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Charge density wave circulating current in NbSe$_3$ rings observed by measuring Shapiro interference

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Abstract. We investigate topological effect of charge density wave (CDW). The Shapiro step measurement was performed on NbSe$_3$ topological (ring) crystals. We found that a beat-like splitting of Shapiro step spectrum. These results suggest that the circulating CDW current flows in the ring, and modifies the CDW fundamental frequency, namely narrow-band-noise frequency.

1. Introduction
The charge density wave (CDW) was originally proposed as a model of superconductivity by Fröhlich in 1954 [1]. The sliding CDW state was discovered in quasi-one-dimensional conductors such as NbSe$_3$ crystals in the 1970's [2], however Fröhlich's CDW supercurrent has yet to be observed because the Fröhlich supercurrent realizes only in an infinite and ideal crystals. Recently, the Hokkaido university group discovered topologically nontrivial loops of the NbSe$_3$ crystals, including rings, Möbius-strips, figure-8s [3, 4] and Hopf-links of crystals [5]. Since the topological crystals of the CDW materials are closed themselves, they are similar to an infinite crystals. Therefore, topological crystals are a candidate system for realizing Fröhlich’s CDW supercurrent.

In this paper, we report the Shapiro interference measurement for ring-shaped crystals of NbSe$_3$ to explore Fröhlich’s CDW supercurrent. Shapiro interference measurement provides detailed information about the CDW current [6, 7, 8]. We found a beat-like structure of the Shapiro step spectrum in the NbSe$_3$ rings. Two possible models are proposed to explain the results.

2. Experimental
The experimental setup was as follows. Ring-shaped crystals of NbSe$_3$ were synthesized by the chemical vapor transportation method [4]. The ribbon-like quasi-one dimensional crystals of NbSe$_3$ form seamless loop crystals by bending and joining naturally under controlled conditions. A ring-shaped crystal with an outer diameter of 120 µm, an inner diameter of 10 µm, and a thickness of 40 µm, was fixed on a sapphire insulative plate by epoxy adhesive. Then two electrical contacts were attached to the surface of the ring using the gold evaporation method and silver conductive paste. Fig. 1 (a) shows an optical microscopy image of a typical thick ring-shaped crystal of NbSe$_3$ equipped with two electrical contacts. The sample was placed on a copper block in an evacuated chamber. We confirmed that the first CDW transition occurred
at 144 K, which corresponded to the results of previous research [9, 10]. For simplicity we measured the Shapiro step spectrum at 120 K because there are two types of CDWs below the second transition temperature ($T_{C2} = 59K$). Moreover, the Shapiro step spectrum can be observed clearly around 120 K since the dc resistance anomaly is at its maximum value. A PID heater control system kept the temperature of the sample within 120±0.1 K. Similar with Josephson junctions, when ac and dc voltages are applied to CDW materials, it is well known that harmonic and subharmonic Shapiro steps are observed in dc current-voltage characteristics [?, ?]. The condition under which a step appears is,

$$\frac{I_{dc}}{2eN} = \frac{p}{q} f_{ex},$$

(1)

where $e$ is the electron charge, $N$ is the number of one-dimensional chains in the crystal, $f_{ex}$ is the frequency of the ac voltage, and both $p$ and $q$ are integers. The left side corresponds to the fundamental frequency of the voltage oscillation, namely narrowband noise produced by the sliding CDW. We measured differential resistance $dV_{dc}/dI_{dc}$ as a function of dc current (time-averaged current) at a constant temperature ($T = 120 K$) and with an ac voltage. Fig. 1 (b) shows the electrical circuit. A Keithley 6220 current source and a Hewlett-Packard HP8712ET network analyzer (frequency range of 300 kHz to 1.3 GHz) were used as dc and ac current sources, respectively. Differential resistance ($dV_{dc}/dI_{dc}$) was measured using a lock-in-amplifier (Stanford Research SR 830DSP) to observe a clear Shapiro step spectrum.

2.1. Results and discussions

The Shapiro step spectrum of the CDW ring was clearly observed. Fig. 2 shows differential resistance $dV_{dc}/dI_{dc}$ as a function of dc current without ac voltage and with an ac voltage of amplitude $V_{ac} = 65$ mV and frequency $f_{ex} = 40, 80, 100, 200,$ and $300$ MHz. The value of the differential resistance at 120 K and $V_{dc} = 0$ mV was 34.7 Ω, which included the contact resistance since this was a two-contact measurement. Since uncondensed electrons remain below the first transition temperature, the resistance of NbSe$_3$ crystals is ohmic at low voltage. The contact resistance is estimated to be about 8 Ω from the temperature coefficient of resistance above the first transition temperature (metallic state). Using the sample dimensions, we estimated that resistivity is of the order of $10^{-3}$ Ωm, on the other hand, the usual resistivity of NbSe$_3$ is of the order of $10^{-6}$ Ωm at $T = 120 K$. Hence only 0.1 % of the one-dimensional chains contributes to conduction. This result suggests that the current is concentrated only near the surface of the crystal.

