Transparent Magnetic State in Single Crystal $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ Superconductors

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Several experimental studies have been reported as evidence of Josephson coupling between the superconducting layers in the highly anisotropic oxide such as the $Bi_2Sr_2CaCu_2O_8$ and $Tl_2Ba_2CuO_6$ systems[1-8]. These include the large penetration depth of 100 μm measured[1], ac and dc Josephson effects[3]. Recently two critical temperatures corresponding to Josephson coupling in between the layers and the Berezinskii-Kosterlitz-Thouless transition in the ab-plane have been directly observed in the transport measurements[7]. If the field is applied parallel to the superconducting layers, the magnetic excitation is not the conventional Abrikosov vortices, but the Josephson vortices which extend λ_{ab} in the c-axis direction and $\lambda_J = \gamma s$ in the plane (s is the interlayer distance, γ is the anisotropy constant). 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 paper, we report an experimental result which illustrates a transition from the Meissner state to the magnetic transparent state in single crystal of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$. Magnetization has been measured as a function of temperature and field in the magnetic field parallel or close to ab-plane geometry. For a fixed magnetic field, the magnetization shows a two-step transition in M(T); for a fixed temperature, the magnetization shows an abrupt change to almost zero value above a characteristic field H_J, an indication of magnetic transparent state. The data of magnetization as a function of field clearly deviates 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[8].

Single crystals of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ are grown using a directional solidification technique[9]. Four crystals are used in the measurement with average dimensions of 1×1×0.02mm. Extensive measurements were made on two samples A and B with T_c of 22.5K and 21K, respectively. Typical magnetic transition widths measured at 1G with zero-field cooling are about 1–2K. Measurements are performed using a Quantum Design magnetometer with low field options. After degaussing and magnet resetting (quenching) the remanent field is typically 5–10mG. All measurements were done with the sample zero-field cooled to a temperature below T_c. Magnetizations in both directions H || ab and H⊥ ab were measured. For samples with ab-plane aligned parallel to the field, misalignment of a few degrees were often observed.

Shown in Figure 1(a) and (b) are plots of magnetization of sample A as a function of temperature in field parallel to c-axis and ab-plane, respectively. In Figure 1(a), the transition temperature T_c is about 22.5K and M(T) is almost flat for T below 20K. The transition width is less than 2K in the applied field of 1G, an indication of high crystal quality. The magnetization in the field close to ab-plane direction is clearly different from that of field parallel to c-axis direction. For an applied field of 10G, there are two apparent transitions. The high temperature transition corresponds to the same T_c as from Figure 1(a), the lower transition starts around 12K and magnetization saturates above 16K.

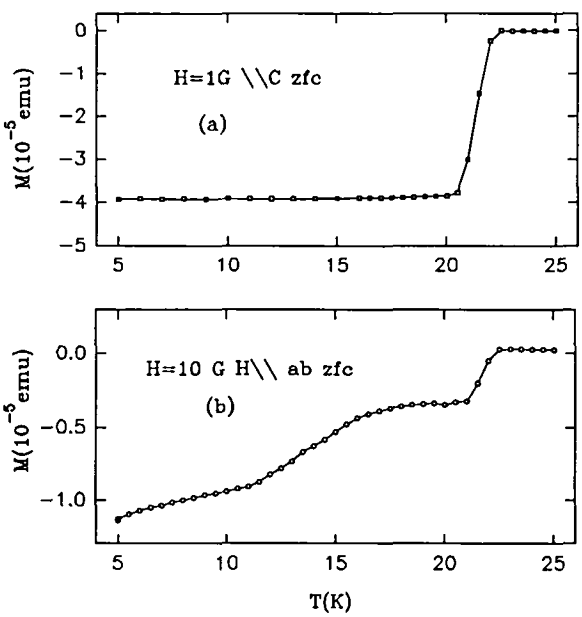

Fig. 1 Temperature dependence of magnetization in field parallel to c-axis (a) and ab-plane (b).

