Attosecond Pump-Probe Measurement of an Auger Decay

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Abstract: We characterize the Auger decay of core-shell excitations in Krypton atoms in an all-photonic fashion, using a novel attosecond EUV pump - attosecond optical probe scheme. An intuitive model, where the optical field acts as a temporal amplitude and phase gate on the EUV excited dipole, is used to retrieve the Auger core-hole decay time.
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1. Introduction

The advent of extreme ultraviolet (EUV) attosecond (1 as=10^{-18}s) pulses and relevant techniques, such as attosecond streaking [1], have opened the way to real-time measurement of Auger decay by tracing the dynamics of the liberated electrons [2]. However, study of electronic processes in the bulk of solids, where photoelectrons can be captured in the crystal lattice or be significantly distorted as they travel their way out, calls for development of full photonic methodologies with adequate temporal resolution. Transient absorption techniques based on attosecond EUV and few femtosecond infrared pulses in gases [3] and solids [4] have allowed first steps in this direction but their temporal resolution is impaired by the few femtosecond duration of the optical pulse.

Here, we present attosecond pump - attosecond probe transient absorption experiments which combine the virtues of an EUV pulse used to electronically excite the atom of Krypton and an attosecond light transient [5,6] used to probe the decay of the excited state. The optical probe acts as a quasi-instantaneous phase and amplitude gate on the coherently radiating electronic dipole. An intuitive model is then used to reconstruct the dipole decay from the pump–probe trace, retrieving both the decoherence times and polarizabilities of the transitions.

2. Attosecond pump-probe experiments

In our experiments we use a ~150 as EUV pulse centered at 92 eV to resonantly excite Krypton (Kr) atoms by promoting 3d core electrons to valence excited levels np, ($n \geq 5$, see process 1 as illustrated in Fig.1A). These excitations will decay due to the Auger filling of the 3d hole, known to last around 8 fs [2]. As the system has been studied both in the spectral and time domain [2,7], it is an ideal candidate to explore this new approach.
We probe the decoherence process (see steps 2' and 2'' in Fig.1 A.) using a delayed multioctave (270-1100 nm) field synthesized light transient of full-width half maximum field intensity duration of 400 as [6], kept at a moderate peak intensity of 10^{13} W.cm^{-2}.

To highlight the changes made by the probing field, the differential absorbance is depicted in the false-color plot of Fig.1.B. To be noted is the sharp rise of the signal when the probing optical field starts overlapping with the exciting pump around the delay 0 fs, and the slow fall at positive delays, mapping the decay of the electronic dipole.

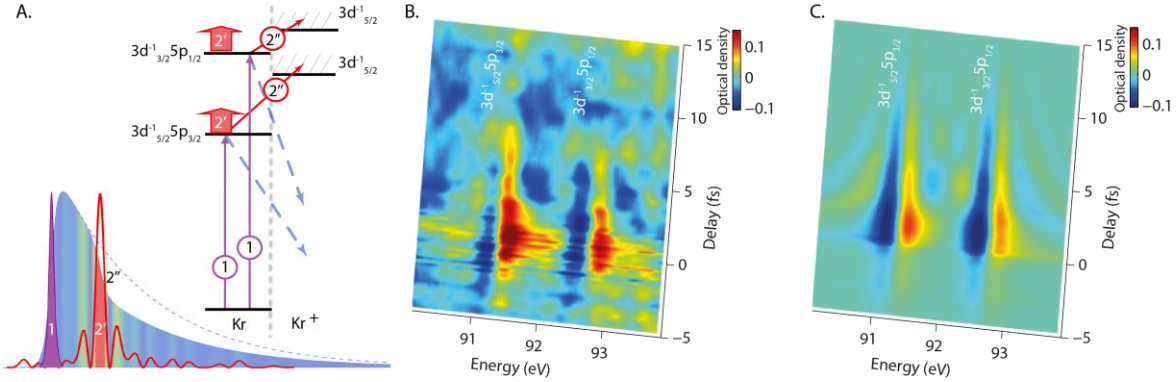

Fig. 1 (A) Energy level diagram and schematics showing the two effects of the probing field on the decaying Auger electronic dipole, Stark phase shifting (2', instantaneous frequency in the color coding) and amplitude depletion (2'') by ionization. **(B)** Experimental differential absorption cross-section spectrogram, comprising 65 pump-probe delay instances, averaged on 30 measurements. **(C)** Reconstruction of the map based on the fitting of a phase and amplitude gated response function

3. Modeling as a phase and amplitude gating of decaying dipoles

Fig.1.A gives an intuitive picture of the gating processes at stake. First in process 2', the probe field has the ability to polarize the dipole and Stark-shift its frequency, thus changing the phase of its oscillations [5, 8, 9]. This constitutes the phase gate. The probe can also directly ionize the promoted electron from the np excited levels, which have an ionization potential limited to a few eVs. This will reduce the amplitude of the dipole, shown as process 2'' with the undepleted time-dependent dipole represented by the dashed line in the graph of Fig.1.A. Both of these gating transformations - phase and amplitude - impact the absorption cross-section in a delay-dependent fashion.

Guided by this intuitive understanding, we model our experiment using for each transition an exponentially decaying response, perturbed by a temporal phase shift and amplitude loss, imposed at the variable pump-probe delay. This model is used to fit the experimental trace of Fig. 1.B. as a whole, using as free parameters the core hole decay time, as well as the polarizabilities and fractional ionization of the two transitions, $3d_{3/2}$-$5p_{1/2}$ and $3d_{5/2}$-$5p_{3/2}$. The fitted spectrogram is shown on Fig. 1.C and agrees well with the experiment, within the noise of our current measurement. Our fit yields a decay time of the core hole of T_{3d} = 8.8 +/-1 fs, compatible with previous high-resolution spectral spectroscopy [7] and a unique time-resolved measurement [2]. In addition, our approach retrieves the optical polarizabilities of the studied transitions, as well as the fractional ionization induced by the transient field itself. As a result, integrated over the entire optical attosecond probe gate, we obtain an accumulated phase shift of $\phi = 0.2\,\pi$ for both lines, and an amplitude depletion on the order of 5% and 15% for the low and high energy lines.

4. Conclusion

We demonstrated a photonic methodology for tracing Auger decay in atoms with attosecond resolution. This method can readily be applied to study various decoherence processes in solid state systems where eV-range spectral linewidths suggest subfemtosecond decays, so far only inferred from time-integrated spectroscopy.

5. References

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