Line-shape broadening of an autoionizing state in helium at high XUV intensity

Lennart Aufleger, Patrick Friebel, Patrick Rupprecht, Alexander Magunia, Thomas Ding, Marc Rebholz, Maximilian Hartmann, Christian Ott and Thomas Pfeifer

Max-Planck-Institut für Kernphysik, Heidelberg, Germany

E-mail: aufleger@mpi-hd.mpg.de, christian.ott@mpi-hd.mpg.de and tpfeifer@mpi-hd.mpg.de

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Abstract

We study the interaction of intense extreme ultraviolet (XUV) light with the 2s2p doubly excited state in helium. In addition to previously understood energy-level and phase shifts, high XUV intensities may lead to other absorption-line-shape distortions. Here, we report on experimental transient-absorption spectroscopy results on the 2s2p line-width modification in helium in intense stochastic XUV fields. A few-level-model simulation is realized to investigate the origins of this effect. We find that the line-shape broadening is connected to the strong coupling of the ground state to the 2s2p doubly excited state which is embedded in the ionization continuum. As the broadening takes place for intensities lower than for other strong-coupling processes, e.g. observed asymmetry changes of the absorption profile, this signature can be identified already in an intermediate intensity regime. These findings are in general relevant for resonant inner-shell transitions in nonlinear experiments with XUV and x-ray photon energies at high intensity.

1. Introduction

Due to the simplicity of its electronic structure, helium is an ideal system to enhance our understanding of fundamental electron–electron interaction. In particular, the doubly excited states have been an excellent playground for theories of multi-electron correlation. With the two pathways of ionization via the doubly excited autoionizing state and via direct ionization into the single-ionization continuum the system acts as a quantum interferometer and hence is sensitive to changes in its pathways. This effect was first described by Fano [1] and is associated with the Fano-profile absorption line shape. The behavior of these doubly excited states in intense fields has been investigated theoretically in the presence of strong near infrared (NIR) and extreme ultraviolet (XUV) radiation fields [2–5].

With high-order harmonic generation sources, the resonant excitation and probe of these transitions under the influence of ultrashort NIR lasers became experimentally accessible. The interest has focused particularly on investigating the laser coupling of two-electron dynamics with intense NIR sources [6–11]. More recently, a study revealed the temporal build-up of the Fano resonance around an isolated doubly excited state [12].

The advent of free-electron lasers (FELs) [13] pushed the limit of multiphoton physics beyond the optical spectral region to the XUV and x-ray regime. Besides the possibility of nonlinear spectroscopy of doubly excited states [14] the availability of intense XUV light sources has enabled the resonant dressing of the doubly excited states directly from the atomic ground state. In dependence of the duration of the stochastic FEL pulses [15] the observation of strong coupling effects via the symmetry distortion of the absorption line shape [16] became possible. The question arises whether the intense field leads to further mechanisms, for example by manipulating the natural lifetime of the autoionizing state. This effect would lead to a change in the width of the absorption line.
Here we present a study on intensity-dependent line-width broadening of the 2s2p autoionizing absorption line in helium driven directly with intense and partially coherent XUV fields from self-amplified spontaneous emission (SASE). This experiment is performed using a transient-absorption spectroscopy (TAS) scheme, an all-optical method in transmission mode, which is sensitive to the interference of the incoming and dipole-emitted light in forward direction [7, 8, 12, 15–17].

2. Ionization mechanism of helium via the 2s2p state

The measurement scheme, which is based on Fraunhofer-type transmission spectroscopy, is illustrated in figure 1. For the case of the lowest dipole-allowed two-electron resonance in helium, XUV fields couple the ground state (1s2) to the doubly excited state (2s2p). The photon energy for this transition (60.15 eV) is above the single-ionization threshold (24.6 eV). Hence, the state is embedded in the continuum as an autoionizing state. With a lifetime of 17 fs the state almost exclusively decays non-radiatively into the single-ionization continuum of the He2+ ground state [6, 18]. A parallel channel of direct ionization from the ground-state helium to the same final state of He2+ exists and the interference of both transition amplitudes leads to the typical Fano absorption line shape in the weak-field limit [1].

SASE-based FEL pulses contain a structure of coherent sub-spikes [19–21]. With the FEL set to a central photon energy of 60 eV at an average bandwidth of 0.4 eV, the duration of the temporal features is about 5 fs according to the Fourier-transform limit, even though the overall pulse duration might be significantly longer. These ultrashort temporal spikes provide very high peak intensities, enabling the strong coupling of the doubly excited state and leading to an asymmetry change of the absorption line shape [15, 16].

