Enhancement of the superconducting transition temperature of $La_{2-x}Sr_xCuO_4$ and $La_{1.875}Ba_{0.125}CuO_4$ bilayers: Bilayer and reference film prepared on the same wafer

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Recently, a paper was published by Yuli *et al.* [Phys. Rev. Lett. **101**, 057005 (2008)], in which a large T_c enhancement was reported in bilayers of the nonsuperconducting $La_{1.65}Sr_{0.35}CuO_4$ and superconducting $La_{2-x}Sr_xCuO_4$ in the underdoped regime, in comparison with the same bare $La_{2-x}Sr_xCuO_4$ film. This result however, was not reproduced in the present experiments and an effort is made trying to resolve this puzzle. The difference in the present study is that both the bilayer and the bare reference film were prepared in the same deposition run on different parts of the same wafer. This is in contrast to the previous experiments where the bilayers and bare reference films were prepared in different runs and on different wafers. Nevertheless, a small but clear T_c enhancement effect of 1.4–2 K is found in the present study in a similar system of $La_{1.65}Sr_{0.35}CuO_4$-$La_{1.875}Ba_{0.125}CuO_4$ bilayers, which is in line with the theoretical arguments presented previously.

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In a recent paper[1] we reported the observation of a large T_c enhancement in bilayers of the nonsuperconducting, heavily overdoped (OD) $La_{2-x}Sr_xCuO_4$ (LSCO) and superconducting underdoped (UD) LSCO, in comparison with the bare film of the latter. The explanation of this result was based on the idea that in the UD regime of the high-temperature superconductors, T_c is determined by phase fluctuations, while pairing without phase coherence occurs in the pseudogap regime at considerably higher temperatures,[2–7] similar to the case of granular superconductors.[8] In the OD regime however, pairing and phase order occur simultaneously, with a robust phase stiffness. Therefore, in the interface of bilayers composed of UD and OD films, one can envision a scenario in which the high phase stiffness of the OD layer locks via Josephson coupling the phases of the preformed pairs in the UD layer. This together with the high pairing in the UD layer is expected to lead to T_c enhancement above that of both components.[9,10] A similar behavior was reported recently by Gozar *et al.*, where interfacial superconductivity with $T_c \sim 38$ K was found in metallic-insulating $La_{1.55}Sr_{0.45}CuO_4$-La_2CuO_4 bilayers, and enhancement of T_c to values of up to 50 K, exceeding that of optimally doped LSCO, was achieved under ozone oxygenation.[11] In a more recent study however, the present authors found a conventional proximity effect with no T_c enhancement in bilayers of superconducting underdoped $La_{1.88}Sr_{0.12}CuO_4$ islands coated with nonsuperconducting overdoped $La_{1.65}Sr_{0.35}CuO_4$.[12] This observation evidently contradicts our previous T_c enhancement results[1] and we therefore decided to test this issue in the present study by repeating some of these experiments. This was done with a better control over the bilayer and reference film properties, but unfortunately, the former results were not reproduced. The results of these experiments will be presented and discussed in the following, including results on $La_{1.65}Sr_{0.35}CuO_4$-$La_{1.875}Ba_{0.125}CuO_4$ bilayers, where a small but reproducible T_c enhancement effect is observed.

As was described in the abstract, the previous bilayers and reference films were prepared on different wafers at different deposition runs.[1] Although efforts were made to keep the same deposition parameters for all deposition runs, some changes in these parameters could occur. For instance, the laser fluence on the target could vary slightly from run to run as well as the actual temperature of the wafer during deposition due to a slightly different thermal contact to the heater block onto which it was clamped. These could lead to small changes in T_c that could not explain the effect observed in Ref. 1. A more serious source for possible T_c variations could be due to deterioration of some of the $La_{2-x}Sr_xCuO_4$ (LSCO*x*, where *x* is in % units) targets which (when detected) were either resintered or newly prepared. Another possible reason for T_c variations could originate in the different (100) $SrTiO_3$ (STO) substrates, which came from different batches or were sometimes acid cleaned from a previous deposition run. One way to avoid most of these uncertainties in the experiment, is to prepare every bilayer together with its reference film on the same wafer in the same deposition run, as was recently done in Ref. 12. This involves two deposition steps as shown schematically in Fig. 1. First, a reference film is deposited over the whole wafer area of 10×10 mm^2 and then the OD cap film is deposited *in situ* on half the wafer while the other half is covered by a shadow mask. This yields a bilayer and its reference film with minimal variation in the deposition conditions. The results in the following were obtained on such samples, where the two halves of the wafers were separated by either wet acid etching of a 2×10 mm^2 strip from the middle of the wafer (see inset drawing in Fig. 2), or by Ar ion beam milling of ten 10×100 μm^2 microbridges, five on each half of the wafer. In addition, gold contacts were prepared by liftoff to reduce the contact resistance in the standard four-probe transport measurements.

