

Attosecond core-exciton dynamics in MgF₂

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Excitons are quasi-particles which can form in solids after interaction with light, deeply influencing their electro-optical properties [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. We applied attosecond transient reflectivity spectroscopy to study the ultrafast response of a MgF₂ single crystal and report the first observation of sub-fs core-exciton dynamics [2]. Comparison with numerical simulations allowed us to disentangle the dual atomic and bulk nature of the exciton which originates competing effects.

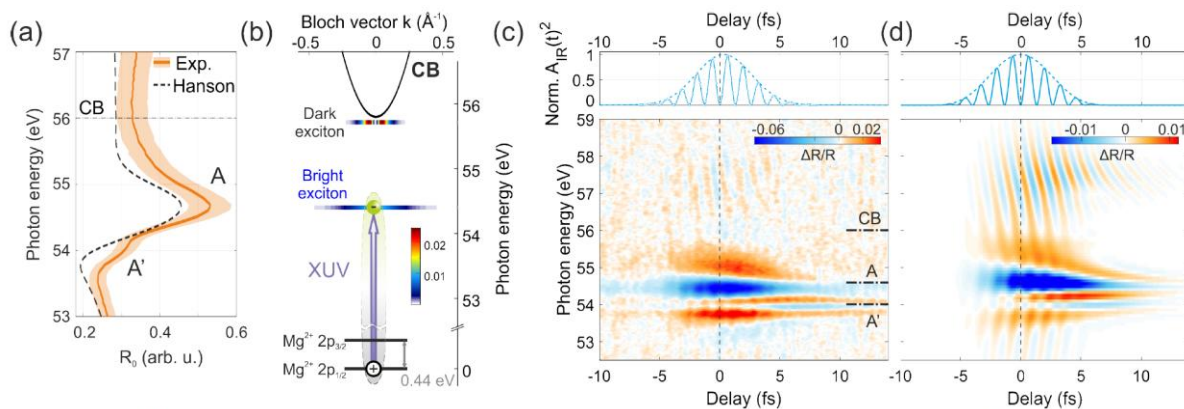


Figure 1 : (a) Static reflectivity of MgF₂ at 73.5° incidence angle. The exciton transitions are marked with A and A'. (b) MgF₂ conduction band and the excitonic states. (c) Experimental differential reflectivity, ΔR . The upper panel shows the square of the measured IR vector potential. (d) Same quantities as in (c) but calculated.

In particular, we used a few-fs infrared pulses (intensity $\sim 10^{12}$ W/cm²) to drive the exciton dynamics in MgF₂ and probed them with isolated attosecond pulses (~ 250 as) around the Mg $2p^{2+} \rightarrow \Gamma^1$ core exciton transitions (Fig. 1 (a), (b)) [3]. The resulting differential reflectivity trace 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 investigate the sub-fs timing of the transient features. This allowed us to 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, providing the community with a possible new lever to develop innovative devices for petahertz excitonics.

[1] L.V. Butov, Superlattices Microstruct. **108**, 2–26 (2017).

[2] M. Lucchini, et al., Nat. Commun. **12**, 1021 (2021)

[3] W. F. Hanson, et al., J. Appl. Phys. **43**, 1661 (1972).

[4] G. D. Lucarelli, et al., Rev. Sci. Instrum. **91**, 053002 (2020).