Further effects beyond the weak-field limit may also affect the width of the absorption line which is at the center of our study. The coupling from the 2s2p doubly excited state back to the 1s2 ground state can influence its natural autoionization lifetime. Furthermore, additional loss channels can occur, for instance due to the direct coupling by the intense electric fields to the n = 2 continuum. Both couplings by the intense XUV fields can change the intrinsic dynamics of the excited state and would manifest in an observable change of the ensemble-averaged absorption line.

3. Experiment

To investigate the width behavior of the absorption line shape under the influence of intense XUV pulses, an experiment was performed at the free-electron laser in Hamburg (FLASH). A home-built XUV-TAS setup was connected to the open-port beamline BL2. The FEL was set to single-bunch mode with 10 Hz repetition rate. An ellipsoidal mirror of 1 m focal length delivered the beam to the target volume and focused it down to an estimated spot size of 10 μm. As the target, helium gas at a path-length-density product on the order of 10^{17} cm^{-2} was used. A flat-field variable-line-space grating spectrally dispersed the transmitted XUV beam and its spectra were measured with a back-illuminated XUV-sensitive Charge-Coupled Device (CCD) camera. A facility-based parasitic reference spectrometer [22] simultaneously recorded single-shot XUV spectra as reference ahead of the TAS setup.
Figure 2. (a) Experimental TAS: absorption lines of the helium 2s2p resonance measured at FLASH with SASE-based XUV pulses at a central energy of 60 eV and 0.4 eV bandwidth for a range of pulse energies (dots). Note: for visualization, the sequential lineouts, starting from 93 μJ, are shifted by an 0.1 offset in OD. The lineouts are fitted with the Fano formula (solid lines, equation (2)). (b) The line widths are extracted from the fit parameters and fitted linearly to indicate the trend.

The pulse energy was measured parasitically with a gas-monitor-detector (GMD) [23, 24] on a single-shot basis. The statistical distribution of the pulse energy averaged to 75 μJ and spanned over several ten μJ. Due to the losses at the XUV optics, the beamline transmission is estimated at 50%, resulting in 36 μJ averaged pulse energy in the target volume. Due to the statistical nature of the SASE FEL, bins on the upper end of the scale, e.g. around 93 μJ, are reached by scattered events when the average GMD energy is set to 75 μJ. To control the pulse energy of the XUV pulses and increase the number of events for energetically lower bins, the pulse energy was dynamically adjusted with a facility-based gas absorber. It was filled with a tunable density of molecular nitrogen, which presents a non-resonant absorber medium in the spectral region of interest.

The resulting averaged absorption spectra (figure 2(a)) show the expected Fano line shape. With increasing GMD pulse energy, starting from the weak-field limit, the amplitude of the resonance reduces significantly. This agrees with previous measurements [15, 16] and is attributed to saturation effects in the target.

To analyze the line-width behavior, the absorption spectra are fitted for quantification with the normalized Fano profile [7]:

\[
\text{OD}(\omega, E_p) = a + \frac{q}{1+q^2} \left( \frac{(q + \epsilon(\omega))^2}{1 + \epsilon(\omega)^2} - 1 \right) + b \tag{2}
\]

with the reduced energy \(\epsilon(\omega) = 2(\omega - \omega_r)/\Gamma\), the asymmetry parameter \(q\), the line width \(\Gamma\), resonance position \(\omega_r\), the amplitude \(a\), and the offset \(b\). The spectrometer resolution of 59 meV full width at half maximum (FWHM) is accounted for by convoluting the Fano line-shape function (equation (1)) with a Gaussian of that width before applying the least-squares algorithm.

The line width starts at about 35 meV in the weak-field limit (figure 2(b)) and increases for higher pulse energy. The increasing decay width \(\Gamma \propto 1/\tau\) indicates a significant change of the 2s2p lifetime \(\tau_{2s2p}\) for XUV intensities beyond the weak-field limit.
4. Simulation

A computational few-level model is employed to investigate the origin of the line-width increase. The time-dependent Schrödinger equation (TDSE) $i \frac{\partial}{\partial t} |\Psi(t)\rangle = H(t) |\Psi(t)\rangle$ is solved by a diagonalization of the three-level Hamiltonian matrix

$$H = \begin{pmatrix} E_e & d_{ge} \cdot E(t) & d_{ge} \cdot E(t) \\ d_{ge} \cdot E(t) & E_e + i \cdot \Gamma_c(t)/2 & V_{CI} \\ d_{ge} \cdot E(t) & V_{CI} & E_e + i \cdot \Gamma_c(t)/2 \end{pmatrix}$$

constituting of only the involved resonant transitions and their coupling to the continuum.

