

# Orientalional phase transitions of a lattice of magnetic dots embedded in a London-type superconductor

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## Abstract

In recent experiments, structured arrays of ferromagnetic nanoparticles in the bulk of a superconductor have been fabricated. We present the theory of orientational phase transitions in a planar regular lattice of nanoscale ferromagnetic particles embedded in a superconductor. In the London approximation, we show that the interactions between ferromagnetic particles can lead to either a parallel or antiparallel spin alignment depending on the ratio of the interparticle distance, the London penetration depth and the temperature. The extension of the results to systems of ferromagnetic nanoparticles with more complicated geometries is discussed.

## 1. Introduction

Recent achievements in nanoscience have demonstrated that fundamentally new physical phenomena can be observed, when systems are reduced in size to dimensions which become comparable to the fundamental microscopic length of a material under study. Superconductivity is a macroscopic quantum phenomenon, and therefore it is especially interesting to know whether this quantum state would be changed or not when the samples are reduced to a nanometer size [1]. Magnetism and superconductivity are two competing collective ordered states in metals. In the case of ferromagnetism, the exchange interactions lead to the parallel alignment of electronic spins, while the electron–phonon interactions in BCS superconductors lead to the spin singlet pairing of electrons. Clearly, these two types of order are generally mutually incompatible. In bulk systems, the frustration between the electron singlet pairing and the spin ordering is resolved by the FFLO (Fulde–Ferrell [2], Larkin–Ovchinnikov [3]) state. The possibility for superconductivity and ferromagnetism to exist in the same volume was proved for magnetic superconductors  $\text{HoMo}_6\text{S}_8$  and  $\text{ErRh}_4\text{B}_4$  [5]. The phase where superconductivity coexists with a magnetic modulated structure, was discovered in works [6, 7].

In recent years, however, a great increase in interest in the coupling between ferromagnetism and superconductivity

in artificially structured systems has arisen. Advances in nanotechnology and microfabrication have made it possible to build hybrid structures containing both ferromagnetic and superconducting components which interact magnetically or via the proximity effect [4, 8–12].

More complex types of structures have also been produced; for example, Moshchalkov fabricated arrays of ferromagnetic nanodots on superconducting substrates [12–14]. The London theory was used previously to describe the vortex structure resulting from such magnetic dipoles [15–17]. A description of similar structures based on the Ginzburg–Landau theory was developed by Peeters [19–22].

Early studies of the influence of ferromagnets on the superconducting state were performed in the 1960s for a dispersion of fine ferromagnetic particles (Fe, Gd, Y) in a superconducting matrix [22, 23]. The number of investigations of S/F hybrids has increased over the last two years [14].

The theory of superconducting systems is faced with a number of important questions which have had no experimental support till now. These questions include, for example, the study of magnetic configurations in a system of magnetic dots positioned in the superconducting matrix.

This problem was solved in the particular case of a nanocomposite with a London-type superconducting matrix [24]. As a result, the formulas defining a configuration

of magnetic fields and the interaction energy for an ensemble of ferromagnetic dots are deduced. It is established that, in the frame of the developed model, orientational phase transitions can occur. The condition for the phase transition for an isolated pair and, in the nearest-neighbors approximation, for a chain of ferromagnetic dots surrounded by a superconductor is determined.

Distinct from the case of a sufficiently simple linear array, the distribution of dots in a planar or three-dimensional lattice retains a more significant arbitrariness for the orientation of magnetic moments. The subject of the present studies is the determination of the equilibrium configurations of the magnetic moments for a planar lattice of magnetic dipoles with a square basic cell embedded into a London-type superconductor. We will show that, depending on the ratio of the lattice constant and the parameter of penetration of a magnetic field into the superconductor, an orientational phase transition between orthogonal and planar distributions of the magnetic moments of dots takes place.

