Anisotropic magnetoresistance of charge-density wave in o-TaS$_3$: 
Anderson localization of soliton

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Abstract

We report magnetoresistance of charge-density wave (CDW) in o-TaS$_3$ whiskers. Anisotropic negative magnetoresistance is found in the nonlinear regime of current-voltage characteristics. The angle dependence of the magnetoresistance exhibits two-fold symmetry, suggesting that two-dimensionality plays an important role for the electric transport phenomenon. This result can be understood qualitatively in terms of Anderson localization of a CDW soliton. From the differences of the offset angle of the magnetoresistance among the samples, it is more plausible to attribute the results to a global property of each sample, rather than symmetry of the crystal structure. We deduce the CDW solitons are randomly distributed in a plane. Our findings are consistent with the previous studies of Aharonov-Bohm oscillations in CDW ring, and also provides a possible reason the oscillations were able to be observed in the experiments.
Necessity of quantum mechanical description has been a key issue to understand electoric transport phenomena of charge-density wave (CDW) \[1, 2\]. Coupling between the quantum phase of electrons and vector potential may be a significant evidence quantum mechanical nature of the particles through interference effects, as demonstrated by Aharonov-Bohm oscillations in CDW of \(\alpha\)-TaS\(_3\) ring crystals \[3\]. These results suggest that the quantum phase of CDW may extend over macroscopic length scale (\(> 10 \mu\text{m}\)). However, it is still unclear what kind of carrier plays a major role for such the quantum interference. In CDW, there are at least three possibilities: uncondensed normal electron, collective motion, and soliton \[4\].

In this article, we report magnetoresistance of CDW in \(\alpha\)-TaS\(_3\) whiskers. We measured resistance in nonlinear regime of \(\alpha\)-TaS\(_3\) at 4.2 K, and observed anisotropic negative magnetoresistance. Since whole Fermi surface of \(\alpha\)-TaS\(_3\) disappears below the Peierls temperature, electric conduction is only due to CDW at low temperatures. Two-fold symmetry of the magnetoresistance suggests that two-dimensionality plays an important role. Since the offset angle of magnetoresistance was different in each sample, our observation must be rooted in macroscopic configuration of CDW solitons. These results can be understood qualitatively in terms of Anderson localization, as a result of quantum interference of a soliton.

Single crystals of \(\alpha\)-TaS\(_3\) were grown by a standard chemical vapor transportation method. \(\alpha\)-TaS\(_3\) crystals were synthesized with a standard chemical vapor transport method. A pure tantalum sheet and sulfur powder were put 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 ribbon-like whiskers. The chain direction of \(\alpha\)-TaS\(_3\) is along to \(c\)-axis, and the flat surface of the ribbon is reported to be \(b\)-\(c\) plane, perpendicular to \(a\)-axis \[5–7\]. The electrodes were made using 50-\(\mu\text{m}\)-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 Ω at room temperature.

Resistance of the sample was measured with a standard four-probe technique. As described in the previous study \[8\], a high-impedance digital voltmeter (Keithley 6512; \(Z_{\text{in}} > 200 \text{TΩ}\)) was exploited. All measurements were performed with constant currents generated by a current source (Keithley 220). 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 to the chain-axis of the
The sample was glued to the sample holder with the ribbon surface facing to the holder. Since Joule heat by eddy current might rise the temperature when the holder was rotated, each measurement was performed after the temperature was settled within 4 mK.

Figure 1 shows a typical temperature dependence of resistance ($R-T$). The cross section of the sample is $15 \times 0.5 \, \mu m^2$. The room temperature resistivity of the sample is $2.8 \times 10^{-6} \, \Omega m$, which is consistent with the previous reports ($\sim 3 \times 10^{-6} \, \Omega m$) [9, 10]. 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 temperature range between 100 K and 200 K, the resistance obeys an Arrhenius law with activation energy of 860 K. Discrepancy from the Arrhenius law is found below 100 K. In the temperature range between 40 K and 100 K, a smaller activation energy ($\sim 200$ K) is applicable, and it turns to be higher ($\sim 400$ K; dotted line in Fig. 1) at temperatures below 40 K. Such the behavior is reproducible and also consistent with the previous report [9].

