

TWO COLOR ATOMIC PHOTOIONIZATION BY FEMTOSECOND PULSES

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Pump-probe processes in the femtosecond domain with a probe VUV and dressing optical pulses are considered: ionization of the Li atom with the continuum coupled to the excited 3d state and ionization of the He atom with the coupled flatcontinua. In the first case, spectra and angular distributions of the photoelectrons were calculated and analyzed for two linearly polarized collinear fields by numerical solution of the non-stationary Schrödinger equation. The spectrum and the angular distributions are controlled by the time delay between the pulses and the two-photon detuning. In the second case, we concentrated on the modulation of the He ionization signal as function of the angle between the field polarization vectors. Within the second order time-dependent perturbation theory for the Gaussian field envelopes, a partly analytical solution was found and used.

Introduction

The advent of free electron lasers and lasers based upon high harmonic generation producing intense femtosecond pulses in the VUV opened up new possibilities in photoionization studies. In particular, the atomic and molecular ionization processes can now be investigated and controlled in the time domain. The corresponding experiments often use the pump-probe method, when two electromagnetic pulses act on a system subsequently. In this contribution, we consider theoretically photoionization in the pump-probe process with one ionizing VUV ('probe') pulse and one coupling optical ('dressing') pulse. The VUV femtosecond pulse transfers an atom from the ground state to the continuum, while the optical pulse couples this region of the continuum either with another continuum or with an initially

unpopulated discrete state, thereby slightly modifying the continuum. There are many publications treating such schemes, although mainly in stationary or adiabatic approaches and for model systems (see, for example, [1,2] and references therein). In this paper, we make theoretical predictions for particular atomic systems, lithium and helium. In the next section, we extend our recent method of the direct numerical solution of the time-dependent Schrödinger equation (TDSE) for hydrogen [3] to the lithium atom. Our goal is to consider the effect of coupling between the ionization continuum and a discrete state by the optical field on the near-threshold ionization from the ground state. In the subsequent section, we analyze the ionization of the helium atom by the VUV pulse far from threshold with the optical pulse, coupling different continua. Such a process has been already studied experimentally and

theoretically [4] concentrating on the ionization cross section. Here we consider a new aspect: the polarization dependence of the ionization cross section. For treating this process, we apply the second-order time dependent perturbation theory in accordance with the characteristics of the pulse parameters used in the experiment. In both processes, involving lithium and helium, we assume moderate field intensities.

Ionization of lithium

In [3] we numerically solved the TDSE for hydrogen in a bichromatic field, when the VUV and the optical pulse frequencies were tuned to couple the same region of the hydrogen continuum with the ground, $1s$, and excited, $n=3$, states, respectively. The optical pulse of higher intensity generates the so-called ‘laser induced continuum structure’ (LICS). It was shown that in the near-threshold region the photoelectron angular distributions (PAD) and the spectral shape of the main photoline could be controlled by the time delay between the two pulses and by the two-photon detuning. A similar scheme for the lithium atom is shown in figure 1. The linear polarizations of the two fields are collinear. The task of solving the TDSE is drastically simplified when a single active atomic electron is considered. This was the exact model for hydrogen, but it is an approximation for ionization from the $2s$ state of lithium. The potential for the valence electron in lithium was taken from the local approximation to the Hartree-Fock potential with additional polarization terms, thereby reproducing with an accuracy of 10^{-2} eV the energies of the Rydberg excited states and the ionization threshold. Calculations showed that the one-electron model with this potential also very well reproduces the experimental photoionization cross section [5] near the threshold. The method of solving the TDSE for the lithium atom initially in the ground state in the two fields with Gaussian envelopes (FWHM = 33 fs) was similar to that for the hydrogen atom and is described in detail in [3].

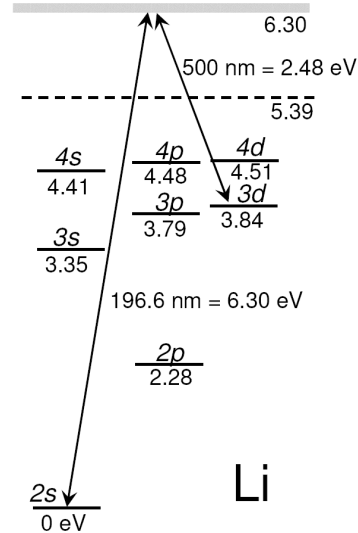


Fig. 1. Scheme of levels and driving fields for Li. Excitation energies in eV are given for the Li states.