To solve the posed problem, we use the results of the previous work [24], according to which the strength of a magnetic field produced by an arbitrary ensemble of point-like magnetic moments is given by the relation

$$\mathbf{H}(\mathbf{r}) = \sum_i \exp(-R_i/\delta) \cdot \left\{ \left( \frac{3\mathbf{R}_i(\mathbf{R}_i \cdot \mathbf{m}_i)}{R_i^5} - \frac{\mathbf{m}_i}{R_i^3} \right) \times \left( 1 + \frac{R_i}{\delta} + \frac{R_i^2}{\delta^2} \right) - \frac{2\mathbf{R}_i(\mathbf{R}_i \cdot \mathbf{m}_i)}{\delta^2 \cdot R_i^3} \right\}, \quad (1)$$

where  $\mathbf{R}_i = \mathbf{r} - \mathbf{r}_i$  and  $R_i = |\mathbf{r} - \mathbf{r}_i|$ . Here,  $\mathbf{H}(\mathbf{r})$  is the magnetic field at the observation point  $\mathbf{r}$ ,  $\mathbf{r}_i$  is the radius-vector of the position of a magnetic dot,  $\delta$  is a parameter characterizing the penetration depth of the magnetic field which can depend, by our assumption, on the temperature, and  $\mathbf{m}_i$  is the magnetic moment of a magnetic dot. We have considered the case where interacting magnetic nanodots are embedded in a bulk superconducting material.

A second source of the interaction between moments is the RKKY interaction modified by the presence of the BCS energy gap  $D$ . It is clear that the range of the dipolar forces is determined by the penetration depth  $\delta$ , while the usual oscillatory power-law RKKY interaction is truncated exponentially on a length scale of the order of  $\xi_0$ . Therefore, the dipolar forces dominate for superconductors in the London limit  $r_0 \ll \delta$ , while the RKKY interaction is more important in the Pippard case  $r_0 \gg l$ , where  $r_0 - 1 = \xi_0^{-1} + l^{-1}$ , and  $l$  is the mean free path [11]. Here, we consider the London limit and neglect the RKKY interaction. Magnetic impurities interacting via the RKKY interaction were considered by Galitski and Larkin [25]. Formula (1) describes the magnetic field strength at an arbitrary distribution of magnetic particles. The model accepted in the present work was earlier discussed in [24]. A single limitation for relation (1) is related to the application of the approximation a magnetic dot. In this case, the characteristic size of a magnetic dot  $d$  must be much less than the penetration depth of the magnetic field ( $\delta \gg d$ ).

It is easy to prove that, in the limiting case where the penetration depth of the field  $\delta$  tends to infinity  $\delta \rightarrow \infty$  (the

transition in the normal state) in the domain adjacent to the ensemble of dots, relation (1) tends to the limit

$$\mathbf{H}(\mathbf{r}) = \sum_i \left( \frac{3\mathbf{R}_i(\mathbf{R}_i \cdot \mathbf{m}_i)}{R_i^5} - \frac{\mathbf{m}_i}{R_i^3} \right)$$

which describes the magnetic field of a system of point-like dipoles in the medium being in the normal state.

In the other limiting case where the distances between dots are significantly greater than the penetration depth  $d \ll \delta \ll R$ , relation (1) reads

$$\mathbf{H}(\mathbf{r}) = \sum_i \frac{\exp(-R_i/\delta)}{R_i \delta^2} \cdot \left( \frac{\mathbf{R}_i(\mathbf{R}_i \cdot \mathbf{m}_i)}{R_i^2} - \mathbf{m}_i \right).$$

## 2. Equilibrium configurations and conditions for the orientational phase transition in a two-dimensional lattice of ferromagnetic dots

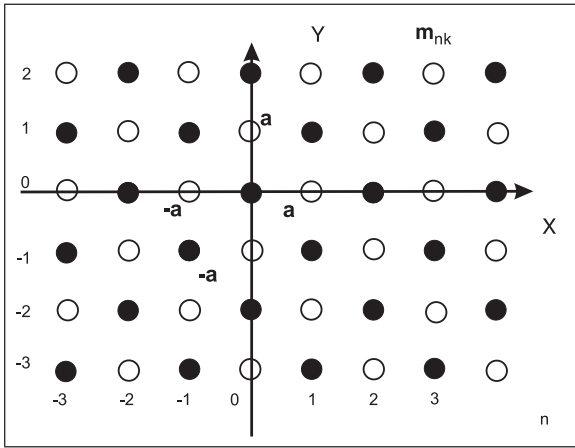
In order to determine the collective state of the magnetic moments in nanocomposite materials with the matrix made of a London-type superconductor, we use the following formula for the energy of magnetic interaction [24]:

$$U = -\frac{1}{2} \sum_i \sum_j \exp(-R_{ij}/\delta) \times \left\{ \left( \frac{3(\mathbf{R}_{ij}\mathbf{m}_j)(\mathbf{R}_{ij} \cdot \mathbf{m}_i)}{R_{ij}^5} - \frac{\mathbf{m}_i\mathbf{m}_j}{R_{ij}^3} \right) \times \left( 1 + \frac{R_{ji}}{\delta} + \frac{R_{ij}^2}{\delta^2} \right) - \frac{2(\mathbf{R}_{ij}\mathbf{m}_j)(\mathbf{R}_{ij} \cdot \mathbf{m}_i)}{\delta^2 \cdot R_{ij}^3} \right\}, \quad (2)$$

