Ultrafast renormalization of the onsite Coulomb repulsion in a cuprate superconductor

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Abstract: Time-resolved x-ray absorption spectroscopy is used to monitor the transient modification of the on-site Coulomb repulsion in the high-temperature superconductor La$_{1.905}$Ba$_{0.095}$CuO$_4$ following an excitation with an ultrashort optical femtosecond pulse. © 2022 The Author(s)

1. Introduction

Intense ultrafast electromagnetic fields are an increasingly important tool to stabilize and control novel emergent phases in quantum materials. Among a variety of nonthermal excitation protocols, a particularly promising route is represented by the direct light-engineering of effective many-body interactions, such as electron hopping amplitudes and local electron-electron repulsion. Achieving a light-induced dynamical renormalization of the screened onsite Coulomb repulsion (“Hubbard $U$”) would have far-reaching implications for high-harmonic generation [1], attosecond spectroscopy [2] and ultrafast magnetism [3] in the solid state. However, experimental evidence for dynamical control over the Hubbard $U$ has so far remained scarce [4], [5].

2. Results and Discussion

Here, we employ time-resolved x-ray absorption spectroscopy (trXAS) to demonstrate the ultrafast renormalization of the Hubbard $U$ parameter in the underdoped cuprate superconductor La$_{1.905}$Ba$_{0.095}$CuO$_4$ (LBCO, $x=9.5\%$) [6]. In our experiments, we excite the LBCO sample with intense femtosecond (50 fs) optical pulses centered at 1.55 eV (see Fig. 1 (a)). The transiently modified local electronic structure is subsequently probed with time-delayed soft x-ray pulses that promote localized core electrons into unoccupied valence states. The light-induced changes of the LBCO’s electronic structure manifest themselves as a pronounced shift of the x-ray absorption maxima associated with transitions to the upper Hubbard bands (UHB), while leaving the transition energy into Zhang-Rice singlet (ZRS) states near the Fermi level unaffected (see Fig. 1 (b) and (c)). This effect is particularly prominent at the Cu $L_3$ edge, where the redshift of the UHB reaches 125 meV, compared to a 63 meV shift at the O $K$ edge. By comparing the experimental results with exact-diagonalization calculations of the time-dependent spectrum of single- and the three-band Hubbard models, we attribute the observed effects to a pump-induced renormalization of the Hubbard $U$ on the Cu sites.
Fig. 1 (a): An LBCO sample is illuminated by an intense optical pump pulse centered at 800 nm, and the resulting changes in the electronic structure are probed at variable time delays $\Delta t$ with soft-x-ray pulses that excite core electrons into unoccupied valence states. (b) and (c): x-ray absorption (XAS) spectra at the oxygen K and the copper L edges, taken before (black, $\Delta t$--1 ps) and after (red or blue, $\Delta t$--0 ps) the arrival of the pump pulse. Vertical dashed lines mark the fitted peak positions of each spectral feature. The shifts of the UHB at both edges is reported within the 95% confidence interval.

Our results represent a first demonstration of dynamically-renormalized Hubbard $U$ in a strongly correlated transition metal oxide and pave the way to a novel strategy for the manipulation of superconductivity, magnetism, as well as to the realization of other long-range-ordered phases in light-driven quantum materials.

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