Crystallization of the ordered vortex phase in high temperature superconductors

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Abstract

The Landau-Khalatnikov time-dependent equation is applied to describe the crystallization process of the ordered vortex lattice in high temperature superconductors after a sudden application of a magnetic field. Dynamic coexistence of a stable ordered phase and an unstable disordered phase, with a sharp interface between them, is demonstrated. The transformation to the equilibrium ordered state proceeds by movement of this interface from the sample center toward its edge. The theoretical analysis dictates specific conditions for the creation of a propagating interface, and provides the time scale for this process.

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The process of formation of various equilibrium phases after a sudden change in the thermodynamic conditions, is a topic of wide theoretical and experimental interest. Obviously, the initial state created immediately after the abrupt change is a non-equilibrium, unstable state. The transformation of this state to the thermodynamic equilibrium state may proceed either homogeneously throughout the entire system, or by nucleation of a spatially localized domain of the equilibrium phase, creating a front which propagates until equilibrium is reached in the entire system. The latter process has been recently observed in the vortex system of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO) crystals. This vortex system exhibits a transition between an ordered and disordered phases, at a temperature-independent transition field $B_{od} \approx 400$ G. High temporal resolution magneto-optical measurements indicated that immediately after a sudden application of external magnetic field $B_a \lesssim B_{od}$, a transient disordered vortex state is created, followed by a nucleation and front propagation of an ordered vortex state. The purpose of this paper is to analyze theoretically the crystallization process of the vortex ordered phase, i.e. the nucleation process, the creation of a front and its motion.

Our analysis is based on the Landau-Khalatnikov (LK) time dependent equation:

$$\frac{\partial \Psi}{\partial t} = -\Gamma \frac{\delta F}{\delta \Psi},$$

(1)

where $\Psi$ and $F$ are the order parameter and the free energy of the system, and $\Gamma$ is the Landau-Khalatnikov damping coefficient. We define the order parameter of the vortex system in a way analogous to the definition of the order parameter in order-disorder transitions.
in atomic solids. In the latter case, the order parameter $\rho_q$ is a set of Fourier components of the atomic density taken at reciprocal lattice vectors $q = G$. In particular, for an ordered lattice phase $\rho_q = \text{const} \neq 0$ at $q = G$, whereas for a disordered state $\rho_q = 0$ for all $q \neq 0$. Extending this approach to the vortex order-disorder phase transition, we note that in small-angle neutron scattering experiments in BSCCO, Bragg peaks are observed at low temperatures and low fields mainly in the first Brillouin zone; these peaks are smeared for fields larger than $B_{od}$. Thus, only one component, $\rho_{G_1}$, is sufficient to completely describe the order parameter, $\rho_{G_1}$ being the value of the Fourier component of the vortex density at the minimal vector of the reciprocal lattice. In order to describe the kinetics of the phase transition we allow the order parameter to be temporally and spatially dependent, $\Psi(r, t) \equiv \rho_{G_1}(r, t)$, assuming that $\Psi(r)$ varies slowly over the inter-vortex distance. The scalar real order parameter $\Psi(r, t)$ so defined, distinguishes between two thermodynamic solid phases of the vortex matter: $\Psi = 0$ for the disordered state and $\Psi = \Psi_0 \neq 0$ for the ordered state.

In the Ginzburg-Landau formalism, the phase transition between the ordered and disordered phases may be described by a free energy density functional $F$:

$$F = \frac{1}{2} D (\nabla \Psi)^2 - \frac{1}{2} \alpha \Psi^2 - \frac{1}{3} \beta \Psi^3 + \frac{1}{4} \gamma \Psi^4$$

(2)

where $\alpha, \beta, \gamma$ and $D$ are the Landau coefficients. These coefficients depend on the vortex-vortex and vortex-pinning interactions, and their evaluation requires a microscopic theory which does not yet exist. Note that Eq. 2 does not describe the whole free energy of the vortex system, but only that part which is varying through the phase transition, i.e. $\Psi$-dependent.

