

Proximity-Induced Pseudogap: Evidence for Preformed Pairs

Ofer Yuli,¹ Itay Asulin,¹ Yoav Kalcheim,¹ Gad Koren,² and Oded Millo^{1,*}

¹*Racah Institute of Physics, The Hebrew University of Jerusalem, Jerusalem 91904, Israel*

²*Department of Physics, Technion-Israel Institute of Technology, Haifa 32000, Israel*

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The temperature evolution of the proximity effect in Au/La_{2-x}Sr_xCuO₄ and La_{1.55}Sr_{0.45}CuO₄/La_{2-x}Sr_xCuO₄ bilayers was investigated using scanning tunneling microscopy. Proximity-induced gaps, centered at the chemical potential, were found to persist above the superconducting transition temperature, T_c , and up to nearly the pseudogap crossover temperature in both systems. Such independence of the spectra on the details of the normal-metal cap layer is incompatible with a density-wave order origin. However, our results can be accounted for by a penetration of incoherent Cooper pairs into the normal metal above T_c .

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Since the discovery of the pseudogap (PG) phase in cuprate high temperature superconductors, it has been deemed that its origin holds key information regarding the pairing mechanism in these materials [1,2]. Despite the intensive study of the subject [3,4], it remains unclear whether the PG phenomenon is related to superconductivity or not. The debate on the origin of the PG revolves around the following two paradigms: (i) It is a superconducting precursor state where electrons pair incoherently above T_c . (ii) The PG bears no direct relation to superconductivity and is a manifestation of a competing order setting in at $T^* > T_c$. The former is based on many reports claiming that the transition temperature for pairing exceeds the phase locking temperature, at least in the underdoped regime [5–8]. The second paradigm points to the various ordered states predicted to compete with superconductivity in strongly correlated systems [9–11] like the cuprates. One way to distinguish between the two is to examine whether unique spectral properties associated with pairing are present in the PG regime. Concomitantly, a recent study of the photoemission spectra in Bi₂Sr₂CaCu₂O₈ single crystals [12], reported that a Bogoliubov-like dispersion curve, typical of superconductivity, was measured above T_c in the gapped antinodal region. The minimal energy value of the leading edge in the energy distribution curve, consistently found at k_F , led the authors to attribute their findings to a finite pairing amplitude existing above T_c rather than to a density-wave ordering (for which the gap minimum does not necessarily reside at E_F).

Another distinctive spectral fingerprint of the superconducting order parameter is its ability to penetrate an adjacent normal-metal via Andreev reflections, a process known as the proximity effect: When a normal-metal (N) and a superconductor (S) are placed in good electrical contact with one another, a holelike quasiparticle with energy smaller than the superconductor gap, Δ , impinging on the $N-S$ interface from the N side, may be retroreflected as an electron, while destroying a Cooper pair on

the S side. This unique process (that can be viewed as a transfer of a Cooper pair from S to N) induces superconducting correlations in N , which are manifested as a gap in the density of states (DOS) of the normal metal [13]. Experimentally, the nature of the proximity effect has been studied mainly at temperatures below T_c . An investigation of the proximity effect above T_c in the PG temperature regime is still lacking and is the focal point of the present study.

In this Letter we report the temperature evolution of the proximity-induced gap in correlation with sample morphology measured on Au/La_{2-x}Sr_xCuO₄ and La_{1.55}Sr_{0.45}CuO₄/La_{2-x}Sr_xCuO₄ bilayers using scanning tunneling microscopy (STM) and spectroscopy (STS). Our main finding, observed on both configurations, is a smooth evolution of the superconductor proximity gap into a proximity-induced PG, as the temperature was raised above T_c for bilayers comprising an underdoped La_{2-x}Sr_xCuO₄ [LSCO(x)] film. The induced gap gradually filled up by spectral weight until it became undetectable very close to the doping dependent T^* value we found via STS for the corresponding bare LSCO film [14]. The magnitude of the proximity gaps exhibited a similar spatial decay as a function of distance from an a axis facet below and above T_c , suggesting a common origin. In contrast, in bilayers comprising an overdoped LSCO layer, the proximity gap disappeared close to T_c . We claim that the observed independence of the $T > T_c$ proximity-gap features on the properties of the normal-metal layer (e.g., its Fermi surface), in particular the fact that the induced gap is always centered at the Fermi energy, E_F , is inconsistent with a density-wave origin for the induced gap. On the other hand, these same results, as well as the spatial dependence of the gap size found above T_c , are indicative of a finite pairing amplitude present at $T > T_c$.

