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# Vortex Molecules in Thin Films of Layered Superconductors

A.I. Buzdin · A.S. Mel'nikov · A.V. Samokhvalov

**Abstract** Both the equilibrium and transport properties of the vortex matter are essentially affected by the behavior of the intervortex interaction potential. In isotropic bulk superconductors this potential is well known to be repulsive and is screened at intervortex distances  $R$  greater than the London penetration depth  $\lambda$ . As a result, in perfect crystals quantized Abrikosov vortices form a triangular lattice. In thin films of anisotropic superconductors this standard interaction potential behavior appears to be strongly modified because of the interplay between the long-ranged repulsion predicted in the pioneering work by J. Pearl and the attraction caused by the tilt of the vortex lines with respect to the anisotropy axes. This interplay results in a new type of vortex arrangement formed by finite-size vortex chains, i.e., vortex molecules. Tilted vortices with such unusual interaction potential form clusters with the size depending on the field tilting angle and film thickness or/and can arrange into multiquanta flux lattice. The magnetic flux through the unit cells of the corresponding flux line lattices equals to an integer number  $N$  of flux quanta. Thus, the increase in the field tilting (or varying temperature) should be accompanied by the series of the phase transitions between the vortex lattices with different  $N$ . A similar scenario should be realized in strongly anisotropic BSCCO high- $T_c$  superconductors where in tilted field a crossing lattice of Abrikosov vortices (the stacks of pancakes in this case) and Josephson

vortices appears. This crossing leads to the zigzag deformation of the pancakes stacks which is responsible for the attraction interaction competing with the long-ranged Pearl's repulsion.

**Keywords** High- $T_c$  superconductors · Pearl's effect · Vortex attraction · Vortex molecules

## 1 Introduction

The physics of vortices in layered high- $T_c$  superconductor is very rich [1]. In particular, it has been noted that in certain directions it is possible to have the long-range attraction between the tilted vortex lines [2, 3]. In bulk anisotropic superconductors this phenomenon is known to result in the formation of vortex chains in the regime of low magnetic fields (see [4] 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  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [5], scanning-tunneling microscopy in  $\text{NbSe}_2$  [6], and Lorentz microscopy measurements in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [7].

The Lorentz microscopy experiments showed that vortex chains formation is absent for slightly tilted vortices [8]. The particularity of the Lorentz microscopy technique is that the magnetic field distribution induced by vortices is probed by the penetrating electron beam and, therefore, it permits to work only with films of thicknesses  $D$  smaller than  $(0.5-1) \mu\text{m}$ . For this case the surface effects modify the intervortex interaction and, as has been demonstrated in the

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pioneering work [9] by Pearl in 1964, the long-range inter-vortex repulsion appears.

If we consider a thin film sample, we get an interplay between two different long-range potentials: (i) attraction of the tilted vortices ( $U_{\text{att}} \sim -1/R^2$ ), and (ii) the Pearl's repulsion ( $U_{\text{rep}} \sim 1/R$ ). By varying either the film thickness or the tilting angle we can modify the balance between these interactions, which should determine energetically favorable vortex configurations in samples with thickness  $d$  comparable to the London penetration depth  $\lambda$  [8]. 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 a 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.

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. The calculations of Buzdin et al. [8] show that the interaction between two tilted straight vortices in thin film may be written as

$$\varepsilon_{\text{int}} \simeq \frac{\phi_0^2}{8\pi^2} \left( -\frac{D_{\text{eff}} \tan^2 \alpha}{R^2} + \frac{2}{R} \right), \quad (1)$$