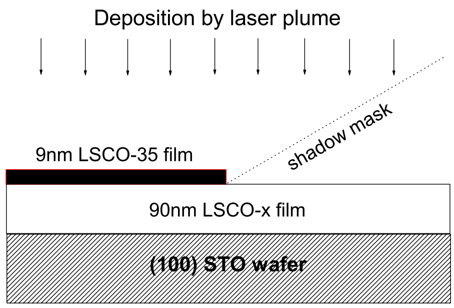

FIG. 1. (Color online) The experimental setup for *in situ* deposition of a bilayer and its reference film on the same wafer.

Figure 2 shows results of the resistance versus temperature of a LSCO35-LSCO12 sample. This sample was separated into its two halves by acid etching in the middle, around the line of mask contact with the wafer, as shown in the inset to this figure. One can see that the bilayer normal resistance is significantly lower than that of the reference film although the thickness of the more conducting LSCO35 layer (9 nm) is only a tenth of that of the base LSCO12 film (90 nm). Their T_c values however, are very close to one another and equal to about 26 K. Clearly, there is no T_c enhancement here, and in order to study the different T_c values in more details, the magnetoresistance (MR) of these samples was measured. Figures 3 and 4 show the results of these MR measurements as a function of temperature at two representative magnetic field values of 0.05 and 0.5 T, respectively. The magnetoresistance at any given temperature is defined here in the standard way as $\mathrm{MR}(H)=R(H)-R(H=0)$. The MR onset signifies the onset of superconductivity at T_c^{onset} with the beginning of flux flow resistance. The MR peak occurs at the maximum flux flow resistance when pinning of vortices starts winning over the flux flow while the MR offset occurs when pinning prevents the flow or creep processes completely. At zero field, it would be convenient to identify $T_c(R=0)$ with the temperature T_p of maximum MR at low magnetic fields although this definition is somewhat arbitrary. Using these definitions, it is obvious from Fig. 3 that T_c^{onset} of the bilayer and reference film are at around 31 and 33 K, respectively. Also the $T_c(R=0)$ values of the bilayer and reference film are found at approximately 26 and 28 K, respectively, so that both T_c values of the reference film are higher by about 2 K than those of the bilayer. Therefore, the results of Ref. 1 [where the $T_c(R=0)$ values of the bilayer and reference film are found at approximately 32 and 21 K, respectively], are not only irreproducible here, but they are also reversed, as a small suppression instead of enhancement of T_c is observed in the bilayer. We note that the present results are also in agreement with those obtained in Ref. 12, where a conventional proximity effect was observed. At a higher field of 0.5 T, Fig. 4 shows that the differences between the corresponding T_c^{onset} and $T_c(R=0)$ of the reference film and the bilayer become smaller (about 1.5 and 0.5 K, respectively) but the above conclusion remains the same. In order to make the comparison with Ref. 1 more complete, a test wafer was prepared with a LSCO35-LSCO12 bilayer deposited over the whole wafer. This yielded a $T_c(R=0)$ value of 26 K, which is exactly the same as the value found on the half-half wafer. Thus, any possibility that the mask in Fig. 1 or the patterning process affect the present results can be ruled out.

