

Ponderomotive dressing of doubly-excited states with intensity-controlled laser light

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Abstract. We laser-dress several doubly-excited states in helium. Tuning the coupling-laser intensity from perturbative to the strong-coupling regime, we are able to measure phases imprinted on the two-electron wavefunctions, and observe a new continuum coupling mechanism.

1 Introduction and Motivation

Fano resonances, described by the formalism of configuration interaction [1] are one of the most fundamental consequences of the quantum behavior of nature. They can also be regarded as a natural quantum interferometer featuring two distinct pathways of photoionization: direct ionization and (auto-)ionization of the system via a metastable (often doubly-excited) state (Figure 1). The interference of these two arms of the interferometer gives rise to asymmetric (Fano) line shapes in the photoionization cross section. The asymmetry parameter q is directly related to the relative transition strength and phase between the two pathways. Especially the metastable arm of this interferometer is of particular interest, as it often describes the correlated motion of two (or sometimes more) electrons.

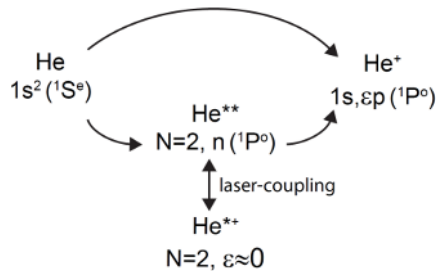


Fig. 1. Illustration of the Fano mechanism and the additional continuum coupling created by a laser field, as applied to the $N=2$ doubly-excited states He^{**}. As we find, these metastable states can be coherently laser-coupled to singly-excited He^{*+} continuum states, where the energy ϵ of the continuum electron is close to zero and thus the electron remains in the vicinity of the helium atom without being ionized. To our knowledge, this important phase-control channel, governing the shape of the Fano resonance, has thus far only been treated as a loss mechanism.

We experimentally observe [2] a significant modification of the Fano asymmetry parameter q (up to negation) as a function of the laser intensity, which we interpret as the selective manipulation of one interferometer arm as further illustrated in Figure 1. As a result of this coupling, both amplitude and phase of the autoionizing state can be significantly modified.

2 Experimental Setup

We use visible (VIS, centered around 730 nm) ultrashort (~ 7 fs) laser pulses to create attosecond-pulsed continuous soft-x-ray (SXR) radiation around the 60-70 eV energy range. The SXR light is transmitted through a 100 mbar helium target and simultaneously excites the full range of dipole-allowed ($^1P^o$) autoionizing states of the $N=2$ series in the presence of the time-delayed VIS pulses. Our high-resolution spectrometer ($\Delta E = 20$ meV @ 60 eV) allows to observe states up to $n = 7$ in the absorption spectrum and gives access to the line shapes of these states. The intensity of the VIS light is finely controlled with a picomotor-driven iris aperture from 0 to several 10^{12} W/cm² peak intensity.

3 Experimental Results and Discussion

Figure 2 shows experimentally observed absorption line shapes of doubly-excited states in helium at various laser intensities in the 10^{12} W/cm² regime. Remarkably, the absorption signatures of all states remain intact, with only slight signal decrease due to ionization, for intensities near 3.5×10^{12} W/cm², far beyond over-barrier ionization intensities. Most interestingly, at intensities higher than 3×10^{12} W/cm², for the highest-lying states, an inversion of the line profile is observed, i.e. the original q -parameter has changed its sign. This shows that in addition to loss, the phase of the doubly excited state is modified by laser coupling, as discussed above.

To understand the phase change we make use of the current understanding of single active electrons interacting with laser light. Electrons close to the ionization threshold can be treated as quasi free and can thus not efficiently be ionized due to momentum mismatch. However they can acquire a phase $\Delta\Phi$, the so-called ponderomotive phase, i.e. the time-integrated kinetic energy the electron acquires during its wiggling motion in the electric field $E(t)$. It is given by

$$\Delta\Phi = \frac{1}{\hbar} \int_{-\infty}^{+\infty} dt' \frac{1}{2} m_e v(t')^2 \quad (1)$$

with

$$v(t) = -\frac{e}{m_e} \int_{-\infty}^t dt' E(t'). \quad (2)$$

Using this formula, for our pulse duration a phase change $\Delta\Phi = \pi$ (thus inverting q) is expected for intensities around 5×10^{12} W/cm², which is in good agreement with the experimentally observed values.

