Evidence of circulating charge density wave current: Shapiro interference in $NbSe_3$ topological crystals

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(Received 6 October 2008; published 28 January 2009)

We investigate the topological effect of charge density wave (CDW). We found a "beat" Shapiro step structure in $NbSe_3$ topological (ring) crystals. We interpreted it as an evidence of the circulating CDW current that flows in the rings and modifies the CDW fundamental frequency, namely, narrow-band-noise frequency. This result also implied that a precursor of Fröhlich's CDW supercurrent emerged in the ring system.

DOI: 10.1103/PhysRevB.79.014304 PACS number(s): 71.45.Lr, 72.15.Nj, 73.23.Ra, 74.50.+r

I. INTRODUCTION

The charge density wave (CDW) was originally proposed as a model of superconductivity by Fröhlich[1] in 1954. The sliding CDW state was discovered in quasi-one-dimensional conductors such as $NbSe_3$ crystals in the 1970s;[2] however Fröhlich's CDW supercurrent has yet to be observed. Recently, the Hokkaido University group discovered topologically nontrivial loops of the $NbSe_3$ crystals, including rings, Möbius strips, figure 8*s*,[3,4] and Hopf links of crystals.[5] The discovery of topological crystals of the CDW materials completely changed the situation. Since one-dimensional chains of these crystals have no edges, they allow the CDW to flow without electrical contacts. Both edges of crystals and electrical contacts have been considered as strong pinning centers that inhibit Fröhlich's supercurrent. Therefore, topological crystals are a candidate system for realizing Fröhlich's CDW supercurrent.

In this paper, we report a phenomenon that is a precursor to Fröhlich's CDW supercurrent. Shapiro interference measurement is a very useful approach for investigating macroscopic quantum states because a Shapiro spectroscopy provides detailed information about the CDW current.[6–8] We found the beat structure of the Shapiro step spectrum only in the $NbSe_3$ rings. The beat signal results from the existence of a circulating CDW current, which modifies the fundamental frequencies of sliding CDW (narrow-band noise) in the right and left arms. The phason damping time of the circulating CDW must be longer than at least 10^{-8} s, which is three orders longer than that of the conventional CDWs in $NbSe_3$ whiskers.[9] The elongation of the damping time suggests that the circulating current is the Fröhlich CDW supercurrent.

II. EXPERIMENT

The experimental setup was as follows. Ring-shaped crystals of $NbSe_3$ were synthesized by the chemical vapor transportation method.[4] The ribbonlike 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. Figure 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.[10,11] 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}=59 K). Moreover, the Shapiro step spectrum can be observed clearly around 120 K since the dc resistance anomaly is at its maximum value. A feedback heater control system kept the temperature of the sample within 120 ± 0.1 K.

The Shapiro interference is a common phenomenon observed with various condensed-matter systems including superconducting Josephson junctions,[12] vortex flows,[13] weak-linked superfluids,[14] and CDW systems. 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.[15,16] The condition under which a step appears is

$$\frac{I_{dc}}{2eN} = \frac{p}{q} f_{ex}, \tag{1}$$

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, narrow-band 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. Figure 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. We used the capacitive high-pass filters and inductive low-pass filters to apply both dc and ac electric fields to the sample. Coaxial cables and microstrip lines were employed to achieve impedance matching of the ac lines. The temperature dependence of the circuit was sufficiently small during the measurement. Differential resistance (dV_{dc}/dI_{dc}) was measured using a lock-in amplifier (Stanford Research SR 830DSP) to observe a clear Shapiro step spectrum. The frequency and amplitude of the

