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Dynamic behaviour of a suspension of superconducting particles in an alternating magnetic field

V I Kalikmanov† and P P J M Schram

Department of Physics, Building W & S, Eindhoven University, PO Box 513, 5600 MB, Eindhoven, The Netherlands

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Abstract. We study the behaviour of a dilute suspension of single crystal high- T_c superconducting particles in an alternating magnetic field $H_c = H_0 \cos \omega \tau$ of intermediate amplitude value H_0 . For the case of the inertia term being neglected and field frequency $\omega < 1$ MHz the analytical solution of the nonlinear equation of rotational motion has been obtained. It is shown that particles rotation $\theta(\tau)$ is squeezed in one of the zones $((\pi/2)n, (\pi/2)(n+1))$, $n = 0, \pm 1, \ldots, \theta(\tau)$ and magnetization $M(\tau)$ are step-like functions of time.

1. Introduction

Recently we have proposed a new class of media—suspensions of high- T_c superconducting particles (SSP) (mean diameter $d \approx 1 \,\mu$ m) dispersed in a solvent at $T < T_c$ [1, 2]. Such media can possess some unusual properties, e.g. an anomalously high diamagnetic susceptibility, magnetization curve that crosses the M = 0 axis (i.e. the behaviour of sSP can be either paramagnetic or diamagnetic depending on applied magnetic field intensity), etc.

The technological aspects of making the particles for a SSP were discussed in [3]. The effective susceptibility of chain and disordered structures of particles in a 'composite suspension' [1] (a SSP with a ferrofluid [4] as a solvent) were calculated by Horvath and Kopcansky [5].

In all the above mentioned papers the internal structure of high- T_c superconducting particles was not taken into account. Meantime it is well known that properties of ceramic and single crystal samples differ essentially. In this paper we shall consider ssp with a disperse phase consisting of single crystal high- T_c particles. In this case an important feature of particles will be a rather high uniaxial anisotropy of the superconducting state parameters: penetration depth λ , coherence length ξ , lower and upper critical magnetic fields H_{c1} and H_{c2} . For example, for YBa₂Cu₃O_{7-x} (a '1-2-3' material) the values of parameters in the Cu–O plane (*ab* plane of a primitive cell) differ from their values along the *c* axis perpendicular to *ab*: $\xi_c = 5.6$ Å, $\xi_{a,b} = 28$ Å, $\lambda_c = 19.6 \times 10^2$ Å, $\lambda_{a,b} = 3.9 \times 10^2$ Å [6].

+ Permanent address: Institute for High Temperatures USSR Academy of Sciences, Izhorskaya Street 13/19, Moscow, 127412, USSR.

8904 V I Kalikmanov and P P J M Schram

Due to the strong uniaxial anisotropy the magnetic moment of a particle in an external magnetic field H_e is not antiparallel to H_e when $H_e > H_{c1}$ (as in the case of isotropic superconductors), and its magnitude depends on the angle θ between H_e and the *c* axis. When H_e is an alternating field this anisotropy results in a specific nonlinear dynamic behaviour of dispersed particles and consequently of the SSP magnetization M. The purpose of this work is to study the behaviour of θ and M in an alternating magnetic field

$$H_{\rm c} = H_0 \cos \omega \tau \tag{1.1}$$

where the amplitude H_0 has an intermediate value:

$$H_{\rm c1} \ll H_0 \ll H_{\rm c2}.\tag{1.2}$$

We note that in a cycle of the field the superconducting material alternately occurs in the Abrikosov vortex phase and in the Meissner phase. The latter occurs when $\omega \tau$ is in the vicinity of points $\pi/2$ and $3\pi/2$; due to the constraint (1.2) these intervals are short compared to the intervals where the vortex phase exists. The dynamic behaviour of a particle will be different for these time intervals.

In section 2 we briefly review the thermodynamic properties of anisotropic superconductors within the frame of the London theory and derive the expression for magnetic energy of a spherical SSP particle, placed in an intermediate magnetic field H_e . Section 3 is devoted to the analysis of the dynamic behaviour of a particle and its magnetization in the field (1.1).

2. Magnetic energy of a ssp particle

Due to the high value of the Ginzburg-Landau parameter $\kappa = \lambda/\xi \ge 1$, the London theory is applicable in a wide domain of fields $H_e \ll H_{c2}$. The anisotropy of a superconductor can be taken into account by introducing the effective mass tensor M_{ik} of Cooper pairs [7, 8] (for '1-2-3' material in the principal axes $M_{aa} = M_{bb} = 8$ electron masses, $M_{cc} = 200$ electron masses [6]).

