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Synopsis   We propose a theory of interferometric measurement of a normal Auger decay width in molecules. Molecular Auger interferometry is based on the coherent phase control of Auger dynamics in a two-colour ($\omega/2\omega$) laser field. We show that, in contrast to atoms, in oriented molecules of certain point groups (e.g. CH_3F) the relative $\omega/2\omega$ phase modulates the total ionisation yield. A simple analytical formula is derived for the extraction of the widths of Auger-active states from a molecular Auger interferogram, avoiding the need of either attosecond or high-resolution spectroscopy.

We present Auger interferometry, i.e. a theory of coherent control of Auger decay and single-photon laser-enabled Auger decay (spLEAD) [1] in ionised molecules belonging to the molecular point groups C_s, C_n, C_{nv}, $C_{\infty v}$, C_{3h}, D_3, D_{3h}, and T_d. It is introduced as a measurement of the total yield of a normal Auger decay (or spLEAD) in oriented molecules as a function of the relative $\omega/2\omega$ phase. Our analytical theory predicts that the Auger decay width can be reconstructed from the relative phase scan of the Auger yield modulation, shown in Fig. 1. The interference contrast $\mathcal{M}$ onto which the decay width is mapped can be maximised at any decay width by controlling the ratio of the ω- and 2ω-field intensities. As a result, the interferometric measurement proposed here is free of the limitations of both high-resolution Auger electron spectroscopy [2] (struggling to characterise decay widths smaller than the instrumental energy resolution) and attosecond time-resolved spectroscopy [3] (struggling to characterise decay rates faster than the available pulse durations). We suggest a method of extracting this information from the Auger interferograms by applying a simple analytical formula

$$\Gamma_1 = \frac{1 - \sqrt{1 - \mathcal{M}^2}}{\mathcal{M}} \frac{d_{01} d_{1E} \mathcal{E}_1^2}{d_{0E} \mathcal{E}_2}.$$

An illustrative example dealing with inner valence hole decay in CH_3F shows that molecular

Auger interferometry is well within the present-day experimental capabilities of the modern FEL facilities, such as FERMI@Elettra. More generally, the same coherent control scheme can be used to study any type of ultrafast hole dynamics, e.g. hole migration [4].
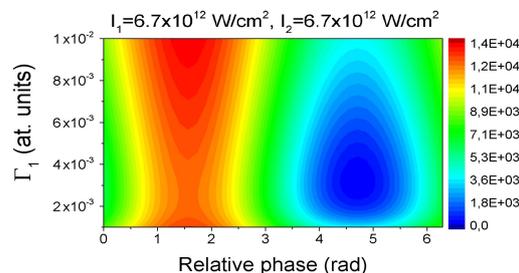

Figure 1. Auger interferograms. The total yield of electrons emitted by a bichromatic laser field for the case of CH_3F^+ as a function of the relative phase ϕ and the AAS width Γ_1 (y-axis).

References

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