Interface states in LaAlO₃/SrTiO₃ as probed by

resonant inelastic x-ray scattering

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Transition metal oxides show a rich 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 the two simple band insulators LaAlO₃ (LAO) and SrTiO₃ (STO) for instance exhibits metallicity, magnetism and superconductivity above a critical film thickness of 4 unit cells (uc). Descrepancies in electron densities as obtained by spectroscopic methods to those determined by Hall measurements on these kind of heterostructures as reported by Berner *et al.* and also the coexistence of superconductivity and ferromagnetism suggest the coexistence of mobile and localized carriers [1]. The latter are often attributed to originate at least partially from oxygen vacancies, and therefore these defects could be responsible for magnetism in this system [2]. Recently Zhou *et al.* correlated two peaks in their resonant inelastic x-ray scattering (RIXS) spectra, which exhibit different hv-dependence, with two types of charge carriers [3]. Nevertheless, the nature of these two loss peaks is still not fully understood.

We performed RIXS on LAO/STO heterostructures with different overlayer thicknesses as well as on samples with an intentionally high concentration of oxygen vacancies at various photon energies across the Ti L-edge to shed light on the coexistence of the two types of Ti 3d carriers (used photon energies are marked in the XAS spectrum of the 6 monolayer LAO/STO sample shown in Fig 1.). When measuring RIXS at the "eg-resonance" (460.1 eV) beside the elastic line, a new inelastic peak appears at an energy loss of about 2.4 eV whose intensity increases with increasing overlayer thickness (Fig. 2). Finite spectral weight can already be observed for the 3 uc thick sample although it is insulating. This is consistent with the finding of a critical thickness for ferromagnetism in this heterostructure by Kalisky *et al.* [2].





Figure 1: X-ray absorption spectra of 6 uc LAO/STO sample. Used photonenergies for RIXS measurements are markes (a) to (o)

Figure 2: RIXS spectra of LAO/STO heterostructures with various film thickness measured at the e_g -resonance

A series of RIXS spectra of the 6 ML LAO/STO sample is shown in Fig. 3. When changing

the incident photon energy from the t_{2g} towards the e_g -resonance the inelastic line splits up into two peaks. While one peak shows up in all spectra at a constant energy loss of about 2.4 eV and can be attributed to localized Ti 3d electrons, the second excitation shifts between an energy loss of 0.5 and 3.5 eV in all spectra. Excitation of non-constant energy loss in RIXS are the response of delocalized electrons, for which the photon absorption and emission are decoupled [4].



Figure 3: RIXS spectra of 6 uc LAO/STO sample measured at various photon energies across the Ti L-edge. Two inelastic signals showing different hv-dependence can be observed.

Figure 4: RIXS spectra of LAO/STO samples grown at different oxygen partial pressure measured 1 eV below the e_g -resonance. An increase of the inelastic signal can be observed, while the ratio between the two signals stays constant.

To see whether we can attribute one of these two types of interface states to oxygen vacancies, we measured samples with an intentionally high concentration of oxygen vacancies by using different oxygen partial pressures during growth. As it can be seen in Fig 4., while the intensity of the inelastic signal increases with increasing oxygen vacancies concentration due to the concomitant electron doping, which is in agreement with conventional photoemission experiments as well as resistivity measurements, the ratio between the two split up inelastic signals seems to stay almost constant. From this observation it is difficult to trace back one or the other peak directly and exclusively to oxygen vacancies.

However, when assigning these two signals to localized and delocalized charge carriers, these results fit to theoretical predictions as well as experimental results that while one of the charge per oxygen vacancy stays localized at the vacancy site, the other charge is delocalized [5,6].

References

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