermally activated quasiparticles as a possible candidate for our observation. If the collective motion of CDW plays a major role for the observed negative magnetoresistance, such a quantum interference would increase the pinning rate of the CDW. In the case of soliton transport, soliton travels along such a closed path may be inactive for a carrier. The negative magnetoresistance with the twofold symmetry is consistent for both cases.

Finally, our observation and interpretation will ease conditions to observe the AB oscillations in CDWs. The previous studies were performed with the ion-beam-radiated NbSe$_3$ [2] and the ring crystal of TaS$_3$ [3]. Tsutoda et al. proposed that a CDW soliton might be confined and move along a single chain whose ends coalesced [3]. This proposal was based on the growth mechanism of the ring crystals [25]. However, the magnetoresistance is interpreted as quantum interference of the CDWs extending over the $b$-$c$ plane, and CDWs can maintain their quantum phase across the chains. Therefore, our observation is not only consistent with the previous reports, but also suggesting the possibility of the AB oscillations in a closed CDW loop realized by other methods.

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