New type of mixed state with positive magnetization in high-temperature superconductors

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It has been observed that the magnetization of high-temperature superconductors cooled in an external magnetic field and held in it at $T = 4.2$ K eventually changes sign, becoming positive. It is suggested that this behavior results from the particular nature of the nucleation and evolution of the vortex structure in the superconductor.

It is well known (Ref. 1, for example) that inhomogeneous type-II superconductors can be put in a metastable state with a positive magnetization M by reducing the external magnetic field. A vivid demonstration of this point comes from experiments on the suspension of a sample under a permanent magnet, which have recently been described both for high-temperature superconductors[2–4] and for Nb_3Sn, a conventional type-II superconductor.[4]

In this letter we are reporting the observation of a phenomenon which is peculiar to high-temperature superconductors: the formation of a state with $M > 0$ in a fixed external magnetic field after a certain time.

In the experiments we measured the force acting on a sample in the nonuniform field of a superconducting solenoid. The use of a string magnetometer of the type in Ref. 5 prevented zero drift of the measurement system during the measurements (up to 4–5 h) and kept the position of the sample unchanged. The regulated current source (with a regulation better than 10^{-4}) and the high inductance of the solenoid (2.82 H) ruled out the possibility of any significant change in the field during the measurement.

When a field is produced by a fixed current, the role of the thermodynamic potential is played by the total free energy of the sample, defined in Ref. 6, which is a functional of the field H_0 which the current would produce in the absence of the sample. The force acting on a sample of volume v along the axis of the solenoid (the z axis),

$$\mathbf{g} = \int \overline{(\mathbf{M}\nabla)}\, \mathbf{H}_0 dv \approx \Big(\overline{M}_z + \gamma \overline{H}_{0z} \frac{\overline{\partial M_z}}{\partial H_{0z}}\Big) \frac{\overline{\partial H_{0z}}}{\partial z} v \mathbf{e}_z,$$

(where we have the value $\gamma < 10^{-9}$ for our experimental conditions), is determined essentially entirely by the magnetization M_z averaged over the volume (at this point we drop the superiod bar meaning an average and also the subscript z).

In this letter we are reporting results on ceramic and single-crystal samples of $YBa_2Cu_3O_{7-\delta}$ (for the single-crystal sample, the c axis was oriented along the field). Qualitatively similar results obtained for other known high-temperature superconductors will be reported in a detailed publication. Experiments were carried out in two regimes: 1) Diamagnetic screening. In this regime, the samples were cooled to $T = 4.2$ K in the absence of a field. A field was then applied, and after it reached a steady state we measured the time (t) dependence of M. 2) The Meissner effect. The samples were cooled in an external field in the range $0.1 < H_0 < 4$ T, held at $T = 4.2$ K, and then heated to $T > T_c$ (the field was constant throughout the cycle; the readings of the magnetometer was taken at intervals of 20 s).

In the diamagnetic-screening regime, we observed (as have Müller *et al.*[7] and Dmitriev *et al.*[8]) a diamagnetic response $-M/H_0 \sim 10^{-2}$, which falls off approximately logarithmically within $\sim 25\%$ of the initial value over 4 h of measurements, for both the ceramic and the single crystal. In a study of the same samples in the Meissner-effect regime, we found a static susceptibility $-M/H_0 \sim 10^{-4}$ immediately after cooling to $T = 4.2$ K (Figs. 1 and 2). As time elapsed, M/H_0 decreased in absolute value to zero, changed sign, and then increased in the region of positive values (see the insets in Figs. 1 and 2). The response to small field variations was $\chi \equiv \partial M / \partial H_0 < 0$.

Figures 1 and 2 show the temperature dependence of M/H_0 for a complete cooling-hold-heating cycle at $H_0 = 1$ T. We see that there is no irreversibility above a certain $T_0 < T_c$. The results do not depend on the size or shape of the samples. A corresponding study of ordinary type-II superconductors (Nb, Nb_3Al, $VN_{0.985}$, and Ti–V alloys) revealed no hint of a time dependence of M in either the diamagnetic-screening regime or the Meissner-effect regime.

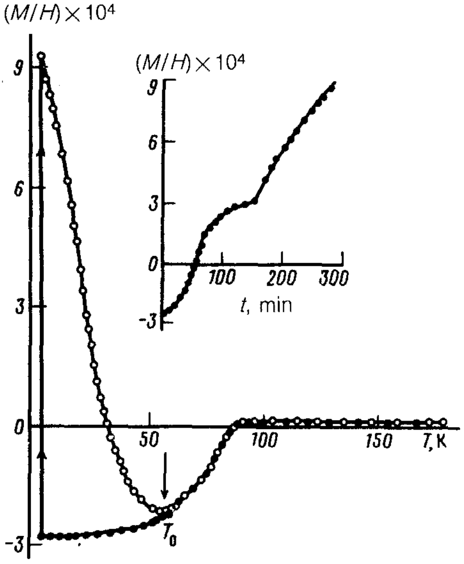

FIG. 1. Temperature dependence of M/H_0 of a ceramic $YBa_2Cu_3O_{7-\delta}$ sample. ●—Cooling in a magnetic field $H_0 = 1$ T; ○—Heating in a field $H_0 = 1$ T after a hold at $T = 4.2$ K for 280 min. The inset shows the time evolution of M/H_0 after cooling in a magnetic field $H_0 = 1$ T at $T = 4.2$ K.