To further test the present results, the same experiment was repeated with a bottom layer of LSCO8 in a LSCO35-LSCO8 bilayer. The half bilayer and half reference film wa-

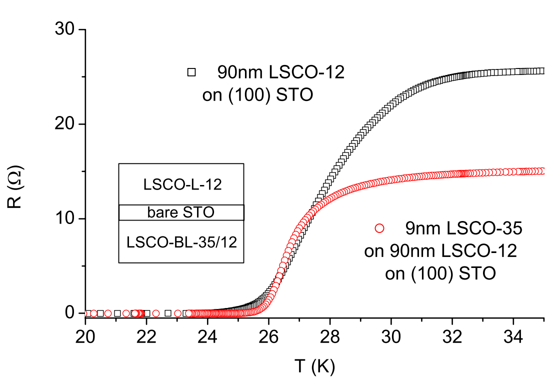

FIG. 2. (Color online) Resistance versus temperature of a LSCO35-LSCO12 bilayer and its reference LSCO12 film on the same wafer. A schematic of the bilayer and reference film layout on the wafer is shown in the inset.

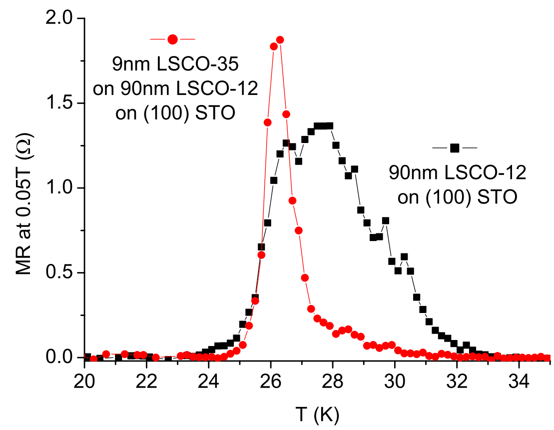

FIG. 3. (Color online) MR at 0.05 T versus temperature of the LSCO35-LSCO12 bilayer and its reference LSCO12 film of Fig. 2.

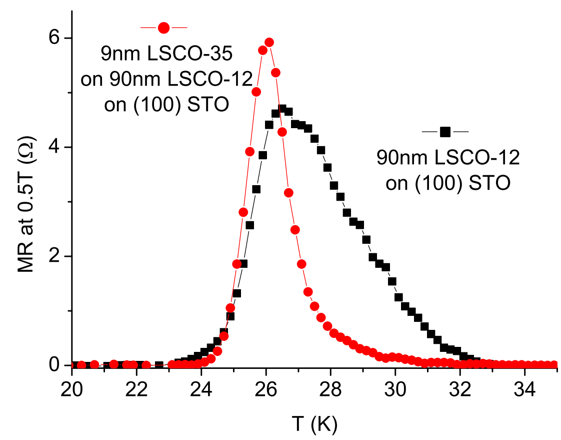

FIG. 4. (Color online) MR at 0.5 T versus temperature of the LSCO35-LSCO12 bilayer and its reference LSCO12 film of Fig. 2.

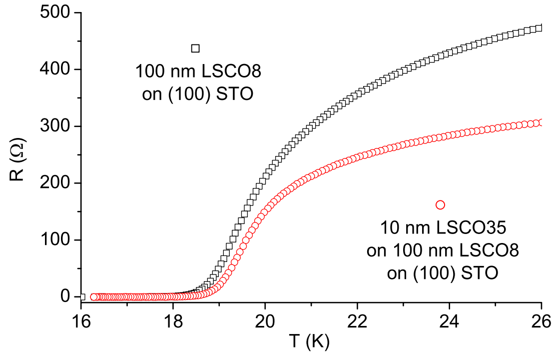

FIG. 5. (Color online) Resistance versus temperature of two typical microbridges, one of the LSCO35-LSCO8 bilayer and the other of the LSCO8 reference film on the same wafer.

fer was patterned this time into ten microbridges of $10 \times 100\ \mu m^2$, five on the LSCO35-LSCO8 bilayer and five on the reference LSCO8 film. The results of the resistance and magnetoresistance versus temperature are shown for two representative microbridges in Figs. 5 and 6, respectively. This time, $T_c(R=0)$ of both the bilayer and reference films are almost the same and equal to about 19 K. This is in contrast to what was found in Ref. 1 where the $T_c(R=0)$ value of the bilayer was 22 K and that of the reference film was 14 K. It is worth noting here that the $T_c(R=0)$ values in the present study (also for the LSCO35-LSCO12 system), are found in between the corresponding values obtained in Ref. 1. It was found that in the same experiment as described in Fig. 1, the use of a LSCO8 target with a longer sintering time affected the results. This yielded a target of greater hardness which produced smoother films with a lower $T_c(R=0)$ value of about 16 K. This should be compared with the 19 K found in the present study while using a softer LSCO8 target, that was also used in Ref. 1 a year earlier. Using this softer target, rougher films are produced with 100–200 nm size grains with 2–3 nm rms roughness for a 100-nm-thick film. This rough morphology certainly increases the *ab* plane coupling at the interface with the cap layer, an important factor that may affect the bilayer T_c.

