Elastic theory of ring-(or cylinder-)shaped crystals is constructed and the generation of edge dislocations due to geometrical frustration caused by the bending is studied. The analogy to superconducting (or superfluid) vortex state is pointed out and the phase diagram of the ring-crystal, which depends on radius and thickness, is discussed.

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What is the lowest energy state of matter? The answer to this fundamental question has been believed to be the “crystalline state” as far as the system has an infinite extension and quantum fluctuation is negligible. Most elements or compounds freeze into crystals when cooled down slowly. Even the electrons form the so-called Wigner crystal if the density is low enough. The basic character of the crystalline state is the breaking of translational symmetry; the system is divided into small fundamental cells. Although some exceptions, such as Penrose lattice or quasicrystals, have been known, this profound form of existence occupies the most intriguing expositions in the “mineralogical department of natural history museum”.

In recent decades, with the help of developed fine processing technology, especially symbolized by the term “nanotechnology”, a new field seems to be added to the studies of crystallography. The topic of this field is the crystallization in nano-scales. A group of materials which is most attracting attention from both basic and applied scientists is evidently Fullerenes and nanotubes, where spherical or cylindrical form of crystals and their subspecies are found to be stable structures of carbon in nano-scales. Tanda et al. have shown that quasi-one-dimensional crystals, such as TaS$_3$ or NbSe$_3$, which are usually grown as needle-shaped crystals, can be made into rings or cylinders by controlling the growing conditions. They have even succeeded in producing single-crystal M"obius strips. These crystals with topologically nontrivial shapes are named “topological matters”. Some other cylindrical crystals were also found recently, such as MoS$_2$. The expanding variety of the topological matters seems to be invoking questions about the nature of crystallization in nano-scales.

The crystal structure largely affects the physical properties of the system and, therefore, these crystals provide excellent experimental environments in investigating topological physics. For example, TaS$_3$ and NbSe$_3$ show superconductivity and charge density wave order under appropriate conditions, and, then, topology-induced novel states are expected. Several theoretical works have been elaborated in this direction, and experimental investigations are under way.

In order to understand the physical properties of the “topological matters”, the knowledge about crystal structure is indispensable. In this paper we study the structures of ring-(or cylinder-)shaped crystals, which we simply call the “ring crystals” hereafter. Special attention is paid to the formation of crystal defects due to the elastic stress which topological matters undergo because of their nontrivial forms. In case of the ring crystals the edge dislocations are likely to be generated because of the bending, as one can see from Fig. 1. In this paper we derive, within the linear elastic theory, the condition for...
the formation of edge dislocations in a ring crystal whose radius and thickness are $R$ and $W$, respectively. Analogy between the superconducting (or superfluid) vortices and the dislocations in a ring crystal is invoked. Our results may be useful in understanding physical phenomena in ring crystals.

**Elastic theory of a ring crystal**

To construct the elastic theory of a ring crystal we start by making a rectangular strip of a crystal into a ring as depicted in Fig. 2, which introduces elastic deformation. We assume that the system is uniform in $z$-direction (along the axis of the cylinder) and suppress the $z$-coordinate hereafter. The free energy is considered to be that for unit length in $z$-direction. The crystal is anisotropic and it is assumed that the elastic coupling in the direction along A-C and B-D is much stronger than that along A-B and C-D (see Fig. 2). This situation is realized in most ring crystals grown until now.

![FIG. 2: Making a rectangular strip of a crystal into a ring. The strip (a) is made into a ring (b) by introducing the elastic deformation. The system is assumed to be uniform in $z$-axis, which is perpendicular to the page.](image)

The elastic free energy of the ring crystal is constructed by examining how the linear element of the original crystal $d^2r = (dx, dy)$ is modified by the bending in the ring crystal. Let us suppose that the position in the original crystal $r = (x, y)$ is moved to $R = (X, Y)$ in the ring crystal. Here orthogonal coordinate system is used. We obtain the following relation,

$$(X, Y) = \left( (r + v) \cos \frac{x + u}{R}, (r + v) \sin \frac{x + u}{R} \right), \quad (1)$$

