Spin-dependent Electron Transfer Dynamics at a Model Interface: Key Role of Bandstructure

Daniel Sánchez-Portal[1]
Moritz Müller, Pedro M. Echenique[1]

[1] Centro de Física de Materiales CSIC-UPV/EHU and DIPC, Paseo Manuel de Lardizabal 5, 20018 Donostia, Spain

daniel.sanchez@ehu.eus

We investigate theoretically the charge transfer dynamics of core-excited Ar on Co(001) and Fe(110). For these systems, recent core-hole-clock measurements [1] of the lifetime of the photo-excited 4s level of Ar* have shown a clear dependence on the spin of the excited electron. Minority electrons in the Ar* 4s resonance decay significantly faster than majority electrons. We investigate such processes using Green's functions techniques on top of semi-local density functional theory (DFT) calculations, explicitly including the effect of the coupling to the semi-infinite substrate [2, 3]. Our calculations agree with the observed behavior and allow analyzing in detail the origin of this phenomenon [4]. To gain a better understanding we have performed a detailed analysis of the energy and distance dependency of the substrate-adsorbate coupling. We have also considered different descriptions of the, inherently ill-defined, wave-packet describing the initial state of the photo-excited electron. The key ingredient behind the observed behavior is the spin-dependence of the bandstructure of the substrate and, in particular, the very different position of the Ar* 4s level within the projected band gap appearing around Gamma in the dispersive bands of Co(001) and Fe(110) for both spin channels.

References

[1] F. Blobner, et al., Phys. Rev. Lett. 112, 086801 (2014)
[2] D. Sánchez-Portal, Prog. Surf. Sci. 82, 313–335 (2007)
[3] M. Müller, et al., J. Phys. Chem. C 122, 7575-7585 (2018)
[4] M. Müller, PhD thesis dissertation; M. Müller, P. M. Echenique, D. Sánchez-Portal, in preparation

Figures

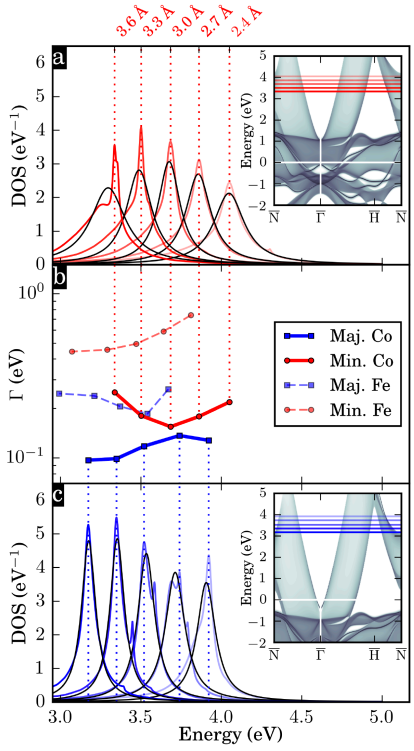

Figure 1. Computed first resonance above the Fermi level for Ar*($2p^{-1}4s$) on Co(0001) for adsorption distances ranging from 2.4 to 3.6 Å in the minority (a) and majority spin channel (c). Different shadings of the colored lines in the plots encode the adsorption heights. The energy positions of the resonance maxima are marked by horizontal lines with respect to the band structure of the Co(0001) surface in the insets, and strongly overlap with the region covered by band gaps around the Γ-point. Panel (b) collects the linewidths Γ(eV) of the Ar* resonances according to a Lorentzian fitting of the peaks (black lines) in (a, c). The spin-dependent linewidths of similar Ar*-resonances on Fe(110) are also shown in (b).