Reversible Magnetization of Irradiated High-T_c Superconductors

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We calculate the free energy and equilibrium magnetization in highly anisotropic layered superconductors with strong defects produced by irradiation. We account for the entropy associated with different configurations of pancakes inside and outside of strong defects. We show how magnetization measurements provide information on pinning energy and how they determine the magnetic fields and temperatures at which pancake vortices are trapped inside strong defects. We discuss also the behavior of magnetization which may signal about decoupling of pancakes inside columnar defects. [S0031-9007(96)00692-8]

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A quest to understand the nature of the vortex state in highly anisotropic layered materials is one of the most topical issues in the current study of high temperature superconductors (HTS). It was proposed that because of the weak coupling Abrikosov vortices break up forming a set of weakly coupled quasi-two-dimensional lattice of pancakelike vortices [1,2]. The thermodynamics of the pancake vortices was first discussed by Glazman and Koshelev [3] who argued the existence of the specific decoupling temperature T_d in the system of Josephson-coupled pancakes, separating a low-temperature domain where pancakes form usual vortex lines from the high-temperature decoupled state, where the coherence in the positions of pancakes along the c direction is lost. Recent neutron structure measurement [4] and measurements of Josephson plasma resonance [5] suggested that pancakes are positioned randomly along the c axis (do not form lines) at all temperatures in fields above 0.05 T. For samples with columnar defects the question is whether such defects help to form lines and push T_d above the irreversibility line. Moreover, although the concept of pancakes was first introduced to describe properties of highly anisotropic Bi-2:2:1:2 compound, recent transport measurements using the dc flux transformer configuration questioned a linear nature of vortices even in less anisotropic Y-Ba-Cu-O crystals [6].

Recently, magnetization and transport measurements on the crystals containing columnar defects were used to clarify the nature of the vortex state in anisotropic HTS. The angular dependence of resistivity and flux transformer experiments in samples with columnar defects evidenced that vortices move as linear objects [7–9]. These results are consistent with the theoretical arguments that columnar defects effectively enhance coupling between layers and correlations along the c direction [10].

It is important, however, to compare the results on nonequilibrium properties with the observation of the thermodynamic properties of highly anisotropic HTS. The strongest support for the discrete nature of the pancake vortices in layered compounds had come from the observations of the so-called crossing point in the temperature dependence of the magnetization at different magnetic fields applied along the c axis: all these curves cross at some temperature T^*. At this point magnetization becomes field independent because the logarithmic field dependence of the mean field magnetization is canceled by the same logarithmic dependence in the entropy contribution of pancakes decoupled along the c axis [11–13]. Recently, the reversible magnetization measurements were performed in crystals with columnar defects for Bi-2:2:1:2 by van der Beek *et al.* [14] and for c-axis oriented Bi-2:2:2:3 tapes by Qiang Li *et al.* [15]. The crossing point behavior was found in these systems in magnetic fields below and above the matching field, but not near the matching field. It is also observed that near the matching field, magnetization exhibits an anomalous dip corresponding to the expulsion of vortices from the sample.

The authors of Refs. [14,15] described the experimental data for reversible magnetization omitting entropy contribution to the free energy. In this Letter we investigate the behavior of the reversible magnetization in layered HTS accounting for entropy associated with different configurations of pancakes. We assume that the interaction of the pancakes is very weak in the temperature interval under consideration and that they can be treated as uncorrelated along the c axis. The model we adapt is complimentary to that explored in Ref. [16], where vortex *lines* were found to form a low temperature Bose glass phase with vortex lines localized near columnar defects. In our model of uncorrelated pancakes we calculate the free energy of the system of pancakes in the London regime, $H_{c1} \ll B \ll H_{c2}$, in the presence of columnar, splayed, or pointlike strong defects produced by irradiation. We show that well below and above matching field entropy contribution leads to the crossing point behavior of magnetization but near the matching field the number of the available states inside strong defects produced by irradiation is restricted, pancake entropy contribution is suppressed, and the crossing point disappears. We show

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also how information on pancake arrangement may be obtained from magnetization measurements.

