Paramagnetic Meissner effect in $YBa_2Cu_3O_7/La_{0.7}Ca_{0.3}MnO_3$ superlattices

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We report the observation of a paramagnetic contribution to the field cooled magnetization that develops below the superconducting transition temperature T_c in measurements performed on $YBa_2Cu_3O_7/La_{0.7}Ca_{0.3}MnO_3$ superlattices. The effect has been detected only for samples whose manganite layers are *magnetically* granular. We discuss our observation of this paramagnetic Meissner effect (*Wohlleben effect*) in terms of an inhomogeneous superconducting state at the interface with the magnetically granular manganite layers.

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A positive field cooled magnetization developing below T_c, the so-called *paramagnetic Meissner* or *Wohlleben effect*, has been occasionally observed in some superconducting samples.[1–3] As it was first found in high-T_c superconductors, an explanation was proposed which considered the existence of an unconventional *d*-wave pairing state leading to the appearance of spontaneous supercurrents in the superconductor due to the presence of "π boundaries",[4,5] which give rise to a paramagnetic contribution to the susceptibility. Following this interpretation, the samples showing a paramagnetic Meissner effect (PME) would consist of a network of Josephson junctions formed at weak links between superconducting grains. The spontaneous orbital supercurrents, which are responsible for the onset of a paramagnetic magnetization below T_c (Ref. 6), are a consequence of the symmetry of the superconducting pairing.[4] Nevertheless, the observation of this effect in different types of niobium samples and Josephson junctions[3,7–9] implies that it is not necessarily linked to an unconventional mechanism for superconductivity. Thus alternative models based on flux trapping and compression effects by Lorentz forces were proposed by different authors.[10,11] An inhomogeneous superconducting transition can be the origin of flux compression. If the edges of the sample become superconducting first, vortices will be excluded from this region. On field cooling, the flux-free region expands, resulting in further flux compression. Once the whole sample becomes superconducting, the compressed flux state gives rise to a paramagnetic signal. Nevertheless, theoretical arguments have been put forward suggesting that flux compression is not essential for the existence of PME.[12]

Thus at present there is no agreement about the origin of this effect. It is worth pointing out that, although the PME appears only in a few samples, a strong sensitivity to the surface microstructure has been reported in every case.[3,13] This suggests that vortex pinning by surface defects, an important source of flux pinning in high-T_c single crystals[14] and epitaxial thin films, must play a significant role in the development of PME.

In this work we report the observation of a paramagnetic Meissner effect in $YBa_2Cu_3O_7/La_{0.7}Ca_{0.3}MnO_3$ (YBCO/LCMO) superlattices. The possible correlation of this behavior with the magnetic state of the manganite layers will be discussed. In light of our results we will consider the plausibility of explanations in terms of current models of the paramagnetic Meissner effect taking into account flux compression effects.

YBCO/LCMO multilayered samples were grown on (100) oriented $SrTiO_3$ substrates using a high-pressure (3.4 mbar) pure oxygen sputtering system at high growth temperatures (900 °C). This technique allows the production of high-T_c epitaxial thin films and superlattices with the *c*-axis oriented perpendicular to the film surface. For the samples studied in this work, x-ray diffraction (XRD) and transmission electron microscopy (TEM) experiments were performed showing atomically sharp interfaces with little step disorder and negligible interdiffusion.[15,16] For every sample the transition temperature was determined from results of supplementary electrical resistivity measurements performed in a closed cycle refrigerator. Hysteresis loops, dc magnetization and ac magnetization versus temperature were measured using a Quantum Design MPMS superconducting quantum interference device magnetometer. dc magnetization measurements were performed following the usual zero field cooled (ZFC) and field cooled (FC-W) procedures. Different runs were performed for magnetic applied fields ranging between 50 and 1500 Oe. All the experiments reported in this work were performed with the field parallel to the surface of the films ($H \| a$-b plane).

In Fig. 1(a) we display the results of ZFC and FC magnetization measurements, performed with a dc magnetic field of 50 Oe, on a $[YBCO_{12u.c.}/LCMO_{10u.c.}]\times 6$ superlattice, which is equivalent to 14 and 4 nm thickness of the YBCO and LCMO layers, respectively. The FC curve displays an *increase* of the magnetization that takes place just below the superconducting transition at T_c (marked by the sharp decrease of the ZFC susceptibility towards negative values).

