Josephson Decoupling in Single Crystal $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ Superconductors

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Magnetization with applied field parallel to the *ab* plane has been measured in single crystals of $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$. The data present the first direct measurement of Josephson decoupling between CuO_2 planes at a critical field H_J. For field below H_J, the Meissner effect is observed; at the critical field H_J, magnetization abruptly changes to zero, an indication of a magnetic transparent state. The results are discussed in terms of a recent theoretical model for extremely anisotropic layered superconductors.

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The physical picture that the CuO_2 planes in high T_c superconducting Tl and Bi compounds are weakly Josephson coupled has been supported by several experiments [1–4], including the recent magnetic and transport measurements on $Bi_2Sr_2CaCu_2O_8$ crystals [5,6]. In a layered superconductor with Josephson coupling between the layers, the magnetic response to field parallel to the layers is determined by the properties of Josephson vortices, instead of the Abrikosov vortices of conventional type-II superconductors. Because of the weak screening effect associated with the Josephson vortices, there have been predictions of magnetic transparent states at magnetic field above a characteristic field H_J, a behavior distinctively different from that of the type-II superconductors.

In this Letter, we report an experimental result which illustrates a transition from the Meissner state to the magnetic transparent state, by measuring the magnetization of $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ single crystals in a magnetic field parallel to the *ab* planes. Below a characteristic field H_J, the magnetization changes linearly with the magnetic field; at the critical field H_J, the magnetization changes abruptly to nearly zero, an indication of magnetic transparent state. The data of magnetization as a function of field clearly deviate from the behavior predicted by the Abrikosov theory for type-II superconductors. Instead, the data fit well into the picture of Josephson decoupling between the CuO_2 layers.

Single crystals of $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ are grown using a directional solidification technique [7]. Four crystals are used in the measurement with average dimensions of $1\times1\times0.02$ mm. Extensive measurements were made on samples A and B with T_c of 21 and 22.5 K, respectively. The magnetic transition widths measured at 1 G with zero-field cooling are about 1 K. Measurements are performed using a quantum design magnetometer with low field options. After degaussing and magnet resetting (quenching) the remanent field is typically 5–10 mG. Magnetizations in both directions $H\parallel ab$ and $H\perp ab$ are measured to obtain demagnetization factors. Samples are placed with the *ab* plane parallel to the magnetic field. Misalignment of a few degrees are often observed, and the data have been corrected using measurements at higher magnetic fields [5].

Shown in Fig. 1(a) are typical data of magnetization versus field for the $H\parallel ab$ plane configuration. Two linear regimes are observed. In the first regime, M is linear in $H(<65$ G), then followed by an abrupt increase in M at a critical field ($H_J=65$ G); in the second regime ($H>100$ G) M is again linear in H. For intermediate field $65<H<100$ G, additional, smaller jumps are typically observed. In the H-descending direction, M is linear all

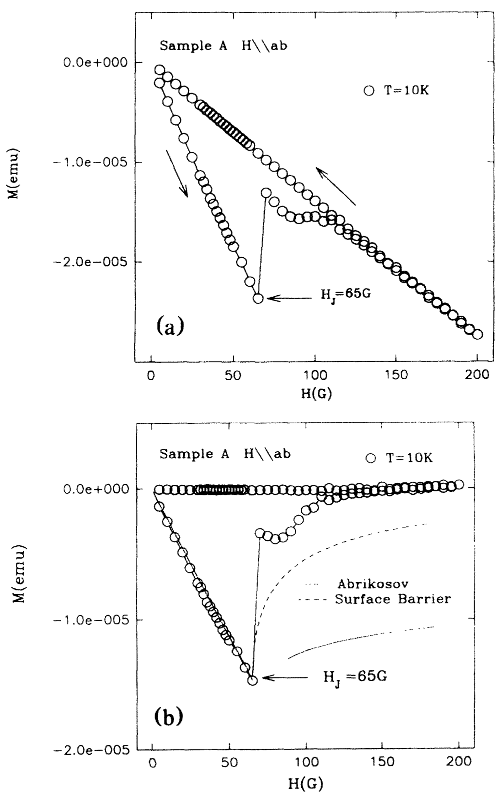

FIG. 1. (a) Magnetization as a function of applied field at $T=10$ K for sample A. (b) Corrected magnetization as a function of applied field at $T=10$ K for sample A. The dashed line is a fit to the surface barrier model and the dotted line is a fit to the Abrikosov theory.

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the way to zero field with the same slope defined in the second regime. This characteristic dependence has been observed in a $Bi_2Sr_2CaCu_2O_8$ single crystal [5] where detailed angular dependence was performed to confirm the fact that the $M(H)$ in the second regime is the contribution of $M_\perp$ (parallel to the c axis) due to misalignment. We have also checked it by varying the sample orientation slightly, and we found the slope of $M(H)$ changes accordingly in the second regime. Typical misalignment is about 2°–6°.

