Very Large Purely Intralayer Critical Current Density in Ultrathin Cuprate Artificial Structures

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Ultrathin artificial high temperature superconducting structures, consisting of $(Ba_{0.9}Nd_{0.1})CuO_{2+x}$ and $CaCuO_2$ layers, were grown by pulsed laser deposition. Intralayer superconductivity at 60 K was obtained for a structure consisting of a single $(CaCuO_2)$ block sandwiched between two $(Ba_{0.9}Nd_{0.1})CuO_{2+x}$ charge reservoir blocks. The purely intralayer critical current density was measured at 4.2 K and resulted to be larger than 10^8 A/cm^2. These findings clearly show that interaction between nearest neighbor $(CaCuO_2)$ layers is not essential for high T_c superconductivity and strongly supports the physical model based on the idea that intralayer interaction alone is responsible for high temperature superconductivity.

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Layer by layer deposition techniques have been recently applied to the synthesis of artificial high temperature superconducting heteroepitaxial structures [1,2]. Such artificial structures are engineered by depositing, in a suitable stacking sequence, infinite layer (IL) blocks and charge reservoir (CR) blocks. The IL block consists, as for conventional cuprates, of CuO_2 planes separated by Ca^{+2} ions. The CR block consists of an epitaxial $Ba_{0.9}Nd_{0.1}CuO_{2+x}$ layer. Charge carriers (holes) are transferred from the CR layers to the IL layers giving rise to superconductivity in the otherwise insulating CuO_2 planes.

Artificial high temperature superconducting structures have allowed a new approach to study the correlations existing between superconductivity and structural features [3–8]. Recently, superconductivity of a single IL layer (intralayer superconductivity) has been reported in ultrathin artificial structures [1]. Such a problem is of crucial importance to understand the physical mechanisms of high temperature superconductivity. Some physical models foresee that intralayer interaction alone can result in high temperature superconductivity [9,10], while other models imply that interlayer coupling is essential for superconductivity [11]. In the past, the occurrence of intralayer superconductivity was investigated in ultrathin films and superlattices made of conventional high temperature superconductors (HTS) materials [12–23]. Experiments were carried out on superconducting $YBa_2Cu_3O_{7-x}$ (YBCO) ultrathin layers, sandwiched between thick nonsuperconducting $PrBa_2Cu_3O_{7-x}$ (PrBCO) layers [19], on $Bi_2Sr_2CaCu_2O_{8-\delta}$ (BSCCO) films with thickness one-half unit cell [21], on YBCO/PrBCO heterostructures, and on organic-chain $[Py - C_nH_{2n+1}]_2HgI_4$ intercalated $Bi_2Sr_2CaCu_2O_8$ [16]. Most of the reports confirmed the occurrence of purely intralayer superconductivity in ultrathin YBCO and BSCCO films containing a single IL block consisting of two CuO_2 planes [18,19,23]. Suppression of interlayer interaction resulted only in a moderate decrease of the transition temperature [18]. On the other hand, a recent report has appeared which cast doubt on this scenario. Magnetization measurements carried out on the organic-chain $[Py - C_nH_{2n+1}]_2HgI_4$ intercalated $Bi_2Sr_2CaCu_2O_8$ have shown a complete disappearance of superconductivity which was ascribed to the drastic increase in the distance of the next-nearest-neighbor CuO_2 planes in these structures [16]. Disagreement with previous measurements by different authors on similar systems were ascribed to the presence of small amounts of superconducting $Bi_2Sr_2CaCu_2O_8$ phase in the intercalated samples. In our opinion, measurements on artificial high temperature superconducting structures, containing a single IL block, definitely prove the existence of purely intralayer high temperature superconductivity: $(Ba_{0.9}Nd_{0.1}CuO_{2+x})_5/(CaCuO_2)_2/(Ba_{0.9}Nd_{0.1}CuO_{2+x})_5$ heterostructures, containing a single IL block, were shown to have T_c of about 55 K. The T_c of these structures resulted to be only slightly reduced (10%–20%) relative to the T_c of the $(Ba_{0.9}Nd_{0.1}CuO_{2+x})_2/(CaCuO_2)_2$ superlattices where adjacent $(CaCuO_2)_2$ blocks result to be well coupled [2].