where $r = r(y)$ denotes the radius of the lattice plane whose $y$-coordinate is $y$ in the original system, and $u = u(x, y)$ and $v = v(x, y)$ are small deviations around the complete ring shape (i.e., dislocation-free ring). We define the metric tensor $g_{ij}$ by $(dR)^2 = \sum_{i,j} g_{ij} dx_i dx_j$, where $g_{ij} = \partial R/\partial x_i \cdot \partial R/\partial x_j$ $(i, j = \{1, 2\}$, and $x_1 = x, x_2 = y$). The elements of the metric tensor are given as

$$
g_{11} \equiv g_{xx} = \left[ \frac{r + v}{R} (1 + u_x) \right]^2 + [v_x]^2, \quad (2)$$

$$
g_{22} \equiv g_{yy} = \left[ \frac{r' + v_y}{R} \right]^2 + \left[ \frac{u_y}{R}(r + v) \right]^2, \quad (3)$$

$$
g_{12} \equiv g_{xy} = \frac{1}{R^2} \left[ (r + v) \left( (1 + u_x) u_y + v_x (r' + v_y) \right) \right], \quad (4)$$

where $r' = dr/dy$, $u_x = \partial u/\partial x \cdots$ etc. In this paper, we assume $r(y) = R + y (-W/2 \leq y \leq W/2)$ for simplicity. Strain tensor $u_{ij}$, which describes the elastic deformation of the crystal, is given by $u_{ij} = \frac{1}{2} (\delta_{ij} - \delta_{ij})$, where $\delta_{ij}$ is the Kronecker’s delta. Assuming an orthorhombic system for simplicity, the elastic free energy is given by

$$F = \int_{-L/2}^{L/2} dx \int_{-W/2}^{W/2} dy \left\{ \frac{1}{2} \lambda_{xxxx} (u_x)^2 + \frac{1}{2} \lambda_{yyyy} (u_y)^2 \right\}$$

$$+ \lambda_{xxyy} u_{xx} u_{yy} + 2 \lambda_{xyxy} (u_{xy})^2 \right\}, \quad (5)$$

where $L = 2\pi R$ is the circumference of the ring [19]. We also assume that the principal axes of the crystal are along $x$- and $y$-axis, which yields $\lambda_{xxyy} = -\lambda_{xyxy}/2$. From now on we denote $\lambda_1 \equiv \lambda_{xxxx}, \lambda_2 \equiv \lambda_{yyyy}$ and $\lambda_3 \equiv \lambda_{xyxy} = -\lambda_{xxyy}/2$.

Next we determine the free energy of the ring crystal within the linear elasticity. To do this we expand the free energy with respect to small parameters, $u_x, u_y, v_x, v_y, y/R$ and $v/R$ to the second order, which yields

$$F = \int d^2r \left[ \frac{\lambda_1}{2} \left( u_x + \frac{y}{R} \right)^2 + \frac{\lambda_2}{2} \left( u_y + \frac{y}{R} \right)^2 \right]$$

$$- \frac{\lambda_3}{2} \left( u_x + \frac{y}{R} \right) u_y \right\}. \quad (6)$$

Here we limit ourselves to the crystals with $W \ll R$. We further neglect $v$-field here, since $\lambda_2 \gg \lambda_3$ in most ring crystals, and dominant elastic fluctuation is expected to arise not from $v$ but from $u$. Then we obtain the following free energy,

$$F = \int d^2r \left[ \frac{\lambda_1}{2} \left( u_x + \frac{y}{R} \right)^2 + \frac{\lambda_3}{2} u_y^2 \right]. \quad (7)$$

By redefining the parameters by

$$\gamma = \sqrt{\frac{\lambda_3}{\lambda_1}}, \quad K = \frac{\lambda_1 \gamma d^2}{(2 \pi)^2}, \quad \bar{y} = \frac{y}{\gamma}, \quad \bar{W} = \frac{W}{\gamma}, \quad A = -\frac{\phi_0 \gamma}{d_x R} \bar{y}, \quad \theta(x, \bar{y}) = \frac{2\pi u(x, y)}{d_x}. \quad (8)$$