As the order-disorder vortex phase transition in BSCCO is field driven, we express the parameter $\alpha$ as a function of $B$:

$$\alpha = \alpha_0 (1 - B/B^*),$$

(3)

where $B^*$ is a characteristic field related to the transition field $B_{od} = B^*(1+2/9\mu)$, where $\mu = \alpha_0 \gamma/\beta^2$. Note that for a second order transition ($\beta = 0$), $B_{od} = B^*$. For a first order phase transition, metastable states of the system are found between $B^*$ and $B^{**} = B^*(1 + 1/4\mu)$. For $B < B^*$ the disordered state is unstable while the ordered state, characterized by

$$\Psi = \Psi_0 = \frac{\beta}{2\gamma} \left( 1 + \sqrt{1 + 4\mu(1 - B/B^*)} \right),$$

is stable. For $B > B^{**}$ the ordered state is unstable, while the disordered state with $\Psi = 0$ is thermodynamically favorable. All the above results are deduced from the conventional Landau theory for phase transitions by replacing temperature with the induction $B$.

In solving Eq. (1) we assume an initial non-equilibrium disordered vortex state ($\Psi = 0$) caused by the rapid injection of the vortices through non-uniform surface barriers. We show that Eq. (1) can describe the nucleation and growth of the vortex ordered state ($\Psi = \Psi_0$). To demonstrate this point we assume an induction distribution with a constant gradient $\ddot{B}/d$, i.e. $B = B_a - \dot{B}(1 - |x|/d)$, where $B_a$ is an applied field and $d$ is half-width of the sample. In this case, Eq. (1) can be solved analytically for both the nucleation and growth processes.
A solution for the nucleation process, i.e. the initial growth of the order parameter Ψ (Ψ close to zero), is obtained by neglecting nonlinear terms in Eq. (1):

$$\frac{1}{\Gamma} \frac{\partial \Psi}{\partial t} = D \frac{\partial^2 \Psi}{\partial x^2} + \left( \alpha_0 - \alpha_0 \frac{B_a - \tilde{B}}{B^*} - \alpha_0 \frac{\tilde{B}}{B^*} \frac{x}{d} \right) \Psi. \quad (4)$$

The boundary conditions dictated by symmetry is \(d\Psi(x, t)/dx|_{x=0} = 0\); also we require that \(\Psi(x, t)\) is a non-diverging function. The solution of Eq. (4) is then:

$$\Psi(x, t) = \sum_{n=0}^{\infty} A_n e^{\Lambda_n t} Ai\left(x/x_s - \varsigma_n\right), \quad (5)$$

where

$$\Lambda_n = \Gamma \alpha_0 \left[ 1 - \frac{B_a - \tilde{B}}{B^*} - \varsigma_n \left( \frac{a_D}{\mu} \right)^{1/3} \left( \frac{\tilde{B}}{B^*} \right)^{2/3} \right]. \quad (6)$$

\(Ai\) is the Airy function, \(\varsigma_n = 0.685, 3.9, 7.06, ...\) are the solutions of \(J_{2/3}(\varsigma_n) = J_{-2/3}(\varsigma_n)\) where \(J_\nu\) is the Bessel function, and \(x_s = \left(DdB^*/\alpha_0\tilde{B}\right)^{1/3} = d \left( a_D B^*/\mu\tilde{B} \right)^{1/3}\). Here \(a_D = D\gamma/\beta^2 d^2\) is a dimensionless exchange coefficient. Note that \(\varsigma_n\) is a constant of order \(n^3\), growing with increasing \(n\).

It is evident from Eq. (4) that only terms with \(\Lambda_n > 0\) play a role in the nucleation process. For \(B_a - \tilde{B} > B^*\), i.e. the entire sample is in a metastable or a stable state, all \(\Lambda_n\) are negative, implying that the nucleation process cannot take place. For \(B_a = \tilde{B}\) the induction at the center of the sample is zero and the rate of the nucleation process is maximum. Relation (6) shows that the exponent with \(n = 0\) yields the fastest nucleation rate, thus governing the nucleation process. This process may thus be approximately described by the first term in Eq. (5). In this approximation, the development of the order parameter during the nucleation process is described by the dashed lines in Fig. 1. Note that the analytical solution (5) describes only the first stages of the nucleation process, where the non-linear terms in Eq. (4) may be neglected. This solution ceases to apply when the value of \(\Psi\) approaches \(\Psi_0\), i.e. after a time period of order \(1/\Lambda_0\). The width of the ordered domain is then \(w \sim x_s (1 + \varsigma_0) \sim x_s\). The condition for appearance of localized domain in the sample center may be then obtained from the inequality \(x_s << d\) or:

