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Interactions of nanoscale ferromagnetic granules in a London superconductor

Sergei P Kruchinin[1], **Yuri I Dzezherya**[2] **and James F Annett**[3]

[1] Bogolyubov Institute for Theoretical Physics, NASU, Metrologichna street 14 b, 252143, Kiev-143, Ukraine

[2] Institute of Magnetism, NASU, Vernadskogo 36, UA-03142 Kiev, Ukraine

[3] University of Bristol, H H Wills Physics Laboratory, Tyndall Avenue, Bristol BS8 1TL, UK

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Abstract

Recent experiments have fabricated structured arrays of ferromagnetic nanoparticles in proximity to a bulk superconductor. We consider the theory of interactions between two nanoscale ferromagnetic particles embedded within a superconductor. In the London limit approximation we show that the interactions between ferromagnetic particles can lead to either parallel or antiparallel spin alignment. The cross-over between these is dependent on the ratio of interparticle spacing and the London penetration depth. We show that a phase transition between spin orientations can occur as temperature is varied. Finally we comment on the extension of these results to arrays of nanoparticles in different geometries.

1. Introduction

Magnetism and superconductivity are two competing collective ordered states in metals. In the case of ferromagnetism the exchange interactions lead to parallel alignment of the electronic spins, while in BCS superconductivity electron–phonon interactions lead to spin singlet pairing of electrons. Clearly these two types of order are generally mutually incompatible. In bulk systems the frustration between electron singlet pairing and spin ordering is resolved by the FFLO, Fulde–Ferrell [1]–Larkin–Ovchinnikov [2], state. However, this has proved elusive experimentally and few examples are known. In particular, the FFLO state appears to be highly sensitive to disorder.

In recent years, however, there has been a great increase in interest in the interactions between ferromagnetism and superconductivity in artificially structured systems. Advances in nanotechnology and micro-fabrication have made it possible to build hybrid structures containing both ferromagnetic and superconducting components, which interact magnetically or via the proximity effect [3]. Superconductor–ferromagnet–superconductor planar structures have been found to show Josephson π-junction behaviour [4–6]. Ferromagnet–superconductor–ferromagnet spin valve structures have also been fabricated [7] with potential applications to spintronics. More complex types of structures have also been fabricated; for example, Lange *et al* fabricated arrays of ferromagnetic nanoscale dots on superconducting substrates [8, 9]. A

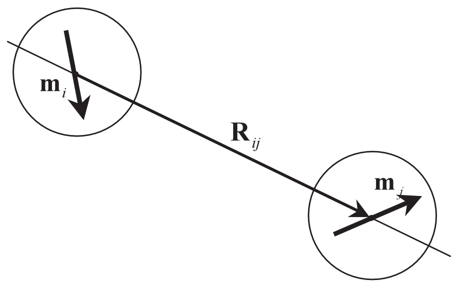

Figure 1. Schematic diagram of a nanocomposite superconductor containing ferromagnetic granules.

description of such structures based upon Ginzburg–Landau theory was developed by Milosevic and Peeters [10].

In this paper we consider the interaction between magnetic nanoparticles embedded within a superconductor, as shown in figure 1. Theoretical studies of such systems can be carried out exactly in two limiting cases depending on the relative magnitudes of the London penetration depth λ and the coherence length ξ. If we consider the nanoparticles to be essentially point-like, on the scale of both these characteristic lengths, then they correspond to effective point-like magnetic moments of the form

$$\mathbf{M}(\mathbf{r}) = \sum_i \mathbf{m}_i \delta(\mathbf{r} - \mathbf{r}_i) \tag{1}$$

where the particle at $\mathbf{r}_i$ has magnetic moment $\mathbf{m}_i$. Interactions between these isolated moments arise: directly from magnetic dipole–dipole forces, modified by the screening of the bulk supercurrents. A second source of interaction between the moments is the RKKY interaction, modified by the presence of the BCS energy gap Δ. It is clear that the range of the dipolar forces is determined by the penetration depth λ, while the usual oscillatory power-law RKKY interaction is truncated exponentially on a length scale of order ξ_0. Therefore the dipolar forces dominate for superconductors in the London limit $r_0 \ll \lambda$, while RKKY interactions are 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 RKKY interactions. Magnetic impurities interacting via RKKY interactions were considered by Galitski and Larkin [12].

