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Effect of quantum fluctuations of vortices on reversible magnetization in high- T_c superconductors

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The field (B) and temperature dependence of reversible magnetization M in Bi (2:2:1:2) single crystals is measured for the orientation of the field along the c axis. The slope of the curve M vs lnB decreases remarkably with field below 70 K. In the mean-field approach this slope should be almost B independent. We explain this change of slope by the quantum fluctuations of vortices. We show that for magnetization quantum fluctuations are important at all temperatures except in a narrow region near T_c .

The effect of thermal vortex fluctuations on the reversible magnetization in highly anisotropic Bi- and TI-based high T_c superconductors was studied previously both experimentally and theoretically. The fluctuations of pancake vortices contribute to the free energy via an entropy term and result in the formation of a crossing point: all M(T) curves for different fields $H_{c1} \ll B \ll H_{c2}$ cross at a point $T = T^*$, which lies close to T_c , and $M(T^*) = T^*/\Phi_0 s$, where s is the interlayer spacing.¹⁻³ This behavior is quite different from that predicted by the mean-field theory of type-II superconductors: in such an approach the absolute value of diamagnetic moment is a decreasing function of the field at $T < T_c$.

Recently Li and Suenaga⁴ found that in the highly anisotropic Bi (2:2:1:2) superconductor the low-temperature behavior of reversible magnetization also deviates from the predictions of the standard (mean-field) London model: the slope of the curves M vs lnB is a decreasing function of the field at all temperatures below 70 K where reversible magnetization can be observed; the decrease as high as 50% was found as the field increases from 0.3 to 7 T. The mean-field London approach predicts this slope to be field independent. The elaborated version of the mean-field approach, which takes into account overlapping of the normal cores, predicts a change of slope in the fields of the order H_{c2} ,⁵ but the scale of fields where changes were observed is by an order of magnitude smaller.

In the following we present experimental data for the reversible magnetization in two samples of Bi (2:2:1:2), one was studied at BNL (the same data as in Ref. 4) and another was studied at LANL. We present also the results for resistivity measurements in conditions close to the flux-flow regime for Bi (2:2:1:2). These data provide information on the viscosity coefficient which characterizes the dynamics of vortices. We explain the observed change of slope in M vs lnB by quantum fluctuations of vortices, i.e., by the contribution of zero point fluctuations of vortices to the free energy of the vortex lattice. The contribution of quantum fluctuations to the free energy depends on magnetic field because frequencies of vortex fluctuations increase with field. Quantum fluctuations depend on the dynamics of the vortex lattice. To describe this dynamics we used the model of overdamped oscillators with viscosity coefficient η and then compare η found from the fit of magnetization with that taken from flux-flow resistivity data.

Note that macroscopic quantum tunneling of vortices was studied previously; it causes the relaxation of nonequilibrium magnetization at very low temperatures.^{6–8} Recently Blatter and Ivlev discussed a possible effect of quantum fluctuations of vortices on the melting line;⁹ the specific heat of the mixed state in type-II layered superconductors was calculated taking into account quantum fluctuations of vortices.¹⁰

The experiments reported here were performed on two Bi-Sr-Ca-Cu-O single crystals grown by the travelingsolvent floating-zone technique, as described by Menken¹¹ and Shigaki et al.,¹² and will be referred to as crystals A and B. Crystal A was a square of dimensions $4 \times 4 \times 0.11$ mm³, weighed 11.4 mg, and had a T_c of 88.1 K, with a transition width of 1.5 K as determined by ac susceptibility. High-resolution electron microscopy performed on representative samples from the same batch confirmed the crystal purity, as only minor traces of the 2:2:0:1 phase could be detected in a concentration of 2 out of 1000 CuO₂ double layers. The composition of crystal A was determined by microprobe analysis to be $Bi_{2.2}Sr_{1.9}CaCu_2O_x$. Crystal B had dimensions $4.1 \times 4.3 \times 0.64 \text{ mm}^3$ and had the composition $Bi_{2.14}Sr_{1.39}Ca_{1.13}Cu_{2.00}O_{7.83}$. Its T_c , as determined by dc magnetometry in a field of 2 Oe, was 84.2 K with a width of 1.5 K. Detailed chemical, crystallographic, and magnetic characterization of crystal B has been published by Shigaki et al.¹²

3507

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FIG. 1. The in-plane resistivity vs magnetic field H || c at different temperatures and high current density 1460 A/cm². The flux-flow regime (resistivity is linear in field) is achieved at T = 70 in fields above 1 T.