The measured magnetic moment M can be written as $M=(M_\parallel \cos\theta + M_\perp \sin\theta)$ with $M_\parallel = (VH\cos\theta/4\pi)/(1-N_{ab})$ and $M_\perp = (VH\sin\theta/4\pi)/(1-N_c)$, where V is the volume of the sample, θ is the angle between the ab plane and the applied field, and N_{ab} and N_c are the demagnetization factors for field along the ab plane and c axis, respectively. For sample A, we obtained $1/(1-N_c) \sim 110$ using the susceptibilities measured on both directions. Shown in Fig. 1(b) is a plot of magnetization along the ab plane after subtracting the contribution of $M_\perp$. The $M_\perp$ contribution is obtained by measuring the slope χ_2 of the second linear regime or the slope of the descending branch. The corrected magnetization is given by $M=M-\chi_2 H$. From the corrected magnetization and χ_2 one can calculate the misalignment angle to be around 3.3°. The corrected magnetization, again, shows a sharp transition in M at $H=65$ G, followed by a plateau and then M goes to zero quickly starting at about $H=85$ G. The jump in M at H_J at $T=10$ K is about 80% of the full Meissner value. The dashed and dotted lines are models to be discussed later. On the descending branch of the magnetization, M is essentially zero, independent of the field value.

Plotted in Fig. 2 is an overlay of magnetization (after subtraction) as a function of field at $T=8$, 10, 14, and 16 K. The overall features are similar, but some differences are observed. For $T=8$ K, the first transition occurs at $H=70$ G, followed by a second transition at about $H=82$ G. The plateau seen in M at $T=10$ K clearly becomes a sharp transition. At $T=14$ K, there are three transitions in $M(H)$, with the largest jump in M occurring at the second transition. The transition is continuous at $T=16$ K. We define the critical field H_J to be the field value at the first jump. As is clear from the data, the critical field decreases with increasing temperatures, and the increase in M at H_J becomes more gradual. For $T>15$ K, no abrupt changes are observed, the overall shapes are similar to the reported data on $Bi_2Sr_2CaCu_2O_8$.

The inset in Fig. 2 is a plot of the first critical field as a function of temperature. For temperature below 12 K, the critical field H_J is almost constant around 70 G, while H_J decreases sharply for T greater than 12 K.

To study the effect of transition temperature on the critical field H_J, we have performed similar measurements on several $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ crystals. Shown in Fig. 3 is an overlay of magnetization as a function of field at different temperatures for sample B. The transition temperature of this crystal is $T_c=22.5$ K, 1.5 K higher than sample A. Again, abrupt changes are observed in the $M(H)$ dependence. At $T=5$ K, several steps are observed with the first critical field around 35 G. The overall shape is similar to the data presented in Fig. 2, except the critical field is much smaller than that of sample A. The magnetization in the descending branch of the hysteresis loop is also zero. Plotted in the inset is the temperature dependence of the first critical field. The temperature dependence is also similar to that of sample A; i.e., H_J saturates at low T and decreases sharply at high T. Notice the saturation value of H_J is considerably smaller than sample A.

The magnetization in the decreasing field direction is almost zero, an effect that has been attributed to surface barriers. In the $H\|c$ direction, this characteristic has

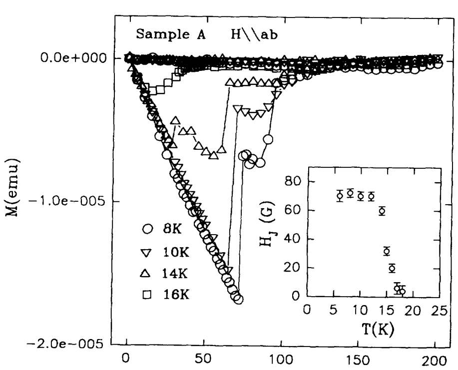

FIG. 2. An overlay of magnetization as a function of field at different temperatures for sample A. $T=8$, 10, 14, and 16 K. The inset is a plot of the critical field as a function of temperature.

