New Crystal Topologies and the Charge-Density-Wave in $NbSe_3$

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Abstract

We successfully synthesized new crystal forms of $NbSe_3$, which is a well-known charge-density-wave conductor. The new forms are basically a loop of diameters 10-200 μm with/without a twist (ex. ring, Möbius strip). Their distinctive topology seems to offer systems for studying possible new quantum phenomena. Samples were obtained by chemical vapor transportation method and identified as $NbSe_3$ crystals by electron diffraction. SEM studies revealed that a loop is formed from a $NbSe_3$ whisker that wraps around a droplet of selenium by forces of surface tension. Also we observed that a twist is introduced during this spooling process. Moreover, we performed transport measurements on the materials and found that, exclusively in samples with a twist, T_{CDW} is lower by a few kelvins than that of $NbSe_3$ whiskers. We suggest that the anomaly originates in local strain due to the twisted topology.

Key words: charge-density-wave; mesoscopic; crystal growth

Since Berry proposed his idea of geometrical quantum phase [1], quantum phenomena based on system topology have been intensively investigated because of their fundamental role in quantum mechanics. Electronic systems in a ring geometry give some examples, e.g. Aharonov-Bohm effect or Aharonov-Casher effect. While such phenomena for free careers are fairly demonstrated, topological effects on correlated electronic systems have turned up as a new interesting problem. Especially, systems that involve macroscopic quantum states, such as superconductivity or charge-density-wave (CDW), are attracting much interest. Possible new macroscopic quantum phenomena were predicted for a Möbius strip superconductor [2] or a CDW ring threaded by a magnetic flux [3].

$NbSe_3$ is a monoclinic ($P2_1/m$) crystal that has a quasi-one-dimensional structure and electric conductivity, which leads to two CDW transitions at 141K and 58K. $NbSe_3$ has been known to form a thin whisker crystal. However, recently we found instability against bending of the matter due to the low-dimensionality results in several new crystal forms with folded structure [4]. The new forms are ring (cylinder), Möbius strip and "figure-of-eight" strip made from $NbSe_3$ whiskers. The topologies of each form can be considered as physical realization of ribbon-knots group in mathematics, which deals with geometries of two-faced, closed loops. So we named the materials topological crystals. The new materials have a size of few μm at their smallest, which is comparable to the CDW coherent length of $NbSe_3$. Thus the new materials can provide mesoscopic CDW systems suitable for investigation of topology-dependent quantum phenomena.

In this paper, we discuss their formation mechanisms and investigate characteristic features of each topological class in their CDW states through transport measurements.

Samples were synthesized simultaneously with standard $NbSe_3$ whiskers by chemical vapor transportation. Reacting condition was as follows: a stoichiometric mixture of selenium (Se) and niobium (Nb) powder was sealed in an evacuated quartz ampoule. Then they were reacted at 740 °C with a temperature gradient of 1°C/cm for few hours - few days. The maximum yield of topological crystals was about a hundred samples per run, that is 0.01% of total yield of whiskers.

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Topological crystals were identified as a single crystal of $NbSe_3$ by means of electron diffraction. It is also revealed that the circumferential direction of a loop crystal is the b-axis (the best-conductive axis).

Fig. 1 shows SEM micrographs of typical samples; (a) a twist-free ring (b) a figure-of-8 strip and (c) a Möbius strip. These crystals are classified by the twists in their peripheries, 0, 2π and π, respectively. The twist number n is introduced, which represents a twist of $n\pi$. The three shapes are referred as $n\pi$ loops hereafter. SEM study revealed a pristine loop formed from a whisker. Fig. 1 (d) shows a $NbSe_3$ whisker attached to a Se drop by surface tension, eventually rapping around the drop. A n=0 loop will appear after coalescence by Van der Waals force of the ends of the whisker. Also we observed twists introduced into whiskers during spooling, which is apparently relevant to formation of $n\neq 0$ loops. The origin of twists is ascribed to shear components in elastic compliance matrix of a monoclinic crystal, which produce a twist in response to a simple bending force.