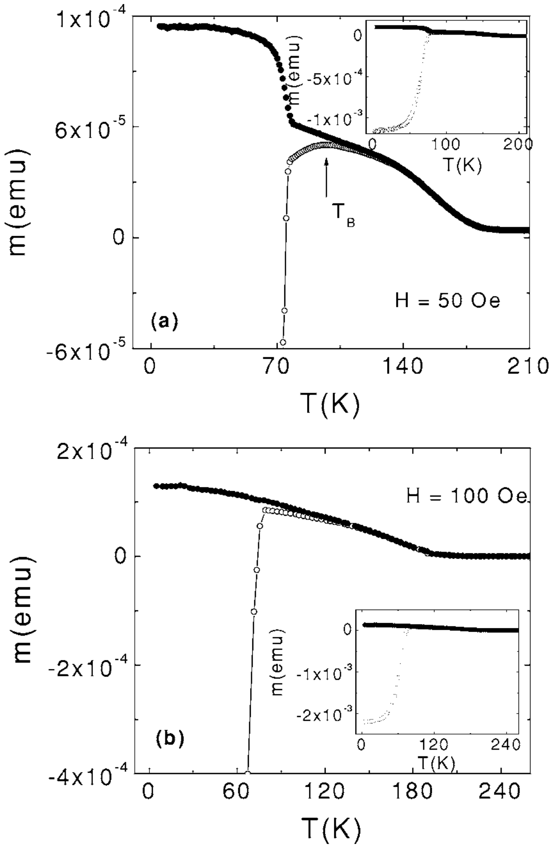

FIG. 1. (a) ZFC (open symbols) and FC (solid symbols) magnetic moment vs T, measured with an applied field of 50 Oe oriented parallel to the sample plane, for the superlattice sample $[YBCO_{12u.c.}/LCMO_{10u.c.}]\times 6$, which shows PME at T_c. Arrow marks the blocking temperature, T_B. (b) ZFC (open symbols) and FC (closed symbols) magnetic moment vs T, measured with an applied field of 100 Oe oriented parallel to the sample plane, for the superlattice sample $[YBCO_{15u.c.}/LCMO_{15u.c.}]\times 6$. This sample does not show PME. Insets show an expanded y axes scale to display the whole superconducting transition.

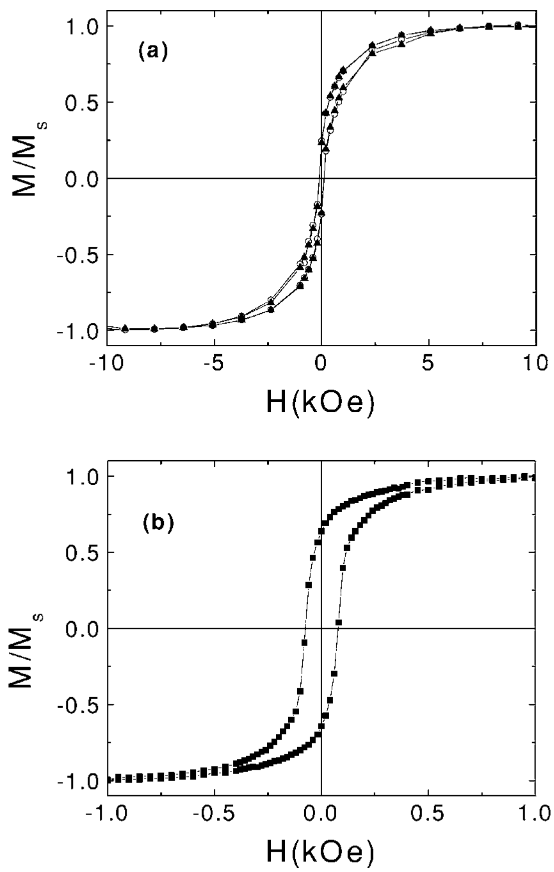

FIG. 2. (a) Hysteresis loops of samples $[YBCO_{12u.c.}/LCMO_{10u.c.}]$ (triangles) and $[YBCO_{8u.c.}/LCMO_{10u.c.}]$ (circles), which show PME, and of sample $[YBCO_{15u.c.}/LCMO_{15u.c.}]$ [Fig. 2(b)] which does not. Notice the different scale of the H axis for both plots.