The Helmholtz free energy of a particle (per unit length in the direction of vortices) is:

$$F = \frac{1}{8\pi} \int \left[\boldsymbol{h}^2 + \bar{\lambda}^2 \boldsymbol{m}_{ik} \left(\operatorname{curl} \boldsymbol{h} \right)_i \cdot \left(\operatorname{curl} \boldsymbol{h} \right)_k \right] \mathrm{d}x \mathrm{d}y \tag{2.1}$$

where h(x, y) is the local magnetic field, dxdy is an element of an area normal to the vortex axis z, $m_{ik} = M_{ik}/\bar{M}$, $\bar{M} = (M_{aa}M_{bb}M_{cc})^{1/3}$ is the 'average mass', $\bar{\lambda} = (\bar{M}c^2/4\pi n_s e^2)^{1/2}$ the 'average penetration depth' (n_s is the number density of superconducting electrons).

In order to make use of equation (2.1) one must express the values of mass tensor m_{ik} in the vortex frame in terms of its values in the crystal frame. The vortex frame (x, y, z) is obtained by a rotation θ of the crystal frame (a, b, c) around the b axis (which coincides with the y axis). So θ is the angle between the c axis of a crystal and the vortex axis (figure 1). The tensor analysis formulae read:

$$m_{xx} = m_1 \cos^2 \theta + m_3 \sin^2 \theta \qquad m_{xy} = m_{yz} = 0$$

$$m_{yy} = m_1 \qquad m_{zz} = m_1 \sin^2 \theta + m_3 \cos^2 \theta \qquad (2.2)$$

$$m_{yz} = (m_1 - m_3) \sin \theta \cos \theta.$$



Figure 1. Mutual orientation of the vortex frame (x, y, z) and the crystal frame $(\hat{a}, \hat{b}, \hat{c})$ of a single crystal SSP particle. **B** is the magnetic induction vector.



Figure 2. Magnetization of a single crystal SSP particle. In the vortex phase M contains a component M_n normal to the vortex axis.

Here we have used the notations: $m_1 = m_{aa} = m_{bb}$, $m_3 = m_{cc}$. Varying F with respect to h(x, y) one can obtain the anisotropic London equations [8].

In view of the periodicity of the vortex lattice it is natural to use the Fourier transformation

$$\boldsymbol{h}_{\mathbf{G}} = \boldsymbol{n}_{L} \int \boldsymbol{h}(x, y) \exp(-\mathrm{i}\boldsymbol{G} \cdot \boldsymbol{r}) \,\mathrm{d}x \,\mathrm{d}y \tag{2.3}$$

where n_L is the vortex density (per unit area), r = (x, y), G are the reciprocal lattice vectors; integration is over a primitive cell. Substitution of the London equations into (2.1) and use of the Fourier transformation (2.3) gives the following result for the free energy density [9]:

$$F = \frac{B^2}{8\pi} \sum_{G} (1 + \bar{\lambda}^2 m_{zz} G^2) [(1 + \bar{\lambda}^2 m_{zz} G_x^2 + \bar{\lambda}^2 m_3 G_y^2)(1 + \bar{\lambda}^2 m_1 G^2)]^{-1}$$
(2.4)

where $B = \phi_0 n_L$ is the magnetic induction, ϕ_0 the flux quantum; summation is over the reciprocal lattice vectors. If H_e is an intermediate field, i.e. $H_{cl} \ll H_e \ll H_{c2}$, then the average vortex spacing L satisfies $\xi \ll L \ll \overline{\lambda}$. We introduce dimensionless reciprocal lattice vectors g = LG and expand the free energy in powers of a small parameter

$$\varepsilon = (L/\bar{\lambda})^2 \tag{2.5}$$

and obtain:

$$8\pi F = B^2 + \varepsilon B^2 \frac{m_{zz}}{m_1} \sum_{g \neq 0} (m_{zz} g_x^2 + m_3 g_y^2)^{-1}.$$
 (2.6)

Replacing summation by integration and introducing a cutoff at $g_{max} = 2\pi L/\xi$ in the logarithmically divergent integral (the Fourier components relative to the interior of the hard core must be excluded—a usual procedure for type 2 superconductors [10]), we obtain:

$$8\pi F = B^2 + (\phi_0/4\pi\lambda^2)(m_1B_a^2 + m_3B_c^2)^{1/2}\ln(\zeta H_{c2}/B).$$
(2.7)