To construct our model we accept that only one pancake can be trapped by the strong pointlike pinning site corresponding to either the strong point defect or the intersection of the heavy ion track with the given layer. This description follows immediately from the results of Ref. [17] that the pinning energy of a vortex inside a hollow cylindrical channel, which models defects produced by irradiation, drops dramatically for a number of flux quanta bigger than one. The concentration of sites available for pancakes is then $B_\phi/\Phi_0 s$, where B_ϕ is the matching field ($B_\phi \ll H_{c2}$) and s is the interlayer spacing. The energy of a pancake trapped inside a defect we denote by $-\epsilon_p$, assuming that all defects are identical. We measure this energy relative to that of a pancake situated far outside of the defect. The pinning energy $\epsilon_p \approx \epsilon_0$, where $\epsilon_0 = \Phi_0^2 s/16\pi^2\lambda_{ab}^2$; see Refs. [16,17]. Pancakes may also occupy positions outside defects (free pancakes). The number of states for free pancakes per unit volume we denote by $B_f/\Phi_0 s$, where B_f is of the order H_{c2} as was supposed in Ref. [12] and calculated as $B_f = H_{c2}(T)[\epsilon_0(T)/2T]$ by Koshelev [18]. Again, each such state may be occupied by a single vortex only. The energy of pancakes in such states we take as zero, ignoring the effect of weak pointlike defects in comparison with those produced by irradiation.

The entropy corresponding to a distribution of pancakes between defects and sites outside of defects is determined by the occupation numbers n_t and n_f, respectively. The entropy per unit volume is given by an expression which corresponds to the statistics of fermions because each state may be occupied by a single vortex only,

$$S(n_t, n_f) = -(B_\phi/\Phi_0 s)[n_t \ln n_t + (1 - n_t)\ln(1 - n_t)] - (B_f/\Phi_0 s)[n_f \ln n_f + (1 - n_f)\ln(1 - n_f)]. \tag{1}$$

The occupation numbers n_t and n_f satisfy the condition

$$B_\phi n_t + B_f n_f = B. \tag{2}$$

For pancake interaction we use the standard mean field energy in the London regime

$$\mathcal{F}_{\rm int}(B) = (\epsilon_0 B/2\Phi_0 s)\ln(\eta H_{c2}/B), \tag{3}$$

where η is a numerical parameter of the order unity. In this approximation energy depends on the concentration but not on the arrangement of pancakes. Thus, we ignore the Josephson interaction of pancakes as well as the correlation part of magnetic pancake-pancake interaction. Later we will discuss the errors caused by this approximation.

The total free energy functional of the system is

$$\mathcal{F}(n_t, n_f) = \mathcal{F}_{\rm int}(B) - (B_\phi/\Phi_0 s)\epsilon_p n_t - TS(n_t, n_f). \tag{4}$$

The equilibrium values of n_t and n_f are determined by minimization of the functional (4) with respect to n_t and n_f under the condition (2). Thus we minimize

$$\Phi(B, n_t, n_f, \mu) = \mathcal{F}(n_t, n_f) - (\mu/\Phi_0 s) \times (B_\phi n_t + B_f n_f - B) \tag{5}$$

with respect to n_t and n_f and then find μ using Eq. (2). The equilibrium occupation numbers are given by the Fermi distribution function,

$$n_f = \{\exp[(\epsilon_m - \mu)/T) + 1\}^{-1}, \tag{6}$$

$$n_t = \{\exp[(-\epsilon_p + \epsilon_m - \mu)/T] + 1\}^{-1}, \tag{7}$$

where $\epsilon_m = (\epsilon_0/2)\ln(\eta H_{c2}/B)$ is the mean field magnetic energy of pancake. The chemical potential is determined by Eq. (2) with n_t, n_f given by Eqs. (6) and (7). We obtain

$$\mu = \epsilon_m - T\ln[hb^{-1}(u + \sqrt{u^2 + pb})], \tag{8}$$

$$u = [1 + (1 - b)p]/2, \qquad p = h^{-1}\exp(\epsilon_p/T). \tag{9}$$

Here $b = B/B_\phi$ and $h = B_f/B_\phi$ are the ratios of sites available outside of defects to those inside. Magnetization is given by the relation

$$M = -\partial\Phi/\partial B = -\mu/\Phi_0 s\,, \tag{10}$$

because it characterizes the change of the free energy with respect to magnetic field (concentration of pancakes). The first term in the right-hand side of Eq. (8) leads to the usual mean field magnetization originating from vortex repulsion, while the second term accounts for pinning and fluctuations in pancake positions. This term results in a positive contribution to magnetization because both pinning and entropy favor the creation of vortices.

