Is the Onset of High-Temperature Superconductivity Associated with a Dimensional Crossover?

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Angle-resolved ultraviolet photoemission spectroscopy (ARUPS) from high-T_c superconductors shows an effective-mass renormalization and intense quasiparticle peaks close to the Fermi energy E_F, which change dramatically with temperature as T_c is crossed. They are attributed to many-body effects, but their precise nature has been controversial until now. We find very similar spectral fingerprints, even with a similar temperature dependence albeit with much higher critical temperature, in a quasi-one-dimensional Br/Pt surface compound. The striking parallels support an interpretation based on spin-charge separation and are consistent with a dimensional crossover taking place at T_c.

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Although an understanding of high-T_c superconductivity is still *in limbo*, much insight has been gained in the past few years from angle-resolved ultraviolet photoemission spectroscopy (ARUPS). The *d*-wave symmetry of the order parameter [1] or the persistence of a pseudogap [2,3] above T_c in underdoped cuprates, for instance, are cornerstones of any future theory of unconventional superconductivity. However, several aspects of the ARUPS results, i.e., the origin of the notorious peak-dip-hump (PDH) feature developing below T_c in the spectral function close to E_F and the absence of well-defined quasiparticle (QP) peaks in some areas of the surface Brillouin zone (SBZ), are still controversial [4]. We observe similar spectral features with an analogous unusual temperature dependence in connection with a structural phase transition in the otherwise unrelated quasi-one-dimensional (quasi-1D) Br/Pt(110) adsorption system. We show that the similarity of the ARUPS data for the two different systems supports an interpretation in terms of spin-charge separation. The anomalous temperature dependence of the spectral function could then indicate a dimensional crossover.

Three hallmarks of the irregular line shapes found in the cuprates are illustrated in Figs. 2 and 3: First, the effective mass of QP bands is strongly renormalized as the band approaches E_F [this gives rise to the "kink" in the band dispersion shown in Fig. 2(a)]. Simultaneously, the corresponding photoemission peak acquires very high intensity [5]. A similar mass renormalization close to E_F is seen on surfaces with strong electron-phonon coupling [6] and is also typical for many-body (MB) effects in strongly correlated materials. Second, weakly dispersing bands close to E_F produce sharp photoemission lines at their minimum (binding) energy positions, but exhibit extreme broadening in between [Fig. 2(c) [4]]. The broadening often exceeds the apparent total dispersion across the zone, implying that the QP picture is not valid in this momentum range [4,7]. Third, an anomalous change in the spectral function is observed around $\overline{M}$ (antinodal direction), as T_c is crossed. While the spectrum is essentially flat above T_c, for $T < T_c$ it develops a sharp, intense peak slightly below E_F [Fig. 3(a)]. Towards higher binding energy the peak is usually accompanied by a dip and a broad hump [8,9] [this PDH feature appearing below T_c is not to be confused with the bilayer splitting observed recently for the Bi(2212) cuprate [10,11]].

We have investigated an adsorbate system exhibiting a pronounced quasi-1D character [12] and found a strikingly similar behavior. The system is obtained by deposition of 0.5 ML of Br onto a Pt(110) surface and subsequent annealing. Thereby the (1×2) missing row reconstruction of the clean surface is lifted and Pt-Br chains self-assemble into a two-dimensionally (2D) ordered $c(2 \times 2)$ structure (Fig. 1). The Pt-Br chains consist of close-packed Pt atom rows as on the clean surface, but every second short-bridge site is occupied by a Br atom [31]. A precision structural analysis [13] using LEED-IV and density functional theory calculations as well as details of the preparation are presented elsewhere [14,15]. The $c(2 \times 2)$ structure might look similar to an ordinary 2D adsorbate structure, but the system exhibits an extreme anisotropy with intrachain interactions being much stronger than interchain interactions. Actually, a charge density wave can be triggered [12], as expected for a quasi-1D system. Raising the