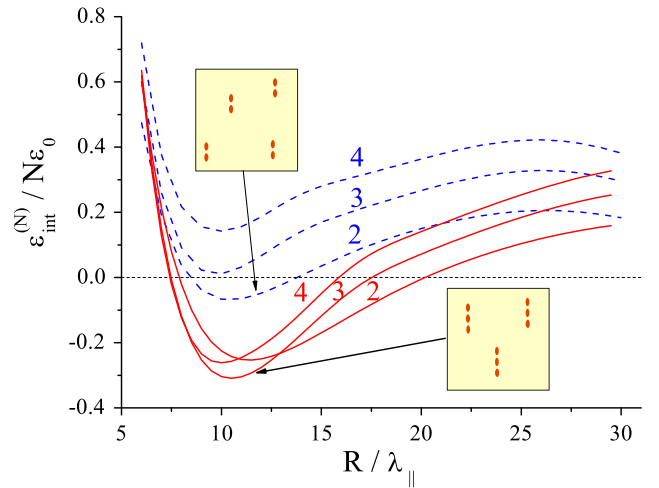
where the effective film thickness is

$$D_{\text{eff}} = D - 2\lambda_{\parallel} \tanh(D/2\lambda_{\parallel}),$$

the angle  $\alpha$  is between the vortex axis and the  $\mathbf{c}$  direction, and  $R$  is the distance between vortices in plane of the film. The formula (1) corresponds to the limit  $\lambda_{\perp} \gg R \gg \lambda_{\parallel}$ . For illustration, several plots of the energy [8] are presented in Fig. 1. We see that depending on the tilting angle the formation of some sort of the vortex molecules—dimer, trimer, etc. occur. When the magnetic field increases, these “molecules” start to build vortex lattice. In fact, the particularity of the intervortex interaction results in the emergence of the flux structures with different number of vortices per unit cell [10].

## 2 Vortex Line in a Finite Stack of SC Layers: Basic Equations

In very anisotropic layered superconductors like BSCCO single crystals ( $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ) the vortex line pierces the film and can be viewed as a string of 2D pancake vortices (PVs): each of these pancakes is centered at the point  $\mathbf{r}_n = x_n \mathbf{x}_0 + y_n \mathbf{y}_0$  in the  $n$ th layer. Within the model of the stack of Josephson-decoupled SC layers, pancakes can interact with each other only via magnetic field. We denote



**Fig. 1** Typical plots of the interaction energy per vortex vs. the inter-vortex distance  $R$  in an equidistant chain of  $N$  vortices: *dashed (solid) lines* correspond to  $D = 3\lambda_{\parallel}$ ,  $\alpha = 78^\circ$  ( $D = 3\lambda_{\parallel}$ ,  $\alpha = 80^\circ$ ). 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*

the interlayer spacing as  $s$  and consider each of the  $N$  layers as a thin film with the thickness  $D$  much less than the London penetration depth  $\lambda$ . General equation for the vector potential  $\mathbf{A}$  distribution in such system reads

$$\text{rot rot } \mathbf{A} = \frac{4\pi}{c} \sum_{n,m=1}^N \mathbf{J}_n^m(\mathbf{r}) \delta(z - z_n), \quad (2)$$

where  $\Lambda = \lambda^2/d = \lambda_{\parallel}^2/s$  is the effective penetration depth in a superconducting film of a vanishing thickness  $d$ , each  $n$ th SC layer coinciding with the plane  $z = z_n = ns$  ( $1 \leq n \leq N$ ). The sheet current at the  $n$ th layer created by the pancake at  $m$ th layer takes the form

$$\mathbf{J}_n^m(\mathbf{r}) = \frac{c}{4\pi\Lambda} [\boldsymbol{\Phi}(\mathbf{r} - \mathbf{r}_m) \delta_{nm} - \mathbf{A}^m(\mathbf{r}, z_n)], \quad (3)$$

where  $\mathbf{A}^m(\mathbf{r}, z)$  is the vector potential induced by the only pancake vortex located in the  $m$ th layer. The vector  $\boldsymbol{\Phi}(\mathbf{r})$  in Eq. (3) is given by the expression

$$\boldsymbol{\Phi}(\mathbf{r}) = \frac{\phi_0}{2\pi} \frac{[\mathbf{z}_0 \times \mathbf{r}]}{r^2}, \quad (4)$$

and  $\phi_0 = \pi \hbar c / e$  is the flux quantum. For the layered system without Josephson coupling, a general expression for the free energy can be written in the form

$$F = \frac{1}{8\pi} \int dV (\text{rot } \mathbf{A})^2 + \left( \frac{4\pi}{c} \right)^2 \Lambda \sum_n \mathbf{J}_n^2(\mathbf{r}) \delta(z - z_n), \quad (5)$$

where the total vector potential  $\mathbf{A}(\mathbf{r}, z)$  and the sheet current in the  $n$ th layer  $\mathbf{J}_n(\mathbf{r})$ , produced by an arbitrary vortex line,

are the sum of the contributions induced by all 2D pancakes:

$$\mathbf{A}(\mathbf{r}, z) = \sum_{m=1}^N \mathbf{A}^m(\mathbf{r}, z), \quad \mathbf{J}_n(\mathbf{r}) = \sum_{m=1}^N \mathbf{J}_n^m(\mathbf{r}).$$