We have found clear indications of this paramagnetic effect in other multilayered samples with similar thickness of the LCMO layers. It is important to note that in single YBCO films of the same thickness as in these superlattices we never observed a paramagnetic enhancement of the magnetization at T_c.

We show in Fig. 1(b) the results of measurements performed on a $[YBCO_{15u.c.}/LCMO_{15u.c.}]\times 6$ superlattice with thicker manganite, with 18 and 6 nm thick YBCO and LCMO layers, for which no PME is observed. Notice that both samples have similar T_c values despite that the thickness of individual layers is different. A comparison between the samples that display the effect and those that do not indicates that the PME seems to occur only for samples showing a marked degree of *magnetic* granularity and/or weakened magnetic properties of the manganite layers. In the case of sample $[YBCO_{12u.c.}/LCMO_{10u.c.}]$, a strong irreversibility between the ZFC and FC curves is observed below $T\sim 100$ K. Similar or even stronger irreversibility is observed for other samples that display PME (not shown), whereas it is much less pronounced for $[YBCO_{15u.c.}/LCMO_{15u.c.}]$ [Fig. 1(b)]. To further illustrate on this point, in Fig. 2 we present hysteresis loops measured at temperatures intermediate between the superconducting and magnetic transition. In Fig. 2(a) we display data corresponding to two samples, $[YBCO_{12u.c.}/LCMO_{10u.c.}]$ and another sample with thinner YBCO but the same manganite thickness $[YBCO_{8u.c.}/LCMO_{10u.c.}]$, both of them showing PME. The loops, measured at $T=85$ K, present features characteristic of granular systems, as a slow approach to saturation and a reduced value of the remanence, which point to a significant contribution to the magnetization coming from superparamagnetic moments of small ferromagnetic clusters. Notice that the FC curve displayed in Fig. 1(a) shows a maximum at a temperature close to 95 K [not observed in Fig. 1(b)], that could be interpreted as the blocking temperature of those superparamagnetic moments. Moreover, these samples show reduced values of the saturation magnetization (as low as 30 emu/cm^3 for sample $[YBCO_{8u.c.}/LCMO_{10u.c.}]$, consistent with coexisting ferromagnetic and nonferromagnetic phases. In contrast, the loop measured at $T=80$ K for sample $[YBCO_{15u.c.}/LCMO_{15u.c.}]$ [Fig. 2(b)] shows a faster approach to saturation as well as higher remanence and saturation magnetization of 100 emu/cm^3. Notice also that the hysteresis loops corresponding to $[YBCO_{12u.c.}/LCMO_{10u.c.}]$ and $[YBCO_{8u.c.}/LCMO_{10u.c.}]$ display irreversibility up to