The pinning ability of defects produced by irradiation is determined by the ratio of trapped and free vortices,

$$A(b, p) = \frac{B_\phi n_t}{B_f n_f} = \frac{(u + \sqrt{u^2 + pb})p}{(u + \sqrt{u^2 + pb}) + pb}, \tag{11}$$

where small term b/h is omitted. We get $A \approx p$ at $p \gg 1$ and for $b \ll 1$, while $A \approx \sqrt{p}$ for $b = 1$ and A vanishes as $1/(b - 1)$ at $b \gg 1$. Pinning is effective if $p \gg 1$. We can estimate h to be in the interval 20–80 depending on B_ϕ and temperature. Thus, pinning is effective at $B \lesssim B_\phi$ if the pinning energy $\epsilon_p(T) \gtrsim 4T$.

Let us discuss the condition under which we can ignore the Josephson interaction in the case of columnar or splayed defects. The energy of Josephson coupling per unit volume lost if pancakes are sitting randomly (do not form lines along defects) is $\Delta F_J = \epsilon_0/2\pi^2\lambda_J^2 s$, where $\lambda_J = \gamma s$ is the Josephson length and γ is the anisotropy ratio. The gain in the free energy due to the increase of entropy for random distribution at $b \ll 1$ is $\Delta F_S \approx (TB/\Phi_0 s)\ln(e/b)$. We estimate the ratio $\Delta F_J/\Delta F_S \approx (\epsilon_0/2\pi^2 T)(\Phi_0/\lambda_J^2 B)/\ln(e/b) < 0.1$ at $T = 20$ K, $B > 0.1$ T, and $b = 0.1$ for Bi-2:2:1:2 with $\gamma \approx 300$, $\lambda_{ab} = 1700$ Å, and $s = 15.6$ Å. Thus, we can ignore the Josephson coupling of pancakes in a quite broad temperature interval when calculating equilibrium magnetization. It is more difficult to estimate accurately the magnetic correlation energy of pancakes, i.e., to go beyond the mean field approach leading to Eq. (3). From our estimate of the Josephson interaction one can assume that the magnetic interaction may be responsible for the formation of lines inside columnar or splayed defects if such a transition occurs.

For unirradiated samples ($\epsilon_p = 0$) we obtain the result

$$M_{\mathrm{un}} = -\frac{\epsilon_0}{2\Phi_0 s}\ln\frac{\eta H_{c2}}{eB_\phi b} + \frac{T}{\Phi_0 s}\ln\frac{h}{b} \qquad (12)$$

found previously in Ref. [12]. This leads to a field independent slope of the magnetization curve vs $\ln B$,

$$\partial M_{\mathrm{un}}/\partial \ln B = [(1/2)\epsilon_0(T) - T]/\Phi_0 s\,. \qquad (13)$$

M_{un} becomes field independent at the temperature $T = T^*_{\mathrm{un}}$ determined by the condition $\epsilon_0(T) = 2T$. This leads to the crossing of all curves $M_{\mathrm{un}}(T)$ for different B at the same temperature $T = T^*_{\mathrm{un}}$. Magnetization at this crossing point is $M^*_{\mathrm{un}} = T^*_{\mathrm{un}}/\Phi_0 s$; see Ref. [13]. The factor $\Phi_0 s$ in Eq. (13) and in the following may be expressed via the experimentally obtained value $T^*_{\mathrm{un}}/M^*_{\mathrm{un}}$. We obtain $T^*_{\mathrm{un}} = T_{c0}[1 - 2T_{c0}/\epsilon_0(0)]$ because of the relation $\epsilon_0(T) \propto \lambda_{ab}^{-2}(T) = \lambda_{ab}^{-2}(0)\,(1 - T/T_{c0})$, where T_{c0} is the mean field critical temperature.

Next we discuss the behavior of magnetization in irradiated samples, $M_{\mathrm{ir}}(B,T)$, determined by Eq. (10).