A further relevant step, in order to investigate the intrinsic intralayer superconducting properties would be the measurement of the critical current density (J_c) in these artificial structures. However, until now, no attempts have been made in this direction. This is in the most part due to both the difficulty to obtain low resistance electrical contacts on the surface of such ultrathin films, which results in a high thermal dissipation even for quite low injected currents, and to the low thermodynamic stability of these materials, which makes it difficult to define a surface geometry for transport J_c measurements. In this paper, to overcome such difficulties, we apply a contactless inductive method to measure the critical current density in structures which contain a

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single IL block. In [24], it was shown that J_c values obtained by this method are nearly the same as those obtained by transport measurements in the zero applied magnetic field.

In this report, we investigate $(Ba_{0.9}Nd_{0.1}CuO_{2+x})_M/(CaCuO_2)_N/(Ba_{0.9}Nd_{0.1}CuO_{2+x})_M$ ($M/N/M$) heterostructures (where a single IL block, consisting of N $CaCuO_2$ unit cells, is sandwiched between two CR blocks $[(Ba_{0.9}Nd_{0.1}CuO_{2+x})_M]$ and $(Ba_{0.9}Nd_{0.1}CuO_{2+x})_2/(CaCuO_2)_2$ superlattices. Artificial structures with $M = 3$, 5, 10 and $N = 1$, 2 were grown by pulsed laser deposition on $SrTiO_3(001)$ substrates. At the end of the deposition procedure, an amorphous protecting layer of electrically insulating $CaCuO_2$ was deposited on the top of the film (deposition temperature lower than 100 °C) [1]. The resistive transition was measured by the standard four probe technique. Critical currents were measured using the inductive method described in [24]. In this technique, a single coil is pressed against the superconducting film. The coil is driven by an audio frequency sine-wave current ($f_o = 1$ KHz) and the third harmonic component is monitored. For practical driving currents, the applied magnetic field parallel to the film surface is quite small (~ 10 mT) and of the same order as the self-field in J_c transport measurements. A driving current flowing in the coil (I_{coil}) gives rise to a small magnetic field which, in turn, induces a shielding current in the film. When the density of the shielding current becomes equal to the critical value of the supercurrent flowing in the film, a third harmonic signal U_{31} is generated. The value of the coil driving current I^0_{coil}, corresponding to the extrapolation to zero of the linear portion of the U_{31} signal, is linked to the value of critical current density of the film by the formula

$$J_c(\mathrm{A/cm^2}) = K \cdot I^0_{coil}(\mathrm{A})/d(\mathrm{cm}), \qquad (1)$$

where d is the film thickness and K is a calibration factor which, in our case, has a numerical value of $1.885 \times 10^3\ \mathrm{cm^{-1}}$. Formula (1) is based on the assumption that $B = 0$ on the reverse side of the film. This is not so obvious in the present experiment because $d < \lambda$ (London penetration length). However, it has been shown in Refs. [24,25] that the field is attenuated by a factor $\exp(-dD/\lambda^2)$ on the reverse side of the film (D is the coil diameter). Thus, screening is essentially complete when $d > \lambda^2/D$ which is a reasonable condition even in our experimental setup. To further increase the reliability of the J_c measurements, an independent calibration of the apparatus was obtained by a 1 μm thick Nb film, whose transport J_c was known, grown on a substrate having identical in-plane dimensions as those used for the artificial structures. Highest T_c values were obtained for the 5/2/5 and 10/2/10 structures. Thinner CR blocks ($M = 3$) resulted in a decrease of T_c possibly because of the occurrence of interface effects at the substrate/film and film/cap-layer interfaces. Furthermore, structures with $N = 1$ were found to have a lower T_c possibly because of the increased role of structural disorder in such thin IL blocks [26]. Therefore we decided to carry out J_c measurements mostly on 5/2/5 structures and 2×2 superlattices. The modulation length Λ of 2×2 superlattice is 14.9 Å (namely, 8.5 Å + 6.4 Å), while the overall thickness of the 5/2/5 structure is 48.9 Å (namely, 21.25 Å + 6.4 Å + 21.25 Å). In Fig. 1, the behavior of resistance versus temperature is reported for a 5/2/5 structure and a 2×2 superlattice. The T_c (zero resistance value) of the superlattice is just above 70 K, while T_c of the 5/2/5 structure is only slightly lower (about 60 K). The normal state resistance of the two samples scales roughly with their thickness. In Fig. 2, we report a typical inductive J_c measurement (at 4.2 K) for a 1 μm-thick Nb film and a 5/2/5 structure, respectively. In this figure, the amplitude of the third harmonic signal U_{31} is reported as a function of the coil current. J_c was estimated, in both cases, using the criterion given in [24], namely, considering the intercept at zero signal of the extrapolation of the linear portion of U_{31} versus I_{coil}. In the case of the Nb film, using (1), we obtained $J_c \simeq 1\ \mathrm{MA/cm^2}$. Such a value is in agreement with an independent estimate of J_c by transport measurements. From Fig. 2, it can be seen that I^0_{coil} for a typical 5/2/5 structure is roughly one-half of the I^0_{coil} value for the Nb film. This leads immediately to