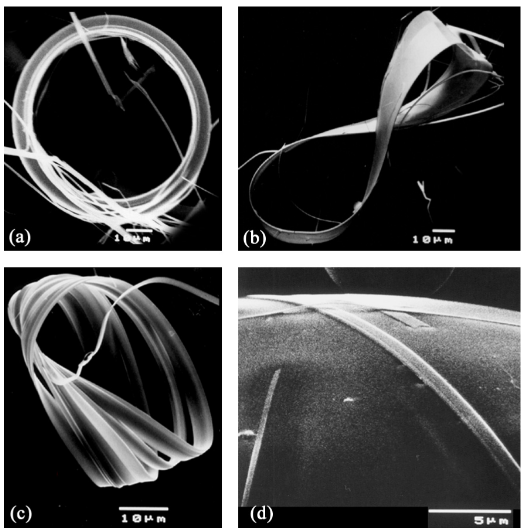

Fig. 1. SEM micrographs of typical topological crystals: (a) ring (n=0 loop), (b) figure-8 strip (n=2 loop), (c) Möbius strip (n=1 loop). (d) shows a drop of Se (the sphere partly shown in the figure) perfectly surrounded by one of the whiskers adhering to the sphere (the strips), which is a pristine ring.

Transport measurements on $n\pi$-twisted loops were performed at temperatures down to 4.2K from room temperature. Silver paste was used to make two electric contacts to samples. Fig. 2 shows typical temperature-dependence of resistivity for loops of n=0, 2 and a whisker. The transitions into CDW state at about 58K and 141K were confirmed for every sample. However, solely n=2 samples showed disagreement with whiskers and n=0 samples about transition temperature T_{Ci} (i=1,2 represent the upper and lower transition). Table 1 summarizes T_{Ci} for all measured samples. Note that the considerable decreases in T_{Ci} (up to 4K) are observed for only n=2 samples. Additionally, value of the decrease varies for each sample. This fact suggests the existence of fluctuation that makes CDW transitions smeared in certain samples.

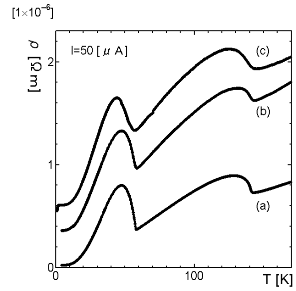

Fig. 2. Temperature-resistivity graph of (a) a whisker, (b) n=0 and (c) n=2 samples.

	whisker	**0πA**	**0πB**	**2πA**	**2πB**	**2πC**
T_{C1}	141.0	142	140.8	139.3	139.9	137.0
T_{C2}	57.6	58	57.4	53.5	56.9	53.4

Table 1
CDW transition temperature of measured samples.

Strain due to bending and twisting is a legitimate suspect of the origin of such topology-dependant and sample-specific behavior, because strain depends on both topology and dimensions of each sample. CDW transition is known to be unstable against modulation of Fermi surface induced by strain. It is demonstrated experimentally that uniaxial strain causes quadratic drop in T_{C2} by an order of -0.4K/GPa^2 for $NbSe_3$ whiskers [5]. Surprisingly, strain in actual samples is estimated at so large value that T_{Ci} is supposed to decrease for even n=0 samples. In fact, strain would exceed elastic limit of week interlayer bonding in $NbSe_3$, so that strain by bending must be relaxed by plastic deformation and not important here. A possible origin of fluctuations in n=2 samples is strain caused by twisting. Unlike the case of bending, it involves shear component, which is harder to be relaxed in $NbSe_3$. We thus suggest that crystal topology affects electrical behavior through elastic and plastic properties.

References

[1] M. V. Berry, Proc. R. Soc. London Ser. A **392** (1984) 45.

[2] M. Hayashi, N. Kawamoto, T. Kuramoto, K. Shigemoto, J. Phys. Soc. Jpn.bf 70 (2001) 3495.

[3] E. N. Bogachek, I. V. Krive, I. O. Kulik, A. S. Rozhavsky, Phys. Rev. B**42** (1990) 7614.

[4] S. Tanda, T. Tsuneta, Y. Okajima, K. Inagaki, K. Yamaya, N. Hatakenaka, Nature **417** (2002) 397.

[5] R. S. Lear, M. J. Skove, E. P. Stillwell, J. W. Brill, Phys. Rev. B**29** (1984) 5656.