Attosecond Dual Nature of Core Excitons

Matteo Lucchini[1,2], Shunsuke A. Sato[3,4], Giacinto D. Lucarelli[1,2], Bruno Moio[1,2], Giacomo Inzani[1], Rocío Borrego-Varillas[2], Fabio Frassetto[5], Luca Poletto[5], Hannes Huebener[3], Umberto De Giovannini[3], Angel Rubio[3], Mauro Nisoli[1,2]

1. Department of Physics, Politecnico di Milano, 20133 Milano, Italy

2. Institute for Photonics and Nanotechnologies, IFN-CNR, 20133 Milano, Italy

3. Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany

4. Center for Computational Sciences, University of Tsukuba, Tsukuba 305-8577, Japan

5. Institute for Photonics and Nanotechnologies, IFN-CNR, 35131 Padova, Italy

Excitons are electron-hole quasi-particles, which strongly influence the electro-optical properties of solids and hold the potential for a variety of applications [1]. Acquiring a detailed understanding of their dynamic nature is thus essential to promote their exploitation in advanced technological areas. In particular, the ultrafast processes unfolding at the femto- and attosecond domain are of primary relevance in view of the desired extension towards the petahertz regime. Here we applied attosecond transient reflectivity spectroscopy to study the ultrafast response of a MgF_2 single crystal and report the first observation of sub-fs core-exciton dynamics [2]. Combined with simulations, our results allowed us to disentangle the dual atomic-solid nature of this fundamental quasi-particle.

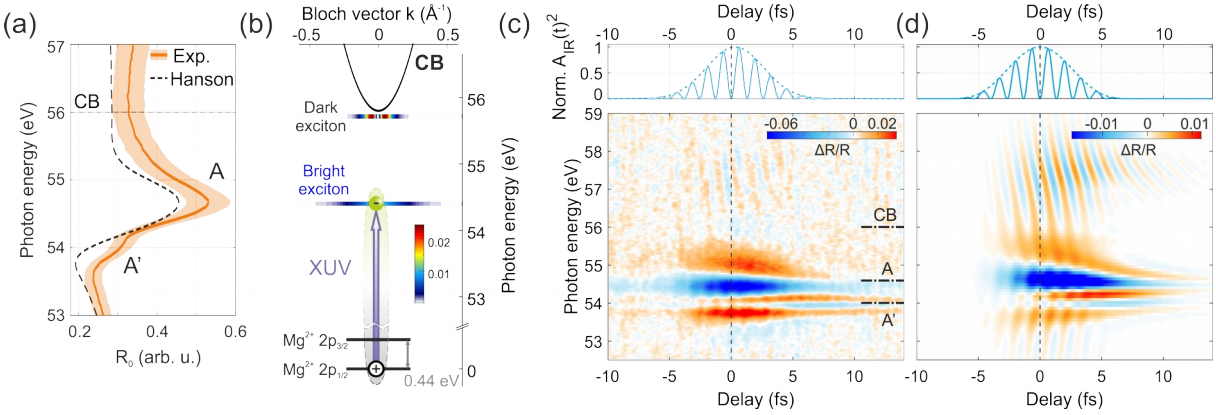

Fig. 1 (a) Static reflectivity of MgF_2 at 73.5° incidence angle measured (orange) or extracted from [3] (black-dashed curve). The experimental reflectivity shows two features around 54.5 eV (marked with A and A'), corresponding to core-exciton transitions. **(b)** MgF_2 conduction band and the excitonic states. **(c)** Experimental differential reflectivity, ΔR. The upper panel shows the square of the measured IR vector potential. **(d)** Same quantities as in (c) but calculated.

Around 54.5 eV, the MgF_2 reflectivity, R_0, is characterized by two peaks (A and A' in Fig. 1(a)), which correspond to the excitation of a Mg $2p^{2+} \rightarrow \Gamma^1$ core exciton [3] (Fig. 1(b)). In the experiment, we used few-fs infrared (IR) pulses (intensity ~10^{12} W/cm^2) to drive the exciton dynamics and we probed the induced modifications of these transitions with isolated attosecond pulses (~250 as). The differential reflectivity, ΔR, defined as the relative difference between sample reflectivity with or without the IR pump, is reported in Fig. 1(c) as a function of the pump-probe delay. We observe a rich dynamics composed by fast oscillations superimposed to a slower, few-fs signal, which is perfectly reproduced by our simulations (Fig. 1(d)). By exploiting our unique beamline [4], we can simultaneously measure the temporal behaviour of the IR vector potential (light-blue curve, upper panel). This allows us (*i*) to obtain an absolute calibration of the delay axis, and (*ii*) to perform a direct comparison with the calculated attosecond dynamics. As a result, we can show that while the few-fs component in ΔR originates from ac Stark shift of the excitonic transitions and can be described within an atomic model, the faster sub-fs component originates from the dispersive nature of the conduction band states, thus requiring a solid-like description. Moreover, we found that the absolute timing of the system optical response can be controlled on attosecond time scale by tuning the exciton binding energy, hence widening our knowledge of solid state phenomena and providing the community with a new lever to develop innovative devices for petahertz excitonics.

References

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35 words abstract Ultrafast core-exciton dynamics was measured in MgF_2 by attosecond transient-reflection spectroscopy. We found that the atomic nature of excitons dominates the few-femtosecond response, while their solid-state nature dictates the attosecond timing of the system.