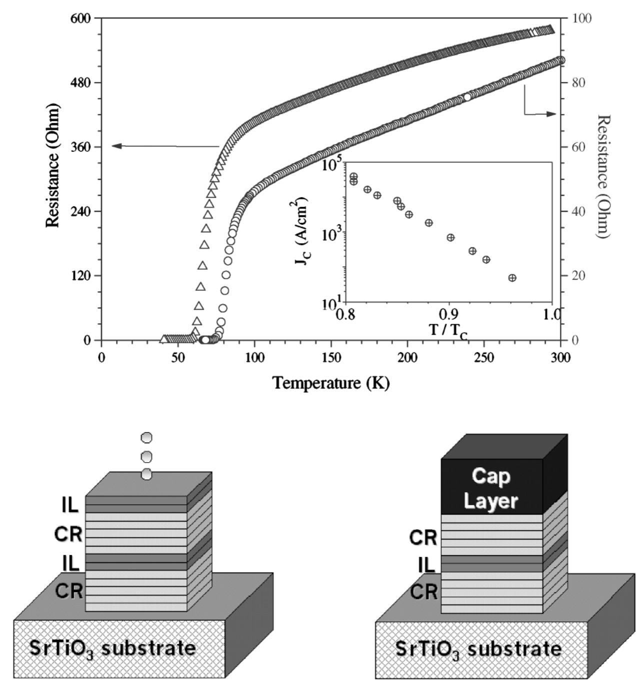

FIG. 1. Resistance vs temperature behavior of a $(Ba_{0.9}Nd_{0.1})CuO_{2+x}/CaCuO_2$ 20-unit-cells-thick 2×2 superlattice (○) and of a 5/2/5 structure (△) is shown. The diagrams corresponding to superlattice and $M/N/M$ structure are roughly sketched below. In the inset, the transport J_c measurement for a 5/2/5 structure in proximity of T_c is shown.