In the London limit, we first derive general expressions for the configuration of magnetic fields and the interaction energy of an ensemble of ferromagnetic granules. Then we consider the case of two interacting nanoparticles. It is found that as a function of temperature an orientation phase transition can take place. The conditions for such a phase transition to occur are derived for a chain of ferromagnetic granules. Finally we comment on the application of these results to determine the equilibrium configurations of more general lattices of ferromagnetic particles.

2. Magnetic field of ferromagnetic inclusions in a London superconductor

We determine the energies of magnetic configurations in superconducting nanocomposite systems by means of Maxwell's equations. The total current, $\mathbf{j}$, includes both normal and superconducting parts,

$$\mathbf{j} = \mathbf{j}_{\mathrm{s}} + \mathbf{j}_{\mathrm{n}}. \qquad (2)$$

The role of the normal currents in the superconductor is negligible since the superconductor exhibits weak magnetic characteristics in the normal state. However, a normal current will be present within the ferromagnetic inclusions. This normal state current can be written in the traditional form:

$$\mathbf{j}_{\mathrm{n}} = \nabla \times \mathbf{M} \qquad (3)$$

where $\mathbf{M}$ is the magnetization of the material. The supercurrent obeys the usual London equation

$$\nabla \times \mathbf{j}_{\mathrm{s}} = -\frac{n_{\mathrm{s}} e^2}{m}\mathbf{B} \qquad (4)$$

in SI units [13], where $\mathbf{B}$ is the magnetic field, n_{s} is the superfluid density, and m and e are the electron mass and charge, respectively.

Combining relations (2)–(4) with the Maxwell equation $\nabla \times \mathbf{B} = \mu_0 \mathbf{j}$, the magnetic field can be found in the form

$$\nabla \times (\nabla \times \mathbf{B}) + \lambda^{-2}\mathbf{B} = \mu_0 \nabla \times (\nabla \times \mathbf{M}), \qquad (5)$$

where λ is the London penetration depth of field in the superconductor. Since $\operatorname{div}\mathbf{B} = 0$, equation (5) can be rewritten

$$-\nabla^2 \mathbf{B} + \lambda^{-2}\mathbf{B} = \mu_0 \nabla \times (\nabla \times \mathbf{M}). \qquad (6)$$

For boundary conditions we assume that the magnetic field in the superconductor vanishes at large distances from the region of the ferromagnetic inclusions. According to these boundary conditions the solution of equation (6) can be expressed as follows:

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi}\int \mathrm{d}^3 r' G(|\mathbf{r}-\mathbf{r}'|)\nabla \times (\nabla \times \mathbf{M}(\mathbf{r}')), \qquad (7)$$

where the Green's function is given by

$$G(|\mathbf{r}-\mathbf{r}'|) = \frac{\exp(-|\mathbf{r}-\mathbf{r}'|/\lambda)}{|\mathbf{r}-\mathbf{r}'|}. \qquad (8)$$

After integrating by parts twice and some steps of algebra this expression can be rewritten in the general form

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi}\int \mathrm{d}^3 r' \exp(-R/\lambda) \cdot \left\{ \left(\frac{3\mathbf{R}\left(\mathbf{R}\cdot\mathbf{M}\left(\mathbf{r}'\right)\right)}{R^5} - \frac{\mathbf{M}\left(\mathbf{r}'\right)}{R^3} \right) \cdot \left(1 + \frac{R}{\lambda} + \frac{R^2}{\lambda^2}\right) - \frac{2\mathbf{R}\left(\mathbf{R}\cdot\mathbf{M}\left(\mathbf{r}'\right)\right)}{\lambda^2 \cdot R^3} \right\}, \qquad (9)$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, $R = |\mathbf{r} - \mathbf{r}'|$.