Shown in Figure 2 is an overlay of magnetization as a function of temperature of sample A at different applied field H=30G, 40G, 50G and 60G. Again, two regimes are clearly distinguishable. In the low temperature regime, the magnetization is nonlinear with the applied field. At the lowest temperature measured (5-6K), magnetization increases in magnitude from 30G to 40G, then decreases at 50G and followed by further increase at H=60G. The magnetization of H=50G and 60G crossover at higher temperature ($\sim$ 8K). In the high temperature regime, the magnetization is relatively flat with respect to temperature. The magnitude of M is quasi-linear with field H. With further increases in temperature the sample becomes normal with paramagnetic susceptibility above T_c. The transition width increases with increasing field.

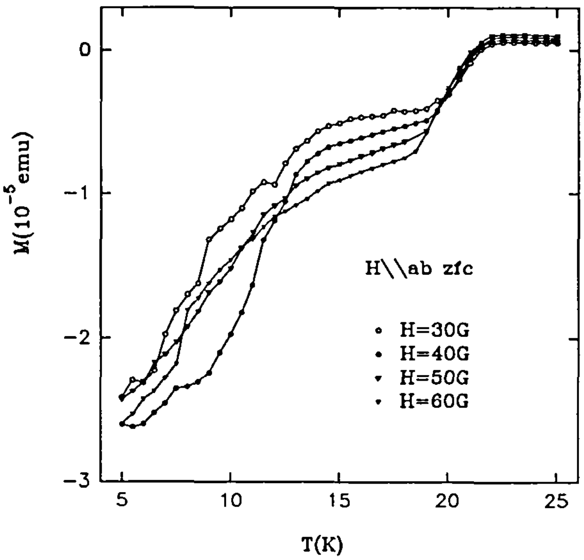

Fig. 2 Temperature dependence of magnetization at different fields H=30G, 40G, 50G and 60G with the field parallel to ab-plane.

The nonlinear field dependence of M observed at low temperature is in contrast with magnetization in the c-axis direction. To understand this anomalous behavior, we have performed detailed magnetization as a function of field at fixed temperatures. For these measurements, the sample is zero-field cooled to a set temperature and M is measured with the magnet set in the non-overshot, persistent mode. Plotted in Figure 3 is a typical magnetization as a function of field for a second sample B of T_c=21K.

Two linear regimes are observed. In the first regime, M is linear in H (<65G), then followed by an abrupt increase in M at a critical field (H_J=65G); in the second regime (H>100G) M is again linear in H. For intermediate field $65G < H < 100G$, additional, smaller jumps are typically observed. In the H-descending direction, M is linear all the way to zero field with the same slope defined in the second regime. This characteristic dependence has been observed in $Bi_2Sr_2CaCu_2O_8$ single crystal[6] where detailed angular dependence were performed to confirm the fact that the M(H) in the second regime is the contribution of $M_\perp$ (parallel to 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 degrees.

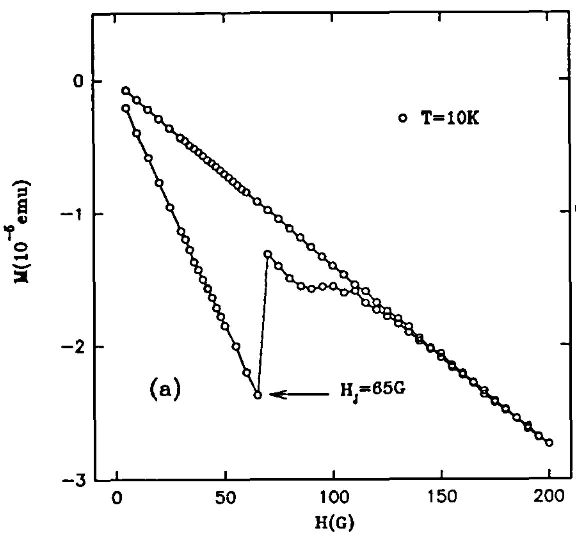

Fig. 3 Magnetization as a function of applied field at T=10K for sample B.