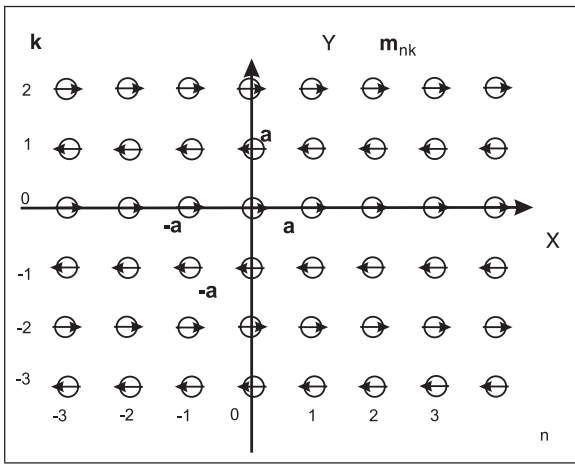
where  $\mathbf{R}_i = \mathbf{r} - \mathbf{r}_i$  and  $R_i = |\mathbf{r} - \mathbf{r}_i|$ . Earlier, this formula was used to determine the conditions for the orientational phase transition for an isolated pair of point-like magnetic moments [24]. However, relation (2) allows one to study the equilibrium states of a wider circle of nanocomposite systems, one of which is a planar lattice of ferromagnetic dots with the square basic cell.

First of all, we note that the realization of one or other magnetic configuration depends on both the competition of diamagnetic effects from the side of the superconducting matrix and the magnetostatic interaction in the system of ferromagnetic dots. At lower temperatures eliminating the thermal disordering of the system, the magnetostatic interaction leads to a correlation of magnetic moments. As the main conditions for the formation of magnetic configurations, we take the equivalence of all sites and the zero value of the net magnetic moment of the lattice.

The planar lattice of dots by itself sets a preferred direction in the space. Therefore, we will separate two configurations from the whole manifold of spatial orientations of magnetic moments. The first configuration is presented in figure 1. It is characterized by the orientations of the magnetic moments of dots which are orthogonal to the base plane. The alternation of the magnetic moments of neighboring dots decreases, to a certain extent, the energy of magnetic interaction. In addition, such a distribution of magnetic moments favors a decrease of the strength of a magnetic field in a system of magnetic dipoles embedded in the superconducting matrix, which is also



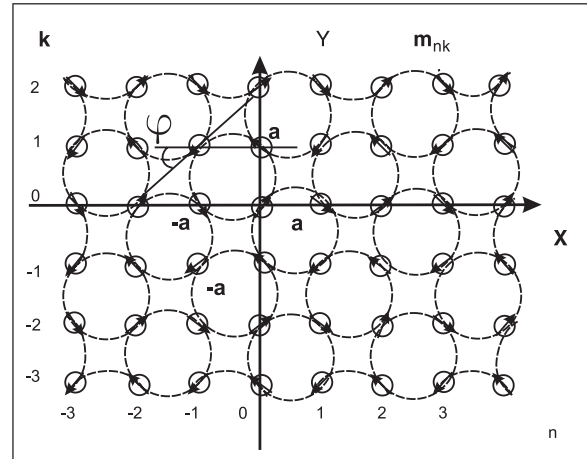
**Figure 1.** Two-dimensional lattice of magnetic dots. Magnetic moments are directed up (filled circles) or down (empty circles);  $a$  is the lattice constant.



**Figure 2.** Two-dimensional lattice of magnetic dots. Magnetic moments are aligned in the plane in the form of chains;  $a$  is the lattice constant.

advantageous from the energy viewpoint. Thus, the given configuration can be considered as a version of the magnetic order.