an estimate of the maximum electrical current I_c^{2D} which can flow in a single IL block without interlayer interaction. We obtained $I_c^{2D} \geq 60\ \mathrm{A\,cm^{-1}}$. However, in order to make a comparison with standard HTS materials, we have to rescale I_c^{2D} for the thickness d of the film. This gives a large enhancement factor for the ultrathin structures depending on which we assume to be the effective thickness, either the whole film thickness or the thickness of the IL block. We find that, even in the most conservative hypothesis, namely, supercurrent flowing in the whole film thickness, $J_c \geq 100\ \mathrm{MA/cm^2}$ for the 5/2/5 structure. Furthermore, we have directly measured the residual field on the reverse side of the sample by a mutual inductance method (inset of Fig. 2). In this geometry, the pickup coil is placed in close contact with the reverse side of the sample. In this experimental configuration the field, well below T_c, is reduced to 25% of its value above T_c. Such a residual field is expected to lead to an overestimate of about 25% of the J_c value. However, such an error is less relevant relative to the intrinsic indetermination of the method. From Fig. 2, it can be seen that the third harmonic voltage does not exhibit a sharp onset, but is rounded to some extent. This effect, already noticed in [24], can cast some doubt on the criterion used to extract J_c. However, we show in Fig. 3 that the behavior of U_{31} versus I_{coil} is roughly the same for the Nb film and the 5/2/5 structure (a different sample): The two curves show a reasonable overlap by simply scaling the respective U_{31} and I_{coil} values. A careful inspection of the curves of Fig. 3 shows that the scaling is quite poor at low current levels. This is more evident on a logarithmic scale (inset of Fig. 3). The different low current behavior probably relates to different low current dissipation mechanisms. However, the inset of Fig. 3 shows that, even if the scaling procedure may lead to overestimate J_c somewhat, the major conclusions of this paper are not substantially modified. We stress here that inductive J_c measurements were carried out on several 5/2/5 samples and that the value quoted is close to the average of the values found. The maximum J_c value found at 4.2 K was $\simeq 500\ \mathrm{MA/cm^2}$. To support the inductive measurements, we also carried out transport measurements (with a $5\ \mu\mathrm{V/cm}$ criterion) in a narrow range below T_c (inset of Fig. 1). The high contact resistance (several hundreds of Ohms) and the lack of a defined pattern on the film surface strongly reduce the effectiveness of this approach. However, the value of about $10^5\ \mathrm{A/cm^2}$ found at $T/T_c = 0.8$, obtained for a sample that, according the inductive method, shoved a J_c value at 4.2 K of about $10\ \mathrm{MA/cm^2}$, gives us further confidence in the inductive measurements. Critical current density measurements were also carried out on 2×2 superlattices. For these samples, the value of J_c, estimated according to the procedure outlined above, does not exceed $50\ \mathrm{MA/cm^2}$ (at 4.2 K). The high J_c values (comparable with the values found in the case of good quality YBCO films) and the circumstance that J_c, for the 5/2/5 structure, was higher relative to the superlattices, at first glance can appear to be surprising. In order to understand the physical origin of these findings, we utilize a classical Landau-Ginzburg approach to critical fields and currents in thin superconducting films. It is now commonly accepted that superconductivity in the IL compounds is obtained by means of doping of the "conducting layer" CuO_2 by carriers coming from the $Ba_{0.9}Nd_{0.1}CuO_{2+x}$ block. Moreover, the superconducting properties of the 5/2/5 structure (consisting of one IL block only) were found to be identical to the 5×2 superlattice [2], that confirms the complete decoupling among the IL blocks in $M \times 2$ superlattice for $M > 5$. On the

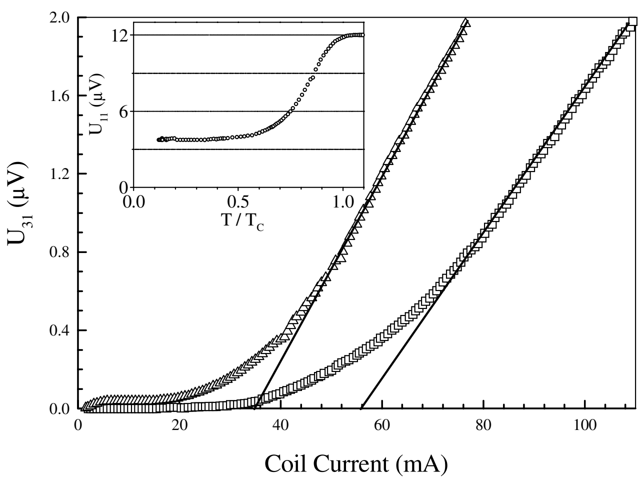

FIG. 2. Third harmonic amplitude as a function of the coil driving current I_{coil} for a 1 μm-thick Nb film (□) and a 5/2/5 structure (△); linear extrapolation for the calculation of I_{coil}^0 is reported also. In the inset, the first harmonic transmitted signal U_{11} is reported as a function of temperature for a 5/2/5 structure.