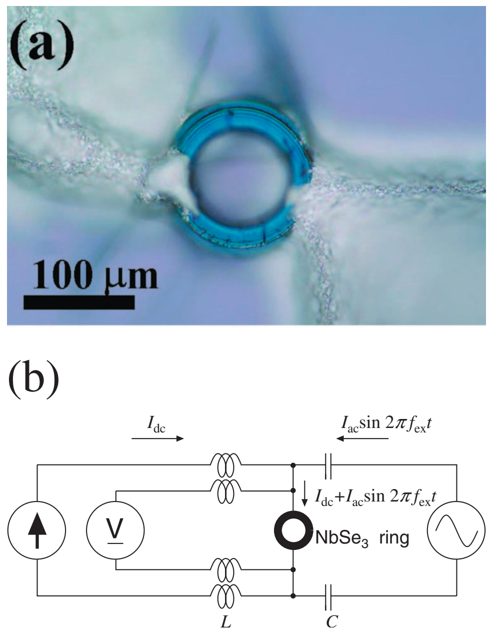

FIG. 1. (Color) (a) Optical microscopy image of a typical ring-shaped crystal of $NbSe_3$ equipped with two electrical contacts attached by silver paste. (b) Electrical circuit for measuring the Shapiro steps (ac-dc interference effect). The thick ring at the center of the circuit is the ring-shaped crystal of $NbSe_3$. Both dc and ac currents are induced into the crystal via the two electrical contacts. Inductive low-pass filters and capacitive high-pass filters separate the dc and ac lines. Differential resistance is measured as a function of dc current. The sample and filters are placed in an evacuated chamber and cooled to 77 K with liquid nitrogen.

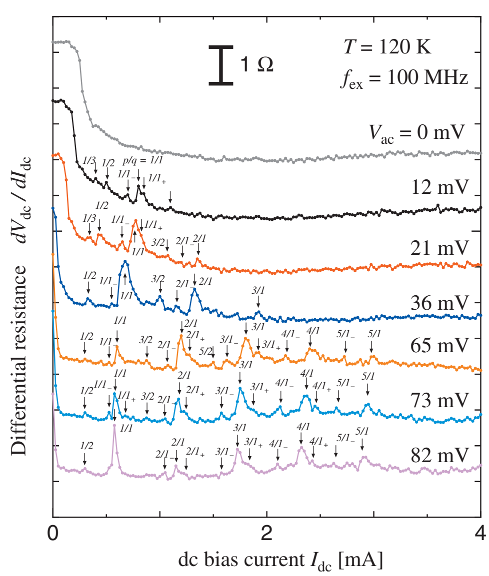

FIG. 2. (Color) dc bias current dependence of differential resistance dV_{dc}/dI_{dc} with applied various ac voltages of $f_{ex}=100$ MHz at $T=120$ K. The ac voltages V_{ac} are 0, 12, 21, 36, 65, 73, and 82 mV from top to bottom. Harmonic and subharmonic Shapiro steps are indicated by arrows and index p/q, where p and q are integers. Moreover, there are small beat peaks (p/q_- and p/q_+) on both sides of the main Shapiro peaks.

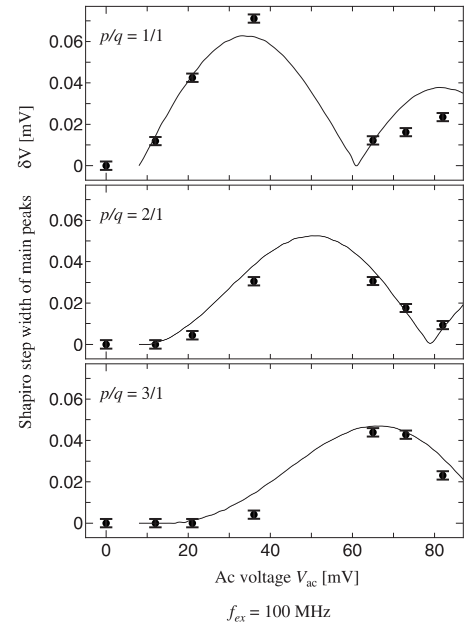

FIG. 3. ac voltage amplitude dependence of Shapiro step width δV of main peaks of (a) $p/q=1/1$, (b) 2/1, and (c) 3/1. The step width is estimated as the product of peak height and half width of half maxim of each peak of Fig. 2. Solid lines are calculation results of amplitudes of Bessel functions $|J_1(x)|$, $|J_2(x)|$, and $|J_3(x)|$, respectively.