Expression (9) determines the magnetic field outside the ferromagnetic inclusions. Since the magnetization of the system $\mathbf{M}(\mathbf{r})$ is defined by (3), it is implied in (9) that $\mathbf{M}(\mathbf{r}) = 0$ outside the volume of the ferromagnetic granules.

3. Magnetic field of ferromagnetic quantum dots in a superconducting nanocomposite material

Assuming that the sizes of the ferromagnetic inclusions are nanometre length scales, they will appear essentially point-like on the scale of the penetration depth λ. In this case we can approximate the magnetization as a sum of point magnetic moments, as shown in equation (1). Using this approximation in the general expression equation (9) we find the result

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi}\sum_i \exp(-R_i/\lambda) \cdot \left\{ \left(\frac{3\mathbf{R}_i\left(\mathbf{R}_i\cdot\mathbf{m}_i\right)}{R_i^5} - \frac{\mathbf{m}_i}{R_i^3} \right) \cdot \left(1 + \frac{R_i}{\lambda} + \frac{R_i^2}{\lambda^2}\right) - \frac{2\mathbf{R}_i\left(\mathbf{R}_i\cdot\mathbf{m}_i\right)}{\lambda^2 \cdot R_i^3} \right\} \qquad (10)$$

where $\mathbf{R}_i = \mathbf{r} - \mathbf{r}_i$ and $R_i = |\mathbf{r} - \mathbf{r}_i|$.

It is apparent from equation (10) that when the temperature of the superconductor approaches T_{c} and the penetration depth $\lambda \to \infty$ then the expression (10) tends to the limit

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi}\sum_i \left(\frac{3\mathbf{R}_i\left(\mathbf{R}_i\cdot\mathbf{m}_i\right)}{R_i^5} - \frac{\mathbf{m}_i}{R_i^3} \right), \qquad (11)$$

which describes the usual magnetic field of isolated dipoles in the normal state medium.

In the other limiting case, if the distance between granules is much more than the penetration depth λ, we have

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi}\sum_i \frac{\exp(-R_i/\lambda)}{R_i \cdot \lambda^2} \cdot \left(\frac{\mathbf{R}_i\left(\mathbf{R}_i\cdot\mathbf{m}_i\right)}{R_i^2} - \mathbf{m}_i \right). \qquad (12)$$

4. Interaction energy of quantum dots in a superconducting nanocomposite material

To determine the collective states of the magnetic moments in superconducting nanocomposite materials in the London limit we make use of the expression for free energy F of the system [14]

$$F = \frac{1}{2\mu_0}\int \mathrm{d}^3r\,\left\{\mathrm{B}^2 + \lambda^2\,(\nabla\times\mathbf{B})^2\right\}. \qquad (13)$$

Integrating by parts and using the Gauss theorem we transform equation (13) to the following form:

$$F = \frac{1}{2\mu_0}\int \mathrm{d}^3r\mathbf{B}\left\{\mathbf{B} + \lambda^2\nabla\times(\nabla\times\mathbf{B})\right\} = 2\pi\lambda^2\int \mathrm{d}^3r\mathbf{B}\cdot\nabla\times(\nabla\times\mathbf{M}). \qquad (14)$$

Integrating by parts and using the Gauss theorem again we transform equation (14) to the form

$$F = -2\pi\lambda^2\int \mathrm{d}^3r\mathbf{M}\cdot\nabla\times(\nabla\times\mathbf{B}). \qquad (15)$$

Then, using the London equation (5) and omitting the magnetostatic self-energy from consideration, we find the interaction energy of magnetic moments. The obtained expression can be used to determine the collective state of magnetization of an ensemble of granules.

$$U = -\frac{\mu_0}{8\pi}\sum_i\sum_j \exp\left(-R_{ij}/\lambda\right)\cdot\left\{\left(\frac{3\left(\mathbf{R}_{ij}\cdot\mathbf{m}_j\right)\left(\mathbf{R}_{ij}\cdot\mathbf{m}_i\right)}{R_{ij}^5} - \frac{\mathbf{m}_i\cdot\mathbf{m}_j}{R_{ij}^3}\right)\cdot\left(1+\frac{R_{ji}}{\lambda}+\frac{R_{ij}^2}{\lambda^2}\right) - \frac{2\left(\mathbf{R}_{ij}\cdot\mathbf{m}_j\right)\left(\mathbf{R}_{ij}\cdot\mathbf{m}_i\right)}{\lambda^2\cdot R_{ij}^3}\right\}, \qquad (16)$$

where $\mathbf{R}_{ij} = \mathbf{r}_j - \mathbf{r}_i$, $R_{ij} = |\mathbf{r}_j - \mathbf{r}_i|$, $i \neq j$.

