Global evaluation of closed-loop electron dynamics in $NbSe_3$ ring

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Owing to the recent progress of material science, various topological structures such as ring, Mebius, and figure of eight have been obtained from quasi-one-dimensional (1D) transition metal (M) chalcogenides (X) of the type MX_3 [1]. Since the correlation length of CDWs in MX_3 is on the order of several μm, the coherent CDWs within a closed-loop chain provide an opportunity for investigating the influence of topology on their dynamics. In this paper, we report a characteristic nonequilibrium electron dynamics associated with a closed-loop topology.

The samples studied here are $NbSe_3$ ring and whisker. Both crystals were grown by the chemical vapor transport method, which usually show a whisker structure. Under controlled conditions, thin whiskers naturally form closed-loop crystals by bending and joining [2]. Several ring crystals are stacked layer by layer, and form a disk-like structure, which we investigated as a closed-loop ring in this measurement (Fig. 1(a)). The whisker crystal has a length of a few mm along the conducting *b*-axis (chain axis) and width of 50 μm. The disk-like ring crystal with a diameter of ~100 μm has a bending *b*-axis along the azimuthal direction. The circumference of the center hole of the disk is around 10 μm, which is comparable to the correlation length of CDWs (>2.5 μm [3]).

When we investigate the globally correlated electron systems such as closed-loop CDWs, non-contact and non-destructive measurements are preferable. Therefore, it is advantageous to probe the CDW dynamics with photoexcitation. A pump-probe method is usually employed in the time-resolved experiments, where optical responses of photoexcited electrons are traced by a probe pulse immediately after an intense pump pulse with a delay time between the two pulses. The pump pulse is utilized to excite the electronic system to a nonequilibrium state while the probe detects changes of the system through the optical properties, such as reflection and transmission. A comprehensive experimental analysis for the ultrafast optical response of phase transition materials has been realized using a successful theoretical model [4,5]. Around phase transition temperature (T_c), the electrons modify their system through the inherent interaction. In the CDW phase transition, the 3D-correlation of the electron density forms a gap around the Fermi surface. The electron relaxation across the gap usually requires longer relaxation time than that of intraband relaxation without gap. At T below around T_c, the re-excitation of electrons by hot phonons becomes efficient owing to a small gap, resulting in a remarkable increase of the relaxation time and thereby a change of the relaxation between below and above T_c: Furthermore, in the vicinity of T_c, the relaxation deviates from the ideal exponential decay, reflecting the order fluctuation.

Figure 1 (b) shows plots of the decay time (τ) as a function of temperature in the ring and whisker [6,7]. Below ~40 K, τ in both samples are nearly constant and are identical. On the contrary, a remarkable difference is seen above 50 K, where the relaxation time in whisker diverges on approaching T_c, while we see a quenching of τ in the ring. For better comparison, simultaneous plots of transient signals with the longest decay time are represented in Fig. 1 (c). Here the data of the ring deviates from the exponential decay, reflecting the quenching of τ in the ring and thereby suggesting the enhancement of the critical Ginzuburg-Landau fluctuations.

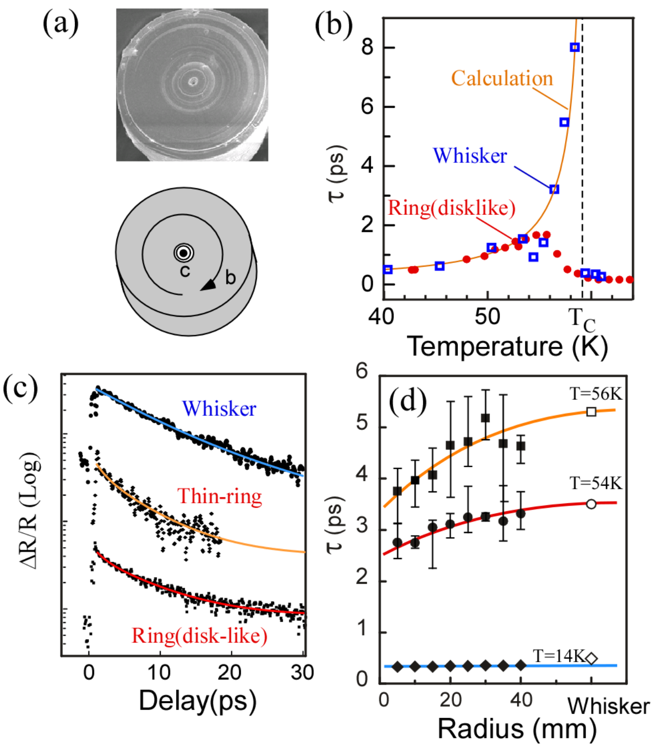

Figure 1. (a) Electron micrograph of a $NbSe_3$ (disklike) ring, and its schematic illustration with crystal axes. (b) *T*-dependences of τ in the whisker and ring. (c) Semi-logarithmic plots of ΔR which are shifted vertically for clarify. (d) Position dependences of τ in the radial direction of the ring. τ is averaged over the orientations of 8-fold rotational symmetry.

Nogawa *et al.* proposed a phase field model for CDWs in ring crystals remarking on the frustration between intra- and inter-chain couplings [8]. By using Monte Carlo simulations, they found that the relaxation function obeys a power-law decay instead of the usual exponential decay. This theoretical result is qualitatively consistent with our experimental results around T_c. although the theoretical model should be valid only when the circumferential length of the chain is smaller than the coherent length of ring CDWs.