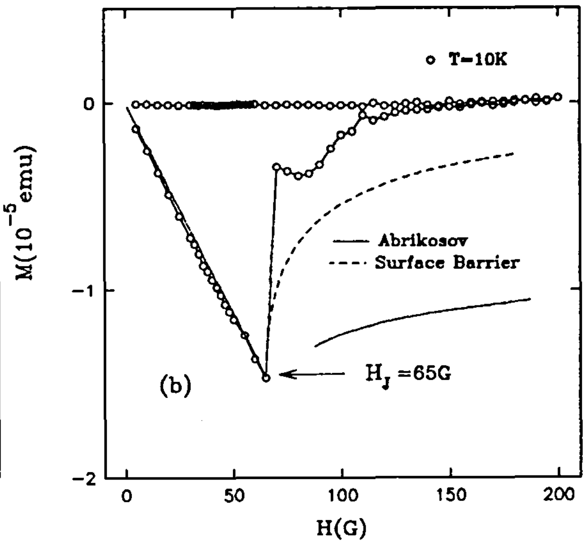

Fig. 4 The corrected magnetization as a function of applied field. The dashed and dotted line are theoretical fits.

The measured magnetic moment M can be written as $M = M_{||}\cos\theta + M_\perp \sin\theta)$ with $M_{||} = (VH\cos\theta/4\pi)/(1-N_{ab})$ 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 B, we obtained $1/(1-N_c) \sim 110$ using the susceptibilities measured on both directions. Shown in Figure 4 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 degrees. The corrected magnetization, again, shows a sharp transition in M at H=65G, followed by a plateau and then M goes to zero quickly starting at about H=85G. The jump in M at H_J at T=10K is about 80% of 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.

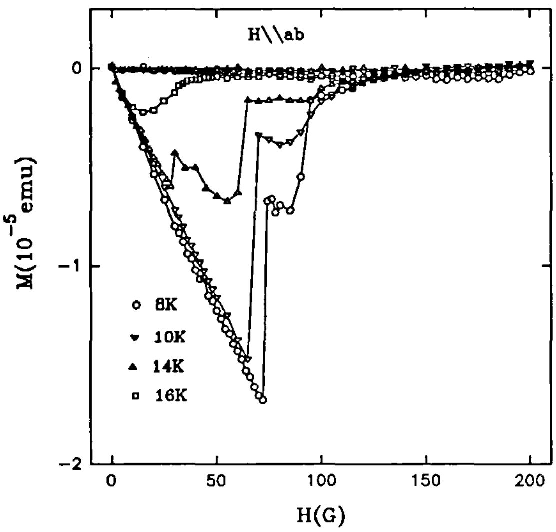

Fig. 5 An overlay of magnetization as a function of field at different temperatures T=8K, 10K, 14K and 16K.

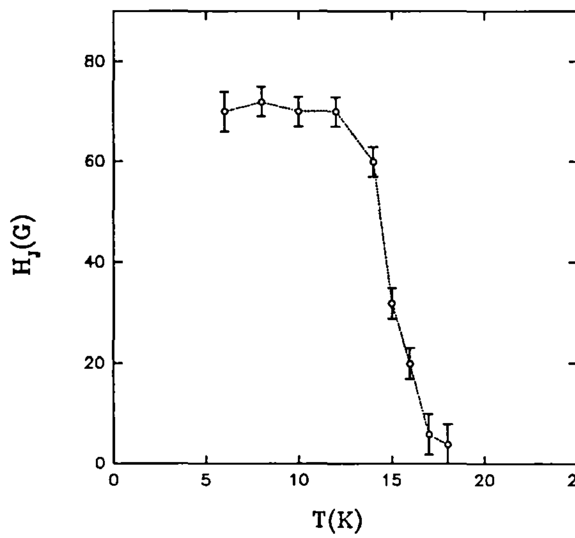

Fig. 6 The first critical field as a function of temperature.