Figure 2 shows our first lithium results for the integral photoelectron spectra with and without the coupling optical pulse. The peak intensities of the probe and the dressing fields were taken as 10^{10} and 10^{12} W/cm^2 , respectively, similar to [3]. The spectrum with the two switched pulses consists of additional lines due to the multiphoton ionization from the ground state, including above-threshold ionization (ATI) peaks. Owing to the smaller ionization potential and larger photon energy of the dressing field, the lithium spectrum, in contrast to hydrogen [3], contains a strong line due to three-photon ionization from the ground state as well as the four-photon ATI peak. The main photoline in lithium changes only slightly when switching on the dressing pulse. The PAD in the main photoline is sensitive to the time delay between the pulses. The PAD from the probe pulse only is described by (atomic units are used here and below)

$$\frac{d^2W}{dEd\Omega_k} = \frac{W_E}{4\pi} [1 + \beta P_2(\cos\theta)], \quad (1)$$

where β is the anisotropy coefficient with the value $\beta = 2$ within our nonrelativistic approach, θ is the angle between the direction of the photoemission with the momentum \mathbf{k} (energy $E=k^2/2$) and collinear polarizations of the VUV and optical fields, $P_2(x)$ is the

Legendre polynomial. Switching on the dressing pulse leads to additional Legendre polynomials of the even order in eq. (1).

Calculations show that the non-negligible contribution only comes from polynomials up to the order six

$$\frac{d^2W}{dEd\Omega_k} = \frac{W_E}{4\pi} [1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta) + \beta_6 P_6(\cos\theta)] \quad (2)$$

with the corresponding anisotropy coefficients β_L . The 6th polynomial corresponds to outgoing f-waves of the photoelectrons. For the main photoline, this is interpreted as population transfer to the f-continuum from the p-continuum through the 3d lithium state (see fig. 1). The delayed optical pulse leads to stronger modification of the PAD than any other time ordering of the pulses. As an example, for a time delay of 40 fs, the coefficients in (2) take the values $\beta_2 = +2.62$, $\beta_4 = +0.87$, $\beta_6 = +0.04$ at the photoelectron energy in the maximum of the main photoline.

main photoline by the time delay of the dressing pulse, which produces the laser induced continuum structure.

Ionization of helium

The direct numerical solving of the TDSE has obvious drawbacks: it is time consuming and the number of spatial dimensions in the problem is critical, the time and space propagation can be performed within certain limits, the dependence of the result on the input parameters (e.g. the field parameters) remains unknown and cannot be established without additional runs of the code. Therefore, it is advantageous to use other, less accurate, methods, which can simplify the problem and even lead to (partly) analytical solutions. One of such situations concerns recent experiments on ionization of the helium atom by a femtosecond pulse of the free-electron laser FLASH at DESY (Hamburg) with a simultaneous action of an infrared dressing pulse [4]. The scheme of the coupling fields is shown in fig. 3. The experiment was performed with peak intensities of 7×10^{11} W/cm² for the free-electron laser and with the frequency-doubled Nd:YLF laser as a source of the coupling optical field. The ionization signal shows only the first sideband, which is two orders of magnitude weaker than the main photoline. It points to the applicability of second-order perturbation theory for the description of the sidebands. We use this approximation for the analysis of the polarization dependence of the sidebands, i.e. the modulation of the sideband ionization signal with changing the angle between the directions of the linear polarization of the two fields. Deviation

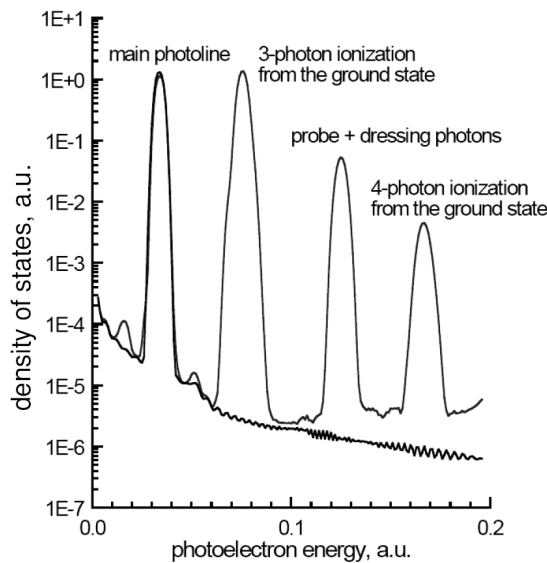


Fig. 2. Examples of the photoelectron spectra of Li with and without the dressing field (see text). The density of states integrated over the whole spectrum, including its discrete part, gives unity.

These our first calculations for lithium, together with the preceding results for hydrogen [3], point to a general character of the phenomenon of controlling the PAD in the

from the axial symmetry, when the fields are not parallel, makes the direct numerical solution of the TDSE very difficult.

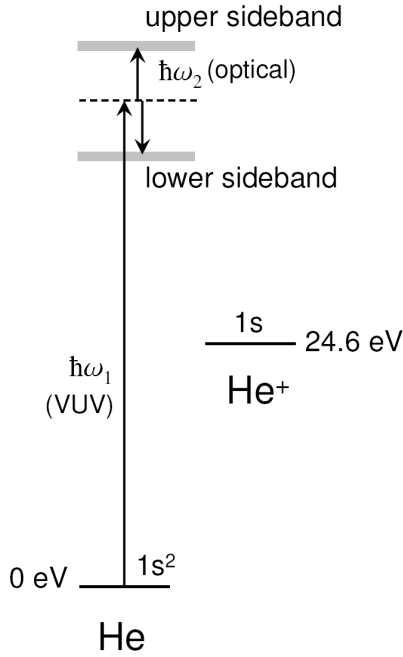


Fig. 2. Examples of the photoelectron spectra of Li with and without the dressing field (see text). The density of states integrated over the whole spectrum, including its discrete part, gives unity.