In the low-field regime, $B \ll B_\phi$, we get

$$M_{\mathrm{ir}} = -\frac{\epsilon_0}{2\Phi_0 s}\ln\frac{\eta H_{c2}}{eB_\phi b} + \frac{T}{\Phi_0 s}\ln\frac{h(p+1)}{b}\,. \qquad (14)$$

The slope $\partial M_{\mathrm{ir}}/\partial \ln B$ is again determined by the right-hand side of Eq. (13), though the value $\epsilon_0(T)$ may be different, because irradiation suppresses T_{c0} and the density of superconducting electrons which determines $\lambda_{ab}^{-2}(0)$. There is again the crossing point in the low-field regime at $T = T^*_{\mathrm{ir}}$. The difference

$$\Delta T^* = T^*_{\mathrm{ir}} - T^*_{\mathrm{un}} = \Delta T_{c0} - [2T_{c0}^2/\epsilon^2(0)]\Delta\epsilon_0(0) \quad (15)$$

may be positive or negative depending on what effect is stronger: suppression of T_{c0} or $\lambda_{ab}^{-2}(0)$.

The important point is that the entropy contribution to the free energy is significant and leads to the crossing point if almost all possible configurations of pancakes inside defects contribute to the entropy. If pancakes form straight lines inside columnar or splayed defects, the entropy is not proportional to the thickness of the sample and may be neglected. In this case magnetization is given by Eq. (10) with $\mu = \epsilon_p$.

The observation of crossing point [14,15] at low fields means that at $T = T^*$ pancakes are positioned randomly in irradiated Bi-2:2:1:2 and Bi-2:2:2:3. In principle, they can align at some lower temperature $T_d(B)$. Such a transition driven by competition between magnetic correlation energy and entropy may result in the change of magnetization, ΔM, of the order $M^*_{\mathrm{ir}}T_d/T^*_{ir}$, though a numerical coefficient may be small. It may result also in the change of slope in the field dependence of magnetization, $\Delta(\partial M_{\mathrm{ir}}/\partial \ln B) \approx T_d/\Phi_0 s$, at $B = B_d$. Anomalies of this type were not observed in Refs. [14,15]. If observed, they would signal the thermodynamic coupling transition in samples with columnar or splayed defects produced by irradiation. Recently, a sharp dynamic coupling transition was observed in flux transformer transport measurements in Bi-2:2:1:2 with columnar defects by Seow *et al.* [9], and the question is if this transition will be seen in reversible magnetization measurements.

Near the field B_ϕ, our model implies that a transition occurs between a state at low fields where pancakes are mobile but randomly distributed among the columnar sites, and one at high fields where they sit predominantly in regions between the columnar sites. Near B_ϕ, the entropy associated with distribution of pancakes among the defects is reduced by the approach of full occupancy. Here the entropy gain that can be obtained from the occupancy of the interdefect regions drives this transition. To show interplay between pinning and entropy let us consider the result used in Refs. [14,15] without taking entropy into consideration, $M_{\mathrm{ir}} = (\epsilon_p - \epsilon_m)/\Phi_0 s$ at $B < B_\phi$ and $M_{\mathrm{ir}} = -\epsilon_m/\Phi_0 s$ at $B > B_\phi$. In this approach $\partial M_{\mathrm{ir}}/\partial \ln B$ is positive independent of temperature, and defects result in the jump of magnetization, $\epsilon_p/\Phi_0 s$, at $B = B_\phi$. Entropy contribution leads to a sign change of $\partial M_{\mathrm{ir}}/\partial \ln B$ at $T = T^*$ and smooths the jump.

In the high-field regime, $B_\phi \ll B \ll H_{c2}$, we obtain from Eq. (10) the same result as for the unirradiated sample because the main part of pancakes sit outside of strong defects. However, this regime may be achieved only at low enough temperatures because $H_{c2}(T)$ drops with temperature while B_ϕ remains constant. Near T_c in strong fields the lowest Landau level approximation may be used [13], but such an approach was not modified yet to account for strong pinning centers.

The simplest procedure to obtain the parameter p from experimental data is to fit the difference between M_{ir} and magnetization $\tilde{M}_{\mathrm{ir}}$ which is a linear extrapolation of M_{ir} vs $\ln b$, Eq. (14), from region $b \ll 1$ to higher fields,

$$\frac{M_{\mathrm{ir}} - \tilde{M}_{ir}}{M^*_{\mathrm{ir}}}\frac{T^*_{\mathrm{ir}}}{T} = f(b,p) = \ln\frac{u + \sqrt{u^2 + pb}}{1 + p}\,, \quad (16)$$

where u is given by Eq. (9).