$$\frac{\tilde{B}}{B^*} \gg \frac{a_D}{\mu}. \quad (7)$$

If this condition is not satisfied, then homogeneous transformation of the unstable phase takes place. Otherwise, a sharp front will develop separating between the nucleating ordered phase and the initial unstable disordered phase, as described above. Thus, when the induction gradient is large enough, we expect the appearance of a sharp interface between the growing stable (ordered) phase and the retreating unstable (disordered) phase.

In describing the growth process, i.e. the movement of the interface between the ordered and disordered phases, non linear terms in Eq. (4) must be taken into account. We express the
linearly varying function $B(x)$ as: $B = B_f + (\tilde{B}/d)(x-x_f)$, where $B_f = (B_a - \tilde{B}) + x_f \tilde{B}/d$ is the induction at the front located at $x_f$. In this notation: $\alpha = 1 - [B_f + (\tilde{B}/d)(x-x_f)]/B^*$. Eq. (1) can be written in the reference frame of an observer moving with the front, by introducing a new variable $\xi = x - x_f(t)$ and defining $x_f(t) \equiv x_0 + \int_0^t v_f(t')dt'$, where $v_f$ is the time-dependent front velocity and $x_0$ is a constant. With the new set of independent variables $(\xi, B_f)$ Eq. (1) becomes:

$$v_f \Gamma \left( -\frac{\partial \Psi}{\partial \xi} + \frac{\tilde{B}}{d} \frac{\partial \Psi}{\partial B_f} \right) = D \frac{\partial^2 \Psi}{\partial \xi^2} + \alpha_0 \left( 1 - \frac{B_f \{x_f(t)\}}{B^*} - \frac{\tilde{B} \xi}{dB^*} \right) \Psi + \beta \Psi^2 - \gamma \Psi^3.$$ (8)

One can solve this equation analytically provided the front width $\Delta << dB_f/\tilde{B}$. In this case, the terms $(v_f/\Gamma)(\tilde{B}/d)(\partial \Psi/\partial B_f)$ and $-\alpha_0(\tilde{B} \xi/dB^*)\Psi$, may be neglected. [12] The solution of Eq. (8) is then [13]:

$$\Psi = \Psi_0 \left\{ 1 + \exp \left( \frac{\xi}{\Delta} \right) \right\}^{-1},$$ (9)

where

$$\Delta^2 = \frac{\Delta_0^2}{2\mu(1-B_f/B^*) + 1 + \sqrt{1 + 4\mu(1-B_f/B^*)}}$$ (10)

is the front width. The front velocity $v_f = dx_f/dt$ is:

$$v_f = v_0 \frac{6\mu(1-B_f/B^*) + 1 + \sqrt{1 + 4\mu(1-B_f/B^*)}}{\sqrt{2\mu(1-B_f/B^*) + 1 + \sqrt{1 + 4\mu(1-B_f/B^*)}}}.$$ (11)

Here

$$v_0 = \Gamma \sqrt{D \beta^2/4\gamma} = \Gamma \alpha_0 d \sqrt{a_D}/2\mu, \quad \Delta_0^2 = 4D \gamma/\beta^2 = 4d^2 a_D,$$ (12)

and $B_f \equiv B_f \{x_f(t)\}$. The solid lines in Fig. 1 show $\Psi$ given in Eq. (9) for different locations of the front $x_f$, describing the propagation of the ordered phase.

We turn now to discuss the front velocity $v_f$, Eq. (11), and the front width $\Delta$, Eq. (10). We first note that $v_f$ and $\Delta$ do not depend explicitly on time or applied field but on $B_f$, the local induction at the front. Several important conclusions may be drawn from these equations:

1) As $B_f$ approaches $B_{od}$ the velocity is approaching zero (the $v_f(B_f)$ dependence is described by the solid line in Fig. 2).

