Resilience of the Mott insulating state of La_2CuO_4 against photodoping

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The parent compounds of high-T_c cuprate superconductors are Mott insulators with antiferromagnetic (AFM) order, where strong correlations between Cu 3d-electrons split the half-filled Cu 3d-band into the upper (UHB) and lower (LHB) Hubbard band, resulting in an antiferromagnetic insulating ground state [1]. The insulating parent (undoped) compounds are characterized by a charge-transfer (CT) gap of about 2 eV between the oxygen-derived p-band, located within the Mott gap, and the upper Hubbard band (UHB). The nature of doping induced changes in the low energy electronic structure by introducing electrons or holes into the CuO_2 planes is, like the problem of high-T_c cuprates itself, still under intense discussion [1-4]. Introducing a few percent of holes (or electrons) into the CuO_2 planes, results in a transfer of spectral weight to lower energies, with the appearance of excitations in the mid-infrared (MIR) range [2]. Upon further doping the MIR peak eventually merges into the Drude-like free carrier peak as doping approaches optimal doping [2]. While numerous models have been put forward to account for the doping evolution of free carrier and MIR response, the consensus is still lacking [5].

Photodoping, i.e., generating electron- and hole-like carriers by absorption of light with frequency exceeding the CT gap, was shown to induce similar changes in low energy excitation spectrum to chemical doping, both by using quasi-continuous [6] or pulsed excitation [7-9], where studies on thick films and single crystals suggest photoinduced metallic state [7-9]. Here, we investigate transient photo-doping phenomena in La_2CuO_4, an archetypal antiferromagnetic Mott insulating parent compound of the cuprate high-T_c superconductor. We capture the dynamics of the transient state by tracking the time-evolution of THz photoconductivity [8] and the changes in the complex dielectric function $\varepsilon(\omega,t)$ [10] in the 0.5 – 2.6 eV range in optically-thin films, following optical excitation across the charge transfer (CT) gap. We cover a large range of excitation densities from 0.001 to ~0.12 absorbed photons [electron-hole (e-h) pairs] per Cu. Analysis of $\Delta\varepsilon(\omega,t)$ demonstrates a pronounced photoinduced reduction of the CT gap, consistent with recent transient X-ray absorption study [11], concomitant with the appearance of mid-infrared absorption and a weak free carrier response, all simultaneously relaxing on a (sub)picosecond scale. Up to the highest excitation densities, where at comparable chemical-doping levels a metallic state is realized, the free carrier contribution remains negligible, underscoring the robustness of the underlying electronic correlations. Recovery dynamics proceeds on the ps timescale and is density-independent up to 0.01 e-h pairs per Cu. The low free-carrier spectral weight and the overall fast recombination suggest that relaxation proceeds via pair-wise recombination of nearly bound e-h pairs through multi-magnon emission [12]. Only at densities beyond 0.01 e-h pairs per Cu the relaxation rate starts to increase with increasing excitation density, which we attribute to partial melting of the antiferromagnetic background and many-body recombination processes. Comparison of the excitation density dependence of the Drude-like and MIR spectral weights suggests the two are intimately related, providing constraints for theories of low-energy excitations in weakly doped cuprates.

References

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