Crossover between magnetic vortex attraction and repulsion in thin films of layered superconductors

A. I. Buzdin,[1] A. S. Mel'nikov,[2] A. V. Samokhvalov,[2] T. Akashi,[3] T. Masui,[4] T. Matsuda,[5] S. Tajima,[4] H. Tadatomo,[4] and A. Tonomura[5,6,7]

[1]*Institut Universitaire de France and Universite Bordeaux I, CPMOH, F-33405 Talence, France*
[2]*Institute for Physics of Microstructures, Russian Academy of Sciences, 603950 Nizhny Novgorod GSP-105, Russia*
[3]*Hitachi High-Technologies Co., Hitachinaka, Ibaraki 312-8504, Japan*
[4]*Osaka University, Toyonaka, Osaka 560-0043, Japan*
[5]*Okinawa Institute of Science and Technology, Kunigami, Okinawa 904-0411, Japan*
[6]*Hitachi, Ltd., Hatoyama, Saitama 350-0395, Japan*
[7]*RIKEN, Wako, Saitama 351-0198, Japan*

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In thin films of anisotropic superconductors, the intervortex interaction may be strongly modified because of the interplay between the long-ranged repulsion caused by an extremely slow decay of the supercurrent induced by a single vortex line (Pearl's effect) and the attraction caused by the tilt of the vortex lines with respect to the anisotropy axes. We present a theoretical analysis and Lorentz microscopy experimental data for high-temperature superconducting cuprates which provide evidence of such interplay. Moreover we report on a theoretical prediction of a very special type of the vortex arrangement—the formation of the vortex molecules.

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I. INTRODUCTION

The understanding of the properties of type II superconductors is ultimately related with the paradigm of "vortex matter." Thermal excitations, vortex pinning, crystal anisotropy, and spatial and time varying magnetic field all reveal a panoply of different transitions in this vortex matter, which makes its physics very rich.[1] Both the equilibrium and transport properties of the vortex matter are affected by the intervortex interaction. In isotropic bulk superconductors it is well known to be repulsive and screened at intervortex distances R greater than the London penetration depth λ. As a result, in perfect crystals quantized Abrikosov vortices form a triangular lattice.

In thin films of superconductors the standard intervortex interaction is modified because of the long-ranged repulsion predicted in the pioneering work by Pearl.[2] This repulsion is caused by an extremely slow decay of the supercurrent j_s induced by a single vortex line:[2] $j_s \propto r^{-2}$, where r is the distance from the vortex center. The intervortex interaction occurs mainly through the magnetic field outside the film and the repulsive potential of interaction between two vortices decays as $\sim 1/R$ in contrast with exponential decay at $R > \lambda$ in bulk superconductors. However, this dramatic difference in the behavior of the interaction potential has not been verified experimentally, because most of the measurable physical quantities do not experience qualitative changes related to Pearl's prediction. In particular, the triangular lattice remains an energetically favorable vortex configuration for any sample thickness d. The goal of the present paper is to demonstrate unconventional effects resulting from the repulsive Pearl interaction. This possibility to probe the Pearl potential is closely related to the phenomenon of the long-range attraction between tilted vortex lines in anisotropic systems.[3–5] To explain qualitatively the origin of this attraction one can note that contrary to the isotropic case the current lines around the tilted vortex are not parallel to the plane perpendicular to the vortex direction. As a result, the magnetic field decays rather slowly with an increase in the distance from the vortex axis. Moreover, in the plane defined by the anisotropy and vortex axes the magnetic-field component perpendicular to the layers changes its sign at large distances. Namely, this circumstance is responsible for the attraction between vortex lines.[3–6] In bulk anisotropic superconductors this attraction phenomenon is known to result in the formation of vortex chains in the regime of low magnetic fields (see Ref. 7 for a review). Indeed, the attraction between two vortices leads to the formation of a vortex pair. Then a third vortex will be attracted by this pair, etc. The interaction between any two vortices in the chain (except the nearest neighbors) is attractive, which stabilizes the chain. These vortex chains have been observed experimentally by the decoration technique in $YBa_2Cu_3O_7$,[8] scanning tunneling microscopy in $NbSe_2$,[9] and Lorentz microscopy measurements in $YBa_2Cu_3O_7$.[10]