2) The motion of the front toward the sample edge is accompanied by an increase of the induction $B_f$ at the front, resulting in a decrease of the velocity with time.

3) The front width $\Delta$ decreases with the increase of $\beta$, implying that for a 'stronger' first order transition the front is steeper. Also from Eq. (10) it is obvious that the exchange coefficient $D$ causes the front to be smeared. In addition, increasing $D$ and/or the damping coefficient $\Gamma$, results in an acceleration of the front motion (see Eq. (11)).
So far we have demonstrated dynamic coexistence of ordered and transient disordered vortex phases, with a sharp interface between them, assuming time-independent induction distribution with a constant gradient. In high-temperature superconductors, however, the induction distribution varies significantly with time due to flux creep. In addition, one may expect different flux creep laws for the different vortex phases. As a result, the nucleation and growth of the ordered vortex phase are manifested experimentally by the appearance of a break in the induction profile and movement of this break toward the sample edge. [3] The location of the break is expected to coincide with the location of the moving front of the order parameter.

To demonstrate this scenario we solved numerically the LK equation (1), allowing for flux creep. We assume a $\Psi$-dependent local current density

$$J(\Psi, t) = J_1(t) \left(1 - \frac{\Psi}{\Psi_0}\right) + J_2(t) \frac{\Psi}{\Psi_0},$$  \hspace{1cm} (13)$$

where $J_1(t)$ and $J_2(t)$ are the current densities in the disordered and in the ordered phases, respectively. [4] Based on the experimental observations [3] we further assume $J_1(t) = J_{01}(1 + t/t_1)^{-\alpha_1}$ and $J_2(t) = J_{02}(1 + t/t_2)^{-\alpha_2}$. The induction $B$ (and thus the coefficient $\alpha$) may now be expressed in terms of the order parameter $\Psi(x)$ by using Maxwell equation:

$$B(x, t) = B_a - 4\pi/c \int_{x}^{d} J(\Psi(y, t), t) dy.$$ 

As before, we assume an initial disordered phase throughout the entire sample, $\Psi(t = 0, x) = 0$.

In order to solve equation (13) numerically we define dimensionless parameters: $b = B/B^*$, $j = 4\pi J d/(cB^*)$, $x' = x/d$, $t' = t \beta^2 \Gamma/\gamma$, $\Psi' = \Psi/\Psi_0(B^{**}) = 2\Psi_1/\beta$. Eq. (13) then becomes:

$$\frac{\partial \Psi'}{\partial t'} = a_D \frac{d^2 \Psi'}{dx'^2} + \mu(1 - b(x'))\Psi' + \frac{1}{2} \Psi'^2 - \frac{1}{4} \Psi'^3 + f'(x', t'),$$ \hspace{1cm} (14)$$

where $f' = 2f/\gamma/(\beta\alpha_0)$ is a dimensionless noise which must be introduced in the numerical solution.

The values of the (dimensionless) parameters used in the numerical calculations are based on experimental measurements. In particular, from the fit of Eq. (14) to the experimental data [3] of $v_f(B_f)$ in Fig. 2 we estimate $\mu = 1.5$, thus $B_{od}/B^* = 1.148$, $B^{**}/B^* = 1.166$. A value of $10^{-4}$ for $a_D = (2\mu v_0/(\Gamma_0 d))^2$ (see Eq. (12)) is estimated from the experiment value $u_0 \approx 20 \mu m/s$ obtained from the same fit; a value of $\Lambda_0 \sim \Gamma_0 \approx 10 s^{-1}$, is estimated from the time elapse between switching-on of the external field and the appearance of a break in the induction profile. A typical value of $d \approx 300 \mu m$ for the sample half width was used. Based on the analysis of magnetic relaxation we take $\alpha_1 = 0.3$, $\alpha_2 = 0.5$. In addition, a noise level of $f'_{\text{max}} = 10^{-4}$ is assumed. The system of equations completed by boundary and initial conditions has been solved numerically utilizing the Euler method. The unit space interval was divided into 200 segments, and time step of $2.5 \cdot 10^{-3}$ (in dimensionless units) was used providing a stability of the numerical procedure. The results for $B_a/B^* = 1.1$ are shown in Fig. 3. The upper case shows the spatial dependence of the order parameter at different times. The nucleation appears at the sample center at $t' \sim 10$, forming a sharp front which propagates toward the sample edge. [15] The lower case of Fig. 3 shows the time evolution of the induction profiles during the nucleation and growth processes. A sharp break in the profiles appears at the location of the front of the order parameter at the
nucleation is completed. As expected, the break in the induction profile and the front of the order parameter move together toward the sample edge. Note that a break in the induction profile can be observed outside the region of phase metastability (i.e., for $B_f < B^*$).