$F$ is rewritten as

$$F = \int_{-L/2}^{L/2} dx \int_{-W/2}^{W/2} dy \frac{K}{2} \left[ \left( \bar{\theta}_x - \frac{2\pi}{\phi_0} A \right)^2 + \bar{y}^2 \right], \quad (9)$$
where $d_x$ and $\phi_0$ are the lattice constant in $x$-direction and the superconducting magnetic flux quantum, respectively. This is nothing but the free energy of an isotropic superconductor\textsuperscript{20}. The superconducting “phase” $\theta$ undergoes the quantization condition $\int_C d\vec{l} \cdot \nabla \theta = 2\pi n_v$, where $C$ is a simple loop and $n_v$ is the total number of vortices enclosed in $C$. The vortices correspond to the edge dislocations in the crystal system. Thus the quantization of vorticity corresponds to the quantization of the Burgers vector. Here the vector potential $\mathbf{A}$ is proportional to $1/R$, which means that the strength of “geometrical” frustration is proportional to the curvature.

**Entry of the first dislocation**

Here we estimate the free energy of a single dislocation and study the condition for the creation of the first dislocation in the system as the strength of frustration is increased. We should note that the “penetration depth” $\lambda$ is infinite in the present system. In a $\lambda = \infty$ (extremely type-II) superconductor, as in the case of rotating superfluid, the lower critical field $H_{c1}$ depends on the system size\textsuperscript{21}. We encounter a similar situation here.

We assume that the dislocation is located at the center of the system ($x = 0$, $y = 0$) and, instead of treating the boundary condition seriously, we employ a rather simple cutoff procedure\textsuperscript{22}. The phase modulation due to the vortex is put as $\nabla \theta = -\hat{z} \times \vec{r}/|\vec{r}|^2$, where $\hat{z}$ is the unit vector in $z$-direction and $\vec{r} = (x, y)$. Substituting this into Eq. (9), we obtain the dislocation free energy $F_d = K \pi \log \sum_{d_y} - \pi^2 K_d R$\textsuperscript{2}. Here the divergence at $y = 0$ is cutoff at $|y| = \bar{d}_y = d_y/\gamma$ ($d_y$ is the lattice constant in $y$-direction). The condition for the dislocation creation (namely, $F_d < 0$) is, then, obtained as

$$R < R_{c1} = \frac{\pi}{2\gamma d_y} \frac{W^2}{\ln(W/d_y)}. \quad (10)$$

**Many dislocations and destruction of elasticity**

Now we study the strongly frustrated regime where many dislocations are created. In this case, the size of the dislocation “core” is important. To study this, we discretize again the $y$-coordinate in Eq. (10) as follows,

$$F = d_y \int_0^L dx \left[ \sum_{j=1}^{N_y} \frac{\lambda_1}{2} \left( \partial_x u_j + \frac{y}{R} \right)^2 \right.
- \left. \sum_{j=1}^{N_y-1} \frac{\lambda_2}{4\pi^2} \left( \frac{d_j}{d_y} \right)^2 \cos \left\{ 2\pi \frac{(u_{j+1} - u_j)}{d_x} \right\} \right]. \quad (11)$$

Because of large anisotropy $\lambda_1 \gg \lambda_3$, the core is strongly elongated along $x$-direction although the size in $y$-direction is comparable to the lattice spacing. Therefore we extract two crystal planes between which the core is located and neglect the coupling to other planes. The solution which minimizes $F$ in Eq. (11) can be obtained analytically as $u_{j+1} = -u_j = (d_x/\pi) \arctan(x/\Lambda)$ (the core is locate between the $j$-th and the $(j+1)$-th plane).

The core size in $x$-direction, $\Lambda$, is given by $\Lambda = d_y/\gamma$ (see Ref.\textsuperscript{23} for a similar calculation). If we consider that the core size in $y$-direction to be $d_y$, the core size in the scaled system (see Eq. (5)) is given by $d_y/\gamma$ in both $x$- and $y$-direction and the core is considered to be isotropic.