If we consider a thin-film sample we get an interplay between two different long-range potentials: (i) attraction of the tilted vortices ($U_{att} \sim -1/R^2$) and (ii) the Pearl repulsion ($U_{rep} \sim 1/R$). The Pearl repulsion always dominates at large distances and, thus, the formation of an infinite vortex chain can become unfavorable. Adding vortices one by one we can find an optimal number of vortices which can be arranged in a chain of finite length. As a result, there appears an intriguing possibility to form a vortex structure consisting of finite size chains, i.e., of vortex molecules. By varying either the film thickness or the tilting angle we can modify the balance between the attractive and repulsive interactions, which should determine energetically favorable vortex configurations.

II. INTERACTION POTENTIAL FOR A VORTEX PAIR

We consider a typical geometry with the anisotropy axis **c** (z axis) oriented perpendicular to the film plane. Our description of the behavior of the interaction potential is based on the standard London theory. Within this model it is possible to find an exact solution describing the field and current distributions for an arbitrary configuration of vortex lines in a film of finite thickness (see Refs. 11–13 for an appropriate calculation method). An anisotropic superconductor in the London limit is known to be characterized by two penetration depths $\lambda_\perp$ and $\lambda_\parallel$ which are, in fact, the lengths of magnetic-field screening by currents flowing in directions perpendicular and parallel to the layers, respectively. In the case of large anisotropy $\lambda_\perp \gg \lambda_\parallel$ we can neglect the currents flowing perpendicularly to the layers when considering rather small distances from the vortex core (i.e., $r \ll \lambda_\perp$). These currents responsible for the weak screening of the parallel magnetic-field component determine the tilt of the vortex line but can be neglected in the calculation of the field distribution. Therefore, for this purpose, we use the model of the noninteracting superconducting layers (CuO planes in the case of YBaCuO).

We restrict ourselves to the case of straight vortex lines parallel to the plane (xz) and tilted at a certain angle γ with respect to the **c** direction. We assume here two vortex lines to be shifted by a certain vector **R** in the plane of the layers. The general equations for the vector potential **A** induced by a tilted vortex line in a layered superconducting system read

$$\text{rot rot}\,\mathbf{A} = \frac{4\pi}{c}\mathbf{j} = \frac{a}{\lambda_\parallel^2}\sum_{n=-N}^{N}[\mathbf{\Phi}_n(\mathbf{r}) - \mathbf{A}]\delta(z - na), \tag{1}$$

where the vector **r** is parallel to the layers, a is the interlayer distance,

$$\mathbf{\Phi}_n(\mathbf{r}) = \mathbf{\Phi}(\mathbf{r} - \mathbf{r}_n), \quad \mathbf{\Phi}(\mathbf{r}) = \frac{\phi_0}{2\pi}\frac{[\mathbf{z}_0,\mathbf{r}]}{r^2},$$

ϕ_0 is the flux quantum. For a tilted vortex line we should put $\mathbf{r}_n = na \tan\gamma\, \mathbf{x}_0$. Solving the above equations in the limit $r \gg a$ we obtain the following expression for the interaction energy of two vortices:

$$\varepsilon_{\text{int}}^{(2)} = \frac{\phi_0^2}{16\pi^3\lambda_\parallel}\int \frac{d^2q}{q^2}\cos\frac{\mathbf{qR}}{\lambda_\parallel}\left(\frac{d}{\lambda_\parallel}\frac{p^2+k^2}{1+p^2} + \frac{2(1-k^2)\{k(1-p^2)\sinh L + (k^2-p^2)[\cosh L - \cos(2pL)] + 2kp\sin(2pL)\}}{\sqrt{1+q^2}(1+p^2)^2[2k\cosh L + (1+k^2)\sinh L]}\right), \tag{2}$$

where

$$L = \frac{d}{\lambda_\parallel}\sqrt{1+q^2}, \quad k = \frac{q}{\sqrt{1+q^2}}, \quad p = \frac{q_x\tan\gamma}{\sqrt{1+q^2}}.$$

The first term in Eq. (2) describes the interaction in the bulk system, while the second term is responsible for the influence of film boundaries.