Plotted in Figure 5 is an overlay of magnetization (after subtraction) as a function of field at T = 8, 10, 14, and 16K. The overall features of M at different T are similar, but some differences are observed. For T=8K, the first transition occurs at H=70G, followed by a second transition at about H=82G. The plateau seen in M at T = 10K clearly becomes a sharp transition. At T=14K, there are three transitions in M(H), with the largest jump in M occurring at the second transition. The transition is continuous at T=16K. We define the critical field H_J to be 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>15K, no abrupt changes are observed, the shapes of M(H) are similar to the reported data on $Bi_2Sr_2CaCu_2O_8$.

Figure 6 is a plot of the first critical field as a function of temperature. For temperature below 12K, the critical field H_J is almost constant around 70G, while H_J decreases sharply for T greater than 12K.

To study the effect of transition temperature on the critical field H_J, we have performed similar measurements on several $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ crystals. Shown in Figure 7 is an overlay of magnetization as a function of field at different temperatures for sample A. Again the abrupt changes are observed in the M(H) at the critical field.

At T=5K, several steps are in fact observed with the first critical field around 35G. At higher temperatures, the similar features to that of Figure 5 are observed, except the critical field is much smaller than that of sample B. The magnetization in descending branch of the hysteresis loop is also zero after correction.

The direct measurement of M(H) is consistent with the results of magnetization as a function of temperature. In Figure 2, the nonlinear field dependence of M seen at low temperature is a direct reflection of abrupt change in M at $H_J \sim 35G$. The crossover between the 50G and 60G data at low temperature indicates the relative contributions due to the jump and $M_\perp$. The quasi-linear M on H demonstrates the fact that at high temperatures the main contribution is from $M_\perp$ when the sample is misaligned.

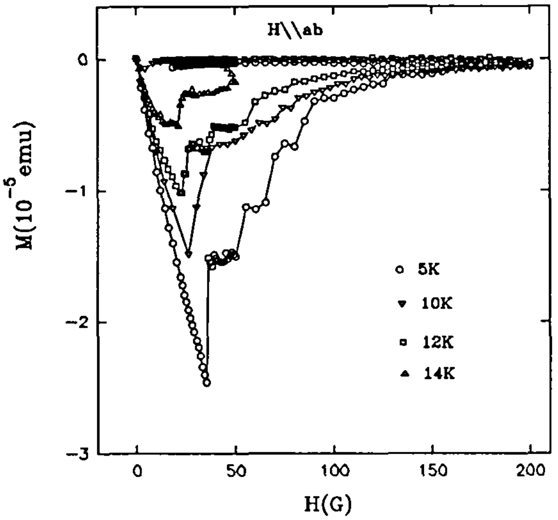

Fig. 7 An overlay of corrected magnetization as a function of temperatures T=5K, 10K, 12K,and 14K for sample A.

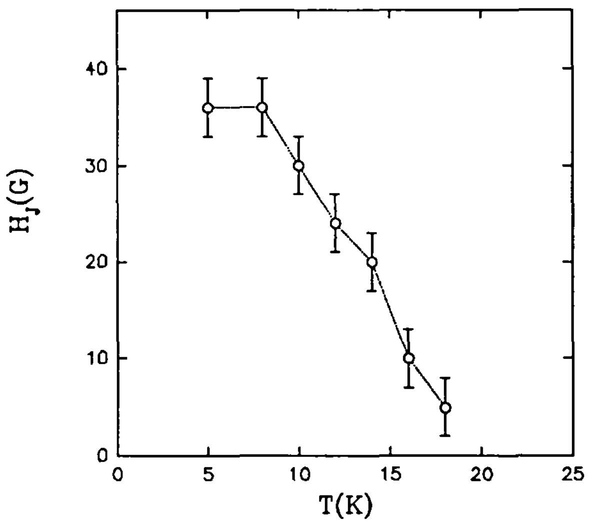

Fig. 8 The first critical field as a function of temperature for sample A.

Plotted in Figure 8 is the temperature dependence of the first critical field. The temperature dependence is also similar to that of sample B, 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 that of sample B.