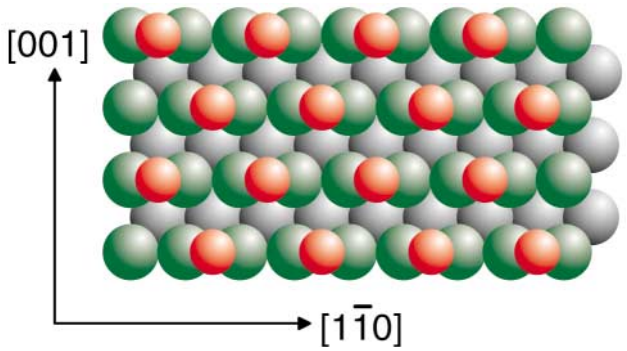

FIG. 1 (color). Ball model of the system $c(2 \times 2)$Br/Pt(110). The Br atoms (red) reside in the short-bridge sites on the closed packed Pt rows (green), second-layer Pt atoms are shown in gray [13].

temperature above 500 K enables the Br atoms to carry out concerted hops along the rows. The Br-Br distances within a row are preserved, but the interchain correlation of Br positions is destroyed. The Pt atom positions are not affected up to temperatures $T < 1000$ K. Note that *the Br coverage remains unchanged*. The structural transition at $T_S > 500$ K is accompanied by a change of the low-energy electron diffraction (LEED) pattern from a $c(2 \times 2)$ to a (1×1) structure.

The experiments were carried out in a UHV system with 5×10^{-11} mbar base pressure. The sample, a Pt(110) single crystal oriented with an accuracy of $<0.5°$, was mounted on a liquid-N_2 reservoir. Resistive heating allowed to heat the sample to 1100 K. The sample cleaning procedure is critical, as even minor C traces prevent perfect ordering. The special procedure used here was described previously [16]. Coverages were calibrated from LEED and temperature programmed desorption spectra. The ARUPS spectra shown here are selected examples from a survey covering almost the entire SBZ. They were recorded by means of a movable hemispherical analyzer with an energy resolution of 60 meV and an angular resolution of $\pm 1°$. A He plasma discharge was used as a photon source ($h\nu = 21.22$ eV).

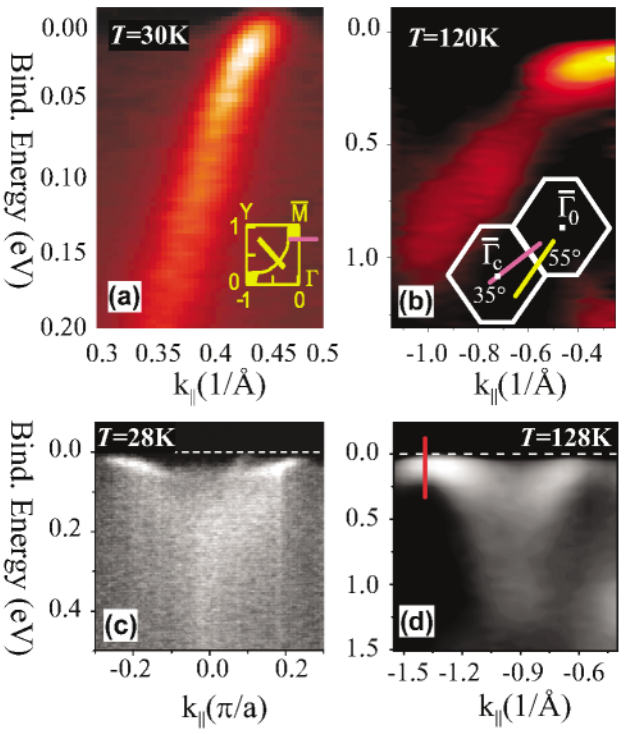

FIG. 2 (color). Contour plots of photoelectron intensity. (a) Data from Ref. [5], overdoped ($T_c = 91$ K) $Bi_2Sr_2CaCu_2O_{8+\delta}$. Wave vectors (**k**) are along the yellow line given in the SBZ in the inset (nodal direction). (b) $c(2 \times 2)$Br/Pt(110), **k** is along the yellow line given in the inset ($\theta = 55°$). (c) Data from Ref. [4], optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$, **k** is along the purple line in the inset of (a). (d) $c(2 \times 2)$Br/Pt(110), **k** is along the purple line given in (c) ($\theta = 35°$). The red line relates to Fig. 3(b). Temperatures at which the spectra were recorded are given on top of each figure.