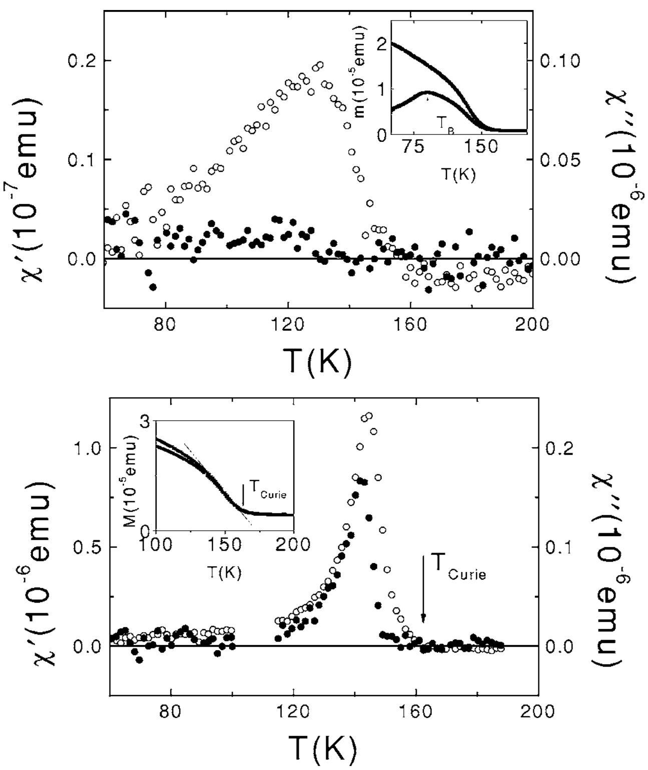

FIG. 3. Temperature dependence of the real (closed symbols) and imaginary (open symbols) components of the ac susceptibility, for $[YBCO_{8u.c.}/LCMO_{10u.c.}]$ (a) and $[YBCO_{15u.c.}/LCMO_{15u.c.}]$ (b) samples, measured with 1 Oe ac field and a frequency of 3 Hz. Insets show ZFC and FC magnetic moment vs T.

field values (5000 Oe) significantly higher than those of $[YBCO_{15u.c.}/LCMO_{15u.c.}]$, which exhibits saturated and reversible magnetization at a field of 1000 Oe. An additional evidence of the relationship between the inhomogeneity of the magnetic state in the manganite layers and the paramagnetic effect in the superconductor is obtained from ac susceptibility measurements, shown in Fig. 3. They were performed at a frequency of 3 Hz with a 1 Oe ac applied field. In the plots we display both components, χ' and χ'', as a function of temperature, for samples with thin manganite layer $[YBCO_{8u.c.}/LCMO_{10u.c.}]$ [Fig. 3(a)], which shows PME below the superconducting transition, and $[YBCO_{15u.c.}/LCMO_{15u.c.}]$ with thicker LCMO layers, which does not [Fig. 3(b)]. Whereas the second plot displays a well-defined peak in both components, due to the Hopkinson effect,[17] in the first one the peak in the real component χ' is extremely broad, and only a very weak upturn of χ'' is observed. The Hopkinson peak in ferromagnets, which occurs at temperatures slightly lower than the Curie temperature T_c, is due to the contribution to the initial magnetization of incoherent rotations of the magnetization as the system approaches the ferromagnetic critical temperature. In homogeneous bulk samples it is a narrow feature. The peaks displayed in Fig. 3(b) occur below the bulk Curie temperature, $T_c = 163$ K, estimated from the dc measurement [see inset to Fig. 3(b)]. The broad peak in χ' observed for sample $[YBCO_{8u.c.}/LCMO_{10u.c.}]$ reflects the ill-defined magnetic transition of a *magnetically* granular or inhomogeneous sample. We want to point out that this sample exhibits a blocking temperature around 90 K. The same blocking temperature was shown by sample $[YBCO_{12u.c.}/LCMO_{10u.c.}]$ which also displays PME effect (see Fig. 1). From the results shown in Fig. 3(a) it can be also concluded that in a sample showing PME effect, the superconducting transition takes place when there is still a significant incoherent movement of domains in the manganite layers.

Our results show a paramagnetic Meissner effect (PME), taking place clearly right at T_c, in some of our YBCO/LCMO superlattices. It seems to appear only when the manganite layers are *magnetically granular*. For all the samples, the magnetization measurements were performed for magnetic fields applied *parallel* to the film surface. The following task is to find out if they are consistent with any of the current models of the paramagnetic effect in superconductors.