Using the gauge  $\text{div} \mathbf{A} = 0$  and the Fourier transform

$$\mathbf{A}(\mathbf{q}, k) = \int d^2 \mathbf{r} dz e^{i\mathbf{q}\mathbf{r} + ikz} \mathbf{A}(\mathbf{r}, z), \quad (6)$$

$$\mathbf{A}_n(\mathbf{q}) = \int d^2 \mathbf{r} e^{i\mathbf{q}\mathbf{r}} \mathbf{A}(\mathbf{r}, z_n), \quad (7)$$

$$\mathbf{J}_n(\mathbf{q}) = \int d^2 \mathbf{r} e^{i\mathbf{q}\mathbf{r}} \mathbf{J}_n(\mathbf{r}), \quad (8)$$

one can rewrite the basic equation (2) in the momentum representation as follows:

$$(q^2 + k^2) \mathbf{A}(\mathbf{q}, k) = \frac{1}{\Lambda} \sum_n (\boldsymbol{\Phi}_n(\mathbf{q}) - \mathbf{A}_n(\mathbf{q})) e^{ikns}, \quad (9)$$

where  $\boldsymbol{\Phi}_n(\mathbf{q}) = \boldsymbol{\Phi}(\mathbf{q}) e^{i\mathbf{q}\mathbf{r}_n}$ . Taking account of the relation

$$2\pi \mathbf{A}_n(\mathbf{q}) = \int dk e^{ikz_n} \mathbf{A}(\mathbf{q}, k),$$

we obtain from (9) the following equations for the Fourier components of the vector potential  $\mathbf{A}_n(\mathbf{q})$ :

$$2q \Lambda \mathbf{A}_n = \sum_m (e^{i\mathbf{q}\mathbf{r}_m} \boldsymbol{\Phi}(\mathbf{q}) - \mathbf{A}_m) e^{-|n-m|qs}. \quad (10)$$

These equations can be reduced to the scalar form

$$f_n + \frac{1}{2q\Lambda} \sum_m e^{-|n-m|qs} f_m = e^{i\mathbf{q}\mathbf{r}_n}, \quad (11)$$

where we introduce new functions  $f_n(\mathbf{q})$ :

$$\begin{aligned} \mathbf{J}_n(\mathbf{q}) &= \frac{c}{4\pi\Lambda} (\boldsymbol{\Phi}_n(\mathbf{q}) - \mathbf{A}_n(\mathbf{q})) \\ &= \frac{c}{4\pi\Lambda} \boldsymbol{\Phi}(\mathbf{q}) f_n(\mathbf{q}). \end{aligned} \quad (12)$$

The solution of the linear system (11) for a fixed distribution of pancakes  $\mathbf{r}_n$  determines the distribution of the vector potential  $\mathbf{A}(\mathbf{r}, z)$  which is created by an arbitrary vortex line in a finite stack of superconducting layers. In the momentum representation the general expression (5) for the free energy of the layered system without Josephson coupling reads

$$F = \frac{1}{32\pi^3 \Lambda} \sum_n \int d^2 \mathbf{q} (\boldsymbol{\Phi}_n(\mathbf{q}) - \mathbf{A}_n(\mathbf{q})) \boldsymbol{\Phi}_n(-\mathbf{q}). \quad (13)$$

For two vortex lines we can write the total vector potential and the total sheet current as superpositions of contributions

coming from the first  $(\mathbf{A}_n^{(1)}, \mathbf{J}_n^{(1)})$  and second  $(\mathbf{A}_n^{(2)}, \mathbf{J}_n^{(2)})$  vortices:

$$\mathbf{A}_n(\mathbf{q}) = \mathbf{A}_n^{(1)}(\mathbf{q}) + \mathbf{A}_n^{(2)}(\mathbf{q}), \quad \mathbf{J}_n(\mathbf{q}) = \mathbf{J}_n^{(1)}(\mathbf{q}) + \mathbf{J}_n^{(2)}(\mathbf{q}).$$

