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Evidence from magnetoresistance measurements for an induced triplet superconducting state in $La_{0.7}Ca_{0.3}MnO_3/YBa_2Cu_3O_{7-\delta}$ multilayers

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Angular magnetoresistance measurements were performed on $La_{0.7}Ca_{0.3}MnO_3/YBa_2Cu_3O_{7-\delta}/La_{0.7}Ca_{0.3}MnO_3$ (LCMO/YBCO/LCMO) trilayers below and above the superconducting transition temperature T_c of the YBCO layer. The conductance of the LCMO layer increases by two orders of magnitude below T_c, while its dissipation is due to vortex motion with highly spin-polarized vortex cores. These results are evidence for induced triplet superconductivity into the ferromagnet over a distance of about 4.7 nm.

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The study of the interaction between ferromagnetism and superconductivity in ferromagnet/superconductor (F/S) heterostructures received increased interest especially after it has been shown that manganites and cuprates have the ability to form high-quality heterostructures such as $La_{0.7}Ca_{0.3}MnO_3/YBa_2Cu_3O_{7-\delta}$ (LCMO/YBCO). The magnetism and superconductivity are modified at the interface of the LCMO/YBCO heterostructure, i.e., carefully controlled interfaces between these two materials give rise to physical phenomena and functionalities, which are not exhibited by either of the constituent materials alone.[1–3] Theoretically, it has been shown that a triplet superconducting condensate could be induced at the interface of the F/S heterostructure, which penetrates into the ferromagnet over long distances.[4–9] Experimentally, long-range proximity effect has been found in LCMO/YBCO heterostructures,[10] but presently there are only speculations on its origin with no experimental evidence which would support the different scenarios put forward. Even though spin-triplet supercurrent has previously been found in completely spin-polarized CrO_2 (Ref. 11) and in Ho ferromagnetic wires,[12] there is no evidence for its presence in heterostructures which contain unconventional superconductors such as the cuprates.

Here we address the origin of the long-range proximity effect found in LCMO/YBCO/LCMO trilayers through out-of-plane angular magnetoresistance measurements. Below the superconducting transition temperature T_c of the YBCO layer, the conductance of the LCMO layer increases by two orders of magnitude, while the dissipation is due to flux vortices that have highly spin-polarized ferromagnetic cores and penetrate into the LCMO layer over about 4.7 nm. These results clearly show that triplet superconductivity is induced into the ferromagnetic LCMO layer.

LCMO/YBCO/LCMO trilayers were grown on (100)-oriented $SrTiO_3$ single crystals. The details of sample preparation were reported elsewhere.[13] The ferromagnetic layers of the trilayer are 40 unit cell (u.c.) (16 nm), while the superconducting layer is 4 u.c. (4.8 nm) (lower inset to Fig. 1). A buffer layer of $PrBa_2Cu_3O_{7-\delta}$ of 6 u.c. was used between the substrate and the first LCMO layer. The LCMO/YBCO interfaces are perfectly coherent and free of disorder.[10] YBCO thin films with a thickness of 12 u.c. (15 nm) were also grown under same conditions. All samples are 1×0.5 cm^2. A current I of 100 μA was applied in the ab plane and the resistance R of the trilayer or thin film was measured using a four-contact method while changing the temperature T, applied magnetic field H, or out-of-plane angle θ defined as the angle between H and the crystallographic c axis of the trilayer or thin film [upper inset to Fig. 2(b)]. The zero-field T_c of the trilayers is 28 K (see upper inset to Fig. 1 for its definition), while of the thin films is 90 K.

Figure 1 is a plot of $R(T)$ of such a trilayer measured in 0 and 14 T. The maximum in $R(T)$ marks the Curie temperature, below which the LCMO layers are ferromagnetic. Notice that the negative colossal magnetoresistance of the two LCMO layers dominates the normal-state $R(T)$ at temperatures lower than the Curie temperature; namely, R decreases substantially with increasing H from 0 to 14 T. Nevertheless, the superconducting transition of the YBCO layer is clearly visible at lower temperatures in the $R(T)$ data measured both in 0 and 14 T (see also the upper inset to Fig. 1).