In case of a type-II superconductor, the order is destroyed by a magnetic field when the distance between vortices becomes comparable to the vortex core radius, and this field strength is called the upper critical field $H_{c2} = \phi_0/(2\pi \xi^2)$, where $\xi$ is the coherence length. In the present case, noting the correspondences,

$$\frac{H}{\phi_0} \leftrightarrow \frac{\gamma}{d_x R}, \quad \xi \leftrightarrow \frac{d_y}{\gamma}, \quad (12)$$

where $H$ is the magnetic field, we obtain the relation $R_{c2} = 2\pi d_y^2/(\gamma d_x)$ corresponding to $H_{c2}$. If $R < R_{c2}$, the frustration due to bending is so strong that the crystal is full of dislocations, whose cores are almost overlapped. In this case, each plane can be almost free from the elastic coupling to neighboring planes. In other words, the inter-plane elasticity is destroyed by the dislocations. If $R_{c2} < R < R_{c1}$, the density of dislocations is low enough and the cores are separated from each other. In this case, the crystal may retain a finite elasticity.

**Phase diagram of ring crystals**

The $R$-$W$ phase diagram of ring crystals is depicted in Fig. 6. As is simply imagined, $R > W/2$ is an absolute requirement for the formation of a ring crystal. The forbidden area is hatched in the figure. It should also be noted that our approach is not reliable in the region too close to $R = W/2$, because of the assumption $R \gg W$.

We have divided the whole region into three “phases” (I ~ III). In the phase I, the crystal is free from dislocations (the “Meissner phase” in superconducting analog). The phase boundary $R_{c1}$ is reentrant as a function of $W$ because of the logarithmic contribution in Eq. (10), although the lower part of the curve is meaningless because $W \leq \sqrt{\pi d_y}$. We denote the smallest $R$ in the phase I by $R_{\text{lim.}}$. In the phase II, the frustration is not too large and the system contains dislocations well separated from each other (the “mixed state”), and the crystal may still hold rigidity. In the phase III, the frustration is large and the dislocation cores are overlapped, destroying the inter-plane rigidity of the crystal (the “normal state”). Possible dislocation pattern in each phase is also depicted in Fig. 6.

The distribution of dislocations are observed directly by scanning tunneling microscope etc. Phonon spectrum measurement is also a powerful tool to investigate the crystal rigidity, since there are drastic changes in elasticity among I ~ III. This may affect the physical properties of the system also. Studies on various ring crystals from such point of view may be an interesting topic in the future\textsuperscript{24} \textsuperscript{25}. Actually, Tanda et al. have observed that the critical temperature of charge density wave transition in NbSe$_3$ is significantly modified in ring crystals, whose relation to crystal defects may be worth investigating.
Now we apply our theory to actual systems. First, we study multi-wall carbon nanotubes, whose elastic constants are approximately those of graphite. From Ref. [20], we read $\lambda_1 = c_{11} \approx 100$, $\lambda_2 = c_{33} \approx 4$, $\lambda_3 = c_{44} \approx 0.02$ (dyn/cm²), and $\lambda_3/\lambda_1 = \gamma^2 \approx 10^{-4}$. Using $d_x = 0.25$ nm and $d_y = 0.34$ nm, we obtain $R_{c2} = 46.2$ nm and $R_{lim} = 395$ nm. When $R < R_{c2}$, the crystal is full of dislocations, and the elastic coupling between graphite sheets is negligible. This is consistent with the fact that thin nanotubes consist of graphite sheets with different chiralities and the inter-plane coupling becomes significant only in thicker tubes ($R > 75$ nm) [27]. However, an accurate treatment of different chiralities based on dislocation theory is a future problem.

In case of transition metal trichargogenides, we study orthorhombic TaS₃, for which the order of $\lambda_1/\lambda_2$ is estimated from Young modulus $E$ and shear modulus $G$ in Ref. [28, 29] as $G/E = \lambda_3/\lambda_1 = \gamma^2 \approx 10^{-2}$. Using $d_x = 0.3$ nm and $d_y = 2$ nm, we obtain $R_{c2} = 133.3$ nm and $R_{lim} = 1138$ nm. Although the phase I may be difficult to realize (e.g., $R \approx 30$ μm and $W \approx 40$ nm), crossover from II to III may be experimentally accessible.

It is also interesting to investigate tubes with $R \approx W$ by numerically minimizing the free energy, Eq. (5) or (6), instead of using superconducting analog. Such study is now prepared.

In summary, we have clarified the distribution of dislocations, in the ring-(or cylinder-)shaped crystals with various radiuses and thicknesses. Our results may be useful for the studies of the physical properties of “topological matters”, and for their future applications to electrical and mechanical devices.

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