The complex eigenenergy of the continuum, in particular its position $E_e = 32.65$ eV and width ($\Gamma_c = 39.73$ eV), as well as the dipole-matrix element ($d_{ge} = 0.6753$ a.u.), which couples the direct ionization from the ground state, are chosen such that the off-resonant absorption cross section of helium is reproduced in this spectral region in the weak-XUV-field limit. While some formulas are given in SI units below, the calculations were performed solely in atomic units.

With the eigenenergy of the ground state set to $E_g = 0$ eV, the values of the excited state, the eigenenergy $E_e = 60.12$ eV, dipole matrix elements ($d_{ge} = -0.04932$ a.u.), and the configuration interaction $V_{CI} = 0.0373$ a.u. coupling the excited state field free to the continuum are determined in a way that the model meets the tabulated values for the resonant 2s2p absorption line shape, again for the weak-field limit [18, 25].

To account for an additional loss channel such as photoionization to the $n = 2$ continuum, the imaginary part is introduced to the eigenenergy of the doubly excited state (equation (3), shaded grey). This acts as an exponential damping factor to the 2s2p-state’s population.

In accordance with the model with lowest-order perturbation theory of a photoionization rate, the latter is directly proportional to the photon flux density. In combination with the photoionization cross section $\sigma$, the loss rate results in

$$\Gamma_\sigma(t) = \sigma \frac{I(t)}{\hbar \omega}. \quad (4)$$

Note that the rate is not constant but depends on the temporally varying intensity envelope of the XUV pulse $I(t) = \frac{1}{2} e^2 |E^+(t)|^2$.

The XUV electric field $E(t)$, with pulses of SASE-like stochastic character, is generated with the partial-coherence model [26]. Starting from an envelope with an average bandwidth of 0.4 eV FWHM and an individual random phase per pulse, the single spectra are Fourier transformed and windowed with a Gaussian intensity envelope of 130 fs FWHM overall pulse duration. The real part $E(t) = \Re \{E^+(t)\}$ of the complex-valued electric field $E^+(t)$ is then utilized.

With this framework, the state vector $|\psi(t)\rangle = [c_g(t), c_e(t), c_c(t)]^T$ — initially fully in the ground state $|\psi(0)\rangle = [1, 0, 0]^T$ — is propagated for each time step $(\Delta t = 0.1$ a.u. $\approx 0.0024$ fs) on a discretized time grid. For this, the state vector is rotated into the diagonal basis and the time-evolution operator $e^{-i \hat{H} \Delta t / \hbar}$ is applied, with $\hat{E}_i$ being the in general complex-valued eigenenergy to the dressed state $i \in \{1, 2, 3\}$.

 Afterwards, the solution of the TDSE, the complex-valued time-dependent dipole moment $d^+(t) = d_{ge} \hat{E}_g(t) \cdot c_g^*(t) + d_{ge} \hat{E}_c(t) \cdot c_c^*(t)$ is Fourier transformed into the spectral domain $\tilde{d}^+(\omega) = \int dt d^+(t) e^{-i \omega t}$. The same applies to the electric field $E^+(t)$ with $\tilde{E}(\omega) = \int dt E^+(t) e^{-i \omega t}$. The optical density is then determined by

$$\text{OD} = -\log_{10} \left( \frac{\left| \Re \{\tilde{E}(\omega) + i \cdot \eta \cdot \tilde{d}(\omega)\} \right|^2}{\left| \Re \{\tilde{E}(\omega)\} \right|^2} \right) \quad (5)$$

with $\eta$ representing the macroscopic particle density, interaction length and other fundamental constants and its order of magnitude $\eta = 10^{-4}$ originating from calibrations of the OD to experimental results.

For the resulting optical density to directly encode the microscopic single-atom response, this value needs to be sufficiently small to suppress propagation effects. Hereby, the interferometric superposition of the input spectrum $\tilde{E}(\omega)$ and the macroscopic polarization response $\eta \cdot \tilde{d}(\omega)$ follow Maxwell’s equations. The accordingly propagated electric fields through the moderately dense target medium [27] are spectrally measured in forward direction. Analog to the experimental analysis, the results are averaged for each fluence for an ensemble of 300 SASE pulses.