N /LSCO(x) bilayers ($N = \text{Au}$ and La_{1.55}Sr_{0.45}CuO₄) with $x = 0.08, 0.10$ (underdoped) and $x = 0.18$ (overdoped) were epitaxially grown on (100)SrTiO₃ wafers by

laser ablation deposition with c -axis orientation perpendicular to the substrate [see schematic illustration in Fig. 1(a)]. The LSCO(x) films were 90 nm thick, and the N cover-layer, grown *in situ* without breaking the vacuum, was ~ 7 nm thick. The *in situ* metallic coating prevented the rapid surface degradation known to occur in LSCO films and enabled high quality STM topographic imaging. The underlying LSCO surface morphology comprised square grains of ~ 100 nm lateral size and 10–20 nm in height, exposing relatively large a -axis facets (see Fig. 1). The gold layer exhibited a granular morphology with a typical grain size of ~ 5 nm and roughness of 1–2 nm [see blow up in Fig. 1(c)]. The bulk superconducting transition temperatures were obtained by four-probe resistance vs temperature [$R(T)$] measurements and are presented in Fig. 1(b). We have also measured the properties of a bare 90 nm $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$ film. The $R(T)$ data showed no sign of a superconducting transition down to 4 K and the tunneling spectra taken at 4.2 K, exhibited gapless Ohmic behavior. Therefore, we conclude that the $x = 0.45$ layer was metallic in the temperature range of our experiments ($T > 4.2$ K).

Our tunneling spectra, namely dI/dV vs V curves, acquired by momentarily disabling the feedback loop, were obtained at specific well-defined locations, thus revealing the local DOS in correlation with the surface topography. In Fig. 2 we present a topographic image taken on an Au/LSCO($x = 0.10$) bilayer at 40 K, well within the PG temperature regime ($T_c^{\text{onset}} = 26$ K). The inset shows a set of dI/dV vs V curves measured consecutively along the

blue arrow, depicting the dependence of the measured gap size on the distance from the LSCO grain boundary, namely, from the interface between an a -axis LSCO facet and the Au layer. As the tip was moved away from the LSCO grain edge, the gap size diminished until near the grain center, far enough from the grain edge, the spectra turned gapless featuring a metallic structureless DOS. Importantly, a similar spatial dependence was found below T_c at ~ 4.2 K. The spatial dependence of our spectra can be understood within the framework of an anisotropic proximity effect expected for d -wave superconductors, as reported by Sharoni *et al.* [15] for Au/YBa₂Cu₃O₇ bilayers: the proximity effect takes place predominantly at $N - S$ interfaces involving a -axis oriented facets, i.e., at the grain boundaries. The spatial evolution of the induced gap showed a behavior similar to the conventional proximity effect, or, in other words, as the STM tip was moved away from the side facet, toward the grain center, a monotonic decay of the gap size was measured. The spectra then turned Ohmic at distances larger than the penetration length which in the dirty limit is given by the expression $\xi_N = (\frac{\hbar v_{\text{FN}} l_N}{6\pi k_B T})^{1/2}$. In our case, $\xi_N \approx 10$ –20 nm at 4.2 K (taking $v_{\text{FN}} = 1.4 \times 10^6$ m/s and the mean free path l_N to be the average Au grain size, ~ 5 nm) which accounts for the Ohmic behavior on top of the grain about 50 nm away from the grain edge [15]. Such behavior agrees qualitatively with the predicted evolution of the DOS calculated by Löfwander [16]. As the temperature was raised, areas exhibiting Ohmic spectra inhabited a larger fraction of the surface. Nevertheless, areas exhibiting pronounced induced gaps were still found as in the 4.2 K case, predominantly near grain edges.