The minimum energy configuration corresponds to $R_y=0$ and in Fig. 1 we present some typical plots of the interaction energy $\varepsilon_{\text{int}}^{(2)}$ vs the distance $R_x=R$. Analyzing the dependence $\varepsilon_{\text{int}}^{(2)}(R)$, one can separate three contributions to the energy of vortex interaction: (i) a short-range repulsion which decays exponentially with increasing intervortex distance R (for $R > \lambda_\parallel$); (ii) an intervortex attraction which is known to be specific for tilted vortices in anisotropic systems; this attraction energy term decays as R^{-2} and strongly depends on the angle γ between the vortex axis and the **c** direction; (iii) long-range (Pearl) repulsion which decays as R^{-1} and results from the surface contribution to the energy. Note that the third term does exist, even for a large sample thickness d (see Ref. 14) although in the limit $d \gg \lambda_\parallel$ it is certainly masked by the dominant bulk contribution. At $R \gg \lambda_\parallel$ the short-range interaction term vanishes and the interaction energy vs R is

$$\varepsilon_{\text{int}}^{(2)} \simeq \frac{\phi_0^2}{8\pi^2}\left(-\frac{d_{\text{eff}}\tan^2\gamma}{R^2} + \frac{2}{R}\right), \tag{3}$$

where the effective film thickness $d_{\text{eff}} = d - 2\lambda_\parallel\tanh(d/2\lambda_\parallel)$.

One can observe here an interplay between the long-range attractive [first term in Eq. (3)] and the repulsive [second term in Eq. (3)] forces. Note that the $\lambda_\parallel$ value increases with an increase in temperature; thus, the effective thickness decreases and the long-range attraction force appears to be suppressed with increasing temperature. For large R the energy is always positive and corresponds to the vortex repulsion similar to the one between the pancakes in a single layer system. With a decrease in the distance R the attraction force comes into play resulting in the change of the sign of the energy at $R^* \simeq 0.5 d_{\text{eff}}\tan^2\gamma$. Such behavior points to the appearance of the minimum in the interaction potential. Certainly, this minimum can exist only if $R^* > \lambda_\parallel$, so that we obtain the following restriction:

$$\tan^2\gamma > \tan^2\gamma_c = 2\lambda_\parallel/d_{\text{eff}}. \tag{4}$$

This criterion gives us a tilting angle interval $\gamma > \gamma_c$ for which the minimum in the interaction potential can exist and the formation of vortex chains can be energetically favorable.

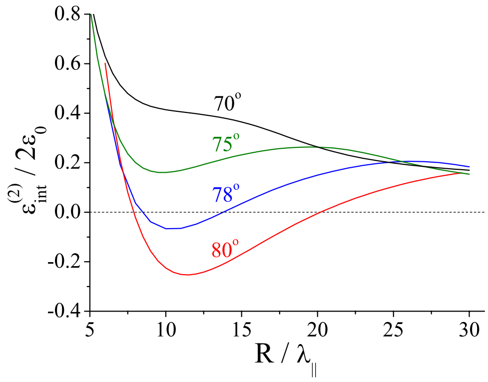

FIG. 1. (Color online) Typical plots of the interaction energy per vortex vs the distance R between two vortices for a film of thickness $d=3\lambda_\parallel$ and different tilting angles $\gamma=70°$, 75°, 78°, 80° ($\varepsilon_0=\phi_0^2/16\pi^3\lambda_\parallel$).

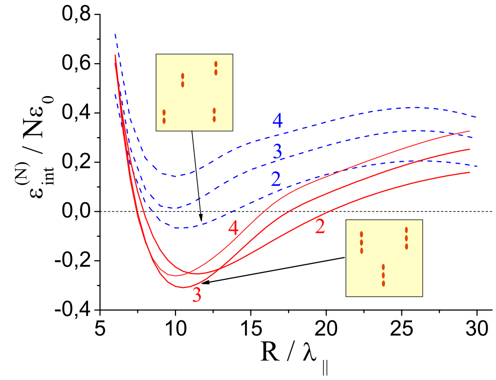

FIG. 2. (Color online) Typical plots of the interaction energy per vortex vs the intervortex distance R in an equidistant chain of N vortices: dash (solid) lines correspond to $d=3\lambda_\parallel$, $\gamma=78°$ ($d=3\lambda_\parallel$, $\gamma=80°$). Here the numbers near the curves denote the vortex number N. Schematic pictures of vortex matter consisting of dimeric and trimeric molecules are shown in the insets. Vortex positions are denoted by filled ellipses.