Measurements of the resistance while the trilayer or the YBCO thin film was rotated out of its ab plane were performed at temperatures both below and above T_c in $H \leq 14$ T. Figure 2(a) shows the flux-flow resistance R_{FF} vs $H\cos\theta$ measured on a YBCO thin film at 88 K, i.e., in its mixed state. The solid curve traces the data taken while scanning H, with $H \| c$ axis ($\theta = 0°$), while the rest of the data are taken while scanning θ in different H. At same $H\cos\theta$ (same out-of-plane field) and up to a certain θ value, $R_{FF}(H\cos\theta)$ data for θ scanning protocol and H scanning protocol scale, while the former data become larger than the latter at larger θ values. The value of θ below which the $R_{FF}(\cos\theta)$ data scale depends on the value of H. The inset to Fig. 2(a) shows that the $R_{FF}(H\cos\theta)$ data for 0 and 60° and scanning H up to 14 T scale; i.e., it shows that the scaling holds at least up to 60° for all the measured H.

This scaling behavior has previously been found in high T_c cuprates, and it is the result of the dissipation of *only* the vortices perpendicular to the CuO_2 planes. Specifically, flux-flow resistance R_{FF} in anisotropic superconductors is given by the functional dependence[14,15]

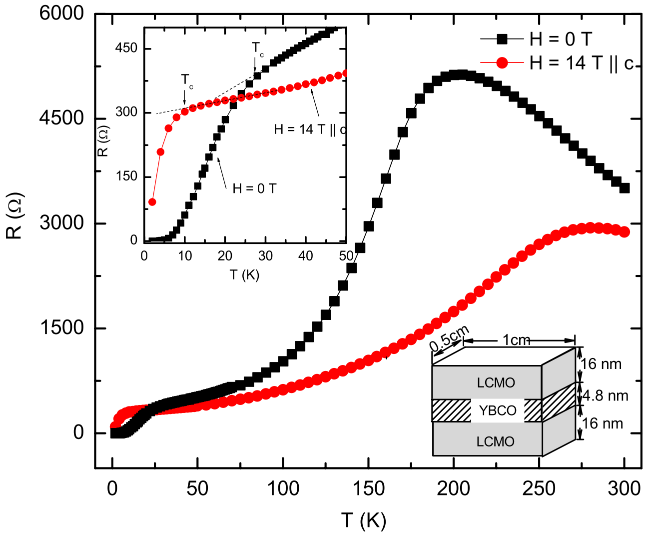

FIG. 1. (Color online) Plot of resistance R vs temperature T for the LCMO/YBCO/LCMO trilayer measured in 0 and 14 T. Upper Inset: $R(T)$ measured in the superconducting transition region. Lower Inset: sketch of the trilayer.

$$R_{FF}=R_n f\left(\frac{H}{H_{c2}(\theta)}\right)=R_n f\left(\frac{H\sqrt{\cos^2\theta+\gamma^{-2}\sin^2\theta}}{H_{c2}(0)}\right), \quad (1)$$

where R_n is the normal-state resistance, the function f gives the percentage of the quasiparticles present in the core of the vortices that dissipate when the vortices move, and γ is the anisotropy of the superconductor. This relationship shows that for an angle θ between H and the c axis, in principle both vortices perpendicular (produced by $H\cos\theta$) and parallel (produced by $H\sin\theta$) to the CuO_2 planes give rise to dissipation. However, for $H\cos\theta \gg \gamma^{-1}H\sin\theta(\tan\theta \ll \gamma)R_{FF}$ is given only by the dissipation of the out-of-plane vortices, hence it scales with $H\cos\theta$, while for $\tan\theta \sim \gamma$ one cannot neglect the $H\sin\theta$ term, i.e., the total dissipation is given by both types of vortices. Hence, R_{FF} scales with $H\cos\theta$ for small θ while the scaling breaks down and the second term in Eq. (1) becomes important beyond a certain θ that depends on γ. Therefore, for the same $H\cos\theta$, the value of R_{FF} that is measured during angle-scanning protocol is always equal to or larger than its value measured during field-scanning protocol with $H\|c(\theta=0°)$.