Figure 2 shows current-voltage ($I-V$) characteristics of the sample at 4.2 K with flowing current $I=2 \, nA$. At this temperature, ohmic resistance exceeds $10^9 \, \Omega$, deduced from the extrapolation of the $R-T$ curve. However, the slope of $I-V$ curve is corresponding to $1.3 \times 10^9 \, \Omega$, which comes from a tiny current accompanied with relaxation. A threshold of nonlinear conduction was not seen in the $I-V$ characteristics at this temperature where thermally activated quasiparticles were almost absent. A slight hysteresis was also observed in the $I-V$ curve in the neighborhood of $I=0$. This phenomenon can be interpreted as rearrangement of the CDW dislocations which hold electric charges, as reported in blue bronze [11]. In the 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 magnetoresistance of the sample observed at 4.2 K. The magnetic field is applied perpendicular to the current flow. $\theta = 0^\circ$ means the field is directed along the sample’s ribbon surface, and at $\theta = 90^\circ$ the field is applied perpendicular to the surface, as shown in the inset. This result shows negative magnetoresistance with anisotropy, and has reproducibility in several samples. We also confirmed that the observed magnetoresistance is independent from the current direction. Moreover, the ratio $V(B)/V(0)$ was found to be constant over the nonlinear regime of the $I-V$ characteristics at the field $B = 5.2 \, T$. As shown in the error bars of 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 CDW’s collective
motion, e.g. narrow band noise. Since no normal carriers in \( \alpha \)-TaS\(_3\) are left at the Fermi surface in the CDW state, and thermally activated quasiparticle is negligible at 4.2 K, we believe that this is an unambiguous evidence of coupling between CDW and magnetic field.

It seems to be contrast to the NbSe\(_3\) case \([12]\), in which a large positive magnetoresistance was observed. However, number of CDW carrier in NbSe\(_3\) was reported to be increased by 5% at the magnetic field of 10 T. Hence the intrinsic magnetoresistance of CDW in NbSe\(_3\) might be hindered by other factors, such as magnetic modification of Fermi surface.

Anisotropy of magnetoresistance provides useful information for understanding the mechanism of the magnetic field dependence. Figure 4 (a) shows angle dependence of the magnetoresistance. This result shows two-fold symmetry of the magnetoresistance. Angle dependence of magnetoresistance was measured for three samples, all of which exhibited the negative magnetoresistance with the two-fold symmetry as shown in Fig. 4. Usually, magnetoresistance is represented with an even function of an applied field. As the first approximation for two-fold 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 an amplitude of magnetoresistance and \( \theta_0 \) is an offset angle. Least square fitting was numerically performed with \( A \) and \( \theta_0 \) being fitting parameters. The solid lines in Fig. 4 are the result of the fit. We measured three samples and this behavior was well reproduced. Residual error of the fit is roughly same as the distribution of observed data, shown as error bars of 2\( \sigma \) in Fig. 4. It is noted that discrepancy from the fitting curve also shows two-fold symmetry.

Moreover, the observed two-fold symmetry of magnetoresistance has a remarkable angle dependence. The minimum of the magnetoresistance differs sample by sample: \( \theta_0 = -7^\circ, 59^\circ, \) and \( 106^\circ \). The difference of the offset angle \( \theta_0 \) suggests the origin of the magnetoresistance. We expect the ribbon surface is perpendicular to \( a \)-axis, as shown in Refs. \([5–7]\), though we did not figure out the crystal axis of each sample. Moreover, since the cross section of an orthorhombic crystal is usually rectangular, it is natural to expect that the difference of the offset angle should fall around \( 90^\circ \), even if the ribbon surface were perpendicular to \( b \)-axis by accident. In fact, the observed angle differences of magnetoresistance are \( 66^\circ \) and \( 47^\circ \). Hence the magnetoresistance might not be related to the crystal structure of the sample.

The observed two-fold symmetry would also be unexpected since the CDW sliding is a one-dimensional phenomenon and the magnetic field is always applied perpendicular to
the current flow. Naively, the response to the magnetic field might be symmetric for the rotation around the chain axis. This result also excludes the possibility of a spin-related phenomenon for a major contribution of the observed magnetoresistance, such like a negative magnetoresistance of TaS$_2$ [13], which is essentially isotropic.

Since the sign of the magnetoresistance was negative, Anderson localization is invoked [14]. In a random medium, a wave interferes with itself, so that randomness can yield a transition from an extended to a localized state. Two-dimensional electron gas is a typical system where Anderson localization can be observed since the critical dimension for noninteracting electron gas is two dimension [14]. Magnetic field affects quantum mechanical phase of an electron, resulting in destruction of the interference of the electron wavefunction [15]. Relative shift of the electron phase $\Delta \psi$ is represented as $\Delta \psi = \frac{e}{h} \int A \, dl$, where $h$ is Planck’s constant, $e$ is a charge quantum of carrier, and $A$ is the vector potential. In the case of a closed path, which surrounds an area $S$, the phase shift is proportional to the flux $\Phi = BS$ where $B = \text{rot} A$ is an applied field. Since Anderson localization results from superposition of possible closed paths of electrons, this leads to an anisotropy of magnetoresistance when the applied field is inclined by an angle $\theta$ to the conducting plane, and effective area becomes $S \sin \theta$. Without spontaneous magnetization [16], 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 $\sin^2 \theta$. This explains the observed magnetoresistance as shown in Fig. 4.