Magnetization measurements were performed on superconducting quantum interference device (SQUID) magnetometers (Quantum Design) between the irreversibility temperature and 200 K with the magnetic field aligned along the crystallographic c axis. The measurements were performed both with fixed temperatures and varying magnetic fields and with fixed fields and varying temperatures. The temperature and field dependence of the magnetization of the sample platform and holder were measured and subtracted. The reversible regime in magnetization was achieved at temperatures above 35 K for the fields in the interval 0.3 T–7 T.

To obtain flux-flow viscosity, we measured the magnetic field dependence $(B||c, 0 \le B \le 7 \text{ T})$ of the in-plane resistivity of a Bi (2:2:1:2) single crystal ($\sim 0.03 \times 0.1 \times 0.023$ cm³). The contacts covered two sides of the crystal for a uniform in-plane current and the current density was ~ 1460 A/cm². As shown in Fig. 1, we were close to the flux-flow regime at T=70 K only in fields above 1 T. The measured resistivity provides only the lower limit for flux-flow resistivity $\rho_{\rm ff}$ because pinning may still be present. Therefore, using the expression $\rho_{\rm ff}=B\Phi_0/c^2\eta$, we extract the upper limit for the viscosity coefficient, $\eta \le 2.3 \times 10^{-7}$ g/cm s at T=70 K.

Measurements of *I-V* characteristics of a Bi (2:2:1:2) single crystal were performed also by Van der Beck, Hagen, and Samoilov¹³ at temperature 58 K in the field B = 1 T and current densities up to 3000 A/cm². They provide a lower limit for flux-flow resistivity $\rho_{\rm ff} \gtrsim 1.66 \times 10^{-5} \Omega$ cm, which determines the upper limit for viscosity, $\eta \lesssim 1.2 \times 10^{-7}$ g/cm s.

These values are about an order of magnitude smaller than the Bardeen-Stephen viscosity $\eta = \Phi_0^2/2\pi \xi_{ab}^2 c^2 \rho_n$, estimated by use of normal state resistivity $\rho_n \approx (4-4.5) \times 10^{-5} \Omega$ cm (as extrapolated from a temperature interval above T_c), and zero temperature correlation length $\xi_{ab}(0) = 20$ Å. The obtained values of η , much smaller than Bardeen-Stephen viscosity, may be explained by the twodimensional (2D) nature of pancake vortices and by the very small region with size $\sim \xi_{ab}$ occupied by the 2D vortex core. In such a region electron scattering is almost absent and we are in fact in the clean limit. (Mean-free electron path l is larger than the size of this region and such average values as l and electron scattering time become meaningless as applied to such 2D regions).

We obtain the contribution of vortex fluctuations to the free energy of the mixed phase in the highly anisotropic, Josephson-coupled layered superconductors using the model of the damped quantum oscillator^{8,9,14} to treat the dynamics of two-dimensional pancake vortices. Note that dynamics of vortex lattice displacements is important to obtain the free energy of the system in the quantum regime in contrast to the classical regime where all the dynamics drops out. We consider a system in field *B* along the *c* axis, assuming $H_{c1} \ll B \ll H_{c2}$.

We start from the Langevin classical equation to describe the vortex displacements $\mathbf{u} = (u_x, u_y)$ (in *ab*-plane):

$$\eta s \dot{\mathbf{u}}(n,\nu) + \hat{\boldsymbol{\epsilon}} \mathbf{u}(n,\nu) = \mathbf{f},\tag{1}$$

$$f_x(t)f_x(t')\rangle = \langle f_y(t)f_y(t')\rangle = 2\eta s T \delta(t-t'), \qquad (2)$$

where n, ν label pancake vortices with coordinates $\mathbf{r}_{n\nu} = (x_{n\nu}, y_{n\nu})$ in the layer *n*, we denote by η the viscosity coefficient, $\hat{\epsilon}$ is the elastic tensor, and **f** is a random force with correlation functions given by Eq. (2). We neglect pinning because its effect seems to be weak, see discussion below. We neglect inertial and Hall drag terms assuming that they are less important than the viscous term.