5. Spin orientation phase transitions in nanocomposite material with arrays of ferromagnetic quantum dots

We begin by studying the magnetic configuration of an isolated pair of magnetic moments. The interaction energy of such a pair can be written as

$$U = -\frac{\mu_0}{4\pi}\exp\left(-R_{12}/\lambda\right)\cdot\left\{\left(\frac{3\left(\mathbf{R}_{12}\cdot\mathbf{m}_1\right)\left(\mathbf{R}_{12}\cdot\mathbf{m}_2\right)}{R_{12}^5} - \frac{\mathbf{m}_1\cdot\mathbf{m}_2}{R_{12}^3}\right)\cdot\left(1+\frac{R_{12}}{\lambda}+\frac{R_{12}^2}{\lambda^2}\right) - \frac{2\left(\mathbf{R}_{12}\cdot\mathbf{m}_1\right)\left(\mathbf{R}_{12}\cdot\mathbf{m}_2\right)}{\lambda^2\cdot R_{12}^3}\right\}. \qquad (17)$$

Let us introduce a coordinate system with origin at the first magnetic moment $\mathbf{m}_1$ and polar axis along the line connecting magnetic moments. In this system the magnetic moments have the components $\mathbf{m}_i = m_i(\cos\varphi_i\sin\theta_i, \sin\varphi_i\sin\theta_i, \cos\theta_i)$, and their interaction energy (17) is written in the form

$$U = \frac{\mu_0}{4\pi}m_1m_2\cdot\frac{\exp\left(-R_{12}\lambda\right)}{R_{12}^3}\cdot f\left(\theta_i, \varphi_i, R_{12}/\lambda\right) \qquad (18)$$

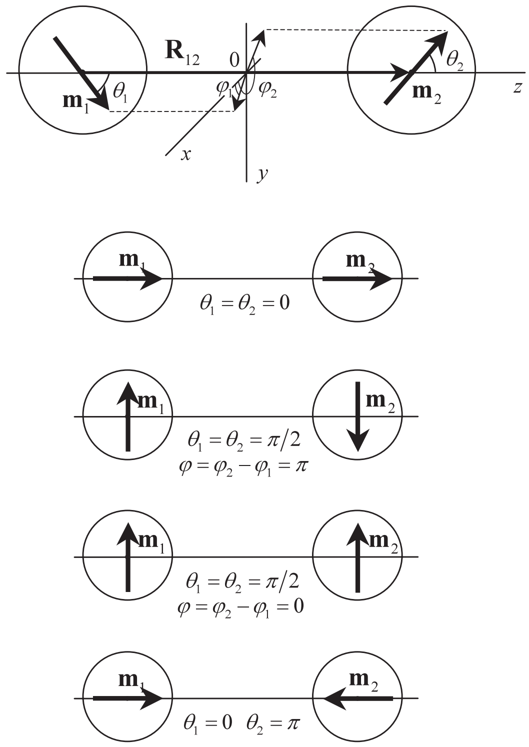

Figure 2. The four energy saddle points of a pair of ferromagnetic quantum dots, as defined in equation (20). Of these four, either 1 and 2 is the ground state, depending on condition equation (22). States 3 and 4 are never stable.

where

$$f\left(\theta_i, \varphi_i, x\right) = -2\left(1+x\right)\cos\theta_1\cos\theta_2 + \left(1+x+x^2\right)\sin\theta_1\sin\theta_2\cos\varphi \qquad (19)$$

and $\varphi = \varphi_2 - \varphi_1$.