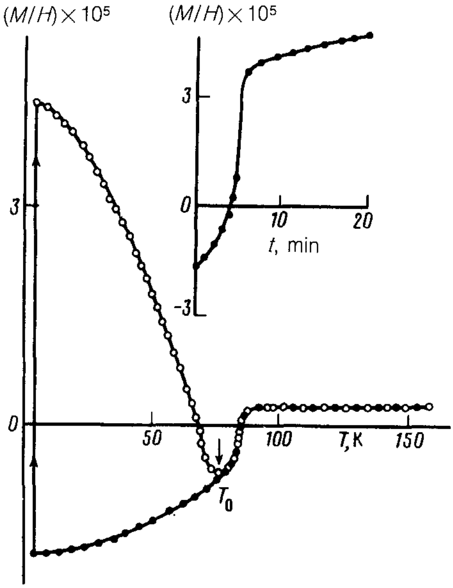

FIG. 2. Temperature dependence of M/H_0 of a single-crystal $YBa_2Cu_3O_{7-\delta}$ sample. ●—Cooling in a magnetic field $H_0 = 1$ T; ○—Heating in a field $H_0 = 1$ T after a hold at $T = 4.2$ K for 15 min. The inset shows the time evolution of M/H_0 after cooling in a magnetic field $H_0 = 1$ T at $T = 4.2$ K.

Turning to a discussion of the results, we first note that the origin of the state with $M > 0$ is not associated with a hysteresis of the magnetization curve. Specifically, as the field is reduced by ΔH_0, the increment in ΔM is bounded by the inequalities $4\pi\Delta M \leqslant |\Delta H| \leqslant |\Delta H_0|/(1-\eta)$, where η is the demagnetizing factor. Since the maximum measured value is $4\pi M/H_0 l \sim 10^{-2} \gg |\Delta H_0|/H_0 \sim 10^{-4}$, a possible instability of H_0 could play a role only for thin-plate samples. That the observed effect is unrelated to the results of Refs. 2–4 is confirmed by the stability of the Meissner effect of ordinary type-II superconductors in our experiments, while a suspension is observed for both those superconductors and high-temperature superconductors.[4]

We believe that the behavior which we have described here, like the "superconductive-glass" effects,[7–9] is a consequence of a well-developed structure of Josephson junctions in the high-temperature superconductors (twinning planes apparently play the role of these junctions in single crystals[10,11]). To demonstrate the point, we adopt the very simple model of an isolated grain with an insulating boundary, at which the condition

$$\partial\psi/\partial n = 0 \qquad (1)$$

holds. During cooling in a field $H_0 \gg \Phi_0/L^2$, where Φ_0 is the flux quantum, and L is the characteristic size of the grain, states with $M > 0$ and $\chi < 0$ can thus form.

We consider a circular cylinder of radius $R \gg \xi(0)$ and of arbitrary length in a longitudinal field $H_0 \ll H_{c2}$ [$\xi(T)$ is the coherence length]. We have in mind the case in which the penetration depth

$$\delta(T) \gg R \tag{2}$$

and the difference between the field and H_0 can be ignored in a first approximation. We also assume that the flux of H_0, divided by Φ_0, through the cylinder satisfies

$$\Phi' \equiv \pi H_0 R^2 / \Phi_0 \ll \kappa^2, \tag{3}$$

where κ is the Ginzburg-Landau parameter. Adopting $(\Phi_0/\pi H_0)^{1/2}$ as a unit of length, we write the Ginzburg–Landau equation for the order parameter as

$$\Delta\psi - 2i\partial\psi/\partial\phi - (\rho^2 + \alpha\tau)\psi - b|\psi|^3 = 0 \qquad (\rho \leqslant \rho_0 \equiv \Phi'^{1/2}), \tag{4}$$

where $\tau = (T - T_{c0})/T_{c0}$, $\alpha \sim H_{c2}/H_0 \gg 1$, and T_{c0} is the temperature of the bulk superconducting transition. Let us find the conditions for the nucleation of a superconducting state in a grain. Setting $\psi \to 0$, and solving (3) without its last term, we find the family of solutions$^{1)}$

$$\psi \sim f(\rho)\exp(im\phi), \tag{5}$$

$$f_m(\rho) = \rho^m \exp(-\rho^2/2) F((2+\alpha\tau)/4,\ m+1, \rho^2),$$

where F is the confluent hypergeometric function, and $m = 0,1,2,\ldots$. States with different values of m are nucleated $\tau_m = 4\alpha^{-1}\varepsilon_m(\Phi') - 2$, where the functions $\varepsilon_m(\Phi')$ are defined by the following equation, which follows from (1) and (6):

$$(m+1)(\Phi' - m)F(\epsilon_m, m+1, \Phi') - 2\Phi'\epsilon_m F(\epsilon_m + 1, m+2, \Phi') = 0. \tag{7}$$