A detailed description of the induced-gap's temperature dependence is portrayed in Fig. 3, which presents tunneling spectra acquired near grain edges in Au/LSCO(x) bilayers

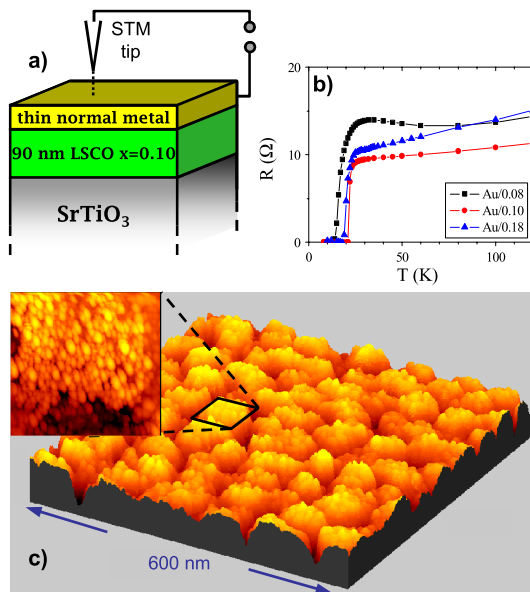


FIG. 1 (color online). (a) Schematic of the experimental setup. (b) $R(T)$ of the Au/La_{2-x}Sr_xCuO₄ bilayers with $x = 0.08, 0.10$, and 0.18 . (c) 600×600 nm² STM topographic image of an Au/La_{1.90}Sr_{0.10}CuO₄ bilayer. The small granular gold is clearly seen on top of the underlying LSCO crystallites.

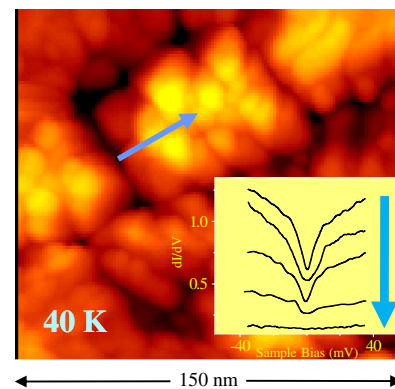


FIG. 2 (color online). 150×150 nm² topographic image of an Au/LSCO($x = 0.10$) film taken at 40 K. The Au coverage is seen as the grainy surface on top of the square LSCO crystallite. Inset: dI/dV vs V curves taken along the arrow indicated in the topographic image. The gap size was found to depend on the distance from the grain edges in a similar manner both below and above T_c . The curves are vertically shifted for clarity.

with $x = 0.08, 0.10$ and 0.18 . To ensure the gaps were proximity gaps, a correlation with local topography was done (similar to the case presented in Fig. 2) thus avoiding confusion with data originating from possibly local low Au coverage. The transition onset temperatures of the bilayers were 24, 26, and 27 K for the $x = 0.08, 0.10$, and 0.18 , respectively, as seen in Fig. 1(c). Nevertheless, and this is the central result of the presented report, the proximity-gap structure did not vanish at T_c for the underdoped bilayers. Moreover, no noticeable change in the measured spectra was observed near $T \sim T_c$ for both underdoped samples. Rather, the gap seems to smoothly fill up, until it disappears completely at $T(x = 0.08) \sim 90$ K and $T(x = 0.10) \sim 50$ K in striking resemblance to T^* values we measured by STS for the corresponding bare underdoped LSCO films [14]. In contrast, no proximity-induced gap was found above T_c in the spectra of the $x = 0.18$ overdoped bilayers [see Fig. 3(c)], in accordance with reports of a conventional normal state in the LSCO overdoped regime [14,17]. We have also measured an underdoped ($x = 0.08$) and overdoped ($x = 0.18$) LSCO, coated by 7 nm of $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$ which replaced the Au as the normal metal. The results were essentially very similar—the gap remained centered at zero bias and disappeared at ~ 90 K for the underdoped bilayer and at T_c for the overdoped layer.