Only for the highest states ($n = 5, 6, 7$) an almost complete inversion of the line shape is observed, while it is incomplete for the lower states ($n = 3, 4$) (see also Figure 2). This is due to the increasingly invalid assumption of a single active and separable quasi-free electron in the lower-lying states, where both electrons have to be treated on equal footing. The state-resolved analysis of the Fano line shapes thus marks an interesting transition between single and two-electron laser-driven dynamics as well as from the classical (ponderomotive, large phase space) to quantum treatment (only a few laser-coupled states).

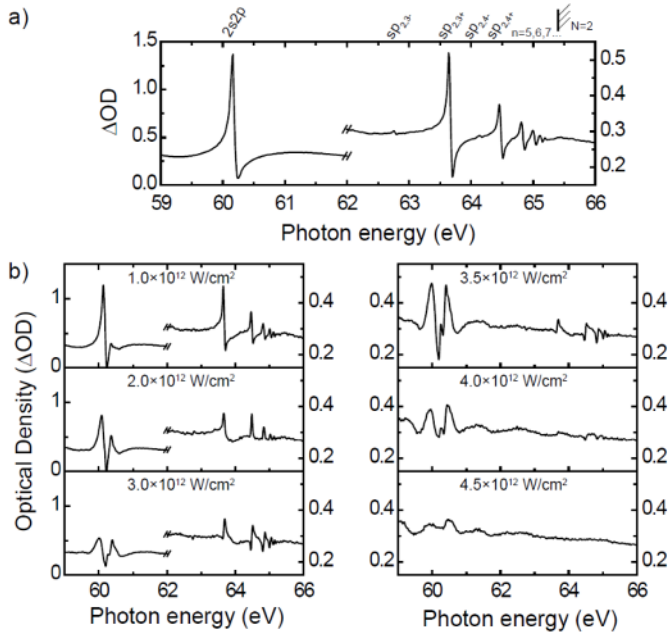


Fig. 2. Experimentally observed laser-dressed absorption line shapes across the $N = 2$ doubly-excited states series in helium. a) Undisturbed Fano line shapes where no VIS laser is present. $sp_{2,n+}$ states are observed up to $n = 7$. b) Absorption line shapes for different VIS laser peak intensities ranging from $1 \times 10^{12} \text{ W/cm}^2$ to $4.5 \times 10^{12} \text{ W/cm}^2$, where the VIS laser pulse was time-delayed at +5 fs after the SXR excitation. All higher resonances ($n \geq 3$) change their shape (up to q -negation) before eventually disappearing, evidencing the laser-induced phase change of the doubly-excited states. Near the $2s2p$ line (around 60 eV), for increasing intensity, we observe the formation and repulsion of an Autler-Townes doublet of coupled autoionizing states, marking the transition from the perturbative to the strong-coupling regime of doubly-excited states in helium.

4 Conclusion and Outlook

We experimentally measure changes both in amplitude and phase of laser-dressed doubly-excited states in helium by making use of the natural Fano-type quantum interferometer. We observe the laser coupling to an additional continuum for states where the second electron is in quantum states with $n \geq 5$. The phase of metastable two-electron states can be controlled by this new mechanism with moderately intense laser fields, with only minor losses due to ionization. These results suggest the possibility of controlling chemical reactions on the electronic time scale by selectively tuning the phases of metastable two-electron transition states governing the rearrangement of molecular bonds, which typically consist of two electrons per bonding orbital.

References

1. U. Fano, Phys. Rev. **124**, 1866-1878 (1961).
2. C. Ott, A. Kaldun, P. Raith, K. Meyer, M. Laux, Y. Zhang, S. Hagstotz, T. Ding, R. Heck, T. Pfeifer (submitted 2012).