These functions are plotted for several values of m in Fig. 3. The value of m, which

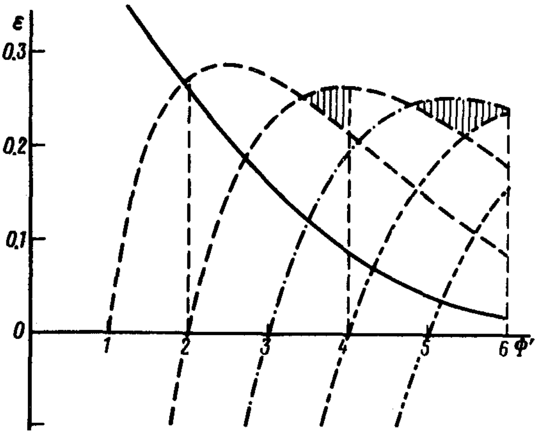

FIG. 3. Curves of $\varepsilon_m(\Phi')$ found through a numerical solution of Eq. (6) for $m = 0,1,\ldots,5$. The curves for $m = 1$–5 intersect the abscissa at $\Phi' = m$. The hatching shows regions in which inequality (10) holds.

corresponds to the maximum value of τ_m, increases with increasing Φ', so states with $m>0$ are nucleated during cooling in sufficiently strong fields ($\Phi' \geqslant 1.92$). Assuming $\Phi' > m \gg 1$, and expanding (7) in $\varepsilon \ll 1$, we easily find the asymptotic expressions

$$\epsilon_m \approx (m/2\pi)^{1/2}(1-m/\Phi')\begin{cases} 1, & \Phi'-m \ll m^{1/2} \\ \frac{1}{2}\exp(-(\Phi'-m)^2/m), & \Phi'-m \gg m^{1/2} \end{cases}. \tag{8}$$

We see from these expressions that τ_m can reach a maximum at $0<\Phi' - m \leqslant m^{1/2}$.

Let us examine the evolution of solutions of (4) of the type in (5) as the temperature is lowered. By virtue of (3) there always exists a region $\alpha^{-1} \ll |\tau| \ll \min(1, (\delta(0)/R)^2)$ in which, on the one hand, the function f is close to its equilibrium value $f_0 = (\alpha|\tau|/b)^{1/2}$ over the greater part of the volume of the grain and, on the other, condition (2) holds and the deviation of the field from H_0 can be ignored. Assuming $\rho \gg |\alpha\tau|^{-1/2}$, we find

$$f_m(\rho) = f_0[1+(2\alpha|\tau|)^{-1}(\frac{m}{\rho}-\rho)^2]. \tag{9}$$

Calculating the magnetization to within some unimportant factors,

$$M \sim \frac{1}{\Phi'} \int^{\rho_\bullet} f^2(\rho)\,(\frac{m}{\rho}-\rho)\rho^2 d\rho \approx \frac{1}{4}\,(2m-\Phi'),$$

we see that this magnetization is positive if

$$m > \Phi'/2, \tag{10}$$

while we have $\partial M/\partial H_0 < 0$. A corresponding estimate of the energy reveals that states with $m>0$ are metastable in the region under consideration.

We thus see that if (10) holds for a state which is nucleated at $T \approx T_c$, a further lowering of the temperature will be accompanied by an evolution of this state to a metastable state with $M>0$. We see from (8) that this clearly is the result if $\Phi' \gg 1$; in practice, (10) holds for all values of $\Phi' > 7.31$.

The meaning of this result is completely clear. In a magnetic field, boundary condition (1) stimulates a nucleation of states in which $|\psi|$ reaches a maximum at a distance $\sim 1[\sim(\Phi_0/H_0)^{1/2}]$ from the surface. In our case, these are states with $m \sim \Phi'$. They correspond to a minimum kinetic energy E_c, since the diamagnetic current at the periphery of a grain is nearly canceled by the vortical current. The contribution of the vortical current to E_c from the inner part of the grain, in contrast, is relatively small. With a further cooling, as $|\psi|$ assumes a constant value f_0 throughout the grain [except in a core of radius $\sim \xi(T) \ll R$], the vortex-current component of M and E_c increases, and a metastable state with $M>0$ forms [an increase in the total field flux through the grain is possible by virtue of (2)]. Presumably, stimulation of the superconducting transition near a twinning boundary [with $\partial(\ln f)/\partial\rho > 0$ at $\rho = \rho_0$] could only strengthen the result.

It appears that this mechanism plays a major role in the effect which we have observed, but the evolution of the magnetization in the region $M > 0$ may also be influenced by other factors, which are ignored in the model of an isolated grain. Foremost among these other factors is the interaction of the phases ψ in different grains as a result of the existence of Josephson junctions. It is possible that the "delay" in the formation of the state with $M > 0$ during cooling is related to that interaction.

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[1)] It is sufficient to consider solutions which are independent of z, since it is easy to see that such solutions correspond to high nucleation temperatures.

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