Previous experimental evidence points to a significant role of the sample surface on the developments of PME in superconductors. PME has been observed to disappear after removing the surface of single-crystal YBCO and Nb samples.[3,13] Theoretically, this is more easily explained in terms of flux trapping and vortex compression models rather than of "π boundaries" in a d-wave superconductor. The first models imply the formation, as a consequence of flux compression, of a giant vortex state at the surface, and thus vortex pinning at the surface must play a role, although this is an unexplored issue. As it was pointed out above, models based in unconventional pairing state do not explain the observation of PME in conventional superconductors. On the other hand, the high-field PME found in melt-textured high-T_c samples has been related to pinning effects due to secondary phases, maybe magnetic.[18] Focusing on our case, the magnetic layers would be responsible for pair-breaking effects and a reduction of T_c at the FM/SC interface due to ferromagnetic/superconducting proximity effect. This provides an interesting scenario for models of the PME relying on flux compression due to an inhomogeneous superconducting transition.[10] Since the bulk of the layers have a higher T_c than the region close to the interface, significant flux trapping (compression) may occur close to the interface, providing an origin for paramagnetic pinning currents as described by Koshelev and Larkin in Ref. 10. However, while PME shows up in samples $[YBCO_{12u.c.}/LCMO_{10u.c.}]$ and $[YBCO_{8u.c.}/LCMO_{10u.c.}]$ with thin (10 unit cells thick) manganite layers, samples with somewhat thicker LCMO (15 unit cells), as shown for sample $[YBCO_{15u.c.}/LCMO_{15u.c.}]$, do not exhibit the PME. It is obvious that any surface effect that could trigger a PME would be dramatically enhanced with respect to bulk and single-crystalline samples. As it has already been pointed out, we only found a clear PME in the more magnetically disordered samples. This experimental fact points to a peculiar interplay between magnetism and superconductivity at the interfaces of SC/FM superlattices as the source of the PME that can be detected in our experimental conditions when the magnetic layers are thin enough. The interface region is always disordered to some extent, and it reflects in the magnetically granular character observed when the manganite layers are thin enough. It is known that epitaxial strain[19] may induce phase separation in manganites,[20,21] and in fact, this is most likely the origin of the depressed magnetization of the thinner LCMO layers in [$YBCO_{12u.c.}$/$LCMO_{10u.c.}$] and [$YBCO_{8u.c.}$/$LCMO_{10u.c.}$] samples. Due to the strain induced phase separation the thinner manganite layers are a disordered mixture of paramagnetic (insulating) and ferromagnetic (conducting) regions, as evidenced by reduced saturation magnetizations as low as 30 emu/cm^3 for sample [$YBCO_{8u.c.}$/$LCMO_{10u.c.}$], to be compared to the 100 emu/cm^3 value found for the thicker manganite layers. As a consequence, there would be a laterally inhomogeneous superconducting state in the region close to the FM/SC interface, with a distribution of T_c values over nanometer length scales. Besides, the ferromagnetic regions close to the interface may act as strong pinning centers. For thicker manganite layers strain relaxation occurs above a critical thickness, so the samples become more (magnetically) homogeneous and the ferromagnetic fraction (magnetization) increases. It seems that the appearance of the PME is related to this lateral magnetic inhomogeneity of the samples. Due to proximity effect the order parameter penetrates into the ferromagnet and superconductivity is depressed within the superconductor over the Ginzburg Landau coherence length.[22,23] For magnetically homogeneous LCMO layers suppressed proximity effect is expected due to the high degree of spin polarization of the manganite,[24] and accordingly, superconductivity will be homogeneous over the YBCO layer. However, for the (laterally) magnetically inhomogeneous thin manganite layers reduced spin polarization is expected and thus proximity effect will take place, especially at the interface with the nonmagnetic regions. As a consequence, superconductivity will be depressed in a nonhomogeneous fashion, providing a feasible scenario for the PME.

In view of our results and the discussion put forward above, it seems very likely that surface pinning effects are at the roots of PME in YBCO/LCMO superlattices, as it has been suggested for other conventional or high-T_c superconductors. In our opinion, it makes models based on flux trapping and vortex compression the most plausible option to explain our experimental observations. The peculiarity of our case is that PME would be triggered by the magnetic properties of the interfaces, not by structural defects or compositional inhomogeneity, as it seems to be the case for melt-textured or single-crystalline high-T_c samples. This is probably also the case of the PME found in Co/Nb multilayers (see Ref. 25).

In summary, we have found clear experimental evidence of a paramagnetic effect in epitaxial YBCO/LCMO superlattices right below the superconducting temperature T_c. The appearance of the effect is related to the degree of magnetic granularity of the manganite layers that gives rise to an inhomogeneous superconducting state in the region close to the FM/SC interface. This leads to flux compression below T_c and a subsequent PME that can be detected if the layers are thin enough.

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