Figure 2(b) is a plot of R vs $H\cos\theta$ measured on a trilayer at 10 $K<T_c$ using the same two protocols discussed above. The resistance for the θ-scanning protocol (symbols) is *less* than the resistance for the H-scanning protocol (solid curve) over a wide $H\cos\theta$ range. This behavior is observed at all T down to the lowest measured value of 1.8 K. This is opposite to what was just discussed for the YBCO thin film [Fig. 2(a)]. In addition, the insets to Figs. 2(a) and 2(b), which represent same measurement for the YBCO thin film and trilayer, respectively, show again the just discussed discrepancy.

Lower R in the θ-scanning protocol than in the H-scanning protocol is incompatible with vortex dissipation [Eq. (1)]. Therefore, the lower R value for the $(60°,2H)$ than

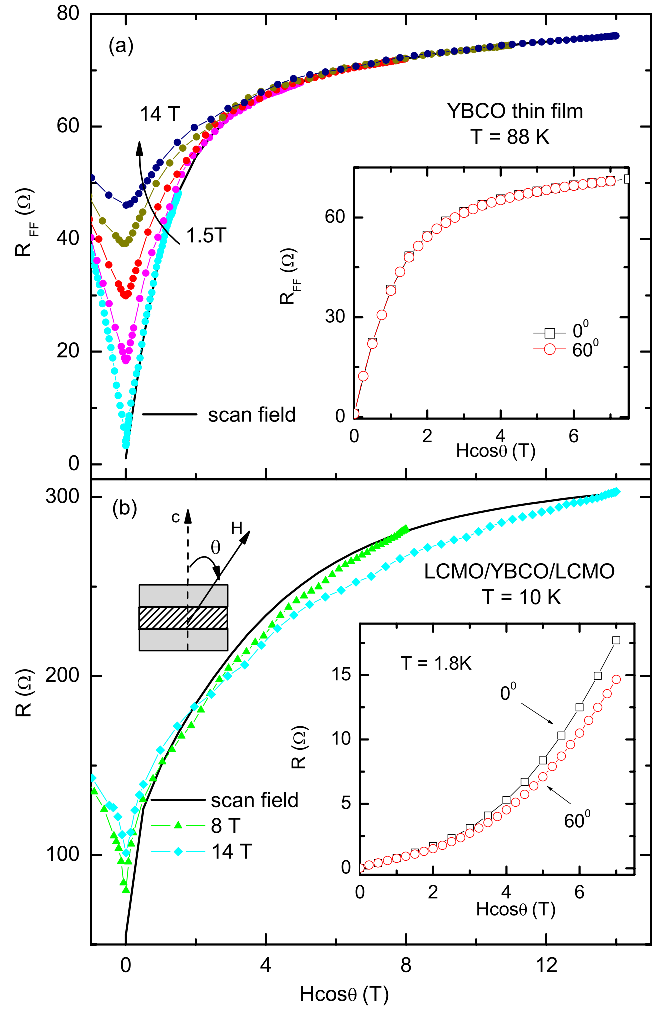

FIG. 2. (Color online) (a) Flux-flow resistance R_{FF} vs the component $H\cos\theta$ of the applied magnetic field H along the c axis $(\theta=0°)$ of a $YBa_2Cu_3O_{7-\delta}$ thin film measured in the mixed state (88 K) while scanning $H\|c$ axis (solid curve) or θ (symbols) in an H of 1.5, 5, 8, 11, and 14 T. Inset: plot of R_{FF} vs $H\cos\dot{\theta}$ measured at 0 and 60° while scanning H up to 14 T. (b) Resistance R vs $H\cos\theta$ of a trilayer measured in the mixed state (10 K) using the same two protocols. Upper Inset: sketch of the trilayer showing the definition of the angle θ. Lower Inset: plot of R vs $H\cos\theta$ measured at 0 and 60° on the trilayer while scanning H up to 14 T.

the $(0°,H)$ data (such that $H\cos\theta$ is same) of inset to Fig. 2(b) could only be the result of the negative magnetoresistance of the ferromagnetic LCMO. This indicates that the resistances of the LCMO and YBCO are *comparable* in the mixed state so that the negative magnetoresistance of LCMO is measurable.