The similar spatial dependence (as a function of distance from the a -axis facets) exhibited by both the proximity-induced PG ($T > T_c$) and the superconductor proximity gap ($T < T_c$), suggests that the PG phase is closely related to superconductivity, e.g., that it may be associated with the existence of uncondensed Cooper pairs above T_c . Nevertheless, the possible induction of a proximity gap by competing orders suggested for the PG cannot be ruled out *a-priori*. These phases, which do not involve Cooper pairing, are generally characterized by an order parameter with nonzero center-of-mass momentum, \mathbf{Q} . In light of our results we need to address the question: can an order parameter with $\mathbf{Q} \neq \mathbf{0}$ induce a gap in a normal metal and if so will its properties comply with our data? We shall first focus for clarity on the phases induced by a nesting condition such as the charge and spin density wave. In a N-DW (DW-density wave of some sort) configuration, normal-metal quasiparticles with subgap energy impinging on the gapped DW interface, change their momenta by the wave vector \mathbf{Q} of the DW pattern in an unconventional reflection process termed a \mathbf{Q} reflection [18,19]. Consequently, a zero charged particle-hole entity with the transferred momentum \mathbf{Q} is created in the DW side. The nesting condition $E(\mathbf{k} \pm \mathbf{Q}) = E(-\mathbf{k})$ responsible for the gap in the DW condensate, induces a sign change of the reflected electron's velocity, at least in the semiclassical limit where $\mathbf{v} = \partial E(\mathbf{k})/\partial \mathbf{k}$. The subsequent mix of \mathbf{k} states with $\mathbf{k} \pm \mathbf{Q}$ in the normal metal can be viewed as a penetration of DW correlations into N . Such a mix

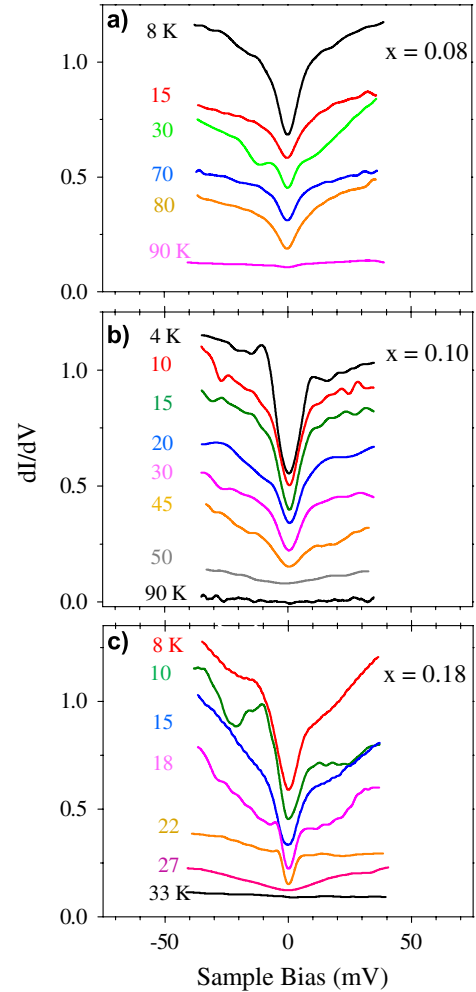


FIG. 3 (color online). Temperature evolution of the local DOS measured on the Au/LSCO(x) bilayer for (a) $x = 0.08$, (b) $x = 0.10$ and (c) $x = 0.18$. The conventional proximity gap found below T_c evolved smoothly to an Ohmic spectra at $T \approx T^*$ of the corresponding bare LSCO for both $x = 0.08$ and $x = 0.10$ and disappeared at $T \approx T_c^{\text{onset}}$ for the $x = 0.18$ bilayer. The curves are vertically shifted for clarity, and normalized to the gap edge value. The vertical axis corresponds to the curve taken at 8 K in (a) and (c) and at 4.2 K in (b).