Calculating the interaction energy  $\varepsilon_{\text{int}}$  of vortex lines we should keep in (13) only the terms which contain the products of fields corresponding to different vortex lines:

$$\begin{aligned} \varepsilon_{\text{int}} &= \frac{1}{32\pi^3 \Lambda} \sum_n \int d^2 \mathbf{q} [(\boldsymbol{\Phi}_n^{(1)}(\mathbf{q}) - \mathbf{A}_n^{(1)}(\mathbf{q})) \boldsymbol{\Phi}_n^{(2)}(-\mathbf{q}) \\ &\quad + (\boldsymbol{\Phi}_n^{(2)}(\mathbf{q}) - \mathbf{A}_n^{(2)}(\mathbf{q})) \boldsymbol{\Phi}_n^{(1)}(-\mathbf{q})]. \end{aligned} \quad (14)$$

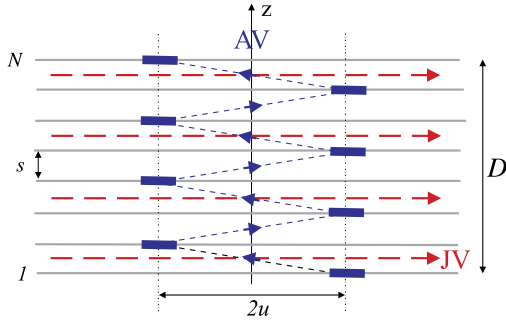
Finally, for the particular case of two identical (parallel) vortex lines which are shifted at the vector  $\mathbf{R} = \mathbf{r}_n^{(2)} - \mathbf{r}_n^{(1)}$  ( $n = 1, N$ ) in the  $(xy)$  plane, we get the following general expression for the interaction energy via the scalar functions  $f_n(\mathbf{q})$ :

$$\varepsilon_{\text{int}}(\mathbf{R}) = \frac{\phi_0^2}{16\pi^3 \Lambda} \int \frac{d^2 \mathbf{q}}{q^2} \cos(\mathbf{q}\mathbf{R}) \sum_n f_n(\mathbf{q}) e^{-i\mathbf{q}\mathbf{r}_n}. \quad (15)$$

The expression (15) and Eq. (11) determine the interaction energy of two identical vortex lines.

### 3 Interaction Potential of Zigzag Vortices

In layered superconductors with a very weak interlayer coupling the application of the magnetic fields tilted from the  $c$  axis provokes the formation of crossing lattice of Abrikosov vortices (AVs) and Josephson vortices (JVs) [11]. The AV is in fact a line of pancake vortices (PVs) interacting with JVs. As has been noted in [12, 13], the perpendicular vortex line formed by the PVs is deformed and attracted by JVs, so the JV stacks accumulate additional PVs. Such decoration of the JVs by the PVs was clearly observed in experiment (see for example [4] and [14]). The interaction between pancakes and in-plane field in the form of JVs is known to produce zigzag deformation of PV stacks (see Fig. 2) [12, 13, 15]. These deformed PV stacks together with JVs could be considered analogous to the tilted vortices in moderately anisotropic superconductors, and they also reveal a long-range attraction between such deformed stacks [16]. Here we study the peculiarities of the interaction of zigzag deformed stacks of PVs caused by the Pearl effect. The zigzag deformation is somewhat larger near the surface (due to the decrease of the stiffness of the PVs stack) but for simplicity we neglect this relatively small effect. Special attention is paid to the conditions under which attraction between PVs in the crossing lattices exists and the dense vortex chains appear in tilted field. Keeping in mind BSCCO, we consider layered superconductors with a high anisotropy ratio



**Fig. 2** Zigzag deformation of the stack of pancakes due to interaction with the Josephson vortices (JVs) in high parallel magnetic field ( $B_x \gg H_0$ ), directed along  $x$  axis. Here  $s$  is a distance between the layers,  $u$  is the amplitude of the zigzag deformation. Josephson vortices are presented by in-plane *dashed arrows* (Color figure online)

$\gamma = \lambda_{\perp}/\lambda_{\parallel} \gg 1$ , when the Josephson core radius  $\lambda_J = \gamma s$  ( $s$  is the interlayer spacing) is larger than an in-plane penetration depth  $\lambda_J \gg \lambda_{\parallel}$ . To describe the intervortex interaction we choose to apply approach of Josephson–decoupled superconducting layers, which is known to be useful in studies of the vortex-lattice structure at low fields.

