The Mixed State of Charge-Density-Wave in a Ring-Shaped Single Crystals

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Charge-density-wave (CDW) phase transition in a ring-shaped crystals, recently synthesized by Tanda et al. [Nature, 417, 397 (2002)], is studied based on a mean-field-approximation of Ginzburg-Landau free energy. It is shown that in a ring-shaped crystals CDW undergoes frustration due to the curvature (bending) of the ring (geometrical frustration) and, thus, forms a mixed state analogous to what a type-II superconductor forms under a magnetic field. We discuss the nature of the phase transition in the ring-CDW in relation to recent experiments.

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I. INTRODUCTION

Charge-density-wave (CDW) has been attracting intense interest from solid state physicists, because of its unique physical properties as an electronic condensate. Especially analogy to superconductivity has been stimulating imagining of various people. Fröhlich paid attention to collective transport (sliding) of CDW and investigated within it the origin of superconductivity before the foundation of BCS theory. Although it has been clarified that the sliding motion of CDW is easily suppressed by inhomogeneities, realization of the Fröhlich’s superconductivity in ultraclean CDW crystals is an intriguing target of experimental trails.

Similarities and dissimilarities between vortices in superconductors and dislocations in CDW systems are also interesting topics of this field. Analogly between the phase slip phenomena via vortex ring in a superconducting wire and the dislocation nucleation in a sliding CDW has been pointed out. Several people attempted to search the counterpart of the mixed state of type-II superconductors in CDW systems. According to Ref. CDW systems can also form a mixed state analogous to that in type-II superconductors when an electric field perpendicular to the chains is applied. This “mixed state” of CDW, however, is formed only in the surface region of the sample within a depth of Debye screening length, which is usually several lattice spacings in most CDW materials. Therefore direct experimental observation of this state is considered to be very difficult.

Recently Tanda et al. have succeeded in synthesizing a ring crystal of CDW material8,9, where the conducting chains are along the azimuthal direction. In a ring crystal, CDW system feels frustration due to bending, just like a superconductor feels frustration under a magnetic field. Thus we can expect that the CDW forms a mixed state as in a type-II superconductor. This fact was first pointed out by Nogawa and Nemoto10 and the dynamics of ring-CDW is investigated in terms of the Monte Carlo simulation on a lattice model of CDW. In their analysis, ring CDW is mapped to a uniformly frustrated XY model.

Experimental investigation of the ring-CDW’s has been carried out by Shimatake et al. They measured the single particle relaxation rate in ring- and whisker-CDW’s using femtosecond spectroscopy and found that quasiparticle density in ring CDW’s shows a stretched exponential decay \( \propto \exp\left\{ -t/\tau \right\}^{0.3} \), whereas it decays exponentially in whisker samples. Moreover relaxation time in ring CDW’s did not show significant divergence at \( T_c \), which is common in whisker samples. Indication of such slow dynamics in ring CDW’s is also found in the simulations by Nogawa and Nemoto, who found a power law decay of relaxation time in ring crystals. Although details of relaxation formula are different, both results suggest the existence of slow relaxation processes in ring CDW’s, which may be due to the dynamics of dislocations intrinsic to ring CDW’s. In this context it is important to study how dislocations intrinsic to ring CDW’s is distributed in the ordered state.

In this paper we first derive the Ginzburg-Landau (GL) free energy of CDW in a ring geometry. Our results are slightly different from uniformly frustrated XY model, or superconductors in a magnetic field, especially when the radial thickness of the ring is large compared to the radius. We then study the mean field behavior of the system in order to clarify the transition temperature of CDW in a ring geometry. We also comment on the analogy to superconductors in a magnetic field, which is useful for thin rings (radial thickness \( \ll \) radius). The relation between our results and some experimental results are also discussed.
II. GINZBURG-LANDAU FREE ENERGY OF A RING CDW