Electron energy distribution curves, measured by ARUPS in the $[1\bar{1}0]$ direction along the chains (azimuthal angle $\theta = 0°$), show normally dispersing bands with no exceptional behavior [12]. For the emission angles deviating from the chain direction shown here, however, anomalous line shapes are observed. The spectra are strongly reminiscent of those measured in high-T_c materials.

The conspicuous similarity of the Br/Pt ARUPS spectra with those of the cuprates is borne out, for example, by the effective-mass renormalization and the intensity enhancement of a band approaching the Fermi energy in both overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ [5] [Fig. 2(a)] and $c(2 \times 2)$-Br/Pt(110) [Fig. 2(b)]. Qualitatively similar mass renormalizations and intensity enhancements are observed for all directions in the SBZ which deviate more than $\sim 10°$ from the Br-Pt-chain direction. Also, an anomalous band broadening observed in optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ [4] [Fig. 2(c)] appears almost identically for a flat band close to E_F in Br/Pt [Fig. 2(d)]. As in the case of $Bi_2Sr_2CaCu_2O_{8+\delta}$, a cut has been chosen which crosses through two parallel Fermi surfaces. Finally, Fig. 3 shows the fading of the QP peak near $\overline{M}$ in optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ as the temperature is raised above T_c [Fig. 3(a)] and a strikingly similar line shape change for Br/Pt as the structural transition temperature T_S is crossed [Fig. 3(b)] [32].

Different models were brought forward to account for the anomalous line shapes in high-T_c cuprates. The disappearance of the QP peak above T_c, for instance, has been attributed to a lifetime catastrophe [17]. It has been assumed that the QP peak is due to paired electrons or electrons dressed by some other (e.g., electron-phonon) interaction. The broadening of the peak into a

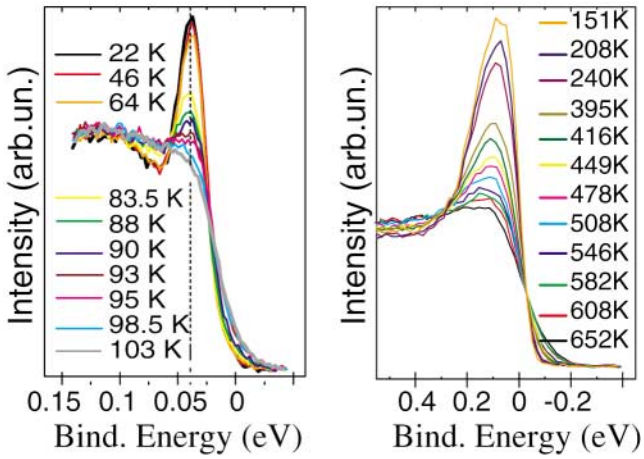

FIG. 3 (color). Temperature evolution of ARUPS energy distribution curves. (a) Data from Ref. [4], optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$. Spectra are angle-integrated between 0.75 and 1 along $\Gamma\overline{M}$. (b) $c(2 \times 2)$Br/Pt(110), the spectra are taken at Γ_c [see SBZ in Fig. 2(b)] and represent cuts along the red line in Fig. 2(d).

quasicontinuum for $T > T_c$ was attributed to a new scattering channel opening up above T_c. As a possible source of the anomalous scattering, Franz and Millis [18] proposed the existence of superconducting phase fluctuations above T_c. A second class of models attributes the peculiar line shapes to spin-charge separation [7,19–23] (see Fig. 4 for a schematic representation of the process). Laughlin [7] pointed out that the PDH feature could be interpreted as a bound state and a scattering resonance, respectively, of two interacting particles, i.e., spinon and holon. An attractive force depending on the center-of-mass momentum could account for the wave vector ($\mathbf{k}$) dependence of the spectra. Within this framework, the fading of the QP peak above T_c signals the disappearance of the attractive spinon-holon interaction for the case of 2D antiferromagnetic ordering. A $\mathbf{k}$ dependent spinon-holon interaction is schematically illustrated in Fig. 4(b) [24]. Accordingly, Carlson, Orgad, Kivelson, and Emery (COKE) [19] interpreted the disappearance of the binding interaction above T_c as a 2D to 1D dimensional crossover. If the interactions in the 2D system are anisotropic, it can be modeled as an ensemble of correlated 1D chains. Raising T above T_c causes k_BT to exceed the interchain interaction energy. Consequently, the chain-chain correlation breaks down and the system reverts to 1D behavior. In 1D, however, the attractive "string" interaction [24] between spinon and holon is absent [see Fig. 4(a)]. Hence, the corresponding QP peak is broadened into a continuum since the photoionization process creates two independent particles in the system [21]. Below T_c, the spinon-holon interaction depends on the orientation of their momentum relative to the chain direction, as can be envisioned from Fig. 4(b). The model therefore naturally accounts for a $\mathbf{k}$ dependence of the linewidth below T_c.