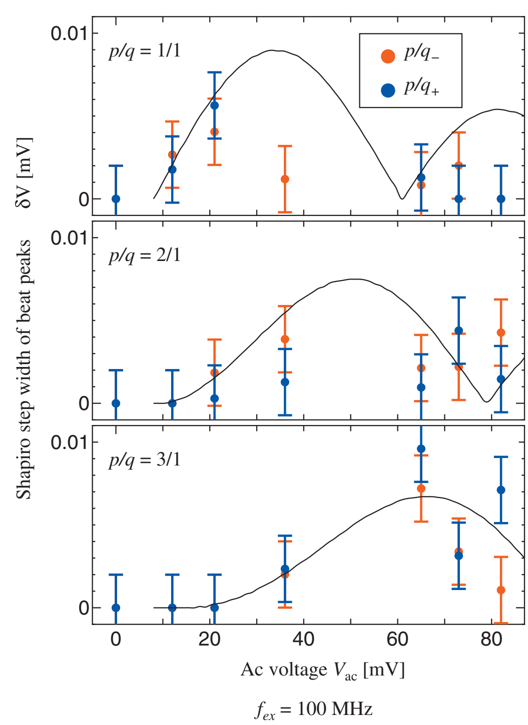

FIG. 4. (Color) ac voltage amplitude dependence of Shapiro step width δV of negative and positive beat peaks of (a) $p/q=1/1$, (b) 2/1, and (c) 3/1. The calculation curves of amplitudes of Bessel functions $|J_1(x)|$, $|J_2(x)|$, and $|J_3(x)|$ are the rescaled ones in Fig. 3.

ac signal for the differential resistance measurement were set at 33 MHz and 1 μA, respectively, which were sufficiently smaller than that of the ac field for producing Shapiro steps.

III. RESULTS AND DISCUSSIONS

The Shapiro step spectrum of the CDW ring was clearly observed. Figure 2 shows differential resistance dV_{dc}/dI_{dc} as a function of dc bias current at applying various ac voltages of 100 MHz at 120 K. In the absence of ac voltage ($V_{ac}=0$ mV), the value of the differential resistance at $V_{dc}=0$ 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} Ω m; on the other hand, the usual resistivity of $NbSe_3$ is of the order of 10^{-6} Ω m 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.

Increasing dc bias current, a clear threshold voltage and current were observed at $V_{th}=9.4$ mV and $I_{th}=87$ μA, respectively. The threshold electric field was estimated at 520 mV/cm, which is similar of order to that of usual $NbSe_3$ crystals (whiskers).[17] In a previous paper, two threshold voltages were observed in a ring-shaped crystal with electrical contacts.[18] 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.

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, 3/2, and 5/2.

Moreover, we found small beat peaks on both sides of the main Shapiro peaks with only the CDW loops. The beat 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 beat peaks at negative and positive sides of the main peaks are indicated by p/q_- and p/q_+, respectively. The beat structures were reproduced in other three ring-shaped crystals with different diameters and thicknesses.

Peak heights of both the main Shapiro peaks and the beat peaks depend on V_{ac}. It is well known that the Shapiro step widths, which are voltage step width in current-voltage characteristics, follow Bessel functions of ac voltage.[7] We estimated the Shapiro step width δV as the product of the peak height and the half width at half maximum of each dV_{dc}/dI_{dc} peak. Figures 3(a)–3(c) show ac voltage dependence of the Shapiro step width of $p/q=1/1$, 2/1, and 3/1, respectively. The solid curves are results of numerical calculation of amplitudes of Bessel functions $J_n(x)$. The experimental data of $p/q=1/1$, 2/1, and 3/1 follow the calculated curves of $J_1(x)$, $J_2(x)$, and $J_3(x)$, respectively. These results correspond to the previous researches.

Figures 4(a)–4(c) show ac voltage dependence of the Shapiro step width of negative and positive beat peaks of $p/q=1/1$, 2/1, and 3/1, respectively. Both beat peaks have similar magnitudes and they also follow the curves of amplitudes of Bessel functions, which are the rescaled ones in Fig. 3. These results imply that there is a common origin of the negative and positive beat peaks.

Figure 5 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 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 the frequency. From Eq. (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 2×10^7 chains contribute to the current. This value corresponds to the order of the effective area estimated from the resistivity.

Figure 6 shows frequency dependence of the peak position I_p, the harmonic and subharmonic peaks, and the negative and positive beat peaks. The peak positions increase with increasing frequency. These features correspond to those of Shapiro steps observed on usual $NbSe_3$ crystals.[16] The beat peak positions I_p are proportional to the frequency in the same way as the harmonic and subharmonic peaks and are independent of V_{ac}. Although some peak heights were small, the positions of beat peaks fall on the same lines in Fig. 6.

What is the origin of the beat peaks? Since the ring sample has two current paths, an imbalance between the currents flowing through the left and right arms would make Shapiro peaks split. However, this model can explain only the presence of two peaks because there are only two kinds of CDW current. It is not consistent with the observation of both one main peak and two beat peaks.