In constructing the free energy describing ring CDW’s we need detailed knowledge about the crystal structure of the ring crystals. We refer to Ref. [13] on this point and summarize the results. According to this reference, crystal defects (edge dislocations) are almost uniformly distributed in ring crystals with diameters $\gtrsim 1 \mu \text{m}$. This means that the chains forming the ring are almost free from elastic stress (stretch or contraction) and can be described by free CDW chains with different lengths depending on the radius. There may be some stretch or contraction of the order of lattice spacing for each chain due to the discrete nature of the lattice, however, this is negligible for rings with realistic diameters $\sim 1 \mu \text{m}$ which is much larger than the unit cell of the lattice.

Then we start from the following GL free energy,

$$ F = \sum_{k=1}^{N_c} \left\{ \sum_{j=1}^{N} F^x_j + \sum_{j=1}^{N-1} F^\text{int}_{j,j+1} \right\}, \quad (1) $$

where the first and the second term correspond to intra-chain and inter-chain contribution, respectively. The number of the chains is denoted by $N$. Summation over $k$ corresponds to the stacking of the layer along the axis of the ring, with $N_c$ being the number of the layers. Since we assume that the system is uniform along the axis, $k$-summation is suppressed hereafter. Here $F^x_j$ is the free energy of the $j$-th chain given by

$$ F^x_j = \int_0^{2\pi R_j} dx \left[ \xi_0^2 |\partial_x \psi_j(x)|^2 + (t-1)|\psi_j(x)|^2 + \frac{1}{2} |\psi_j(x)|^4 \right]. \quad (2) $$

where $R_j$ is the radius of the $j$-th chain given by $R_j = R + d (j - N/2)$ ($R$ is the radius at the center of the width and $d$ is the inter-chain spacing), $\xi_0$ is the coherence length of CDW at zero temperature $T = 0$, and $t$ is the reduced temperature $T/T_c$ with $T_c$ being the transition temperature in the bulk. $\psi_j(x)$ is the order parameter on the $j$-th chain.

The inter-chain coupling term is constructed in the following way: We write the order parameter as $\psi_j \equiv |\psi_j| e^{i\phi_j(x)}$. Then the local charge density modulation due to CDW can be written as $\delta \rho^\text{local}_j(x) \propto |\psi_j(x)| \cos\{2k_F x + \phi_j(x)\}$, and the inter-chain coupling is given in the following form,

$$ F^\text{int}_{j,j+1} = -\frac{R_j + R_{j+1}}{2} \int_0^{2\pi} d\theta J_0 |\psi_j(R_j \theta)| |\psi_{j+1}(R_{j+1} \theta)| \cos\{2k_F R_j \theta + \phi_j(x)\} \cos\{2k_F R_{j+1} \theta + \phi_{j+1}(x)\} \quad (3) $$

where $J_0$ is a constant. CDW usually prefers $\pi$-phase shift between neighboring chains due to, for example, Coulomb repulsion. We have beforehand taken it into account in the definition of the order parameter, and the minus sign of Eq. (3) is due to this. Noting the equality,

$$ \cos\{2k_F R_j \theta + \phi_j(x)\} \cos\{2k_F R_{j+1} \theta + \phi_{j+1}(x)\} = \frac{1}{2} \cos\{2k_F d \theta + \phi_j(x) - \phi_{j+1}(x)\}, \quad (4) $$

where $d = R_{j+1} - R_j$, and neglecting the first term of the right hand side as the negligible fast oscillation, we obtain,

$$ F^\text{int}_{j,j+1} \approx -\frac{R_j + R_{j+1}}{2} \int_0^{2\pi} d\theta J_0 |\psi_j(R_j \theta)| |\psi_{j+1}(R_{j+1} \theta)| \frac{1}{2} \cos\{2k_F d \theta + \phi_j(x) - \phi_{j+1}(x)\} \times \left| \psi_j(R_j \theta) e^{ik_F d \theta} - \psi_{j+1}(R_{j+1} \theta) e^{-ik_F d \theta} \right|^2. \quad (5) $$

In the last line, we added some terms proportional to $|\psi_j|^2$, in order to subtract the effect of the renormalization of $T_c$.