This simulation is performed for a set of fluence values, corresponding to the experimental settings. Since neutral, ground-state beryllium has a similar atomic structure as the 2s2p excited helium, its photoionization cross section will serve as an initial estimate of the order of magnitude of $\sigma$, with
Figure 3. Few-level-model simulations of the helium 2s2p resonance with SASE-based XUV pulses at 59.7 eV central energy and 0.4 eV FWHM average spectral bandwidth. (a) The simulation was performed for a range of fluences with a loss-channel cross section \( \sigma = 0.336 \text{ Mbarn} \). The lineouts of the absorbance (thick & translucid) over photon energy show the characteristic Fano absorption line shape, which changes with increasing fluence from 0.001 J cm\(^{-2}\) (top) towards 35 J cm\(^{-2}\) (bottom). For illustration purposes, the sequential lineouts starting from 35 J cm\(^{-2}\) are shifted by a 0.1 offset in OD. Furthermore, the fit of the Fano profile to each absorption line is illustrated (thin & opaque). (b) Line width extracted from the Fano fit (equation (2)). In addition to the value of ground state beryllium \( \sigma_{\text{Be}} = 0.336 \text{ Mbarn} \) for the loss channel cross section, the dataset shows the result for values \( \sigma \in \{0, 0.5, 1.5\} \cdot \sigma_{\text{Be}} \). A linear fit (dashed) indicates the tendency.

\[ \sigma_{\text{Be}} = 0.336 \text{ Mbarn} \] at 60 eV photon energy [28]. The absorption lines show a decreasing amplitude with higher fluence (figure 3(a)). This is consistent with former [15, 16] and the experimental results (figure 2(a)) and explained by the depletion of the ground state. Furthermore, an increase in line width is visible for an increasing fluence.

For quantification, the line shape is fitted with the Fano-profile (equation (2)) analog to the experimental data. The extracted line width parameter (figure 3(b), green dots) confirms a general increase of the profile width with rising fluence by about 20 meV, comparable to the observed broadening in the experiment.

As the simulation is repeated for a set of cross sections \( \sigma \in \{0, 0.5, 1.5\} \cdot \sigma_{\text{Be}} \) to investigate the influence of a possible radiation-induced photoionization decay, the results show an increased line width with higher fluence proportional to the cross sections. As both quantities, the cross section as well as the intensity, have the same relation to \( \Gamma_{e}(t) \) (equation (4)), the line-width broadening due to this loss channel is linear for both quantities.

At the same time it is obvious that the dominant feature of the broadening in general is acting on the line width even without the loss-channel cross section (figure 3(b), blue dots). The utilized computational model has no other field dependencies apart from the off-diagonal dipole-matrix elements. Thus, the single possible way of influencing the intrinsic dynamics in this simulation is the coupling of the doubly excited state back to the ground state. This means, that the line-width broadening can be dominantly associated to the increased coupling of those two states, indicating an effectively shortened lifetime of the excited state with higher field strength.

5. Conclusion

We have shown that intense resonant XUV fields lead to a broadening of the 2s2p transition in helium. The origin of this line-shape change has been investigated and while a contribution of an additional loss channel through coupling of the doubly excited state to the \( n = 2 \) continuum has been considered, the major contribution within this model can still be attributed to the strong coupling of the doubly excited state back to the ground state. Consequently, the effective lifetime of the doubly excited, autoionizing state can be manipulated using exclusively strong coupling XUV pulses. Kanter et al. [29] made similar observations on the Auger resonances. In our case, however, a single-photon two-electron transition is investigated which is only possible due to electron correlation. As this effect is observed at intensities where symmetry distortions are not yet acting on the absorption line shape itself [15], it has to be considered for all experiments that drive resonant coupling at high intensity. Future investigations could further benefit from the superradiance mode of a seeded FEL [30], providing new opportunities for driving this fundamental correlated two-electron transition with well defined few-femtosecond XUV pulses.

Our findings contribute to the interpretation of transient-absorption spectra at FELs in general. They also extend the line-broadening mechanism from the optical to the all-XUV regime. Furthermore, this method is not limited to the fundamental system of helium, but can help to understand ultrafast ionization...
dynamics in heavier atoms and molecules, as well as solid-state systems. In the near future the insights of this study will also become important for XUV tabletop sources, as they are on the brink of achieving higher XUV intensities [31, 32], closing the gap and reaching into the low intensity regime of FELs, where this effect becomes relevant.

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**Data availability statement**

The data that support the findings of this study are available upon reasonable request from the authors.

**ORCID iDs**

Lennart Aufleger [https://orcid.org/0000-0003-4903-3981](https://orcid.org/0000-0003-4903-3981)

Patrick Friebel [https://orcid.org/0000-0001-9589-9933](https://orcid.org/0000-0001-9589-9933)

Patrick Rupprecht [https://orcid.org/0000-0002-6491-793X](https://orcid.org/0000-0002-6491-793X)

Marc Rehholz [https://orcid.org/0000-0002-3035-1412](https://orcid.org/0000-0002-3035-1412)

Maximilian Hartmann [https://orcid.org/0000-0002-6494-9368](https://orcid.org/0000-0002-6494-9368)

Christian Ott [https://orcid.org/0000-0002-5312-3747](https://orcid.org/0000-0002-5312-3747)

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