In the following, we eliminate the contribution of YBCO to the measured dissipation in order to obtain the dissipation in the LCMO layer. The thickness of each layer of the trilayer is much smaller than its length or width, therefore the current density is homogenous and its conductance R^{-1} is the sum of the conductances R_L^{-1} and R_Y^{-1} of the LCMO and YBCO layer, respectively. Since the resistance of YBCO for $\theta=0°$ and 60° scales with $H\cos\theta$ in the mixed state [inset to Fig. 2(a)] and its magnetoresistance is negligible in the nor-

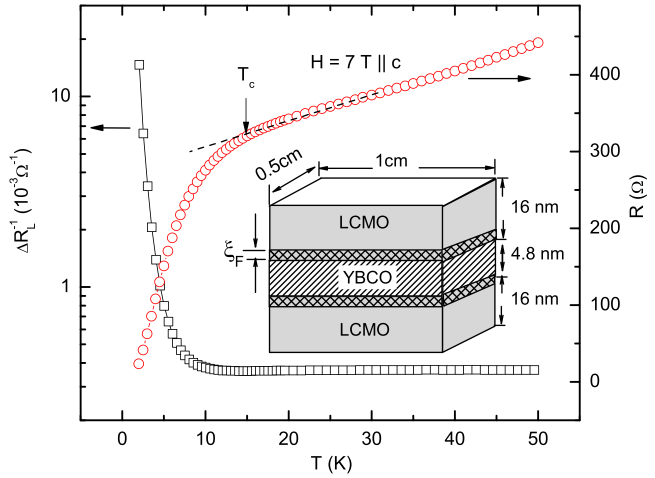

FIG. 3. (Color online) Plots of the temperature T dependences of ΔR_L^{-1} as defined by Eq. (2) and of its resistance R measured in 7 T applied along the c axis. Inset: sketch of the trilayer.

mal state, we eliminate the contribution of YBCO to the measured R^{-1} of the trilayer by taking the difference between the trilayer conductances measured at $(60°,2H)$ and $(0°,H)$, such that $H\cos\theta$ is same; i.e.,

$$\begin{aligned}\Delta R_L^{-1}(H,T) &= R^{-1}(60°,2H) - R^{-1}(0°,H) \\ &= [R_Y^{-1}(60°,2H) + R_L^{-1}(60°,2H)] \\ &\quad - [R_Y^{-1}(0°,H) + R_L^{-1}(0°,H)] \\ &= R_L^{-1}(60°,2H) - R_L^{-1}(0°,H). \end{aligned} \tag{2}$$

Hence, $\Delta R_L^{-1}(H,T)$ only reflects the transport properties of the LCMO layers of the trilayer. We note that the assumption $R_Y^{-1}(0°,H)=R_Y^{-1}(60°,2H)$ for the YBCO of the trilayer is based on the finding[16] that the YBCO in the trilayer is even more two dimensional than the YBCO thin film, hence the scaling holds over an even wider θ range for the former.

Figure 3 is a plot of $\Delta R_L^{-1}(T)$ obtained from data measured at $(60°,14\ \text{T})$ and $(0°,7\ \text{T})$. Notice that $\Delta R_L^{-1}(T)$ is positive over the whole measured T range, it is almost constant for $T>T_c$, and it increases by almost two orders of magnitude for $T<T_c$. The *positive* $\Delta R_L^{-1}(T)$ for both $T>T_c$ and $T<T_c$ shows that the conductance of LCMO increases with increasing H which reflects the negative magnetoresistance of the ferromagnetic LCMO layers. The two orders of magnitude increase in $\Delta R_L^{-1}(T)$ below T_c reflects the presence of superconductivity in the LCMO layers. Under this scenario, superconducting pairs are induced into the LCMO layers over an effective penetration depth ξ_F, producing a significant increase in the conductance of these layers (see inset to Fig. 3 for a sketch) most likely through the normal metal/superconductor proximity-induced Josephson effect.[17] Moreover, as shown below [Eq. (3) and the discussion that follows], the dissipation in this ξ_F layer below T_c is due to flux vortices with ferromagnetic cores.

An enhanced conductance has previously being observed in F/S heterojunctions[18,19] (with F=Co or Ni). Bergeret *et al.*[5] explained these experimental data based on triplet pairing induced in the ferromagnet. Therefore, the present enhancement of conductance of LCMO is consistent with triplet superconductivity induced in LCMO.