Another idea is that the CDW current flows inhomogeneously in the samples. For example, difference in current density, the inner and outer circumferences made a difference in current density along radial direction and derived several Shapiro peaks. However, it is not consistent with our results because the beat peaks were also commonly observed in all other three samples with different dimensions, although the inhomogeneity must be strongly dependent on thickness and radius. Moreover, the magnitude of the step widths of both beat peaks (see Fig. 4) supports that the pairs of the beat peaks originate from a common mechanism. Therefore, it is reasonable to consider that the beat peaks are caused by an interference effect associated with the ring topology and not with inhomogeneous current flow.

We propose a plausible model that the beat structures of the Shapiro step spectrum result from a circulating current of the CDW as shown in the inset of Fig. 7. When the circulating CDW current flows in the ring, the resonance condition of the Shapiro step spectrum is modified as

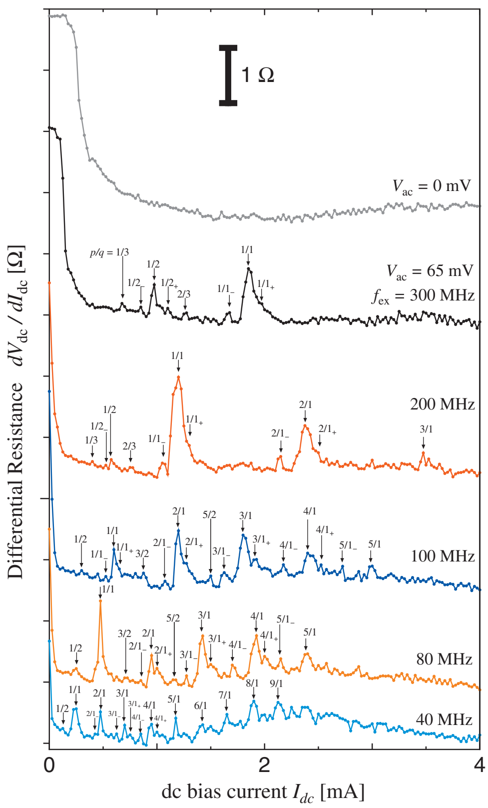

FIG. 5. (Color) Differential resistance dV_{dc}/dI_{dc} versus dc current I_{dc} at T=120 K without ac voltage and with ac voltage of amplitude V_{ac}=65 mV and frequency f_{ex}=40, 80, 100, 200, and 300 MHz, respectively.

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

where $\pm\delta I$ is the additional current flowing through the right and left arms. Since a temporal circulating current δI flows through the ring, 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 peaks appear on both sides of the main peaks. Figure 7 shows the slope of each line in Fig. 6 as a function of index p/q. The slopes of the main and two beat peaks are proportional to p/q. The coefficient of p/q of the main peak is 5.964 $\times 10^{-3}$ mA/MHz, and the coefficients of the beat peaks are 5.348×10^{-3} and 6.256×10^{-3} mA/MHz. Since the slopes of the beat peaks fall on the same lines, the circulating current δI is proportional to dc bias current I_{dc}. The difference between the coefficients of the negative beat peaks and the main peaks is greater than that between the positive beat peaks and the main peaks. In the model, the beat peaks must appear symmetrically. Since we observed symmetric beat peaks with other ring samples, the asymmetry may not be an essential property. Although this simple model does not completely explain the asymmetry of peak position of negative and positive beat peaks, it is sufficiently consistent with experimental results of frequency dependence and ac voltage dependence of the beat peaks.

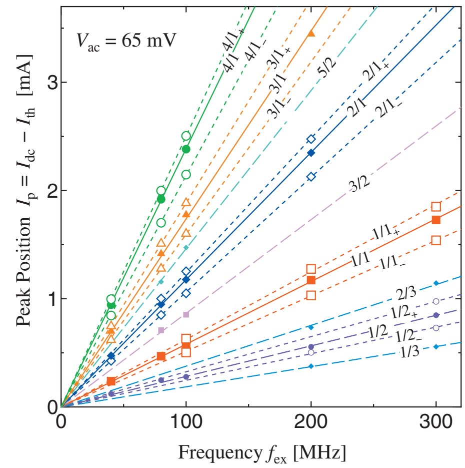

FIG. 6. (Color) Frequency dependence of peak position I_p=I_{dc} $-I_{th}$ of each step. The peak positions of the harmonic, subharmonic, and beat peaks are proportional to frequency. The slopes of each line are shown as a function of the index p/q in Fig. 7.