Next we gauge transform the order parameter as $\psi_j(x) \rightarrow \psi_j(x) e^{ik_F d \theta j}$ and obtain the resulting free energy as,

$$ F^x_j = R_j \int_0^{2\pi} d\theta \left[ \xi_0^2 \left| \left( \frac{1}{R_j} \frac{\partial}{\partial \theta} - \frac{2k_F d j}{R_j} \right) \psi_j(R_j \theta) \right|^2 + (t-1)|\psi_j(R_j \theta)|^2 + \frac{1}{2} |\psi_j(R_j \theta)|^4 \right], \quad (6) $$

and

$$ F^\text{int}_{j,j+1} = \frac{R_j + R_{j+1}}{2} J_0 \int_0^{2\pi} d\theta |\psi_j(R_j \theta) - \psi_{j+1}(R_{j+1} \theta)|^2. \quad (7) $$

FIG. 1: Geometry and parameters of our system. The $z$-axis is perpendicular to the sheet.
Since we are interested in the behavior of CDW near $T_c$, we have disregarded the effects of electrostatic screening. In order words, the scalar potential does not appear in our equations. This is an over simplification when we go to lower temperatures, where CDW order is well developed and charge modulation is significant. This point will be discussed further in the end of this paper.

The boundary condition for $\psi_j(x)$ in the ring geometry is given by

$$\psi_j(0) = \psi(R_j), \quad \partial_x \psi_j(0) = \partial_x \psi(R_j).$$

These conditions impose $\phi_j(R_j) - \phi_j(0) = -4\pi k_F R_j + 2\pi N$, where $N$ is an integer. This gives rise to the phenomena similar to the Little-Parks effect in superconducting rings. For systems much larger than the Fermi number, the inter-chain term can be rewritten as

$$F_{\text{int}} = \frac{R_j + R_{j+1}}{2R} \int_0^{2\pi R} dx' \left[ \frac{(R_j)}{4} \psi_j'(x') - \psi_j'(x') \right]^2.$$

The inter-chain term comes from the fact that inner chains have smaller lengths. This can make the thermodynamic properties of ring CDW’s different from those in superconductors in a magnetic field especially when the ring is thick $W \sim R$. This point should be carefully studied in further analysis.

### III. CASE OF A THIN RING $W \ll R$: A SUPERCONDUCTING ANALOGY

Although thick rings ($W \sim R$) are a little different from superconductors in a magnetic field, as we have seen above, the analogy to superconductors is still useful for thin rings ($W \ll R$). For thin rings, the free energy is simplified to

$$F = \frac{1}{d} \int_0^{2\pi R} dx' \int_{R-W/2}^{R+W/2} dy' \left[ \frac{x_0^2}{\gamma} \left( \psi(x', y) \right)^2 + \frac{\gamma^2}{\gamma} \psi(x', y) \right]^2 + (t - 1) \left( \psi(x', y) \right)^2 + \frac{1}{2} \left( \psi(x', y) \right)^4,$$

where $\gamma$ is defined by $J_0/4 = \gamma^2 x_0^2/d^2$. This is actually the continuum version of the lattice model previously introduced by Nogawa and Nemoto.

We further change the coordinates as $y' = y/\gamma$ so that the system is isotropic in both $x$- and $y$-direction and obtain

$$F = \frac{\gamma}{d} \int_0^{2\pi R} dx' \int_{(R-W/2)/\gamma}^{(R+W/2)/\gamma} dy' \left[ \frac{x_0^2}{\gamma} \left( \psi(r') \right)^2 + (t - 1) \left( \psi(r') \right)^2 + \frac{1}{2} \left( \psi(r') \right)^4 \right],$$

where $r' = (x', y')$ and $\gamma(r') = 2 \left( \frac{\gamma y'}{R} - 1 \right) k_F e_x$ with $e_x$ being the unit vector in $x$-direction.