One possible origin for the discrepancy between the present results, where no T_c enhancement was observed, and the previous results of Ref. 1, is the high sensitivity of the T_c enhancement effect to the interface properties. As was theoretically discussed in Refs. 10 and 18, the interface attributes crucially affect the T_c of a superconductor-normal bilayer. In particular, the T_c of such a bilayer may be either enhanced, reduced, or remain unchanged with respect to that of the bare superconductor layer. It is therefore possible that the above discrepancy results from fluctuations in the interface properties. Another possible reason for our failure to reproduce the previous results, following the discussion above, is the different substrate properties between the two sets of measurements that may yield different strain-induced changes in T_c.[13–15] In particular, some of the substrates used in Ref. 1 could have caused relaxation of the tensile-strain-induced decrease in T_c in the bilayers, in particular, at the interface, and thus to an apparent enhancement effect that is not related to any fundamental electronic effect suggested in Ref. 1. Note that the enhanced bilayer transition temperatures never exceeded the corresponding bulk transition temperatures so this scenario is plausible as well. Unfortunately, we could not characterize the quality of the relevant interfaces at the atomic scale. We do not believe that different overoxygenation conditions can account for the different results since in both cases growth procedures that give rise to over-oxygenation[16] were not used. An important question one should address now relates to the origin of the systematic doping dependence of the T_c enhancement reported in Ref. 1, in relation to the above two scenarios. In the first, interface property related scenario, the special doping dependence can be understood within the model presented in Ref. 1, based on the inhomogeneous, granularlike structure of the cuprates high-temperature superconductors.[17] As the doping is decreased, the areas of high local pairing become more spatially separated and higher quality interface with the metallic layer is needed to Josephson couple them and enhance T_c. At very low doping these local high-pairing regions are separated by larger distances to be efficiently coupled, resulting in a small enhancement only. At low interface quality, the enhancement effect should quench all over the underdoped regime. In addition, within the slave-boson mean-field-theory model of Goren and Altman,[18] the shape of the bilayer superconducting dome structure (including the T_c enhancement and the shift of the dome peak or the apparent "optimal doping" level) strongly depends on the coupling. This in turn, may be related to the degree of in-plane coupling between the *ab* planes of the two layers, which could be affected by the substrate properties that influence the interface morphology in the two sets of experiments.

The systematic doping dependence of the bilayer T_c found in Ref. 1 can be accounted for also within the second scenario of the bilayer strain, proposed above. It was clearly demonstrated that the tensile-strain-induced reduction in T_c of LSCO films on STO takes place mainly in the underdoped regime and is quite negligible in the overdoped regime.[13] Therefore, a relaxation of this effect, due to substrate quality, would lead to an apparent enhancement only in the underdoped regime. The small enhancement, considering the error bar, reported in Ref. 1 for the LSCO6 bilayer may be due to the fact that in this case the reference and bilayer films were grown on substrates of the same batch at consecutive growth runs.

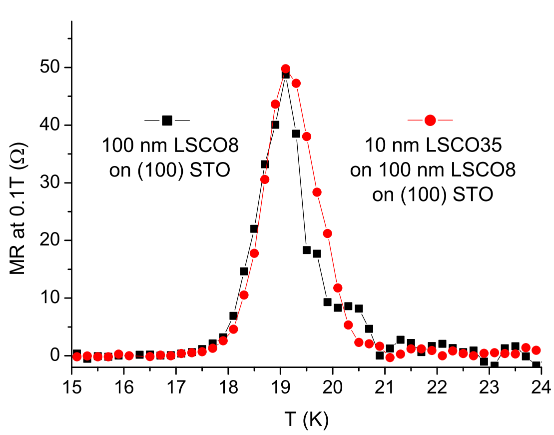

FIG. 6. (Color online) MR at 0.1 T versus temperature of the microbridges of Fig. 5.