The theoretical predictions described above are confirmed experimentally in BSCCO crystals. In particular, breaks in the induction profiles were recorded following a sudden application of external field of intensity close to $B_{od}$. This break moves toward the sample edge at a velocity which depends only on $B_f$, the value of the induction at the break. Thus, the dependence of $v_f$ on $B_f$ is not affected by magnetic relaxation. As shown in Fig. 2, the analytical curve (solid line) of $v_f(B_f)$, Eq. (11), is in good agreement with the experimental results (open symbols). Moreover, numerical results (solid symbols in Fig. 2) for $v_f(B_f)$ for different applied fields also show a good agreement with the analytical curve demonstrating that magnetic relaxation does not affect the dependence of $v_f$ on $B_f$.

Finally, we note that two velocities govern the vortex dynamics in the process of the phase transformation: The interface velocity $v_f$, and the flux velocity, $v_F$, due to creep. The effect of the latter on the shape of the interface must be taken into account in close vicinity of $B_{od}$ where $v_f \rightarrow 0$.

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[14] A more general approach to this problem should be based on introduction of different $E(j)$ characteristics for the different phases and a solution of appropriate Maxwell equations. We solved our system using this approach too, and obtained similar results.

[15] The numerical solution does not exhibit breaks in the profiles if the magnetic relaxation is too fast (e.g., $J \sim \exp(-t/t_0)$ while $t_0 < < 1/\Lambda_0$) or if the critical current is too low ($j_{01} < j_{02} \lesssim a_D/\mu$). This is because the requirement for a relatively large gradient, mandatory for the front development (Eq. 7), is not satisfied in these cases. Thus, in $YBa_2Cu_3O_{7-\delta}$, for which $\tilde{B} < < B_{c1} \sim B^*$, an homogeneous phase transformation is expected rather than front propagation.
Figure captions

Figure 1. Nucleation and growth of the order parameter. The nucleation process is demonstrated by the dashed curves calculated from Eq. (5) for \( A_n = A_0 \delta_{n,0} \) at times \( \Lambda_0 t = 8.12, 9.86, 11.02 \). The solid lines, describing the growth process, are calculated from Eq. (6) at different locations \( x_f/d = 0.3, 0.5, 0.7, 0.9 \).

Figure 2. Experimental data (open symbols) of \( v_f(B_f) \) for different applied fields, taken from Ref. [3], a fit (solid line) to Eq. (11) and results of numerical solutions (solid symbols) for the indicated applied fields. The fit was done with two fitting parameters \( v_0 \) and \( \mu \), after \( B_{od} \) was estimated to be approximately 400 G, corresponding to an induction value where the velocity is going to zero.

Figure 3. Order parameter (upper case) and induction profiles (lower case) for \( j_1 = 1.7(1 + t')^{-0.3} \) and \( j_2 = 1.5(1 + t')^{-0.5} \). The profiles are shown for dimensionless times \( t' = 1, 4, 9, 10, 11, 12, 14, 16, 18, 22, 27, 35, 44, 54, 65, 79, 99 \).
Giller et al.  Fig. 1
Fig. 2: Theoretical fit of the data from Giller et al. showing the relationship between $v_f$ [µm/s] and $B_f$ [G] for different $B_a/B^*$ values, with $B_a$ being in units of [G].
Giller et al.  Fig. 3