We note that our model is oversimplified in two main aspects. First, it assumes all pinning centers to be identical, while in reality there is distribution of pinning energy due to variation in defect size. It leads to the smoothing of change in magnetization near B_ϕ. Second, the model does not take into account the correlation energy of pancake interaction. It is unclear now how this effect may change the field dependence of magnetization.

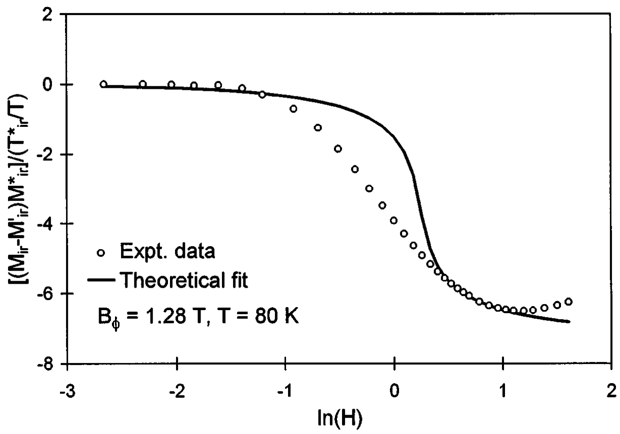

FIG. 1. The function $f(b)$ given by Eq. (16) at $p = 1220$ and experimental data for the left-hand side of Eq. (16) in Bi-2:2:2:3 tapes at 80 K and matching field 1.28 T; see text.

The function $f(b)$ at $p = 1200$ and experimental data of Qiang Li *et al.* [15] for $(M_{\mathrm{ir}} - \tilde{M}_{\mathrm{ir}})T_{\mathrm{ir}}^*/M_{\mathrm{ir}}^*T$ in Bi-2:2:2:3 at $T = 80$ K and $B_\phi = 1.28$ T are shown in Fig. 1. Respectively, the dependence of $M_{\mathrm{ir}}(b)$ is shown in Fig. 2. The shift in the drop of experimentally found magnetization to lower fields in comparison with theoretical prediction seems to be caused by the distribution of pinning energy. Taking $h \approx 30$ we estimate $\epsilon_p \approx 1000$ K at $T = 80$ K which is in rough agreement with estimate $\epsilon_p \approx \epsilon_0$. For the Bi-2:2:1:2 sample with $B_\phi = 1$ T studied by van der Beek *et al.* [14] we estimate $p \approx 10$ at $T = 74$ K.

In conclusion, we propose the model which allows us to estimate quantitatively the effect of strong pinning centers produced by irradiation on the thermodynamics of the vortex state. We show how the parameter p describing trapping ability of pinning centers may be obtained from magnetization measurements. We argue that experimental results [14,15] for reversible magnetization in irradiated samples of Bi-2:2:1:2 and Bi-2:2:2:3 show that pancake vortices at $B_\phi = 1$ T do not form lines at temperatures above 70 K and magnetic fields above 0.1 T. We propose to observe the aligning of pancakes above irreversibility (if it occurs) by magnetization measurements.

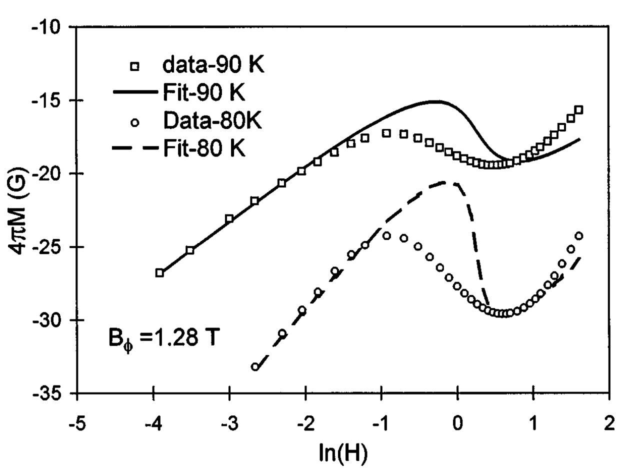

FIG. 2. Field dependence of reversible magnetization in irradiated superconductor: theoretical curves, Eq. (10), with $p = 765$ and 1220 and experimental data for Bi-2:2:2:3 tape at 90 K and 80 K at matching field 1.28 T; see text.

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