At this stage, we see a nice analogy to the free energy of a superconductor in a magnetic field. The magnetic field $H$ corresponds to the curvature of the ring $1/R$ as

$$\frac{2\pi H}{\phi_0} \Leftrightarrow 2k_F \gamma.$$ 

From this analogy we can deduce the transition temperature of CDW in a ring crystal. We note that a ring CDW corresponds to a type-II superconductor, not type-I, since there is no mechanism to screen the “curvature” (or bending) of the ring in the CDW system. In other words, there is no Meissner phase.

Since the transition temperature of a type-II superconductor under a magnetic field $H$ is given by

$$T_{c2}(H) = T_c(0) \left( 1 - \frac{H}{H_{c2}(0)} \right),$$

...
where \( H_{c2}(0) = \phi_0/(2\pi\xi_0^2) \) is the upper critical field at \( T = 0 \) (Note that this formula is valid if \( H < H_{c2}(0) \). In that sense, \( H_{c2}(0) \) is the critical field at \( T = 0 \) obtained from the linear extrapolation from \( T \sim T_c \)), the CDW transition temperature of a ring crystal of radius \( R \) is given by

\[
T_{c2}(R) = T_c(\infty) \left( 1 - \frac{R_{c2}}{R} \right),
\]

where \( R_{c2} = 2k_F\gamma\xi_0^2 \) and \( T_c(\infty) \) is the transition temperature of the whisker sample.

Here we apply our results to actual NbSe_3 ring crystal. We put \( 2k_F \approx 4.3nm^{-1} \), \( \xi_0 \approx 6.1nm \) and \( \gamma \approx 1/10 \sim 1/100 \) for NbSe_3, and then we obtain \( R_{c2} \approx 160 \times 100 \gamma nm = 1.6 \sim 16nm \).

### IV. NUMERICAL STUDY OF LINEARIZED GINZBURG-LANDAU EQUATION

We study the linearized GL equation for the cases of arbitrary \( R \) and \( W \) and determine the CDW transition temperature. We start by disregarding the quartic term of the order parameter in Eqs. (10). By introducing the Fourier transformation,

\[
\psi_j(x) = \frac{1}{\sqrt{2\pi R}} \sum_{n = -\infty}^{\infty} \tilde{\psi}_j(n)e^{in\alpha x/R}.
\]

the free energy \( F \) is rewritten as

\[
F = \sum_{n = -\infty}^{\infty} \sum_{j = 1}^{N} \left[ \frac{R}{R_j} \xi_0^2 \left( \frac{n}{R} + \chi(R_j) \right)^2 + \frac{R_j}{R} (t - 1) \right] |\tilde{\psi}_j(n)|^2 + \sum_{n = -\infty}^{\infty} \sum_{j = 1}^{N-1} \frac{\gamma^2\xi_0^2 R_j + R_{j+1}}{2R} |\tilde{\psi}_j(n) - \tilde{\psi}_{j+1}(n)|^2.
\]

We put \( \tilde{\Psi}_j(n) \equiv \sqrt{R_j/R}\tilde{\psi}_j(n) \), and the above equation becomes

\[
F = \sum_{n = -\infty}^{\infty} \sum_{j = 1}^{N} \left[ \left( \frac{R}{R_j} \right)^2 \xi_0^2 \left( \frac{n}{R} + \chi(R_j) \right)^2 + (t - 1) \right] |\tilde{\Psi}_j(n)|^2 + \sum_{n = -\infty}^{\infty} \sum_{j = 1}^{N-1} \frac{\gamma^2\xi_0^2 R_j + R_{j+1}}{2} \left| \frac{\tilde{\Psi}_j(n)}{\sqrt{R_j}} - \frac{\tilde{\Psi}_{j+1}(n)}{\sqrt{R_{j+1}}} \right|^2
\]

\[
\equiv \sum_{n = -\infty}^{\infty} \sum_{j,k = 1}^{N} \tilde{\Psi}_j^*(n) \cdot \{M_{jk}(n) + (t - 1)\delta_{jk}\} \cdot \tilde{\Psi}_k(n).
\]

