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NiO: Magnetic coupling and ultrafast electron dynamics in thin films

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Charge excitation across an electronic band gap plays an important role in optoelectronics and light harvesting. In contrast to conventional semiconductors, studies of above-band-gap photoexcitation in strongly correlated materials are still in their infancy. One prominent example is the antiferromagnetic oxide NiO, a material that is also used in spintronics and for which the ultrafast electron dynamics and the coupling between charge transfer and antiferromagnetic spin system is largely unexplored. Here we present a study by combining scanning tunnelling microscopy (STM) and spectroscopy (STS) with time-resolved two-photon photoemission (2PPE) for NiO(001) ultrathin films with special emphasis on the electronic response upon optical excitation [1-4].

At the surface of epitaxial NiO(001) films grown on Ag(001), we find series of well-defined image potential states below the vacuum level with film thickness dependent lifetimes in the range of 30 to 120 fs [2]. In contrast to these rather long lifetimes, we find an ultrafast ($\lesssim 10$ fs) relaxation for electrons that are excited just across the charge-transfer gap into the conduction band, which corresponds to the upper Hubbard band of a charge-transfer insulator [3]. We identified an ultrafast relaxation of the initial excitation into long-lived many-body in-gap states. Remarkably, the spectral weight of these in-gap states displays coherent THz oscillations up to 2 ps at low temperature [3]. The frequency of these oscillations corresponds to the strength of the antiferromagnetic superexchange interaction in NiO and their lifetime vanishes slightly above the Néel temperature. These observations indicate a strong coupling of the excited states to the antiferromagnetic spin system and pave the way for addressing antiferromagnetic spin-spin correlation in oxides on the ultrafast time scale [3].

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