Our further consideration is based on two assumptions:

(i) for each vortex we choose the centers of pancakes to be positioned along the zigzag line and put

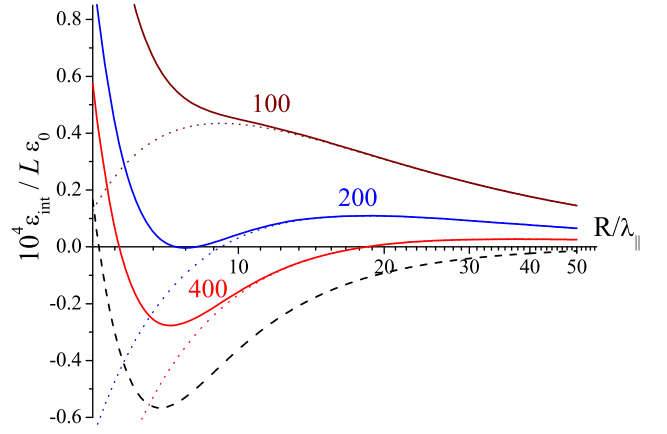
$$\mathbf{r}_n^{(1)} = \pm u \mathbf{x}_0, \quad \text{if } n - \text{odd/even} \quad (16)$$

$$\mathbf{r}_n^{(2)} = \mathbf{r}_n^{(1)} + \mathbf{R};$$

(ii) we restrict ourselves by the continuous limit assuming  $qs \ll 1$  and  $q_x u \ll 1$ . Solving Eqs. (11) and (15) (the details of the calculations will be published elsewhere) we can obtain the following expression for the interaction energy of two identical zigzag pancake stacks which are shifted at the vector  $\mathbf{R} = R \mathbf{x}_0$  in the  $(xy)$  plane:

$$\begin{aligned} \varepsilon_{\text{int}}(R) &= \frac{\phi_0^2}{32\pi^2 \Lambda s} \left\{ D \left[ 2K_0\left(\frac{R}{\lambda_{\parallel}}\right) + K_0\left(\frac{R-2u}{\lambda_{\parallel}}\right) \right. \right. \\ &\quad \left. \left. + K_0\left(\frac{R+2u}{\lambda_{\parallel}}\right) + \ln\left(\frac{R^2-4u^2}{R^2}\right) \right] \right. \\ &\quad \left. + \frac{2}{\lambda_{\parallel}^2} \int_0^{\infty} \frac{dq}{(q^2 + \lambda_{\parallel}^{-2})^2} \right. \\ &\quad \left. \times \frac{2J_0(qR) + J_0(q(R-2u)) + J_0(q(R+2u))}{1 + q/\sqrt{q^2 + \lambda_{\parallel}^{-2}} \tanh^{-1}(D\sqrt{q^2 + \lambda_{\parallel}^{-2}}/2)} \right\}, \quad (17) \end{aligned}$$

where  $J_0$  and  $K_0$  are the Bessel and modified Bessel functions of zero order, respectively. The first term in Eq. (17) describes the interaction of the zigzag stacks in the bulk