In the Fourier representation

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$$\mathbf{u}(\mathbf{k},q) = \frac{\Phi_0}{B} \sum_{n,\nu} \mathbf{u}(n,\nu) \exp(i\mathbf{k} \cdot \mathbf{r}_{n\nu} + iqn)$$
(3)

the elastic tensor becomes diagonal for transversal (t) and longitudinal (l) components of the vortex displacements:

$$\epsilon_l(k,q) = c_{11}k^2 + c_{44}Q^2, \qquad (4)$$

$$\epsilon_t(k,q) = c_{66}k^2 + c_{44}Q^2. \tag{5}$$

Here c_{66} , c_{11} , and c_{44} are the flux lattice shear, compression, and tilt moduli. $Q^2 = 2(1 - \cos q)/s^2$. In the limit of high anisotropy, $\gamma \ge \lambda_{ab}/s$, they are given by¹⁵

$$c_{66} = \frac{\Phi_0^2}{(8\pi\lambda_{ab})^2}, \qquad c_{11} = \frac{B\Phi_0}{4\pi\lambda_{ab}^2k^2}, \qquad (6)$$

per unit length of vortex, λ_{ab} is the penetration length. The term with $c_{44} \propto 1/\gamma^2$ can be neglected in the following calculations because $c_{44}Q^2 \ll c_{11}K_0^2$, $c_{66}K_0^2$, where $K_0^2 = 4\pi B/\Phi_0$. Momenta **k** and *q* are restricted to the first Brillouin zone: $|\mathbf{k}| \ll K_0$ and $|q| \ll \pi$.

The free energy density, F(T,B), of the vortex lattice is the sum of the mean-field result and the contribution from vortex fluctuations F_n :

$$F(T,B) = \frac{\Phi_0 B}{32 \pi^2 \lambda_{ab}^2} \ln \frac{\beta H_{c2}}{B} + F_v(T,B),$$
(7)

where we use the London result for the mean-field contribution. β is a numerical coefficient of the order unity. The contribution F_v of vortex lattice excitations has to be obtained using the action, S, for a damped oscillator⁸ corresponding to the classical Eq. (1): EFFECT OF QUANTUM FLUCTUATIONS OF VORTICES ON ...

$$S = \sum_{i=t,l} \sum_{\mathbf{k},q} S\{\mathbf{u}(i,\mathbf{k},q)\},\tag{8}$$

$$\mathbf{u}_m = \sum_m \mathbf{u}(\tau) \exp(-i\omega_m \tau),$$

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$$S\{\mathbf{u}(i,\mathbf{k},q)\} = \frac{nsB^{-}}{2T\Phi_{0}^{2}} \sum_{m} [\eta(\omega_{m})|\omega_{m}| + \epsilon_{i}(\mathbf{k},q)]|u_{m}(i,\mathbf{k},q)|^{2}, \qquad (9)$$

where $\omega_m = 2 \pi m T/\hbar$ are Matsubara frequencies. In the action we use the frequency-dependent viscosity $\eta(\omega_m) = \eta_0 \alpha(\omega_m)$, where $\alpha(\omega_m) = [1 + (|\omega_m|/\Omega)^n]$ and *n* is an integer, to introduce a smooth cut-off frequency Ω for summation over ω_m (see Ref. 8 for detailed discussion on the meaning of this cut-off). One can argue that Ω should be determined by the energy gap, $\Omega \sim \Delta(T)/\hbar$, because oscillations of a vortex with higher frequencies result in effective excitation of quasiparticles, and the vortex as an object becomes meaningless.

Using Eq. (9) we obtain the following relations for the free energy F_v :¹⁶

$$F_{v}(T,B) = \frac{1}{2s} \sum_{i} \int \frac{d\mathbf{k}}{(2\pi)^{2}} \int \frac{dq}{2\pi} F_{i}(\mathbf{k},q), \qquad (10)$$

$$\frac{2\Phi_0^2}{B^2s}\frac{\partial F_i(\mathbf{k},q)}{\partial \epsilon_i(\mathbf{k},q)} = \langle |\mathbf{u}(i,\mathbf{k},q)|^2 \rangle = \sum_m \frac{\int \mathscr{D}\mathbf{u}(i,\mathbf{k},q) |u_m(i,\mathbf{k},q)|^2 \exp[-S\{\mathbf{u}(i,\mathbf{k},q)\}/\hbar]}{\int \mathscr{D}\mathbf{u}(i,\mathbf{k},q) \exp[-S\{\mathbf{u}(i,\mathbf{k},q)\}/\hbar]} = \frac{T\Phi_0^2}{sB^2} \sum_m \frac{1}{\eta(\omega_m)|\omega_m| + \epsilon_i(\mathbf{k},q)} \,.$$
(11)