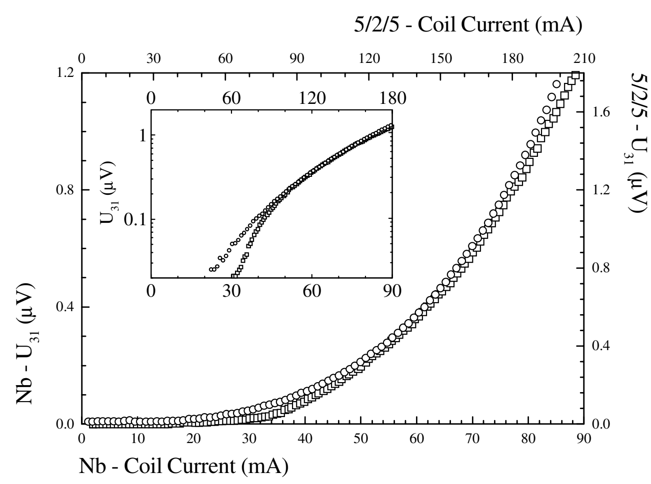

FIG. 3. Scaling of the third harmonic signal for a 1 μm-thick Nb film (□) and a 5/2/5 structure (○). In the inset, the same scaling is shown in a logarithmic plot.

other hand, anisotropy measurements on 2×2 superlattices showed the existence of a strong coupling between nearest neighbor IL blocks. Therefore it seems reasonable to assume that the effective coherence length along the perpendicular direction ξ_c is of the same order of the thickness of a whole 5/2/5 structure [27]. It is well known (see, for example, [28]) that the boundary conditions for the order parameter drastically change the value of critical field H_c, of a thin superconducting film, in comparison with its bulk value $H_{\mathrm{c.m.}}$. For instance, for thin superconducting film ($d \ll \lambda$) of the first kind ($\kappa < 1/\sqrt{2}$),

$$H_c = 2\sqrt{6} H_{\mathrm{c.m.}} \frac{\lambda}{d}. \tag{2}$$

In our case, we deal with a superconductor of the second kind ($\kappa \gg 1$), but the magnetic field is weak and is applied along the layers, so the situation does not much differ from the case of the first kind superconducting film. It is possible to show, in the framework of the Ginzburg-Landau formalism, that even in this case the critical field of a thin film exceeds noticeably its bulk value ($d \ll \xi_c$), being several times larger even for $d \sim \xi_c$ [28]. The increase of the critical field, in accordance with the Silsby rule, is directly related to the increase of the critical current. Physically, it can be said that the requirement of the zero flow at the film boundary ($d\Psi/dz|_{\pm d/2} = 0$) results in an increase, in the interface region, of the value of the order parameter itself in comparison with the value obtained applying periodic boundary conditions (superlattice). As a result, the value of critical current in the monolayer structure can considerably exceed (by a factor $2\sqrt{6}\xi/d$) that of the corresponding superlattice. Of course, the above discussion does not pretend to explain the numerical ratio between J_c values found for the 5/2/5 structure and the 2×2 superlattice but to show only that a moderate increase of J_c can be foreseen in the case of an ultrathin film of very good quality relative to a thicker film of comparable quality.

In conclusion, we have shown that ultrathin artificial structures, containing a single IL layer, cannot only show T_c values exceeding 60 K (purely intralayer superconductivity), but can also support a critical current density of the same order as conventional bulk HTS compounds. Namely, J_c values obtained for 5/2/5 heterostructures at $T = 4.2$ K and zero applied magnetic field are very high ($J_c \gtrsim 10^8$ A/cm^2) and comparable with those reported for good YBCO films at the same temperature. Such a result is even more surprising if we consider that artificial heterostructures have lower T_c values if compared with YBCO (60 and 92 K, respectively). Furthermore, J_c results to be somewhat higher in the case of artificial structures relative to $(Ba_{0.9}Nd_{0.1}CuO_{2+x})_2/(CaCuO_2)_2$ superlattices which have an isostructural IL block. This effect can be explained in a simple Landau-Ginzburg model which takes into account surface effects on the order parameter. The results obtained lead us to conclude that the interlayer coupling between nearest neighbor $CaCuO_2$ layers is negligible for high T_c superconductivity and confirm that intralayer interaction alone can be responsible of high temperature superconductivity.

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[27] Such evaluation gives $\xi_c \sim 1$–2 nm which is noticeably larger than the widely reported values of $\xi_c \leq 0.3$ nm. The latter estimate is deduced from the measurements of H_{c2} near T_c and then by some conventional model (which may not be valid for HTS) is extended to low temperatures. This procedure is not self-consistent since a ξ_c value smaller than the interatomic distance contradicts the basic assumptions of the conventional theory of superconductivity.
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