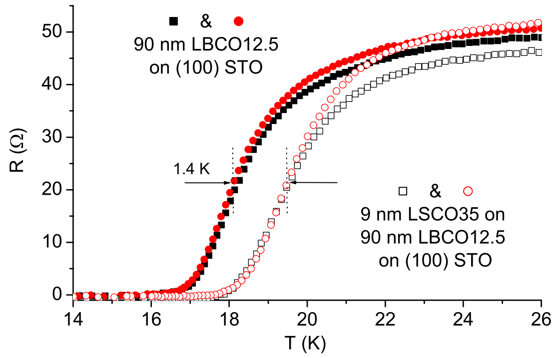

FIG. 7. (Color online) Resistance versus temperature of four typical $10\times100\ \mu m^2$ microbridges, two of the LSCO35-LBCO12.5 bilayer and two of the LBCO12.5 reference film on the same wafer.

After showing that the T_c enhancement results of Ref. 1 were not reproduced in the present study, we decided to check whether this effect appears in $La_{1.875}Ba_{0.125}CuO_4$ (LBCO12.5)-LSCO35 bilayers. In LBCO films T_c has a deep minimum at 1/8 doping level when the films are grown on STO[19] and thus the enhancement effect may be more robust. The experiment was thus repeated with a LSCO35-LBCO12.5 bilayer and its reference LBCO12.5 film on the same wafer. The results were obtained on ten microbridges as before, five on each half of the wafer, and with low-resistance gold contacts. Figure 7 shows the resulting R versus T of four representative microbridges, two on each half of the wafer, and Fig. 8 shows the corresponding magnetoresistance results at 0.05 T. A small but clear T_c enhancement effect is now observed where the T_c^{onset} and $T_c(R=0)$ of the bilayer are higher than those of the reference film by 2 and 1.4 K, respectively. This T_c enhancement effect is also robust and not affected by different annealing processes at 400 °C in 1 mTorr to 0.8 atm oxygen pressure. After vacuum annealing though (of 5×10^{-6} Torr), the bilayer and reference film have a metal to insulator transition, where they both become insulating at low temperatures. Thus the oxygen content at the interface of the bilayer is not affecting the T_c enhancement effect. Moreover, this experiment was repeated on an-

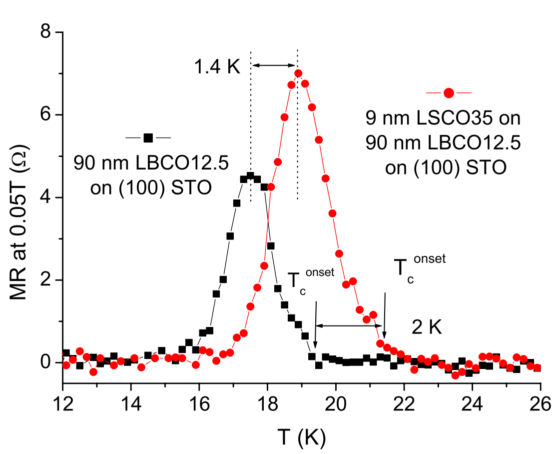

FIG. 8. (Color online) MR at 0.05 T versus temperature of two of the microbridges of Fig. 7.

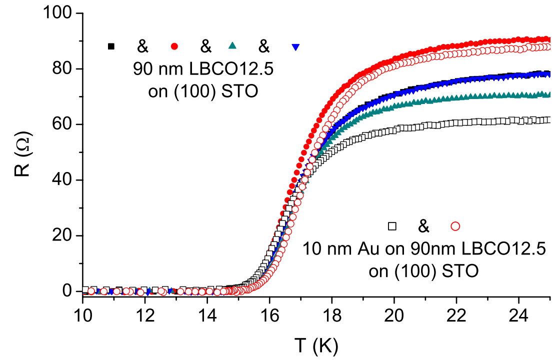

FIG. 9. (Color online) Resistance versus temperature of two typical microbridges of the Au-LBCO12.5 bilayer and four microbridges of the LBCO12.5 reference film on the same wafer.

other wafer with the patterning as shown in the inset to Fig. 2 (no microbridges) and the results were perfectly reproducible. Note that the T_c of our bare LBCO12.5 film is higher compared to that reported by Sato *et al.*,[19] possibly due to a higher disorder in our laser ablated films as compared to the electron-beam coevaporated films used in Ref. 19. Larger enhancement could possibly be obtained in films showing a stronger dip in T_c. It should be noted, however, that this small T_c enhancement may result form other effects such as strain or dopants migration at the interface. Nevertheless, the lack of T_c enhancement in the LSCO35-LSCO12 bilayers (Figs. 2–4), is a good reference indicator to the quality of the interface also in the LSCO35-LBCO12.5 case.