III. VORTEX MOLECULES

Even in the large angle regime ($\gamma > \gamma_c$) the formation of infinite chains can be questioned for rather thin films. The point is that, despite the fact that two vortices attract each other, further increase in the number of vortices arranged in a chain can be energetically unfavorable because of the slower decay of the repulsive force compared to the attractive one. In this case, for rather thin samples, there appears an intriguing possibility to observe vortex chains of finite length, i.e., vortex molecules. The vortex molecule cohesion energy is given by the expression

$$\varepsilon^{(N)}_{\rm int} = \sum_{i>j} \varepsilon^{(2)}_{\rm int}(R_{ij}), \tag{5}$$

where N is the number of vortices in the molecule and R_{ij} are the distances between ith and jth vortices in the chain molecule.

Shown in Fig. 2 are typical plots of the interaction energy per vortex vs the intervortex distance R for equidistant vortex chains with different N numbers. The energetically favorable number of vortices in a molecule grows as we increase the film thickness and/or the tilting angle because of the increasing attraction term in the pair potential $\varepsilon^{(2)}_{\rm int}$. Shown in the insets of Fig. 2 are schematic pictures of vortex matter consisting of dimeric and trimeric molecules. Finally, for rather thick samples with $d \gg \lambda_\parallel$ we get a standard infinite chain structure typical of bulk systems. Note that the formation of an infinite vortex chain may be considered in some sense as a polymerization of the vortex molecules. Certainly, the crossover from the vortex molecule state to the infinite chain structure is strongly influenced by the increase in the vortex concentration governed by the component of the external magnetic field perpendicular to the film. Indeed, one can expect such a crossover to occur when the mean intervortex spacing approaches the molecule size. Thus, the vortex molecule state can appear only in a rather weak perpendicular field when its observation can be complicated, of course, by the pinning effects.

IV. EXPERIMENTAL OBSERVATION OF THE ATTRACTION-REPULSION CROSSOVER IN YBaCuO FILMS

To provide experimental support for our theoretical findings we present here the results of Lorentz microscopy measurements of vortex structures in thin YBaCuO films. In this method[15,16] the magnetic-field distributions induced by vortices in thin films are probed by the penetrating electron beam. Therefore, perpendicular and tilted vortex lines can be observed as different projection images. This technique, owing to the low penetration power of the existing 1 MV field-emission beam, permits us to work with films of thicknesses smaller than (0.5–1) μm. It is, therefore, "par excellence" an ideal tool to study the vortex structures in thin films. The vortex observation was carried out by means of the Lorentz-Fresnel (out-of-focus) method in YBaCuO films of thickness $d \simeq 0.5$ μm, placed in a tilted magnetic field. When a superconducting thin film having vortices is tilted, electrons passing through vortices inside the film are phase shifted, or deflected, by the magnetic field of the vortices. The vortices have quantized magnetic flux of ϕ_0, and therefore create the phase shift π to an illuminating electron wave due to the Aharonov-Bohm effect. The vortices can be observed in an out-of-focus plane, the phase shift is transformed into an intensity change, and the vortex appears as a pair of bright and dark contrast features. When a vortex line is tilted, the image is elongated (for more details about the experimental method, see Ref. 10).

The above theoretical analysis gives us two theoretical predictions: (i) we find the crossover between different intervortex interaction regimes and demonstrate that the formation of chains occurs only for the rather large tilting angles; (ii) we predict possible formation of the vortex molecules. As for the very special type of arrangement formed by vortex molecules, its observation is possible only at rather low vortex concentrations, i.e., when the magnetic-field component perpendicular to the film plane is very weak. At the moment we cannot reach this regime in the present experimental