The integer \( n \) indexes the mode of the order parameter. All the eigenvalues of \( M_{jk}(n) \) are positive and we denote the smallest one by \( \lambda_{\text{min}}(n) \). The mode indexed by \( n \) becomes unstable when \( \lambda_{\text{min}}(n) + t - 1 < 0 \) is satisfied. Below this temperature \( \tilde{\Psi}_j(n) \) (or \( \tilde{\psi}_j(n) \)) tends to have a nonzero expectation value and the nonlinear term of GL free energy becomes relevant. The mode which becomes unstable at the highest temperature dominates the real CDW transition. Therefore, by diagonalizing \( M_{jk}(n) \) and comparing the eigenvalues of all possible \( n \)’s, we can estimate the CDW transition temperature.

In Figs. 2 and 3 we have shown the log-log (natural logarithm) plot of the shift of the transition temperatures of several systems as a function of the radius of outermost chain \( R_{\text{out}} = R + W/2 \). (\( \Delta T_c \) is the decrease of the transition temperature from the bulk \( T_c \).) Fig. 2 is for thick samples where \( W \sim 2R \) (\( W = 2(R - d) \) to be more precise). In this case, the ring looks almost like a disk. Fig. 3 is for thin samples where we put \( W = R/5 \). (a) and (b) in each figure are data for different coherence length \( \xi_0 \). (a) is for \( \xi_0 = 5d \) and (b) is for \( \xi_0 = 20d \). From these figures, we can see that amount of \( \Delta T_c \) is inversely proportional to \( R_{\text{out}} \) for samples with larger diameters.

We write \( T_c(R_{\text{out}}) = T_c(1 - R_c/R_{\text{out}}) \) and estimate \( R_c \) from the numerical data. The results are shown in the table below. \( R_c \) is obtained from the fitting in the figures. If the fitting line is \( y = \alpha x + \beta \) where \( \alpha \approx -1 \), we obtain \( R_c = e^\beta \). It can be seen that \( R_c \) is not close to \( R_{c2} \) which is estimated from Eq. (15). We found that \( R_c \) is rather close to \( R_{c3} \) which corresponds to the third critical field \( H_{c3} \) of type-II superconductors. According to Ref. (14), \( R_{c2} \) and \( R_{c3} \) are related by \( R_{c3}^2 = 0.59R_{c2}^2 \). \( H_{c3} \) of a type-II superconductor gives the critical field of surface nucleation of superconducting order in a magnetic field. Therefore \( R_c \) in a ring CDW may also correspond to the same phenomenon. This is confirmed by examining the order parameter which develops at the transition temperature. In Fig. 4 we have shown the order parameter at the critical temperature in case of Fig. 2 (b). Even though the radius is changed, CDW order starts developing always from the outer edge of the sample and it tells us that the CDW transition is of surface nucleation type.

From the above argument we obtain a rough picture of CDW transition in the ring crystals: At \( T = T_{c2} = T_c(1 - R_{c2}/R) \), CDW order starts nucleation at the outer surface of the sample and, at \( T = T_{c3} = T_c(1 - R_{c3}/R) \), CDW order prevails in the entire sample.