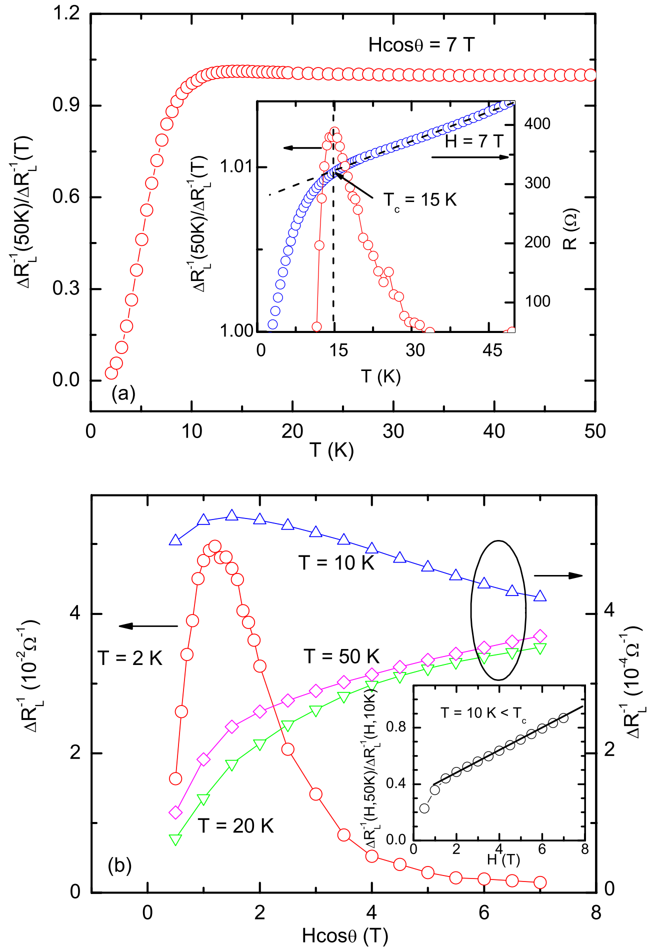

FIG. 4. (Color online) (a) Plot of $\Delta R_L^{-1}(50\ \text{K})/\Delta R_L^{-1}(T)$ as defined by Eq. (2) vs temperature T measured in $H=7\ \text{T}$. Inset: plots of $\Delta R_L^{-1}(50\ \text{K})/\Delta R_L^{-1}(T)$ (left y axis) and resistance of the trilayer R (right y axis) vs T in the transition region, both measured in 7 T. (b) Plot of ΔR_L^{-1} as defined by Eq. (2), measured with H up to 14 T and at four different T. Inset: plot of $\Delta R_L^{-1}(H,50\ \text{K})/\Delta R_L^{-1}(H,T)$ vs H.

Recent neutron reflectometry measurements revealed two possible magnetic profiles at the YBCO/LCMO interface:[1] (a) a magnetic moment induced in the YBCO layer, which is antiparallel to the ferromagnetic moment in the LCMO, and a 1 nm thick suppressed ferromagnetic moment in the LCMO layer or (b) a magnetic dead layer (zero net magnetic moment) in the LCMO layer near the YBCO/LCMO interface. The present finding of a *negative* magnetoresistance below T_c in the LCMO layer is consistent with option (a).

If, as shown by Fig. 3, normal-state ferromagnetism and superconductivity coexist in the layer of thickness ξ_F below T_c, a reasonable scenario is that the dissipation in this region is given by flux vortices with ferromagnetic cores; i.e., based on Eq. (1), the resistance $R_{L,sc}(T)$ of the LCMO region of thickness ξ_F at $T<T_c$ and for $\theta\leq 60°$ is given by

$$R_{L,sc}(H,T,\theta) = \frac{d}{\xi_F} R_{L,n}(H,T) f(H\cos\theta,T), \qquad (3)$$

where f scales with $H\cos\theta$ (Ref. 20) and $(d/\xi_F)R_{L,n}$ is the normal-state resistance of the layer of thickness ξ_F, with $R_{L,n}$ the normal-state resistance of the whole LCMO layer of thickness $d=16$ nm. In the next three paragraphs we show that the experimentally-measured dissipation in the LCMO layers at $T<T_c$ is, indeed, vortex dissipation given by Eq. (3).