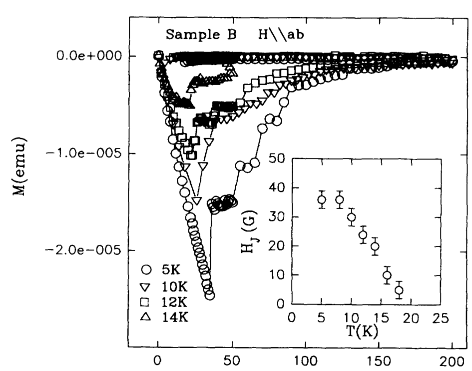

FIG. 3. An overlay of magnetization as a function of field at different temperatures for sample B. $T=5$, 10, 12, and 14 K. The inset is a plot of the critical field as a function of temperature.

only been observed at high temperatures on high quality single crystals $YBa_2Cu_3O_7$ [8] and $YBa_2Cu_4O_8$ [9], where bulk pinning can be neglected. For field parallel to the *ab* plane, this effect has been observed in several systems [5,10] such as $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CuO_6$, and $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$. The origin of this behavior may be due to the fact that there is little or no bulk pinning for vortex motion along the *ab* planes; vortices only see a surface barrier. The surface current decreases to zero quickly in the descending branch and remains zero when the applied field is further reduced, such that vortices can flow out freely and the overall magnetization is almost zero.

The jump at H_J in $M(H)$ in the ascending field direction is in sharp contrast with the magnetic behavior of classical type-II superconductors. The magnetization of conventional type-II superconductors can be well described by the Abrikosov theory. At field slightly larger than the lower critical field H_{c1}, the magnetization is obtained by neglecting interaction between vortex lines,

$$4\pi M = \frac{2\phi_0}{\sqrt{3}\lambda^2}\left\{\ln\left[\frac{3\phi_0}{4\pi\lambda^2(H-H_{c1})}\right]\right\}^{-2} - H\,.$$

In the case of high T_c cuprate, the very large value of the Ginzburg-Landau parameter κ renders the use of this result to magnetic field very close to H_{c1} ($H_{c1} < H < H_{c1} + 10$ G). For intermediate fields, the magnetization is given by $4\pi M = -H_{c1}\ln(\beta' l/\xi)/\ln(\lambda/\xi)$ where $\beta' = 0.231$, and l is the lattice constant given by $B = 2\phi_0/\sqrt{3}l^2$ for a triangular lattice [11]. Assuming $\kappa = \lambda/\xi \sim 20$ and $H_{c1} = 65$ G, the equilibrium magnetization can be calculated numerically as a function of field. The result is shown as the dotted line in Fig. 1(b). The experimental data clearly reject the use of this model.

As evidenced by the zero magnetization in the descending branch of the hysteresis loop, the entrance field is determined by Bean-Livingston surface barriers [12]. The magnetization for field greater than the penetration field H_p and assuming no bulk pinning, has been discussed in several recent articles [8,13]. By modeling an Abrikosov lattice in the bulk of the sample and vortex-free zone near the surface due to surface barriers, the magnetization is derived and given by $-4\pi M = H - \sqrt{H^2 - H_p^2}$. The result is plotted as the dashed line in Fig. 1(b). Again, the model deviates from the magnetization data. It is to be noted that the expression above is derived for a conventional type-II superconductor, where the flux lattice is the Abrikosov lattice.

The abrupt change that occurred at H_J strongly indicates the inadequacy of the use of Abrikosov's theory in the magnetization of $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ crystals in the $H \| ab$ direction. Since the $M_\perp(H)$ clearly exhibits the Meissner effect in the perpendicular direction, the jump in $M_\|(H)$ shows directly magnetic decoupling between superconducting layers at the critical field H_J.

To describe the vortex state of the Josephson coupled superconducting stacks, one has to solve the coupled Lawrence-Doniach equations [14]. In the case where the applied field is tilted to the superconducting layers, several theoretical models are proposed [15–18]. Recently it has been shown that there are two distinct vortex structures depending on the anisotropy constant $\gamma = \lambda_J/d$, where λ_J is the Josephson penetration depth and d is the separation between the superconducting layers. If $\lambda_J < \lambda_{ab}$, the theory suggests a tilted vortex line structure, where different segments of vortex lines parallel to the *ab* planes are connected by pancakes residing in the *ab* planes [15,16]. If $\lambda_J > \lambda_{ab}$, the model predicts that coexisting sets of perpendicular and parallel vortices should exist when the field is applied close to the *ab* plane [16–18]. The Abrikosov vortices are due to $H_\perp$, and Josephson vortices are due to $H_\|$. The vortices due to $H_\|$ and $H_\perp$ act independently of each other. The complete Meissner effect is possible only if $H_\| < H_{c1}^\|$ and $H_\perp < H_{c1}^\perp$. For $H_\| > H_{c1}^\|$, $H_\|$ penetrates into the layers between the CuO_2 planes almost completely, creating the so called magnetic transparent state [17,18]. Using the Lawrence-Doniach model, the magnetization has been calculated for extreme cases when H is either very small (close to H_{c1}) or very large [16]. The small field limit has no practical bearing because the Josephson penetration depth is extremely large, such that one has to take interaction between vortices into account even when field is very small. For large magnetic field, the magnetization is given by $M = -(\phi_0/32\pi^5\lambda_{ab}\lambda_c)[H_0/H]^3$ for $H \gg H_0$, where $H_0 = \phi_0/\gamma d^2$. The magnetization is essentially zero in comparison with the Meissner value, in agreement with prediction of the transparent magnetic state.