**Fig. 3** Typical plots of the interaction energy per layer  $\varepsilon_{\text{int}}(R)/L\varepsilon_0$  vs. the distance  $R$  between two zigzag deformed stacks of pancakes for in-plane magnetic field  $B_x = H_0$ . The numbers near the curves denote the values of film thickness  $d/\lambda_{\parallel}$ . The *dashed line* shows the interaction energy between two zigzag vortices in bulk ( $D \rightarrow \infty$ ) layered SC. *Dotted lines* show long-range part of interaction energy (18). Here  $\varepsilon_0 = \phi_0^2/32\pi^2 \lambda_{\parallel}$ ,  $s = 0.01\lambda_{\parallel}$ ,  $\gamma = 300$

layered system [16], while the last term describes the long-range Pearl repulsion which decays as  $1/R$  and results from the surface contribution to the energy. In Fig. 3 we present some typical plots of the interaction energy per one layer  $\varepsilon_{\text{int}}/N$  vs. the intervortex distance  $R$  for different thickness  $D$  of the film. Neglecting the effect of the zigzag deformation  $u \ll \lambda_{\parallel}$  on the Pearl repulsion, the long-range part of interaction energy (17) between two zigzag PV stacks is given once again by the formula (1) where  $D_{\text{eff}} \equiv D$  and  $\tan \alpha$  must be substituted by the amplitude of a zigzag deformation  $\delta = u/\lambda_{\parallel} = H_0 \lambda_{\parallel} / B_x \lambda_J$ :

$$\varepsilon_{\text{int}}(R) \simeq \frac{\phi_0^2}{8\pi^2} \left( -\frac{D}{R^2} \delta^2 + \frac{2}{R} \right). \quad (18)$$

One can observe an interplay between the long-range attractive and the repulsive forces between two zigzag deformed PV stacks, similar to the one between two tilted vortices in layered [8] or anisotropic [10] SC films. Certainly, in a bulk sample ( $D \rightarrow \infty$ ) the Pearl term in (18) vanishes, and at long distances the dominant interaction between the zigzag PV lines is an attraction [16]. As a result, there always exists the minimum of the interaction potential, which realizes at  $R_m \sim 2\lambda_{\parallel} \ln(1/\delta)$ . However the second term in (18) is very important even for a large sample thickness: for large  $R$  the energy  $\varepsilon_{\text{int}}$  is always positive and corresponds to the vortex repulsion. With a decrease in the distance  $R$  the attraction force comes into play and can result in the change of the sign of the energy at  $R_0 = D\delta^2/2 \gg \lambda_{\parallel}$ , while short-range repulsion is still weak. Such behavior should be accompanied by the appearance of the minimum in the interaction potential if  $R_0 > R_m$ . This condition gives us the estimate of the critical film thickness  $D^*$  for which the minimum in the interaction

potential can exist and the formation of vortex chains can be energetically favorable:

$$D^* \sim 4 \left( \frac{B_x}{H_0} \right)^2 \frac{\lambda_J^2}{\lambda_{||}}. \quad (19)$$

So, in the presence of a dense lattice of Josephson vortices, the AVs penetrate in the form of chains only for a large film thickness  $D > D^*$ . Otherwise, if  $D < D^*$ , the formation of the usual Abrikosov lattice of zigzag deformed stacks of PVs occurs. It is interesting to note that following (18) the intervortex attraction increases near  $T_c$ , when  $\lambda_{||}$  becomes large. This is in contrast with the moderately anisotropic case—Eq. (1) when  $D_{\text{eff}}$  decreases near  $T_c$  and the repulsion prevails.

#### 4 Discussion

If the in-plane, magnetic field  $B_x$  is small the crossings between JVs and PVs are rare and the attractive part of the intervortex interaction is weak. On the other hand, if the in-plane field is large,  $B_x \gg H_0 = \phi_0/\gamma s^2$ , the rhombic lattice of JVs is so dense that the currents of adjacent JVs overlap and this decreases the zigzag deformation of the PV stacks [15]. Optimal regime for the long-range attraction corresponds to  $B_x \sim H_0$ . This case of the intervortex interaction is presented in Fig. 3. We should note that the Pearl interaction plays an important role even for rather thick film—indeed, for the most favorable case  $B_x \sim H_0$  it completely masks attraction for the film thicknesses smaller than 0.01 mm (if we take the BSCCO case with  $\gamma \sim 300$ ,  $\lambda_{||} = 0.1 \mu\text{m}$ ).

We may conclude that the formation of the vortex molecules and the exotic vortex lattices with different number of vortices per unit cell should occur in the films with

the thickness  $D \sim (10\text{--}100)\lambda_{||}$ , and therefore the most suitable technique to observe these structures seems to be the scanning tunneling microscopy, scanning Hall-probe or decoration.

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