Equations (2) and (3) give

$$\frac{\Delta R_{L,n}^{-1}(H,T)}{\Delta R_{L,sc}^{-1}(H,T)} = \frac{d}{\xi_F} f(H,T,0°) = \frac{R_{L,sc}(H,T,0°)}{R_{L,n}(H,T)} \qquad (4)$$

for measurements at $(60°,2H)$ and $(0°,H)$, i.e., for all the data of the figures that follow. This relationship shows that the experimentally obtained $\Delta R_{L,n}^{-1}(H,T)/\Delta R_{L,sc}^{-1}(H,T)$ [left-hand side of Eq. (4)] is the normalized out-of-plane vortex dissipation in LCMO [right-hand side of Eq. (4)]. Since $\Delta R_{L,n}^{-1}$ is almost T independent (see Fig. 3), we plot in Fig. 4(a) $\Delta R_L^{-1}(50\text{ K})/\Delta R_L^{-1}(T)$ vs T measured at $(60°,14\text{ T})$ and $(0°,7\text{ T})$, i.e., $H\cos\theta=7$ T. The resistance of LCMO is almost constant at $T>T_c$, it displays a sharp peak at T_c [see inset to Fig. 4(a)], and it decreases significantly at $T<T_c$, typical to the resistance of a superconductor in the mixed state due to vortex dissipation. A similar peak at T_c has also been found in other F/S heterostructures, and it has been attributed to an interface resistance between the ferromagnet and the superconductor.[19,21]

Based on Eq. (4), $\Delta R_{L,sc}^{-1}(H)$ should be a nonmonotonic function of H for $T<T_c$ since $1/f(H\cos\theta,T)$ should decrease and $\Delta R_{L,n}^{-1}(H)$ increase (negative magnetoresistance in the LCMO layers, see Fig. 1) with increasing H. This result is, indeed, shown by Fig. 4(b). At $T>T_c$ (e.g., 20 and 50 K), $\Delta R_L^{-1}(H\cos\theta)$ increases with increasing H, and it is almost T independent. At $T<T_c$, $\Delta R_L^{-1}(H\cos\theta)$ becomes nonmonotonic with H. Hence, Fig. 4(b) shows that Eq. (4), hence the assumption given by Eq. (3) is correct.

Further support of the fact that the dissipation in LCMO for $T<T_c$ is due to vortices and given by Eq. (3) is provided by the inset to Fig. 4(b); $f(H,T,0°)$ should increase and be linear in H up to a certain H value. The inset to Fig. 4(b) is a plot of the H dependence of $\Delta R_L^{-1}(H,50\text{ K})/\Delta R_L^{-1}(H,10\text{ K})$, which is proportional to $f(H,10\text{ K},0°)$ [see Eq. (4)], obtained by scanning H at 0 and 60° and at 10 and 50 K. Indeed, f increases with H and is linear in H.

In summary, the previous three paragraphs showed that dissipation measured in the LCMO layer at $T<T_c$ is vortex dissipation, given by Eq. (3). The picture that emerges is that at $T<T_c$ the resistance of the LCMO of thickness ξ_F is due to the dissipation of highly spin-polarized quasiparticles [polarization $P\geq 0.8$ (Refs. 22–24)] present in the out-of-plane vortex cores. This is the second evidence for triplet superconducting condensation present in LCMO. In fact, Ping Niu and Xing[25] recently showed that triplet superconducting correlations appear in this system for $P\geq 0.8$.

As a note, Eq. (4) [with the value 0.49 of the normalized resistance at 2 T from inset to Fig. 4(b), $d=16$ nm, and $f\approx H/H_{c2}\approx 1/7$] gives $\xi_F\approx 4.7$ nm at 10 K. This estimate is in excellent agreement with a previously reported value[10] and further shows the consistency of the whole analysis presented here.

In summary, the conductance of the LCMO layers of LCMO/YBCO/LCMO trilayers increases by two orders of magnitude below T_c of the trilayer, which is evidence that triplet correlations are induced in LCMO over an effective penetration depth ξ_F producing this significant increase in conductance. The dissipation present in the LCMO layers of thickness $\xi_F\approx 4.7$ nm at 10 K is due to vortex motion with highly spin-polarized quasiparticles in the vortex cores. This further shows that triplet superconducting pairs are induced in the LCMO layers.

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