Molecular Auger Interferometry

M Khokhlova[1]*, B Cooper[2], K Ueda[3], K C Prince[4], P Kolorenč[5], M Ivanov[1,6] and V Averbukh[1]

[1]Blackett Laboratory, Imperial College London, London SW7 2AZ, UK
[2]Department of Physics and Astronomy, University College London, London WC1E 6BT, UK
[3]Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan
[4]Elettra-Sincrotrone Trieste SCpA, Basovizza-Trieste 34149, Italy
[5]Charles University, Institute of Theoretical Physics, V Holešovičkách 2, Prague 180 00, Czech Republic
[6]Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Strasse 2A, Berlin D-12489, Germany

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].

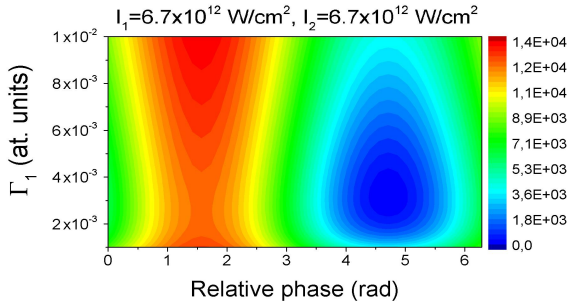

Figure 1. Auger interferograms. The total yield of electrons emitted by a bichromatic laser field for the case of CH_3F^+ as a function of the relative phase ϕ and the AAS width Γ_1 (y-axis).

References

[1] Cooper B and Averbukh V 2013 *Phys. Rev. Lett.* **111** 083004; Iablonskyi D *et al* 2017 *Phys. Rev. Lett.* **119** 073203
[2] Jurvansuu M, Kivimäki A and Aksela S 2001 *Phys. Rev. A* **64** 012502
[3] Drescher M*et al* 2002 *Nature* **419** 803
[4] Calegari F *et al* 2014 *Science* **346(6207)** 336-339; Kraus P M *et al* 2015 *Science* **350(6262)** 790-795

*E-mail: m.khokhlova@imperial.ac.uk