Differentiating the function $f(\theta_i, \varphi_i, x)$ with respect to angular variables and equating to zero we find, that there are four possible stable energy configurations

$$\theta_1 = \theta_2 = 0; \quad \theta_1 = \theta_2 = \pi \quad 0 \leqslant \varphi < 2\pi \qquad (20a)$$

$$\theta_1 = \theta_2 = \pi/2 \quad \varphi = \pi \qquad (20b)$$

$$\theta_1 = \theta_2 = \pi/2 \quad \varphi = 0 \qquad (20c)$$

$$\theta_1 = 0, \quad \theta_2 = \pi \quad 0 \leqslant \varphi < 2\pi \qquad (20d)$$

which are illustrated in figure 2.

Further analysis shows that the configurations (20c) and (20d) are saddle points, not energy minima. Evaluating second derivatives of (19) we obtain the stability condition of the configuration (20a):

$$\left(\frac{R}{\lambda}\right)^2 - \frac{R}{\lambda} - 1 \leqslant 0. \qquad (21)$$

This implies that ferromagnetic ordering of the pair of magnetic moments is possible if

$$\frac{R}{\lambda} \leqslant \frac{1}{2}(1+\sqrt{5}). \qquad (22)$$

It turns out that if condition (22) is violated then the alternative configuration (20*b*) is the stable energy minimum. We can conclude that when the temperature changes and the penetration depth parameter, $\lambda(T)$, varies in such a way as to break condition (22) then the ground state orientation will change from (20*a*) to (20*b*).

This result readily generalizes to ordered arrays of ferromagnetic granules, and so we conclude that orientational phase transitions are possible in systems of quantum dots in a superconducting matrix. For example, it is clear that condition (22) can be applied to linear chains of quantum dots. On the other hand, the results for square or cubic lattices remain to be determined.

6. Conclusions

We have considered the interactions between nanoscale magnetic dots embedded within a bulk superconducting material. Our approach is valid for materials which are well described by the London limit $r_0 \ll \lambda$, for RKKY interactions are negligible. We have shown that depending on the dimensionless parameter R/λ different stable ground states occur, and so as a function of temperature orientational phase transitions will take place for periodic arrays of such quantum dots. Of course our calculation does not include all types of interactions which define the orientation of magnetic moments in the space. In particular, we neglect the energy of magnetic anisotropy of the granules, which is defined by the shape of the granules or the type of their crystal lattice. However, when the granules are close to spherical in shape and the crystal lattice of the ferromagnet has cubic symmetry, then equation (16) will be essentially exact.

In the experimental systems studied by Lange *et al* [8, 9], a square array of Pt/Co magnetic nanodots was deposited on the surface of the superconductor Pb, which is type I ($\kappa = 0.48$). The dots were about 0.26 μm in diameter and they were deposited on a grid of spacing 0.6 μm. For Pb the penetration depth is 39 nm at low temperatures, and so this array was in the limit $R > \lambda$ and the dot–dot interaction would be expected to correspond to the antiferromagnetic alignment shown in the second case in figure 2. Increasing temperature, the transition to ferromagnetic alignment would occur according to equation (22) at $\lambda = 0.36R$, i.e. 219 nm. Using the Casimir form $\lambda(T) = \lambda(0)(1-t^4)^{-1/2}$ with $t = T/T_c$ this would occur at $T = 7.14$ K, compared to $T_c = 7.2$ K. Therefore the experimental conditions for the transition to be observed are certainly feasible. Of course, for an exact comparison with theory in this case our theory should be generalized to deal with magnetic particles near the surface rather than embedded in the bulk of the superconductor.

Of course it would also 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 [12]. Expression (17) can also be used to study, by numerical methods, the magnetic configurations and orientation phase transitions in ensembles of nanogranules. It is possible to determine the conditions of orientation transformations in analytical form for ordered structures (a chain of granules, plane and volume lattices). To the extent that the magnetic subsystem state of a specimen at phase transitions is changed, then this phenomenon can be experimentally observed as a change in magnetic susceptibility in the region of low field values.

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