The dimensional crossover proposed by COKE is not necessarily associated with superconductivity. Therefore this model is likely to apply for the $c(2 \times 2)$-Br/Pt(110) system: The intrachain and interchain interaction are indeed substantially different [12] and the fading of the QP peak [Fig. 3(b)] is associated with the loss of chain-chain correlation at the structural transition of the Br/Pt(110) system. In Fig. 5, we compare the binding energy of the QP peak with the intensity of the $c(2 \times 2)$ LEED spot. The fading of the centered LEED spot in the $c(2 \times 2)$ structure, which characterizes the structural transition, is shown in Fig. 5(a). Both binding energy of the QP peak and LEED intensity show a critical behavior at the transition

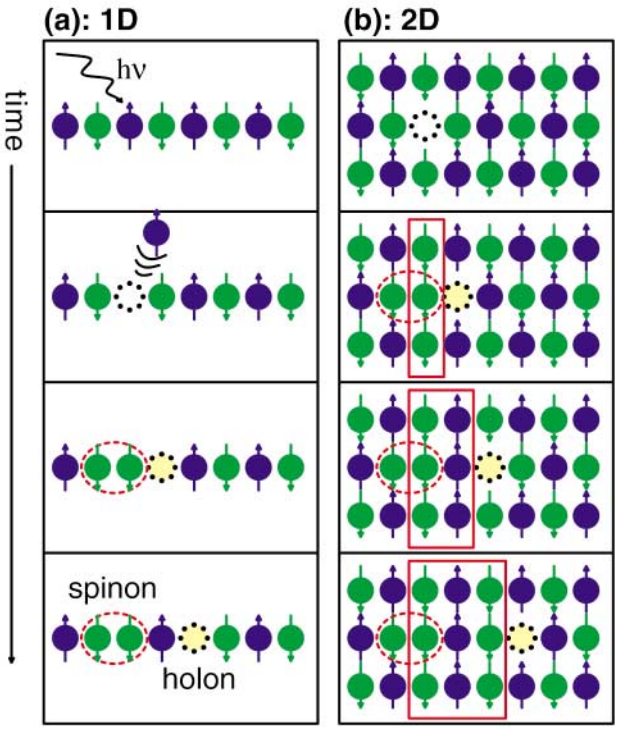

FIG. 4 (color). Simplified picture of spin-charge separation in the limit of infinite on-site Coulomb repulsion U (antiferromagnetic ground state). (a) In 1D, only the first hop of the charge vacancy creates a magnetic excitation (spinon). Additional hops do not require energy, spinon and holon move independently. (b) In 2D, the number of magnetic excitations (red boxes) increases with the distance between spinon and holon. Spinon and holon motion are therefore coupled.