A configuration of the second type is shown in figure 2. It is characterized by the distribution of magnetic moments in the base plane of the lattice such that the magnetic moments are aligned as magnetic chains with alternating directions of the magnetic moments. Here, like the configuration presented in figure 1, the main requirement, namely the equivalence of the states of magnetic dots, is satisfied. A similar distribution also decreases the energy of the magnetostatic interaction, on the one hand, and on the other favors a decrease in the strength of a magnetic field in a superconductor. At the same time, the planar orientation of magnetic moments has the basic distinction from the orthogonal one. For example, by means of a continuous change of the magnetic moments in the base plane, the configuration in figure 2 can be transferred in the structure shown in figure 3. Such a system is characterized by a coherent rotation of magnetic moments by an angle  $\pm\varphi$  relative to the principal directions of the lattice. In this case, both the modulation of the direction of moments at the



**Figure 3.** Part of the lattice with a modulated planar distribution of the magnetic moments ( $a$  is the lattice constant). Circles stand for magnetic dots, and the arrows on them indicate the directions of magnetic moments  $\mathbf{m}_{nk}$  in the base plane ( $n, k$  are the spatial indices of magnetic dots). The angle  $\varphi$  defines a deviation of the moments of magnetic dots from the principal direction of the lattice. The states of all dots in the given configuration are equivalent. The net magnetic moment is zero. The separated circles denote schematically magnetic vortices. The dotted lines are tangents to the directions of the magnetic moments at sites of the lattice.

sites of the lattice and some increase in the energy of the magnetic chains occur. These processes are accompanied by the formation of magnetic vortices in cells, which promotes a decrease in the magnetic interaction energy. The states of separate magnetic dots in the lattice remain equivalent at the zero total magnetization. Thus, the questions arise; how the energy of the array of magnetic moments in the base plane shown in figure 3 depends on the angle  $\varphi$ , and to which value it is equal in the equilibrium state.

To answer these questions, we consider relation (2) for the interaction energy and reduce it to a single sum by virtue of the fact that the states of the magnetic dots are equivalent. In this case, in order to calculate the energy of the lattice, it is sufficient to determine the energy of a single magnetic dot, e.g.,  $\mathbf{m}_{0,0}$ , located at the origin of the coordinate system and then to multiply the result by the total number of magnetic points  $N$ .

Relation (2) becomes significantly simpler:

$$U = -\frac{N}{2} \left\{ 3 \left( 1 - \frac{\partial}{\partial \alpha} \right) + \frac{\partial^2}{\partial \alpha^2} \right\} \times \sum_i^N \exp(-\alpha \cdot r_i / \delta) \frac{(\mathbf{r}_i \cdot \mathbf{m}_i)(\mathbf{r}_i \cdot \mathbf{m}_{0,0})}{r_i^5} + \frac{N}{2} \left\{ 1 - \frac{\partial}{\partial \alpha} + \frac{\partial^2}{\partial \alpha^2} \right\} \sum_i^N \exp(-\alpha \cdot r_i / \delta) \frac{\mathbf{m}_i \cdot \mathbf{m}_{0,0}}{r_i^3}. \quad (3)$$

By writing formula (3), we used the method of differentiation with respect to the parameter  $\alpha$  which should be set equal to unity after the calculations. The index  $i$  stands for the summation over all sites of the lattice.

At the summation in relation (3), it is convenient to introduce the pair of indices  $(n, k)$  defining the position of a site in the lattice (figure 3) instead of the running index of magnetic dots  $i$ .

It is easy to see that the system represented in figure 3 possesses the translational invariance with a period of  $2a$  so that

$$\mathbf{m}_{n,k} = \mathbf{m}_{n+2l,k+2p}, \quad l, p = \pm 1, \pm 2, \dots \quad (4)$$

The lattice has only four types of magnetic dots differing from one another by a spatial orientation of magnetic moments. Their vector components depend on the angle  $\varphi$  in the following manner:

$$\begin{aligned} \mathbf{m}_{n,k} = \mathbf{m}_{n+2l,k+2p}, \quad \mathbf{m}_{2l,2p} = \mathbf{m}_{0,0} &= m \begin{pmatrix} \cos \varphi \\ \sin \varphi \\ 0 \end{pmatrix}, \\ \mathbf{m}_{2l+1,2p} = \mathbf{m}_{1,0} &= m \begin{pmatrix} \cos \varphi \\ -\sin \varphi \\ 0 \end{pmatrix}, \\ \mathbf{m}_{2l,2p+1} = \mathbf{m}_{0,1} &= m \begin{pmatrix} -\cos \varphi \\ \sin \varphi \\ 0 \end{pmatrix}, \\ \mathbf{m}_{2l+1,2p+1} = \mathbf{m}_{1,1} &= m \begin{pmatrix} -\cos \varphi \\ -\sin \varphi \\ 0 \end{pmatrix}, \\ l, p = \pm 1, \pm 2, \dots & \\ l, p = \pm 1, \pm 2, \dots & \end{aligned} \quad (5)$$