Results of the previous researches for the dynamical property of CDWs in $NbSe_3$ rings suggested the existence of a circulating current mode. Okajima *et al.*[18] found several anomalous dV_{dc}/dI_{dc} steps in the sliding state. In our previous paper,[11] we found that the pinning frequency of a ring is smaller than that of a cut ring from the investigation of the frequency dependence of ac conductivity of a CDW ring in the pinning state. These results implied that the CDW dynamics in $NbSe_3$ rings is different from conventional dynamics due to the circulating CDW current mode.

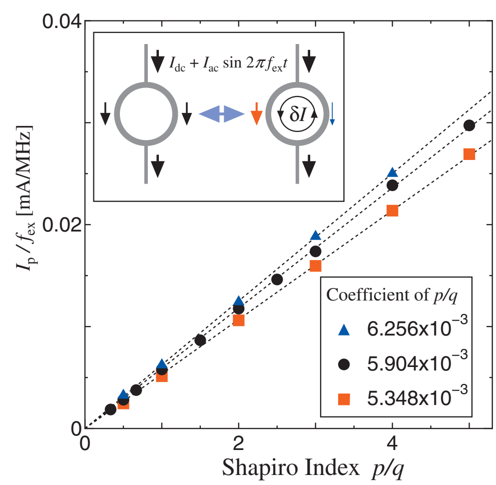

FIG. 7. (Color) Slopes of each peak position versus frequency I_p/f_{ex} as a function of index p/q. Circles, squares, and triangles indicate the main peaks (harmonic and subharmonic peaks), negative beat, and positive beat peaks, respectively. Each I_p/f_{ex} is proportional to p/q. The coefficients of p/q for each line are shown in the box. They were calculated by the least-squares method. The inset shows a model of the circulating current to explain the beat structure of the Shapiro step spectrum. If the circulating current is temporarily excited by an external electric field, it modifies the currents through the right and left arms as $I\pm\delta I$. As a result, fundamental frequency of each arm is modified and two beat peaks are produced around the main peaks.

The circulating current is possibly Fröhlich's supercurrent. The experimental results show that the circulating current is proportional to the external dc current because the peak positions of the negative and positive beat peaks are proportional to the index p/q. A possible mechanism for exciting the circulating current is as follows. When dc voltage is applied, a CDW phase slip occurs at the electrical contacts. Since the electrical contacts were actually not completely symmetrically attached, the electric-field imbalance induces a circulating current of CDW. The current must flow for at least 10^{-8} s, which is the ratio of the circumference of the ring-shaped crystal and the CDW phase velocity. This time is three orders longer than the phason damping time of $NbSe_3$ whiskers ($\tau \sim 10^{-11}$ s).[9] Experimental[10,19] and theoretical[20,21] researches of the CDW transition temperatures in topological crystals concluded that phase fluctuation is enhanced by the closed-loop topology. The enhanced fluctuation allows a CDW in each one-dimensional chain to move independently. Hence, the damping time becomes longer than that observed with conventional measurements, and the CDW reverts to the type that Fröhlich[1] considered.

Shapiro step spectrum measurement allows us to detect the circulating current. A conventional dc current measurement cannot detect the circulating current because, although a superconducting current circulates in the rings, the total time-averaged current does not change. Moreover, since a CDW is a kind of electron crystal, the electrical contacts essentially change the dynamic property of the CDW. In principle, a CDW system has the inertial and elastic factors and quantum tunneling property.[22] Those factors play important roles in the dynamics of a CDW supercurrent system. Investigations of the CDW dynamics with the topological crystals will reveal the hidden intrinsic properties of the CDWs.

IV. CONCLUSION

In conclusion, we observed clear harmonic and subharmonic Shapiro step spectra and a beat spectrum around the main Shapiro peaks in differential resistance-dc current characteristics. The beat spectrum suggests that a circulating CDW current flows in the ring. This is a sign of Fröhlich's CDW supercurrent. The realization of Fröhlich's CDW supercurrent will open a different field of condensed-matter physics that may be associated with the supersolid state of ^{4}He (Ref. 23) and have applications to macroscopic quantum coherent devices working at high temperatures.

ACKNOWLEDGMENTS

We thank K. Yamaya, K. Ichimura, M. Oda, H. Orihara, O. Matsuda, V. Osipov, D. Churochkin, M. Tsubota, J. Hara, T. Tsuneta, T. Toshima, and K. Shimatake for fruitful discussions, helpful suggestions, and technical support. This research was supported by the Japan Society for the Promotion of Science and the 21 COE program on "Topological Science and Technology" from the Ministry of Education, Culture, Sport, Science and Technology of Japan.

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