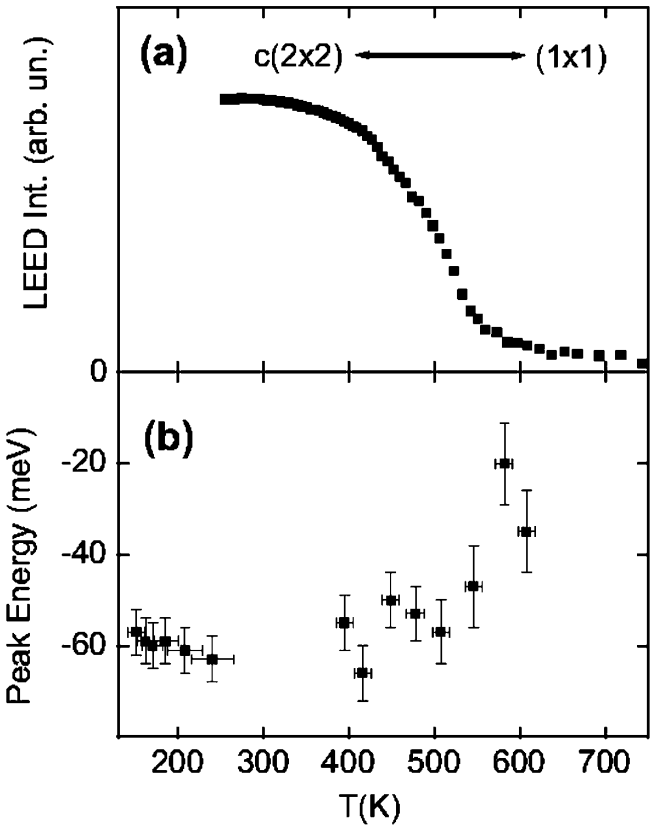

FIG. 5. 0.5 ML Br/Pt(110). (a) Temperature dependent intensity of the center spots of the $c(2 \times 2)$ phase. The fading of the centered spots while raising the temperature from 450 to 550 K indicates a loss of the chain-chain correlation and, hence, a 2D to 1D dimensional crossover. (b) Temperature dependent energy shift of the peak shown in Fig. 3(b).

temperature T_S. The spectral weight and the width of the QP peak [33] (not shown) in the system Br/Pt(110) parallel the behavior which was found in the cuprates [25,26], suggesting a similar underlying mechanism for both systems.

Applicability of the COKE model to high-T_c cuprates may be more controversial, although spin-charge separation is inherent also to the resonating valence bond (RVB) model [22] and appears to be a widely accepted paradigm [27]. Anisotropy of interactions within the CuO planes, in contrast, is under dispute, although the observation of striped phases is cited in support of this view [27]. In the context of the RVB theory [22] and the interlayer tunneling mechanism of high-T_c superconductivity [28], a 2D to 3D dimensional crossover is envisaged at T_c, whereas in the present 2D Br/Pt(110) system the crossover is obviously from 1D to 2D. To our knowledge, there is no prediction of a spectral function for the 2D to 3D case, so that we cannot entirely rule out a possible 2D to 3D dimensional crossover in the cuprates.

The interpretation of the ARUPS spectra given above should be contrasted to the other possible alternatives: Given the structural transition temperature $T_S > 500$ K, BCS electron-pair formation and any explanation directly relating the QP peak to superconductivity can be excluded in the present system. Examining Fig. 2(b) in detail reveals an onset of the mass renormalization at a binding energy of $\sim$200 meV, ruling out electron-phonon interaction as the dominant MB effect. In contrast, both the peculiar **k** dependence and the critical behavior of the QP peak at the phase transition are incorporated in the "dimensional crossover model."

In summary, the appearance of surprisingly similar MB effects in high-T_c cuprates as well as in the present quasi-1D system Br/Pt(110) and, in particular, the similar critical temperature behavior of QP peaks seen in ARUPS argue for a common underlying mechanism in both systems, i.e., spin-charge separation. The observation of a dimensional crossover on the Br/Pt(110) surface at the same temperature demonstrates the possibility that also in the cuprates the critical temperature behavior may be due to a 1D to 2D dimensional crossover rather than directly to superconductivity. This would put the cuprates in line with organic superconductors, where superconductivity is also observed only above a critical coupling strength between the quasi-1D molecular stacks [29] and T_c depends critically on the dimensionality of the compound [30].

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[32] For the present Br/Pt system, we cannot define a doping level, because the underlying metal acts as a charge reservoir. The charge actually residing within the relevant surface state bands can therefore not be determined from a simple electron count, but only from a complete Fermi surface mapping.
[33] Binding energy, width, and spectral weight of the QP peak were derived from a line fit to the energy distribution curves. Fit model: Lorentz peak on a constant background multiplied by the Fermi function and convoluted with the analyzer resolution. The spectral weight is linearly increasing with decreasing temperature, whereas the inherent width (FWHM) decreases only from 320 meV above 500 K to a constant 250 meV below 300 K.