Here $B_a = B \sin \theta$, $B_c = B \cos \theta$ are the projections of the magnetic induction vector **B**

on the crystal axes, parameter ζ depends on the type of the vortex lattice. The quantity $\phi_0/4\pi\lambda^2$ is of the order of H_{c1} and in the field domain under consideration is small with respect to both *B* and *H*. Therefore we can replace *B* under the ln sign in equation (2.7) by H_e and obtain:

$$8\pi F = B^2 + 2H^* (m_1 B_a^2 + m_3 B_c^2)^{1/2}$$
(2.8)

$$H^* = (\phi_0 / 4\pi \bar{\lambda}^2) \ln(\zeta H_{c2} / H_e).$$
(2.9)

Using the thermodynamic relation $H = 4\pi \partial F / \partial B$ we can derive magnetic field H inside the particle:

$$H_a = B_a + H^* m_1 \sin \theta / \sqrt{m(\theta)} \qquad H_c = B_c + H^* m_3 \cos \theta / \sqrt{m(\theta)}$$
(2.10)

where

$$m(\theta) = m_1 \sin^2 \theta + m_3 \cos^2 \theta. \tag{2.11}$$

Then the reversible magnetization $M = (B - H)/4\pi$ can be easily obtained:

$$M_{a} = -(1/4\pi)H^{*} m_{1} \sin \theta / \sqrt{m(\theta)} \qquad M_{c} = -(1/4\pi)H^{*} m_{3} \cos \theta / \sqrt{m(\theta)}.$$
(2.12)

In general, vector M is not directed along the vortex axis: $M = M_p + M_n$. It has a component M_n normal to it. The specific feature of this problem is also the dependence of a particle magnetic moment on the angle between field and the crystalline axis c.

In the field domain under consideration we can neglect demagnetization effects and therefore magnetic energy of a spherical particle of radius R can be expressed in the following form:

$$U = -M \cdot H_e^{\frac{4}{3}} \pi R^3. \tag{2.13}$$

In the same approximation we can consider that the direction of vortices (B) coincides with H_c (figure 2). Then equation (2.13) reads:

$$U = \frac{1}{3}R^3 H^* H_e \sqrt{m(\theta)}. \tag{2.14}$$

It can be seen that the magnetic energy minimum corresponds to $\theta = \pi/2$.

In the field domain $H_e \leq H_{cl}$ the situation is different: M is always antiparallel to H_e and the demagnetization effect is important. In this domain

$$M = -(1/4\pi)H_{\rm e}/(1-D) \tag{2.15}$$

$$U = \frac{1}{3}R^3 H_e^2 / (1 - D)^2$$
(2.16)

where D is the demagnetization factor (D = 1/3 for a sphere); the magnetic energy does not depend on θ .

Finally we shall consider the domain of fields slightly larger than H_{c1} : $H_c \ge H_{c1}$. In this case the vortex density n_L is low, vortices are separated by more than λ and only a few of nearest neighbours are important. For an *isotropic* superconductor the following expression for magnetic induction is valid [11]:

$$B = (2\phi_0/\sqrt{3}\,\lambda^2) [\ln(3\phi_0/4\pi\lambda^2(H-H_{\rm cl}))]^{-2}.$$
(2.17)

Here $H = H_e - 4\pi D M$ is the magnetic field inside the particle. In our case in this field domain demagnetization effect is still important while anisotropy effect is small and we can approximately take it into account by substituting the mean penetration depth $\overline{\lambda}$

instead of λ in (2.17). Then using the relation $B = H_e + 4\pi(1 - D)M$ we obtain the following transcendent equation for magnetization:

$$4\pi M = -\frac{H_{\rm e}}{1-D} + \frac{2\phi_0}{\sqrt{3}\,\bar{\lambda}^2(1-D)} \left[\ln \frac{3\phi_0}{4\pi\bar{\lambda}^2(H_{\rm e} - 4\pi DM - H_{\rm cl})} \right]^{-2} \tag{2.18}$$

defining M as a function of H_e . Now we can calculate the magnetic energy:

$$U = (-M \cdot H_e + 4\pi DM^2)(4\pi/3)R^3.$$
(2.19)

So for $H_e \ge H_{c1}$ magnetization remains approximately antiparallel to H_e , its value is given by the solution of (2.18); therefore the magnetic energy does not depend on θ as in the pure Meissner phase.