The experimental results clearly demonstrate the complete Meissner state for $H < H_J$. The linear field dependence of M in the descending branch and the overlapping $M(H)$ at high field indicate $H_\perp < H_{c1}^\perp$. For $H > H_J$, there are only Josephson vortices parallel to the *ab* plane and $M_\perp$ is still in the Meissner state. The jump in $M(H)$ for $H > H_J$ indicates the transition to the magnetic transparent state in Josephson coupled layered superconductors.

The jump in $M_\|(H)$ can also be qualitatively understood from the field dependence of critical current of a single Josephson junction: $J_c(H) = J_0 \sin(\pi\phi/\phi_0)/(\pi\phi/\phi_0)$ where $J_c(H)$ is the field dependent critical current, J_0 is the maximum zero-field critical current, and ϕ is the magnetic flux through the junction. The critical current is almost zero when there is a finite number of Josephson vortices threading through the junction. Assume the magnetization is proportional to the critical current, M should become zero as Josephson vortices penetrate into the junction. For stacked layers with Josephson coupling, the field dependence of J_c should not be the same as that of a single Josephson junction, rather one expects a stronger reduction in $J_c(H)$ with increasing H. In this picture, the experimentally observed smaller jumps in the magnetization data thus would indicate the presence of superconducting layers with different Josephson couplings.

In the presence of surface barriers, the penetration field H_p is not the lower critical field H_{c1}. For conventional superconductors, H_p is of the order of thermodynamic critical field H_c. In the case of Josephson coupling between layers, the penetration field has been derived to be [19]

$$H_p \approx \phi_0/4\pi\lambda_{ab}\gamma d \,, \tag{1}$$

where λ_{ab} is the penetration depth in the ab plane. Using the values of $\lambda_{ab}=1000$ Å [20], $H_p=70$ G for sample A and $d=12$ Å, one obtains $\gamma=400$. For sample B, γ is 200 using the same λ_{ab} and $H_p=35$ G. The large value of γ is consistent with that of other high T_c cuprate systems such as $Bi_2Sr_2CaCu_2O_8$ where γ of 700 is obtained.

The anisotropy constant γ obtained above is very different between the two $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ single crystals. Crystal B has a transition temperature $T_c=22.5$ K and crystal A has a slightly lower T_c of 21 K; however, the γ is different by a factor of 2. In terms of Josephson coupling strength η, where $\eta=\hbar^2/4d^2m_c=\hbar^2/4d^2\gamma^2m_{ab}$, η increases by a factor of 4 in the lower T_c sample. The increase in η is qualitatively consistent with Ambegaokar and Baratoff's result on Josephson junction between two bulk superconductors [21]. The coupling strength is given by $\eta=[\pi\Delta(T)/2eR_n]\tanh[\Delta(T)/2kT]$, where $\Delta(T)$ is the gap function and R_n is the normal state tunneling resistance. From the resistivity measurements in the electron-doped system [22], it is found that the normal state resistivity along the c axis decreases with decreasing T_c. The lowest resistance is found when the material is no longer superconducting. The exact dependence of R_n on T_c is not clear; however, with increasing T_c, a decreasing η is expected.

The difference in magnetic transition around the critical field between the $Nd_{1.85}Ce_{0.15}Cu_2O_{4-y}$ and $Bi_2Sr_2CaCu_2O_8$ crystals is not clear. One possibility is that the $Bi_2Sr_2CaCu_2O_8$ crystals studied are very thick (0.4 and 0.9 mm), which may contain a broad distribution of η [5]. A broad distribution in the coupling strength will smear out the transition. Another possibility is that there are two inequivalent layers in $Bi_2Sr_2CaCu_2O_8$ system, one layer is between CuO_2 planes and the other is between the double layers of CuO_2. Such inequivalent planes have been argued to be capable of hosting stable vortices in between the planes [23]. While this inequivalent plane model may not apply to the $Tl_2Ba_2CuO_6$ system, structural disorders in that system may offer another explanation.

In summary, we have reported an experimental measurement of an abrupt Josephson decoupling between the CuO_2 planes at the critical field H_J parallel to the ab planes. The abrupt increase in $M(H)$ at H_J is incompatible with the conventional Abrikosov theory. For field $H_\parallel > H_J$, the magnetic field has a complete penetration in between the superconducting layers—a magnetic transparent state. The coupling strength η decreases with increasing T_c among the samples studied. A more detailed study of η as a function of T_c will help to elucidate the mechanism of superconductivity in the layered superconductors.

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