inevitably yields a gap centered at the corresponding energy $E(\mathbf{k})$, as was discussed in detail by Kanigel *et al.* [12] for the cuprate normal state. For the special case of $\mathbf{Q} = 2\mathbf{k}_F$, the induced gap will open at E_F in accordance with our results. However, in view of this proposed scenario we need to recall that the gap remained centered at zero bias when the Au was replaced by a different normal metal, $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$, albeit now obviously $\mathbf{Q} \neq 2\mathbf{k}_F$. Such robust behavior of the gap location implies the induced gap in N does not stem from a nonzero momentum order. Note that the latter argument does not require the nesting condition assumed above. Any phase which mixes \mathbf{k} and $\mathbf{k} \pm \mathbf{Q}$ states in N (e.g., a d -density-wave order) will eventually induce a gap removed from the chemical po-

tential upon changing the Fermi surface in N . In contrast, Andreev reflections, unlike \mathbf{Q} reflections, involve particle-hole mixing with zero momentum ($\mathbf{Q} = 0$) and accordingly, the induced gap will *always* be centered at E_F regardless of the normal-metal details. We thus conclude that our findings comply better with the preformed pairs scenario for the PG compared to any of the suggested competing orders.

The question now is whether a system comprising Cooper pairs lacking global phase coherence can undergo an Andreev reflection. The answer entails a solution of the Bogoliubov–de Gennes equations in the presence of thermal phase fluctuations as was done by Franz and Millis [20]. Following the framework put forward in Ref. [6], the normal state was modeled as a plasma of unbound vortex-antivortex pairs (associated with the Kosterlitz-Thouless transition). Each vortex is surrounded by a circulating supercurrent which leads to a Doppler shift in the local quasiparticle excitation spectrum of $\Delta E = \hbar \mathbf{k} \cdot \mathbf{v}_s$ where \mathbf{v}_s is the local superfluid velocity, which is related to the order parameter phase by $\mathbf{v}_s(\mathbf{x}) = \hbar \nabla \theta(\mathbf{x}) / 2m$. The local DOS was calculated by averaging over the phase fluctuations assumed to vary in space slower than the superconductor coherence length, ξ_s . Choi *et al.* [21] extended this calculation to the PG regime and predicted an enhancement of the differential conductivity due to Andreev reflected Cooper pairs which lack long-range phase coherence. This work established the finite probability for the occurrence of Andreev reflections by preformed pairs. We note, however, that the effect of such reflections on the DOS in the normal metal has not been calculated yet.

It should be noted that in a previous Andreev spectroscopy study of LSCO in the PG regime, no surface Andreev bound states were found above T_c [22]. This observation may appear to contradict ours. However the conditions for the formation of these bound states, a process that involves alternating Andreev and normal reflections [23,24], are probably more stringent compared to gap induction which requires a single Andreev reflection. This notion is supported by the commonly observed disappearance of the zero-bias conductance peak [13] associated with the Andreev bound states even at $T < T_c$ [22,25].

In conclusion, we have observed a proximity gap above T_c in the DOS of a normal-metal overcoating an underdoped LSCO film. The induced gap survives up to a temperature close to the PG onset temperature T^* , suggesting a common origin for both phenomena. The similar spatial dependence of the proximity induced PG and the superconductor proximity gap indicate that the origin of the PG is related to superconductivity. Moreover, the gap minimum was always measured to be at E_F , even for two very different N layers, Au and $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$, a finding difficult to reconcile with a PG order parameter characterized by $\mathbf{Q} \neq 0$. The predicted ability of Cooper pairs to induce correlations in a normal-metal in the presence of

(coherence breaking) phase fluctuations leads us to ascribe the PG to a state consisting of Cooper pairs with short-range phase coherence.

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*milode@vms.huji.ac.il

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