where  $m$  is the modulus of the magnetic moment of a site. After the substitution of (5) in (3) and the summation over sites of the unbounded lattice, we get the following significant result. It turns out that the interaction energy of the system of magnetic dots (see figure 3) does not depend on the angle  $\varphi$  and is determined by the relation

$$\frac{U_{\parallel}}{N} = \frac{m^2}{a^3} \cdot F(a/\delta), \quad (6)$$

where  $N$  is the number of sites of the lattice,  $m$  is the magnetic moment of a dot, the factor  $m^2/a^3$  on the right-hand side has a dimension of energy and characterizes the interaction energy in the system of magnetic dots,  $F(a/\delta)$  is the energy characteristic of a magnetic state which is a universal function of a single parameter and determines the dependence of the energy on both the period and the field penetration depth at the distribution of magnetic moments in the base plane of the lattice. It can be represented in the form of a sum

$$\begin{aligned} F(a/\delta) &= \frac{\mu_0}{4\pi} \left( -1 + \frac{\partial}{\partial \alpha} + \frac{\partial^2}{\partial \alpha^2} \right) \cdot \frac{1}{8} \\ &\times \sum_{l=1}^{\infty} \sum_{p=0}^{\infty} \frac{\exp(-2\alpha(a/\delta)\sqrt{l^2+p^2})}{(l^2+p^2)^{3/2}} \\ &- \left( -1 + \frac{\partial}{\partial \alpha} + \frac{\partial^2}{\partial \alpha^2} \right) \cdot \frac{1}{4} \\ &\times \sum_{l=-\infty}^{\infty} \sum_{p=-\infty}^{\infty} \frac{\exp(-\alpha(a/\delta)\sqrt{(2l+1)^2+(2p+1)^2})}{((2l+1)^2+(2p+1)^2)^{3/2}} \\ &- \left( 3 - 3\frac{\partial}{\partial \alpha} + \frac{\partial^2}{\partial \alpha^2} \right) \cdot \frac{1}{2} \\ &\times \sum_{l=-\infty}^{\infty} \sum_{p=-\infty}^{\infty} \frac{((2l+1)^2-(2p)^2) \cdot \exp(-\alpha(a/\delta)\sqrt{(2l+1)^2+(2p)^2})}{((2l+1)^2+(2p)^2)^{5/2}}. \end{aligned} \quad (7)$$

Thus, the state of the system is degenerated in the parameter  $\varphi$  in the presence of a tough correlation of the mutual orientations of moments of the ensemble of magnetic dots. The energies of the configuration shown in figure 2 and the ensemble with a modulated distribution of the magnetic moments (see figure 3) coincide. In turn, the determination of the energy of magnetic interaction for the configuration possessing the orthogonal orientation of magnetic moments (figure 1) requires a smaller amount of calculations, because the first sum in formula (3) vanishes.

The result of the calculations can be represented in the form

$$\frac{U_{\perp}}{N} = \frac{m^2}{a^3} \cdot \Phi(a/\delta), \quad (8)$$

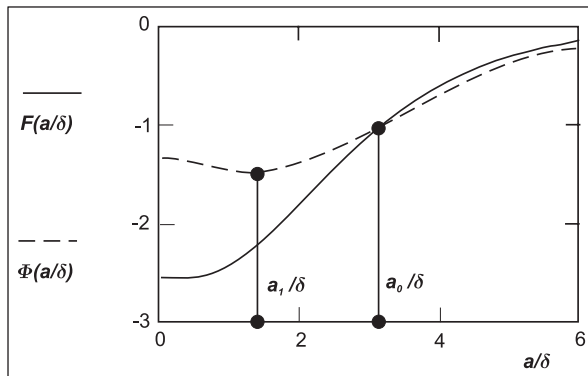
where the factor  $m^2/a^3$  characterizes, like that in relation (6), the magnetic interaction energy,  $\Phi(a/\delta)$  is the energy characteristic of the magnetic state which is a universal function of the single parameter and determines the dependence of the energy on both the period and the field penetration depth at the distribution of magnetic moments which are perpendicular to the base plane of the lattice. This function can be represented in the following form:

$$\begin{aligned} \Phi(a/\delta) &= \left( 1 - \frac{\partial}{\partial \alpha} + \frac{\partial^2}{\partial \alpha^2} \right) \cdot \frac{1}{2} \\ &\times \left\{ \sum_{l=1}^{\infty} \sum_{p=0}^{\infty} \frac{4 \cdot \exp(-\alpha \cdot (a/\delta) \cdot \sqrt{2 \cdot l^2 + 2 \cdot p^2})}{(2 \cdot l^2 + 2 \cdot p^2)^{3/2}} \right. \\ &\left. - \sum_{l=-\infty}^{\infty} \sum_{p=-\infty}^{\infty} \frac{\exp(-\alpha \cdot (a/\delta) \cdot \sqrt{2 \cdot (l+1/2)^2 + 2 \cdot (p+1/2)^2})}{(2 \cdot (l+1/2)^2 + 2 \cdot (p+1/2)^2)^{3/2}} \right\}. \end{aligned} \quad (9)$$

The difference in the energies of the system of magnetic moments with planar (6) and perpendicular (8) orientations relative to the lattice plane is determined by the ratio of the functions  $\Phi(a/\delta)$  and  $F(a/\delta)$ . The calculation of the functions  $\Phi(a/\delta)$  and  $F(a/\delta)$  on the basis of relations (7) and (9) is not a difficult task and can be realized with any mathematical software. The results are presented in a graphical form in figure 4. In figure 4, we represent the plots of the energy characteristics for two different states of the magnetic lattice versus the ratio of the parameter of a cell and the penetration depth of the magnetic field,  $a/\delta$ . The limit  $a/\delta \rightarrow 0$  corresponds to the transition of the matrix into the normal state.

It is obvious that the lattice with the planar orientation of magnetic moments (figures 2 and 3) possesses a lower energy in the normal state at  $a/\delta = 0$ . Therefore, the state with the perpendicular direction of moments (figure 1) cannot be realized at all in the absence of a superconductor. As the temperature decreases, and the penetration depth diminishes gradually, the parameter  $a/\delta$  begins to grow. When this parameter attains the value  $a/\delta \approx 3.3$ , the configuration with the orthogonal orientation of magnetic moments (figure 1) becomes more advantageous in energy, and the orientational phase transition occurs in the system. At a decrease in the temperature, a similar scenario of events completely corresponds to a reorientation of the magnetic moments of an isolated pair of magnetic points which was considered in the previous work [24].





**Figure 4.** Plots of the energy characteristics  $F(a/\delta)$  and  $\Phi(a/\delta)$  of states of the lattice with the normal and planar orientations of magnetic moments, respectively. Values of  $F(0)$  and  $\Phi(0)$  correspond to the transition of the superconducting matrix into the normal state.

Our approach is valid for materials which are well described by the London limit  $r_0 \ll \delta$ , RKKY interactions being negligible. In the experimental systems studied by Moshchalkov [9, 10], a square array of Pt/Co magnetic nanodots was deposited on the surface of the Pb-based superconductor which is type I ( $\kappa = 0.48$ ). The dots were about  $0.26 \mu\text{m}$  in diameter and occupied the lattice with a spacing of  $0.6 \mu\text{m}$ . We showed in [24] that the spin-orientational phase transition for Pb will be observed at the temperature  $T = 7.14 \text{ K}$ , whereas  $T_c = 7.2 \text{ K}$ .

A certain interest can be attracted to the question about the magnetic ordering in a three-dimensional lattice of magnetic points. The answer can be obtained from the condition of minimum for the potential energy of the magnetic interaction (2), the formula for which is deduced in work [24]. However, the arbitrariness in the orientations of magnetic moments of a 3D system is much greater. Therefore, the determination of steady three-dimensional configurations is a promising task of the given trend. Of course, it would be interesting in future to generalize our results to superconductors in the Pippard limit, in which RKKY interactions between the quantum dots will dominate over dipolar forces [25].

### 3. Conclusion

In conclusion, we note that an analogous phase transition can be expected to occur in a planar lattice with a rectangular basic cell. The only difference will consist in the elimination of the degeneration relative to the directions of magnetic moments in the base plane.

Of course, the direct observation of a phase transition will be hampered, because the problem involves a magnetic lattice surrounded by a superconductor. However, a similar orientational transformation must also happen in a lattice

applied on the surface of a massive superconductor, though values of the parameter  $a_0/\delta$  will be different in this case.

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