

Mott-Hubbard excitons in edge-sharing CuO₄ chains

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Y. Matiks, P. Horsch, B. Keimer, and A. V. Boris

Max Planck Institute for Solid State Research, Heisenbergstr. 1, DE-70569 Stuttgart, Germany

Compounds composed of chains of edge sharing Cu²⁺O₄ plaquettes have the peculiar property that the magnitude of the nearest-neighbor hopping matrix element along the chains is anomalously small due to the orthogonality of the 2p σ orbitals on oxygen ions adjacent to the Cu ion. By virtue of their exceptionally narrow electronic bandwidths, these compounds provide a highly favorable platform for the investigation of exciton formation and the interplay between spin and charge correlations in cuprates. We have performed a comprehensive ellipsometric study of the charge excitations across the optical gap in high-quality LiCuVO₄ [1], NaCu₂O₂ [2], Li₂CuO₂, CuGeO₃ and α -CuV₂O₆ single crystals. In all these compounds, the excitonic states associated with the Zhang-Rice singlet state were assigned. For photon polarization along the chains, the data reveal a weak but well-resolved two-peak structure forming the absorption edge whose spectral weight is strongly enhanced upon cooling near the magnetic ordering temperature. These bands were identified as exciton doublets, originating from the long-range Coulomb interaction between the nearest and the next-nearest-neighbour Cu sites along the chains. These results have not only persuasively demonstrated the formation of the Mott-Hubbard excitons, but also quantified characteristic energy scales, such as the local Hubbard U (2.55 – 4.3 eV) and long-range Coulomb V (0.8 – 1.6 eV) interactions.

[1] Y. Matiks, P. Horsch, R. K. Kremer, B. Keimer, A. V. Boris, Phys. Rev. Lett. 103, 187401 (2009)

[2] Y. Matiks, A. N. Yaresko, K. Myung-Whun, A. Maljuk, P. Horsch, B. Keimer, A. V. Boris, Phys. Rev. B 84, 245116 (2011)

Primary author: BORIS, Alexander (Max Planck Institute for Solid State Research)

Presenter: BORIS, Alexander (Max Planck Institute for Solid State Research)

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