To test this result further, another control wafer was prepared with a gold normal metal instead of the LSCO35 cuprate as the cap layer, following our previous experiment.[1] Again, this wafer was prepared in the geometry of Fig. 1, and patterned into ten microbridges of $10\times100\ \mu m^2$, five on each half of the wafer. The highly conducting gold film was deposited on half of the wafer, on the LBCO12.5 film, under vacuum and at 150 °C. Under these conditions, the LBCO12.5 layer did not lose oxygen and the gold layer did not form spherical grains as it generally does at higher temperatures due to surface tension. In the Au-LBCO12.5 half of the wafer, the gold was removed by ion beam milling from the contacts area to avoid shorts, and then gold contacts were prepared by liftoff using a new layer of gold. The results of the resistance versus temperature of two microbridges of the Au-LBCO12.5 bilayer and four microbridges of the reference LBCO12.5 film are shown in Fig. 9 while the corresponding magnetoresistance versus temperature of the two bilayer microbridges and one reference film microbridge are shown in Fig. 10. First, we note that $T_c(R=0)$ of the reference LBCO12.5 film in Figs. 9 and 10 is lower by about 1 K as compared to that of Figs. 7 and 8. This is due to some ion milling damage or loss of oxygen in the film which was not reannealed at 450 °C, as is generally done, to avoid the formation of ball-like grains in the nearby gold layer. Second, one observes that this time, the bilayer and reference film have almost the same T_c^{onset} and $T_c(R=0)$ values at 18.5 ± 0.5 and 16.3 ± 0.3 K, respectively. The spread of the T_c values here is due to the typical spatial inhomogeneities of the films on the wafer. Clearly, the gold-cap layer in the Au-

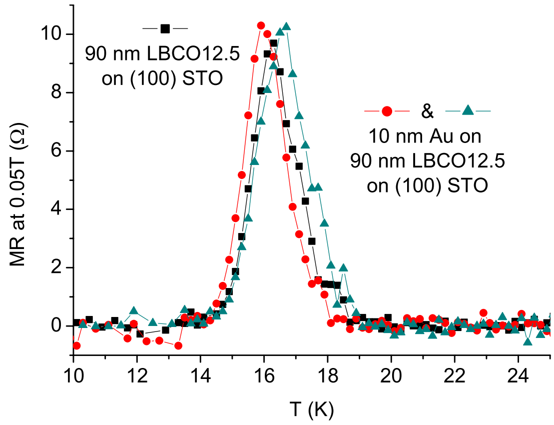

FIG. 10. (Color online) MR at 0.05 T versus temperature of three of the microbridges of Fig. 9, two of the bilayer, and one of the reference film.

LBCO12.5 bilayer does not lead to any $T_c(R=0)$ enhancement effect within the spread of the experimental data. This result is also consistent with Ref. 1, where no enhancement of T_c was found in an Au-LSCO10 bilayer, a result that was attributed to differences in the Fermi wave vectors and lattice structures of the two layers. The T_c enhancement effect of Figs. 7 and 8 in the LSCO35-LBCO12.5 system is therefore resulting from the OD cuprate-cap layer, as was originally suggested and explained from theoretical arguments in the introduction and in Ref. 1. As noted above, however, the possibility that T_c was enhanced due to strain or dopants migration effects at the interface cannot be ruled out.

In conclusion, the T_c enhancement results of Ref. 1 in LSCO35-LSCO12 and LSCO35-LSCO8 bilayers were not reproduced in the present experiments, where the bilayers and reference films were grown in the same deposition run. This may possibly reflect the strong dependence of the bilayer T_c on the interface properties, as predicted in previous theoretical studies, or on the properties of the individual films that are substrate dependent. However, in the LSCO35-LBCO12.5 system a small but clear T_c enhancement effect is observed, possibly in line with theoretical arguments predicting this behavior.

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