RESONANT INELASTIC X-RAY SCATTERING ON γ-Al₂O₃/SrTiO₃-HETEROSTRUCTURES

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Transition metal oxides show a vast variety of physical properties due to the interplay of cooperating and competing microscopic degrees of freedom. Recently, with the enormous progress in pulsed laser deposition and molecular beam epitaxy it has become possible to combine several oxides in epitaxially grown heterostructures in order to control these properties or even create novel functionalities. The interface between LaAlO₃ (LAO) and SrTiO₃ (STO) for instance hosts a two dimensional electron system (2DES) exhibiting ferromagnetism and superconductivity although both constituents are wide band gap insulators. The observation that both interface magnetism as well as conductivity only appear above a critical LAO overlayer thickness of 3 and 4 unit cells (uc), respectively, has been related to an electronic reconstruction resulting from the polar discontinuity between LAO and STO. Descrepancies in electron densities determined by spectroscopic methods to those determined by Hall measurements on these kind of heterostructures as reported by Berner et al. suggest the coexistence of mobile and localized carriers and has been demonstrated by Zhou et al. [1,2]. Nevertheless, the origin of these two types of carriers is still not fully understood. In particular, ferromagnetic properties are often at least partially attributed to oxygen vacancies and/or defects in the STO substrate [3]. Very recently, Chen et al. observed the formation of a 2DES exhibiting very high electron mobilities and charge carrier concentrations in the also polar/non-polar heterostructure γ -Al₂O₃/SrTiO₃ (GAO/STO) above a critical thickness of 1 uc of GAO [4]. Here it is supposed, that the 2DES is stabilized by oxygen vacancies in the STO side of the interface which is in glaring contrast to the LAO/STO heterostructure system.



Figure 1: XAS spectrum of 2.5 uc GAO/STO sample, Ti L-edge

To elucidate the influence of oxygen vacancies in such kind of heterostructures and to learn more about the driving mechanism in the two above mentioned cases, we performed resonant inelastic x-ray scattering (RIXS) on GAO/STO heterostructures with different overlayer thicknesses at various photon energies across the Ti L-edge (used photon energies are marked in the XAS spectrum of the 2.5 uc GAO/STO sample shown in Fig 1.). When measuring RIXS at e_g -resonance ($\approx 459.9 \text{ eV}$), beside the elastic peak, a new inelastic line from an intermediate state of the type $t_{2g}^{l}e_{g}^{l}$ or e_{g}^{2} appears at an energy loss of about 2.4 eV which intensity increases with increasing overlayer thickness (Fig. 2, left). Interestingly, finite spectral weight can already be observed for the insulating 1 uc sample. In comparison with our previous RIXS measurements on LAO/STO, this fits to the observation of an inelastic signal appearing also for the insulating, but magnetic 3 uc sample and therefore is a first indication for local moments also in GAO/STO heterostructures. Nevertheless, the existence of magnetism has not been proved, yet. When changing the incident photon energy from the e_{g} - towards the t_{2g} -resonance the inelastic line splits up into two peaks showing different hv-dependence. While one peak emerges in all spectra at a constant energy loss of about 2.4 eV and therefore can be attributed to localized Ti 3d carriers, the second line shows up between 1.4 eV and 2.6 eV in all spectra (see Fig. 2, right). Such kind of excitations of non-constant energy loss in RIXS are the response of delocalized electrons, for which the photon absorption and emission are decoupled [5]. This is direct evidence for the coexistence of two types of charge carriers in GAO/STO similar to LAO/STO although the driving mechanism leading to the 2DES appears to be different.

Assuming that no electronic reconstruction is taking place in the GAO/STO system and the 2DES is just stabilized by oxygen vacancies, our results imply, that also in LAO/STO the electrons supplied by oxygen vacancies are partially localized and therefore influence the magnetic as well as the electronic properties.





Figure 2: left: RIXS spectra of GAO/STO heterostructures with various film thickness measured at eg-resonance, beside the elastic line and inelastic signal can be observed whose intensity increases with film thickness. right: RIXS spectra of 2.5 uc GAO/STO heterostructure for various photon energies, inelastic line splits up into two components showing difference dependence on the used photon energy

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