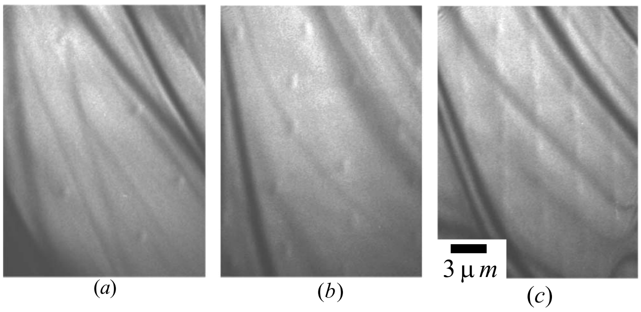

FIG. 3. Lorentz microscopy images for YBaCuO films at temperature $T=5.7$ K, the field tilting angle $\gamma_H=85°$ and absolute magnetic-field values (a) $B_0=10$ G, the vortices are slightly tilted and the repulsive interaction dominates; (b) $B_0=20$ G, vortices are more tilted and long-range attraction comes into play; (c) $B_0=60$ G, vortices are strongly tilted and well pronounced chain structures are observed.

setup and conditions. Still we managed to confirm the first theoretical prediction and observe clearly the attraction-repulsion crossover performing the field cooled experiment at temperature $T=5.7$ K for a fixed external magnetic-field direction and increasing absolute value of the field B_0 (see typical images in Fig. 3). We believe that in the field cooled experiment we can observe the vortex arrangements which form at a certain melting temperature T^* close to T_c and remain frozen during the temperature decrease. In the vicinity of T_c pinning effects should be weak and, thus, the observed vortex configurations in our experimental conditions can be controlled by the intervortex interaction rather by the pinning effects. Certainly, in this case an appropriate estimate for the penetration depth $\lambda_\parallel(T^*)$ can be several times larger than $\lambda_\parallel(0)$.

The increase in the field value B_0 causes an increase in the tilting angles of vortex lines. It is worth noting that the angle γ may differ strongly from the tilting angle γ_H of the applied external field. To find the relation between the angles γ and γ_H we should compare the energy difference between tilted and perpendicular vortex lines,[17] with the work of Lorentz forces acting on pancakes and rotating the vortex line in the presence of screening currents induced by the external magnetic-field component B_x parallel to the layers. Assuming $d<\lambda_\parallel$ we get

$$\gamma \simeq \frac{4\pi d^2 B_x}{3\phi_0} = \frac{4\pi d^2 B_0 \sin \gamma_H}{3\phi_0}, \qquad (6)$$

where B_0 is the absolute value of the external field. Therefore, the vortex attraction prevails at higher B_0 values, while at low fields the attraction force is overcome by Pearl's repulsive effect and the vortex chains are expected to disappear. Our vortex images clearly show that vortex lines at constant orientation of applied field are more tilted when the magnetic-field strength increases. At low fields which correspond to small γ values the vortex chains are completely absent [see Fig. 3(a)] while at rather high fields [see Fig. 3(c)] $B_0>B^*$ the formation of vortex chains appears to be energetically favorable. Taking $\lambda_\parallel(T^*)\sim 1$ μm for YBaCuO we can use Eq. (4) to estimate the critical angle γ_c for our films: $\tan \gamma_c \sim 1$. Using now Eq. (6) (see below) we get a rough estimate for the critical field $B^* \sim (10-100)$ G which is in good agreement with experimental data.

Our experiments were performed at constant orientation of the applied magnetic field. Therefore, by varying the magnetic field we change both the tilting angle and vortex concentration. Apparently in the case presented in Fig. 3(a), the tilt of the vortices is small (their images are not elongated) and the potential of the intervortex interaction corresponds to the curves presented in Fig. 1 with $\gamma<78°$, i.e., with energy minimum at an infinite distance. For the higher magnetic fields [see Figs. 3(b) and 3(c)] the tilt of the vortices is larger: their images are clearly elongated and they form chains. At the same time as the vortex concentration is high we cannot expect to observe the molecules (in this regime the average distance between vortices must be much larger than the size of the molecule). To observe the vortex molecules it would be preferable to change only the parallel component of the magnetic field, in order to vary the vortex tilting angle but not to affect the vortex concentration (which must be very low to avoid the intermolecule interaction).

V. CONCLUSIONS

To sum up, we have revealed a crossover from vortex repulsion to vortex attraction in thin superconducting films with the increase in the tilting angle. We also predict an unconventional type of vortex arrangement specific for thin-film samples of anisotropic superconductors and formed by vortex molecules. We believe that further development of the experimental technique will allow one to observe these unusual vortex configurations.

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