| Fig. | W | \( \xi_0/d \) | \( k_F/d \) | \( \gamma \) | \( R_{c2}^d/d \) | \( R_{c3}^d/d \) | \( R_c/d \) |
|-----|---|----------|-------|------|----------------|----------------|-------|
| 2 (a) | 2 x R | 5 | 2 | 0.1 | 10.0 | 5.90 | 5.46 |
| 2 (b) | 2 x R | 20 | 2 | 0.1 | 160 | 94.4 | 87.3 |
| 3 (a) | R/5 | 5 | 2 | 0.1 | 10.0 | 5.90 | 5.96 |
| 3 (a) | R/5 | 20 | 2 | 0.1 | 160 | 94.4 | 95.7 |

TABLE I: \( R_{c2}^d \) is obtained from Eq. (15), \( R_{c3}^d = 0.59R_{c2}^d \) (see text). \( R_c \) is obtained from the fitting in Figs. 2 and 3.
FIG. 2: Log-log plot of the outermost radius $R_{\text{out}}$ vs. the shift of the transition temperature in thick ($W \sim 2R$) rings (or cylinders). (a) $\xi_0 = 5d$, (b) $\xi_0 = 20d$. Lines are the fitting line to the linear part of the data. $R_{\text{out}}$ is in the unit of $d$.

FIG. 3: Log-log plot of the outermost radius $R_{\text{out}}$ vs. the shift of the transition temperature in thin ($W = R/5$) rings. (a) $\xi_0 = 5d$, (b) $\xi_0 = 20d$. Lines are the fitting line to the linear part of the data. $R_{\text{out}}$ is in the unit of $d$.

V. DISCUSSION

Let us compare our theory with the experimental results obtained by Tsuneta et al.\cite{Tsuneta15}.

Tsuneta et al.\cite{Tsuneta15} found that the transition temperature estimated from the anomalies in resistivity does not change significantly between whisker and ring crystals, although significant decrease is observed in “figure-eight” (twisted ring) crystals. As for the ring crystals, their data are in good agreement with our result, since the radiiuses of the ring crystals used in the experiment may be mostly above 10$\mu$m and then, according to our theory, the suppression of transition temperature is about $T_c R_{\text{out}}/R \sim 0.001 \times T_c \sim 0.1K$. This is smaller than the sample-to-sample fluctuations. In contrast to this, in figure-eight crystals, the transition temperature is suppressed about 1K. This significant suppression compared to rings is worth noting. Although detailed effects of the twisting is not clear at this stage, we consider that such a large suppression of $T_c$ due to geometrical frustration is unexpected from the naive intuition based on the present analysis. The explanation of this phenomenon needs further studies, both from theoretical and experimental sides.

Here we comment on the lower temperature side. The free energy, Eqs. (10) and (11), is valid only in very vicinity of $T_c$, since the effect of electrostatic screening (Debye screening) is neglected. The dislocations in CDW give rise to the polarization of charge arising from $\partial_x \phi_j(x)$, and thus undergoes the electrostatic screening due to the quasiparticles and the condensate (phasons). Since the electrostatic screening is strong in most CDW materials, this neglect is serious. A free energy which includes the screening effect is given, for example, in Ref.\cite{6}, according to which, the interaction between dislocations is not the type of anisotropic GL theory but shows complicated dependence on direction and scale. In order to clarify the CDW order in ring crystals at much lower temperature than $T_c$, a study based on such accurate free energy is indispensable, which may need further numerical studies. We leave this for future study.

It should also be noted that in realistic CDW materials the effects of thermal fluctuation are essential. As is well known, the real transition temperature of CDW is much lower than the mean-field value which is estimated from the CDW energy gap. This is considered to be due to the renormalization of $T_c$ by the thermal fluctuations. In order to obtain more reliable results, we need to go beyond
FIG. 4: The amplitude of the order parameter at the CDW transition temperature for samples with several different radii. Each peak corresponds to solutions for different radius. The peak is always located at the outer edge of the sample. The inset is the magnification of one of the data. Bold line shows the outer edge.

VI. SUMMARY

We have derived the free energy which describes the CDW order in ring crystals and, based on it, calculated the mean-field transition temperature. We have discussed several physical properties of ring CDW based on the obtained free energy.

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