3. Behaviour of a dilute ssp in an alternating magnetic field

We consider a dilute SSP placed in an alternating magnetic field $H_e = H_0 \cos \omega \tau$, $H_{cl} \ll H_0 \ll H_{c2}$. Neglecting interparticle interactions we shall study the behaviour of a single particle of the suspension. Its Lagrangean L takes the form:

$$L = \frac{1}{2}I(\theta')^2 - U$$
 (3.1)

where $I = (8/15)\pi\rho_s R^5$ is the momentum of inertia of the particle, ρ_s its density, $' = \partial / \partial \tau$. We omitted in the Lagrangean terms connected with translational degrees of freedom because they do not contribute to the rotational motion we are studying.

The Lagrange equation of motion in the presence of viscous dissipation reads:

$$I\theta'' - \partial U/\partial \theta = -8\pi\eta R^3 \theta'. \tag{3.2}$$

The right-hand side of this equation represents the viscous torque acting on the spherical particle [12], η being the solvent viscosity. This expression for the viscous torque is valid when

$$(\rho_{\text{solv}}\omega/2\eta)^{1/2}R \ll 1 \tag{3.3}$$

where ρ_{solv} is the solvent density. Taking into account the characteristic values of the parameters $R \sim 0.1 \,\mu\text{m}$, $\eta \sim 10^{-3} \,\text{g/cm s}$, $\rho_{\text{solv}} \approx 0.8 \,\text{g/cm}^3$ we obtain the limitation on the field frequency:

$$\omega \leq 1 \text{ MHz}.$$

If we introduce the dimensionless time $t = \omega \tau$ equation (3.2) can be written as:

$$I\omega^2\ddot{\theta} - \partial U/\partial\theta + 8\pi\eta R^3\omega\dot{\theta} = 0 \tag{3.4}$$

where $\dot{\theta} = d\theta/dt$.

We consider the case when the inertia term in equation (3.4) can be neglected compared to the viscous one (the general case will be considered elsewhere). It implies the following inequality:

$$|I\omega^2\ddot{\theta}| \ll |8\pi\eta R^3\omega\dot{\theta}|. \tag{3.5}$$

We shall discuss its validity below. As we are interested in periodical solutions $\theta(t)$ we shall discuss the behaviour of θ for $0 \le t \le 2\pi$. It was noted in section 2 that there were two different superconducting phases at time interval $(0, 2\pi)$: Abrikosov vortex phase

and Meissner phase with the expressions for magnetic energy U given by equations (2.14), (2.16) and (2.19). So equation (3.2) in the dimensionless form reads:

$$\dot{\theta} = \begin{cases} 0 & \text{for } t_{c} < t < \pi - t_{c} \\ & \& \pi + t_{c} < t < 2\pi - t_{c} \\ & (\text{Meissner phase}) \\ -(1/\beta)f(\theta) \cos t & \text{otherwise} \\ & (\text{Abrikosov phase}). \end{cases}$$
(3.6)

Here $t_c = \arccos(H_{c1}/H_0)$, $f(\theta) = \sin 2\theta/\sqrt{1 + (\gamma^2 - 1)\cos^2\theta}$, $\beta = 48\pi\eta\omega/H^*H_0$ $\gamma^{-1/3}(\gamma^2 - 1)$; we have introduced the anisotropy coefficient $\gamma = (m_3/m_1)^{1/2} > 1$ and used an obvious equality $m_1^2m_3 = 1$. We shall search for the solution $\theta(t)$ of equation (3.7) and then join it with the solution θ = constant of equation (3.6) by continuity.

Analysing equation (3.7) one can see that the range of θ values in the $\theta - t$ plane is divided into zones bounded by the straight lines $\theta = (\pi/2)n$, $n = 0, \pm 1, \ldots$, corresponding to zeros of function $f(\theta)$. If $(\pi/2)n < \theta(t = 0) < \pi/2(n + 1)$ for arbitrary n, then the trajectory $\theta(t)$ will be totally situated in the zone $((\pi/2)n, \pi/2(n + 1))$ and will never intersect its boundaries.