The abrupt change in the magnetization at the critical field is clearly different from that of conventional type-II superconductors. The magnetization of conventional type-II superconductors can be well described by the Abrikosov's theory[10]. At field slightly larger than the lower critical field H_{c1}, the magnetization is obtained by neglecting interaction between vortex lines $4\pi M = \frac{2\phi_o}{\sqrt{3}\lambda^2}\{\ln[\frac{3\phi_o}{4\pi\lambda^2(H-H_{c1})}]\}^{-2} - H$. 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} ($H_{c1} < H < H_{c1}+10G$). For intermediate fields, the magnetization is given by $4\pi M = -H_{c1}\frac{\ln(\beta' l/\xi)}{\ln(\lambda/\xi)}$ where β'=0.231, and l is the lattice constant given by $B = 2\phi_o/\sqrt{3}l^2$ for a triangular lattice[10]. Assuming $\kappa = \lambda/\xi \sim 20$ and H_{c1}=65G, the equilibrium magnetization can be calculated numerically as a function of field. The result is shown as the dotted line in Figure 4. The jump in M(H) clearly rejects the use of this model.

As suggested by the zero magnetization in the descending branch of the hysteresis loop, the entrance field is determined by Bean-Livingston surface barriers[11]. In the presence of surface barriers, the penetration field is not the lower critical field H_{c1}, but comparable to the thermodynamic critical field H_c. The surface barrier is a result of attractive interaction between an image vortex with the entering vortex. Strong evidence has been found in the field parallel to the c-axis direction in high quality single crystal materials[12, 13, 14] The magnetization for field greater than the penetration field H_p and assuming no bulk pinning, has been discussed in several recent articles[12, 15]. 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 Figure 4. Again, the model deviates from the magnetization data. It is to be noted that the expression above is derived for conventional type-II superconductors, where the flux lattice is the Abrikosov lattice.

The abrupt change occurred at H_J strongly indicates the inadequacy of the use of Abrikosov's theory in the magnetization of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ crystals in the H $\parallel$ 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.

For weakly Josephson coupled layered superconductors, the critical current in the c-axis direction, which provides magnetic screening when field is in the ab-plane, is determined by the coupling strength. For small applied field, complete screening of external field is possible. At the critical field, the Josephson coupling breaks down, magnetic field penetrates into the sample. If the layers are effectively decoupled, a transparent magnetic state is expected, due to the fact that the 2D superconducting layers can not support screening current by themselves.

The jump in $M_{||}(H)$ can also be qualitatively understood from the field dependence of critical current of a single Josephson junction: $J_c(H) = J_o \sin(\frac{\pi\phi}{\phi_o})/(\frac{\pi\phi}{\phi_o})$

where $J_c(H)$ is the field dependent critical current, J_o 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 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.

To describe the vortex state of the Josephson coupled superconducting stacks quantitatively, one has to solve the coupled Lawrence-Doniach equations[16]. In the case where the applied field is tilted to the superconducting layers, several theoretical models are proposed. Recently it has been shown that there are two distinct vortex structures depending on the anisotropy constant $\gamma = \lambda_J/s$, where λ_J is the Josephson penetration depth and s 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[17, 18]. 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. 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[18, 19, 20].

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 difference in magnetic transition around the critical field between the $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ and the $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CuO_6$ crystals is not clear. One possibility is that the $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CuO_6$ crystals studied are very thick[6, 21]. A broad distribution in the coupling strength will smear out the transition. This picture is supported by a recent report which shows a similar abrupt change in M(H) in another $Tl_2Ba_2CuO_6$ crystal[22].

In summary, we have presented an experimental measurement of an abrupt

Josephson decoupling between the CuO_2 planes at the critical field H_J parallel to ab-planes. The abrupt increase in M(H) at H_J is incompatible with the conventional Abrikosov theory. For field $H_{||} > H_J$, the magnetic field has a complete penetration in between the superconducting layers–a magnetic transparent state. A more detailed study of H_J as a function of T_c will help to elucidate the mechanism of superconductivity in the layered superconductors.

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