The crossover between coherent and incoherent dynamics is experimentally demonstrated in Fig. 1 (d), where we plot τ obtained at various positions within the disk. A remarkable deviation from the mean-filed (MF) *T*-dependence is seen around the center region while the decay approaches MF behavior with the position away to the outer region. The position dependence thus supports the enhanced phase fluctuation in the coherent CDWs associated with the closed-loop topology. The result also suggests that the dynamic correlation length reaches as long as several tens of micron within the ring.

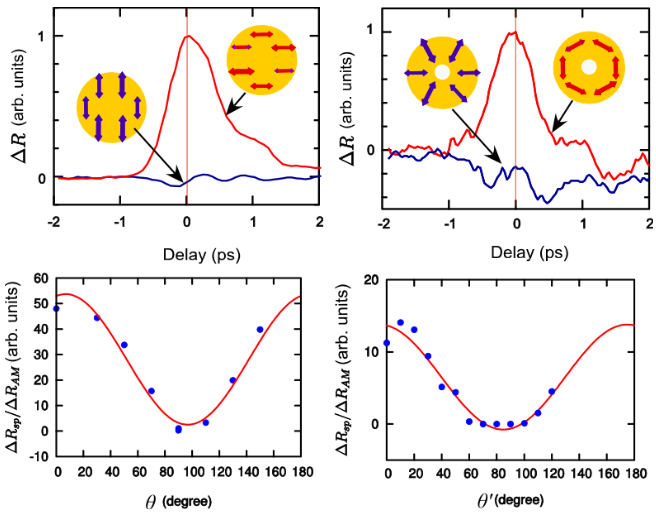

Figure 2. Transient Δ*R* (upper) and polarization (lower) for (left) a whisker crystal measured by the linear polarization with various *θ* and (right) a ring crystal measured by the polarization vortex with various *θ*′ [10].

Next, we demonstrate a global evaluation of closed-loop CDWs in the ring using polarization vortex (PV) pulses. What we can obtain by using the optical vortices is the information on closed-loop phase and/or polarization coherence of electrons [9]. Figure 2 shows the results of transient Δ*R* measured by probe pulse with various polarization distributions and angles [10]. The transient responses of the whisker sample measured by linear polarization show high polarization and thus reflect the 1D character. On the other hand, the responses of the ring show similar polarization only when we probe the nonequilibrium electrons with PV pulses. In the right row of Fig. 2, *θ'* indicates the rotation angle of the local field polarization; *θ'*=0 and 90 correspond to azimuthal and radial polarizations, respectively (see the illustrations in the insets of upper figure). It is significant to remember that the relative position between PV and center hole of the ring is critical for the Δ*R* polarization. We carefully adjusted the optical vortex onto the center hole of the ring. In order to quantitatively compare the data sets, we show the polarization of the exponential decay component as a function of *θ* or *θ'* in the lower part of Fig. 2, where we estimate the electron polarization from the ratio between the amplitudes of the single particle (ΔR_{sp}) and amplitude mode (ΔR_{AM}) contributions. Because of the fully symmetry of the CDW oscillation, this method allows to cancel the remnant polarization of the experimental setup, thus being more accurate than the comparison of the absolute values of Δ*R*.

The results in both data clearly indicate the polarization of the electron relaxation whose magnitudes are near unity. Therefore we conclude that the ring sample exhibits a well-defined azimuthal electron polarization. Although this conclusion is easily deduced from the geometrical structure, our measurement verifies that the CDW electrons in the ring maintain their 1D character, and so that the technique using polarization vortex has the ability to globally evaluate the electron dynamics in closed-loop systems.

In summary, we have investigated nonequilibrium electron dynamics in $NbSe_3$ ring to study the topological effects on CDWs. A power-law relaxation was found around T_c, suggesting a phase vortex lattice transition characterized by the closed-loop topology. We have also evaluated the global dynamics in terms of radial and azimuthal characteristics of the ring.

References

[1] S. Tanda, T. Tsuneta, Y. Okajima, K. Inagaki, K. Yamaya, N. Hatakenaka, Nature 417, 397 (2002).
[2] T. Tsuneta, and S. Tanda, J. Crystal Growth 264, 223 (2004).
[3] E. Sweetland, C-Y. Tsai, B. A. Wintner, and J. D. Brock, Phys. Rev. Lett.65, 3165 (1990).
[4] J. Demsar, K. Biljakovic, and D. Mihailovic, Phys. Rev. Lett.83, 800 (1999).
[5] V.V. Kabanov, J. Demsar, B. Podobnik, and D. Mihailovic, Phys. Rev. B59, 1497 (1999).
[6] K. Shimatake, Y. Toda, and S. Tanda, Phys. Rev. B73, 153403 (2006).
[7] K. Shimatake, Y. Toda, and S. Tanda, Phys. Rev. B75, 115120 (2007).
[8] T. Nogawa and K. Nemoto, Phys. Rev. B 73, 184504 (2006).
[9] Y. Ueno, Y. Toda, S. Adachi, R. Morita, T. Tawara, Opt. Express 17, 20567 (2009).
[10] Y. Tokizane, K. Shimatake, Y. Toda, K. Oka, M. Tsubota, S. Tanda, and R. Morita, Opt. Express 17, 24198 (2009).