Function $f(\theta)$ can be expressed in the following form:

$$f(\theta) = (1/\gamma)\sin 2\theta/\Delta(\theta)$$
(3.8)

$$\Delta(\theta) = \sqrt{1 - (1 - 1/\gamma^2) \sin^2 \theta}.$$
(3.9)

Then equation (3.7) reads:

$$\dot{\theta} = -(1/\beta\gamma)[\sin(2\theta)/\Delta]\cos t. \tag{3.10}$$

Taking the time derivative from both sides of equation (3.10) we find that the neglect of the inertia term (inequality (3.5)) results in the following condition:

$$\frac{\gamma^{-1/3}(\gamma^{2-1})}{360\pi} \cdot \frac{H^* H_0 \rho_s R^2}{\eta^2} \ll 1.$$
(3.11)

It is satisfied for $H^* \sim 10^2$ Gs, $H_0 \sim 10^3$ Gs, $\gamma \sim 4 - 8$. Equation (3.10) has the solution:

$$\frac{1}{2}\ln\frac{1-\Delta}{1+\Delta} + \frac{1}{2\gamma}\ln\frac{\Delta+1/\gamma}{\Delta-1/\gamma} = -\frac{2}{\beta\gamma}\sin t + C$$
(3.12)

where constant C depends on the initial conditions $\theta_0 = \theta(t = 0)$. In order to study the properties of the solution obtained we shall introduce a new variable

$$y = (1 - \Delta)/(1 + \Delta).$$
 (3.13)

It follows from the definition of $\Delta(\theta)$ that

$$0 < y < y_{c} = (\gamma - 1)/(\gamma + 1) < 1.$$
(3.14)

Substituting (3.13) into (3.12) we obtain:

$$y^{\gamma}[(\gamma + 1) - y(\gamma - 1)] = a - by$$

$$a = p(t)(\gamma - 1) \qquad b = p(t)(\gamma + 1) \qquad p(t) = \exp[2\gamma C - (4/\beta)\sin t]. \qquad (3.15)$$



Figure 3. Geometrical interpretation of equation (3.15).



Figure 4. Rotation of a particle. $\beta = 4.28 \times 10^{-2}$, $\gamma = 4.5$, $\theta_0 = \pi/10$.

A geometrical interpretation of equation (3.15) is presented in figure 3. Function $f_1(y) = y^{\gamma}[(\gamma + 1)-y(\gamma - 1)]$ has a single minimum in the point $y_{\min} = 0$ and a single maximum in the point $y_{\max} = \gamma/(\gamma - 1) > 1$. Straight lines $f_r(y) = a - by$ with different a and b have negative slopes and intersect the y axis in one point $y = y_c < 1$. So for each value of t equation (3.15) has the unique solution $y(t) \in (0, y_c)$. From equation (3.13) we find:

$$\sin^2 \theta = [\gamma^2/(\gamma^2 - 1)](1 - \Delta^2).$$

The behaviour of $\theta(t)$ for $\beta = 4.28 \times 10^{-2}$, $\gamma = 4.5$ and $\theta_0 = \pi/10$ is shown in figure 4. Particle rotation is squeezed between $\theta = 0$ and $\theta = \pi/2$ and represents a step-like function of t. The particle spends most of the time in positions with fixed θ orientations. Changes in orientations occur very rapidly.

Next we consider the magnetization behaviour. Due to the fact that particles are considered to be independent we shall be interested in magnetization M of one arbitrary particle. We must distinguish between the vortex phase and the Meissner phase.

In the Meissner phase M is antiparallel to H_e and demagnetization is important. If M_p is the projection of M on the direction $H_e(t=0)$, then in accordance with equation (2.15):

$$4\pi M_{p}(t) = -\frac{3}{2}H_{0}\cos t.$$

For $H_e \ge H_{c1}$ the expression for M_p is given by equation (2.18). In the developed vortex phase $(H_{c1} \le H_e \le H_{c2})$ the corresponding quantity can be derived from equation (2.12):

$$4\pi M_p(t) = \pm H^* \gamma^{2/3} \Delta[\theta(t)].$$

The behaviour of $M_p(t)$ is shown in figure 5 for the same values of the parameters as used in figure 4. It is also a step-like function of time in the vortex phase and is nearly linear in the Meissner phase (due to $H_{c1} \ll H_0$). It should be noted that the sharp corners in figures 4 and 5 are not realistic. The neglected inertia term and frequency dependent part of the viscous torque can be expected to have a smoothing influence.

In conclusion we have studied the behaviour of dilute SSP with single crystal high- T_c superconducting particles in an alternating magnetic field of intermediate amplitude





value. For the case of the inertia term being neglected and field frequency $\omega < 1$ MHz the analytical solution of the nonlinear equation of rotational motion has been obtained. Particles rotation $\theta(t)$ takes place in one of the zones $((\pi/2)n, (\pi/2)(n + 1)), n = 0, \pm 1, \ldots, \theta(t)$ and magnetization M(t) are step-like functions of time.

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