Figure 1. (a) Optical microscopy image of a typical ring-shaped crystal of NbSe$_3$ equipped with two electrical contacts attached by silver paste. (b) Electrical circuit for measuring of the Shapiro steps (ac-dc interference effect). The thick ring at the center of the circuit is the ring-shaped crystal of NbSe$_3$. Both dc and ac currents are induced into the crystal via the two electrical contacts.
Differential Resistance $dV_{dc}/dI_{dc}$ versus dc current $I_{dc}$ at $T = 120$ K without ac voltage and with ac voltage of amplitude $V_{ac} = 65$ mV and frequency $f_{ex} = 40, 80, 100, 200,$ and 300 MHz, respectively. Harmonic and subharmonic Shapiro steps are indicated by arrows and index $p/q$, where $p$ and $q$ are integers. Moreover, there are small beat-like peaks ($p/q$ and $p/q$) on both sides of the main Shapiro peaks.

In the absence of ac voltage ($V_{ac} = 0$ mV), a clear threshold voltage and current were observed at $V_{th} = 9.4$ mV and $I_{th} = 87 \mu$A, respectively. The threshold electric field was estimated at 520 mV/cm, which is similar order to that of usual NbSe$_3$ crystals (whiskers) [11]. In the previous paper, two threshold voltages was observed in a ring-shaped crystal with electrical contacts [12]. However, we observed only one threshold current. This result reflects the fact that the two electrical contacts were symmetrically attached to the ring.

When the ac voltage increases, the ohmic region decreases and the Shapiro steps appear as $dV_{dc}/dI_{dc}$ peaks. Fig. 2 (b) shows many sharp peaks. Almost all the large peaks can be classified by index $p/q$, where $p$ and $q$ are integers. When $p/q$ is an integer, the peak corresponds to a harmonic Shapiro step. On the other hand, when $p/q$ is a rational number the peaks are called subharmonic steps. We also observed subharmonic peaks at $p/q = 1/3$, $1/2$, $2/3$, $3/2$, and $5/2$. The CDW current $I_{CDW}$ is roughly proportional to $I_p = I_{dc} - I_{th}$, where $I_{dc}$ is the dc current.
value at the Shapiro peak and $I_{th}$ is the threshold current of each ac voltage, hence the peak position $I_p$ must be proportional to frequency. From equation (1), we can roughly estimate the effective number of the one-dimensional chains $N$ from the current value at the first peak ($p/q = 1$). Then we find that about $2 \times 10^7$ chains contribute to the current. This value corresponds to the order of the effective area estimated from the resistivity.

Moreover, we found small beat-like splitting of the peaks on both sides of the main Shapiro peaks. The beat-like peaks are completely different from usual subharmonic peaks because the distance between the main peak and the beat peak is increased when the dc bias current is increased. Therefore, the peak cannot be classified as subharmonic.

The imbalance between the currents flowing through the left and right arms can explain the presence of two peaks, however, it cannot explain the observation of both one main peak and two beat peaks. Two possible models can be proposed. The first one is that the CDW current density is inhomogeneous. Due to the curvature of the ring-shaped crystals, the electric field might be inhomogeneously applied the CDWs. Hence, the CDW velocity may distribute at the part of the crystals. The current density distribution will derive the splitting of the Shapiro steps. The second model is that the beat structures of the Shapiro step spectrum result from a circulating current of the CDW. When the circulating CDW current flows in the ring, the resonance condition of the Shapiro step spectrum is modified as

$$\frac{I_{dc} \pm \delta I}{2eN} = \frac{p}{q} f_{ex},$$

(2)

where $\pm \delta I$ is additional current flowing through the right and left arms. Due to the circulating current $\delta I$, the CDW current flowing in the right arm must decrease (or increase) and that flowing in the left arm must increase (or decrease). Then the beat-like peaks appear on the both sides of the main peaks.

2.2. Conclusion

In conclusion, we observed a clear harmonic and subharmonic Shapiro step spectrum and a beat-like splitting of the Shapiro peaks in differential resistance-dc current characteristics. The beat spectrum suggests that few types of CDW current density exist. As a possible model, the circulating CDW current flowing the rings can explain the splitting of the steps.

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