Using Eqs. (7), (10), (11) we get

$$\frac{dM}{d \ln B} = \frac{\Phi_0}{32\pi^2 \lambda_{ab}^2} - \frac{T}{2s\Phi_0} \sum_m \left\{ \frac{\omega_c [\omega_c + 2|\omega_m|\alpha(\omega_m)]}{[\omega_c + |\omega_m|\alpha(\omega_m)]^2} + \frac{\omega_s}{\omega_s + |\omega_m|\alpha(\omega_m)} \right\}.$$
(12)

Here we introduce the characteristic frequencies in the problem: the maximum frequency of the shear excitations, $\omega_s = B\Phi_0/16\pi\lambda_{ab}^2\eta_0$, and that of compression excitations (at $k=K_0$), $\omega_c \approx 4\omega_s$.¹⁷

The classical expression corresponds to the term with m=0, it was derived previously in Ref. 2. The classical result is valid for temperatures $\hbar\Omega < 2\pi T$ or approximately $\Delta(T) < T$. The quantum correction (terms with nonzero m) is important practically at all temperatures except in a narrow interval near T_c . In contrast, such dynamic effect as quantum tunneling becomes observable at very low temperatures, below several K only. In the classical region Eq. (12) gives the crossing point: at the temperature T^* given implicitly by $T = \Phi_0^2 s/32 \pi^2 \lambda_{ab}^2(T)$ the magnetization $M^* = M(T^*)$ becomes field independent.

Let us consider results for T=0. At any *n* we obtain the following asymptotic at low fields $B \ll B_O$:

$$\frac{dM}{d \ln B} = \frac{\Phi_0}{32 \pi^2 \lambda_{ab}^2} - \frac{9\hbar\Omega}{8\pi s \Phi_0} \frac{B}{B_Q} \ln \frac{B_Q}{B} \,. \tag{13}$$

Here $B_Q = 4\pi \lambda_{ab}^2 \eta_0 \Omega / \Phi_0$ is the scale of magnetic field which characterizes the quantum correction. In the large *n* limit we obtain

$$\frac{dM}{d \ln B} = \frac{\Phi_0}{32\pi^2 \lambda_{ab}^2} - \frac{\hbar\Omega}{2\pi s \Phi_0} f\left(\frac{B}{B_Q}\right),$$
$$f(x) = 2x \ln\left(1 + \frac{1}{x}\right) + \frac{x}{4} \ln\left(1 + \frac{4}{x}\right) - \frac{x}{1+x}.$$
 (14)

The theoretical curves based on Eq. (12) for T=35 (55) K with parameters n=4, $\hbar\Omega=1200$ (1100) K, $\eta_0=0.48\times10^{-7}$ (0.43×10^{-7}) g/cm s, $\lambda_{ab}=2100$ (2300) Å, and experimental results for sample A are shown in Fig. 2. We take s=15.5 Å. Here are shown also the theoretical curves for T=35 (54) K with parameters $\hbar\Omega=830$ K, $\eta_0=0.52\times10^{-7}$ (0.4×10^{-7}) g/cm s, $\lambda_{ab}=2040$ (2250)



FIG. 2. The dependence $dM/d \ln B$ vs B; for sample A (open symbols) for T=35 K (circles) and T=55 K (squares); for sample B (closed symbols) for T=35 K (circles) and T=54 K (squares). The solid lines are the theoretical fits for (from top to bottom): sample B (T=35 K), sample A (T=35 K), sample B (T=54 K), and sample A (T=55 K).

Å, and experimental results for the sample B. The experimental results and fitting parameters for these two samples are quite close. We can conclude that the observed dependence of M on B is consistent for both samples. The fitting values of η_0 are close to those estimated from flux-flow resistivity. The fitting parameters for Ω are 3-4 times larger than $\Delta(0) \approx 3T_c$ (Ref. 18) and seem to be reasonable taking into account that data in the limited interval of fields below 7 T do not allow determination of n and Ω very accurately.

In the above calculations we neglected the Josephson interlayer coupling. As the Josephson coupling becomes stronger, the elastic modulus c_{44} increases, as do vorton frequencies. This results in an increase of the quantum correction to the magnetization. However, the modifications are minor as long as the system remains in the Josephson regime $(\gamma \gg \xi_{ab}/s)$ and $c_{44}Q^2$ remains much smaller than $c_{11}K_0^2$.

