Intra-atomic angular momentum induced attosecond delays in solid state photoemission

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Attosecond EUV pulses allow investigating electron dynamics with attosecond temporal resolution by recording shifts of the photoelectron streaking in a simultaneously present IR field. The origin of the relative photoemission delays is not yet fully understood demonstrating our limited understanding of the photoemission process. As demonstrated here attosecond time-resolved photoemission from the van der Waals crystal WSe$_2$ (Fig. 1, lower panel) combined with theoretical modelling shows that intra-atomic effects such as the centrifugal barrier and other intra-atomic corrections significantly affect the relative delays between different emission channels. Identification of this effect [1] is enabled both by the long-term stability of the inert WSe$_2$ surfaces and by the detection of four qualitatively different emission channels (VB, Se 4s, W 4f, and Se 3d core levels).

The observed delays cannot be attributed to photoelectron propagation in the solid (Fig. 1, upper right panel) but intra-atomic corrections (Fig. 1, upper left panel) must be taken into account to match the experimental results. State-of-the-art photoemission models emphasize the translational invariance in the solid and include scattering by the surface. In contrast, the effect of the significant intra-atomic delay is related to the fact that the photoelectron initially experiences an “atomic” spherically symmetric environment. Only after some time, as the wave propagates to neighboring atoms, the photoelectron feels the structure of the crystal.

This sheds new light on the fundamental mechanism involved in the photoemission process and shows that a refined model of photoemission accounting for these local effects is needed to correctly model the kinematics of the photoemission process.