Therefore, two-fold symmetry in magnetoresistance is a natural consequence of the Anderson localization picture.

In fact, motion of individual electrons was shown to be inapplicable to charge transport in $\alpha$-TaS$_3$ at this temperature. As we mentioned above, no free electron remains in CDW state of $\alpha$-TaS$_3$ because all the electrons at the Fermi surface contribute to form CDW, and thermally excited quasiparticles are negligible at 4.2 K (Figs. 11 [2]). The anisotropic magnetoresistance was observed at the nonlinear $I$-$V$ regime, where CDW carries electric charge. It is known that the gradient of CDW phase corresponds to electric charge [4]. Miller Jr. et al. explained the Aharonov-Bohm oscillations in terms of quantum dynamics of a kink of CDW phase, namely, CDW soliton which can respond to the applied field [1, 2]. Recently, it is proposed that the CDW solitons are randomly distributed over the sample [17]. This model explains the synchrotron X-ray spectra of $\alpha$-TaS$_3$ have both commensurate and incommensurate peaks [18].
We assume that Anderson localization of a CDW soliton occurs. It is known that Anderson localization is not limited to electron systems, but can occur in any kind of waves in random media, e.g. photon [19], radio wave [20], sound [21], and matter wave [22]. Our assumption of the Anderson localization of a CDW soliton provides a straightforward explanation. As revealed by the experiment of the CDW Aharonov-Bohm oscillations, CDW solitons are accompanied with quantum phase. When the magnetic field is applied, the quantum phases of two wave functions shift relatively each other, and the system becomes delocalized. Consequently, negative magnetoresistance occurs.

Now we remind that the offset angle depends on the sample. This observation implies that the direction of a conducting plane is independent to the crystal axes. Synchrotron study of o-TaS$_3$ revealed coexistence of commensurate and incommensurate CDWs [18]. Since the incommensurate peak in X-ray diffraction disappears below 50 K, we expect all CDWs are commensurate at 4.2 K. When the current flows in the CDW, it is plausible to a discommensuration-like structure generates on a certain plane. The angle between the plane and the crystal axis may be determined by distribution of the CDW solitons.

Our discussion provides the reason why Aharonov-Bohm oscillations are able to be observed in the CDW rings. The ring crystal is thought to be grown on a droplet of chalcogen, guided by surface tension [23]. This growth mechanism is plausible since a ring surrounding a droplet has the smallest surface tension. If the CDW soliton must be confined with a chain as proposed in Ref. [3], the proposed model seems too strict because a chain should atomically coalesce when a ring crystal is grown. However, our study implies that the quantum phase of CDW can interfere within a conducting plane. This loosens necessary conditions for formation of a CDW loop.

Finally, we would like to point out some topics related with our result. Quantitative study of the magnetoresistance (Fig. 3) remains unresolved. This requires further modelling for dynamics of CDW solitons than that proposed in Refs. [1], [2] where randomness of the solitons is not involved. Degrees of freedom in configuration of CDW soliton may be the source of low temperature specific heat [24] which shows extended relaxation. This also suggests the randomness plays a important role in CDW dynamics at low temperatures.

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FIG. 1: Temperature dependence of $o$-TaS$_3$. Discontinuity is due to change of measurement current. Dotted line is an Arrhenius fit with the activation energy of 400 K to see if the low temperature data were not rounded by nonlinearity.
FIG. 2: Current-voltage ($I-V$) characteristics of the $o$-TaS$_3$ sample observed at 4.2 K. The $I-V$ curve is significantly nonlinear. A slight hysteresis is found near $I=0$ as shown by the arrows.
FIG. 3: Magnetoresistance of the α-TaS$_3$ sample observed at 4.2 K. θ = 0 (open circles) means the magnetic field is parallel to the current flow, and at θ = 90° (solid circles) the field is perpendicular to the ribbon face, 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 ±2σ, where σ is a standard deviation of each data.
FIG. 4: Angle dependence of the magnetoresistance in the field $B = 5.2$ T observed at 4.2 K. The error bars represent $\pm 2\sigma$, same as Fig. 3. Two-fold symmetry is clearly exhibited. The solid lines are results of the least-square fit to the formula $\Delta R = -A \sin^2(\theta - \theta_0)$, where the offset angle $\theta_0 = -7^\circ$ and $59^\circ$ in the panels (a) and (b), respectively. The other sample exhibits $\theta_0 = 106^\circ$ (not shown in the figure.)