Pinning does not contribute to the mean-field part of $dM/d \ln B$, which is the first term in (12). The reason can be seen from the form of the mean-field free energy [first term in (7)], where the logarithmic dependence on the magnetic induction comes from the logarithmic interaction between vortices at short distances; the structure of the lattice enters only through the numerical factor β which drops out when calculating $dM/d \ln B$. In the contribution of quantum fluc-

tuations to $dM/d \ln B$, pinning may be ignored because the main contribution to the free energy comes from vortex displacements with large momentum. They are affected weakly by pinning. For this reason, pinning was neglected in calculations of $dM/d \ln B$.

Note that above irreversibility line vortices are likely to be in a liquid phase. Though our calculations are done for a lattice, they are valid in a liquid phase as well because fluctuations with large momenta $k \approx K_0$ contribute mainly to $dM/d \ln B$; they are determined by short-range order in the *ab* plane.

In conclusion, we present the experimental results for reversible magnetization of the vortex lattice which show a change of slope in the dependence M vs $\ln B$ at high fields and temperatures well below T_c . We explain this change of slope as arising due to the quantum, zero point fluctuations of vortices. The main parameter of the vortex dynamics, the viscosity coefficient, is estimated from flux-flow resistivity. The obtained estimate reproduces correctly the scale of the quantum fluctuation contribution to equilibrium magnetization.

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- ¹P.H. Kes *et al.*, Phys. Rev. Lett. **67**, 2383 (1991); K. Kadowaki, Physica C **185-189**, 2249 (1991); C. Opagiste *et al.*, J. Alloys Compounds **195**, 455 (1993); F. Zuo *et al.*, Phys. Rev. B **47**, 8327 (1993); J.H. Cho *et al.*, Physica C **212**, 419 (1993).
- ²L.N. Bulaevskii, M. Ledvij, and V.G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).
- ³Z. Tešanović et al., Phys. Rev. Lett. 69, 3563 (1992).
- ⁴Qiang Li et al., Physica B **194–196**, 1501 (1994).
- ⁵Z. Hao and J. Clem, Phys. Rev. B 43, 2844 (1991); Phys. Rev. Lett. 67, 2371 (1991).
- ⁶L.I. Glazman and N.Ya. Fogel, Fiz. Nizk. Temp. **10**, 95 (1984) [Sov. J. Low Temp. Phys. **10**, 51 (1984)].
- ⁷M.P.A. Fisher, T.A. Tokuyasu, and A.P. Young, Phys. Rev. Lett. 66, 2931 (1991); G. Blatter, V.B. Geshkenbein, and V.M. Vinokur, *ibid.* 66, 3297 (1991); J. Tejada, E.M. Chudnovsky, and A. Garcia, Phys. Rev. B 47, 11 552 (1993).
- ⁸A.O. Caldeira and A.J. Leggett, Ann. Phys. (NY) **149**, 374 (1983).
- ⁹G. Blatter and B. Ivlev, Phys. Rev. Lett. 70, 2621 (1993).

- ¹⁰L.N. Bulaevskii and M.P. Maley, Phys. Rev. Lett. **71**, 3541 (1993).
- ¹¹M.J.V. Menken, Ph. D. thesis, University of Amsterdam, 1991.
- ¹²I. Shigaki et al., Jpn. J. Appl. Phys. 29, L2013 (1990).
- ¹³C.J. Van der Beck, C.W. Hagen, and A.V. Samoilov (private communication).
- ¹⁴L.P. Gor'kov and N.P. Kopnin, Usp. Fiz. Nauk 116, 413 (1975)
 [Sov. Phys. Usp. 18, 496 (1975)].
- ¹⁵E.H. Brandt, J. Low Temp. Phys. 26, 709 (1977); 28, 291 (1977);
 A. Houghton, R.A. Pelkovits, and A. Sudbo, Phys. Rev. B 40, 6763 (1989);
 L.I. Glazman and A.E. Koshelev, *ibid.* 43, 2835 (1991).
- ¹⁶ For n = 1 the free energy F_v can be expressed as integral over real frequencies, see Ref. 8. It was used in Ref. 10 to calculate the low-temperature specific heat.
- ¹⁷E.H. Brandt, Physica C 162-164, 257 (1989).
- ¹⁸T. Hasegawa, H. Ikuta, and K. Kitazawa, in *Physical Properties of High Temperature Superconductors III*, edited D.M. Ginzberg (World Scientific, Singapore, 1992).