In second-order perturbation theory, the ionization probability is given by

$$\frac{dW}{dE} = \frac{W_E}{4\pi} \sum_{\ell LM} \left| \langle E(L_f \ell) LM | U^{(2)} | 0 \rangle \right|^2, \quad (3)$$

where L_f is the orbital angular momentum of the residual ion, the summation is taken over the partial waves of the photoelectron (ℓ) and the total orbital momentum of the system ion + photoelectron and its projection (LM). We assume Gaussian envelopes for the electromagnetic pulses. After identical transformations within the rotating-wave approximation and summing over magnetic quantum numbers, the result can be cast into the form

$$\frac{dW}{dE} = \frac{1}{6} \left[3(S_1 + S_2) + (2S_0 - 3S_1 + S_2) \cos^2 \theta \right] \quad (4)$$

Equation (4) provides the explicit dependence of the sideband intensity on the angle between the polarizations. When an electron is ionized from the initial atomic S state, like in helium, the coefficient S_1 vanishes. For the helium case

$$S_\ell = \frac{R^2}{3(2\ell+1)} \exp \left[-2\xi^2 \left(\frac{t_p^2}{a^2} + \frac{b^2 D^2}{4} \right) \right] \times \left| \sum \int dE' \omega(z) D_\ell(E, E') D(E') \right|^2. \quad (5)$$

Here a and b are the time widths of the optical and the VUV pulses (FWHM = $2a\sqrt{\ln 2}$ and $2b\sqrt{\ln 2}$), respectively; $\xi^2 = a^2/(a^2+b^2)$; t_p is the time delay of the VUV pulse with respect to the optical one; $R = \pi ab E_1 E_2 / 8$ with E_1 and E_2 being the Gaussian envelope field amplitudes of the VUV and the optical pulse, respectively; $D = (E - E_0) - (\omega_1 \mp \omega_2)$ is the two-photon detuning, where E_0 is the energy of the ground state and ω_1 (ω_2) is the frequency of the VUV (optical) photon; summation/integration is taken over the intermediate discrete and continuum states;

$\omega(z) = \exp(-z^2) \operatorname{erfc}(-iz)$ is the Faddeeva function of the complex argument

$$z = \frac{a}{2\xi} (\xi^2 D - \delta) + i \frac{\xi t_p}{a}, \quad \text{where } \delta = E' - E_0 - \omega_1.$$

The reduced dipole matrix elements are defined as $D(E') = \langle E' p || p || 0 \rangle$ and

$$D_\ell(E, E') = \langle E \ell || p || E' p \rangle, \quad \text{where } E \ell \text{ denotes electron in continuum with the energy } E \text{ and the orbital angular momentum } \ell \text{ } (\ell = 1 \text{ for } Ep), \text{ and}$$

$$\langle E \ell || p || E' \ell' \rangle = -iB \int_0^\infty P_{E\ell}(r) Q P_{E'\ell'}(r) dr \quad \text{with}$$

the operator $Q = \frac{d}{dr} + \frac{C}{2r}$ and $P_{E\ell}(r)$ being the radial wave function of the electron. The

parameters B and C take the values $B = \sqrt{\ell'+1}$, $C = -2\ell'$ for $\ell = \ell'+1$ and $B = -\sqrt{\ell'}$, $C = 2\ell'+2$ for $\ell = \ell'-1$. Calculating the dipole matrix elements between

the continuum states is a non-trivial task. For this purpose we use a recipe developed in [6] and present the corresponding integral in the form

$$I(E\ell, E'\ell') \equiv \int_0^\infty P_{E\ell}(r) Q P_{E'\ell'}(r) dr = F_1(E'\ell', E\ell) \delta(E'-E) + \wp \frac{F_2(E'\ell', E\ell)}{E'-E} \quad (6)$$

with easily calculated smooth functions $F_1(E'\ell', E\ell)$ and $F_2(E'\ell', E\ell)$ (see [6] for details).

With additional assumptions, a further simplification of the dynamical factors S_ℓ is possible. Our two principal assumptions are:

(a) The optical pulse is much longer than the VUV pulse.

(b) Ionization proceeds far from the threshold, where the continuum is smooth and the matrix elements do not change much over the interval that gives the dominating contribution to the amplitude.

The properties and the asymptotic form of the $\omega(z)$ function are used in further transformations. Keeping the leading terms, the final result can be cast into the form (for the upper sideband)

$$S_\ell = \left(\frac{E_1 E_2}{4} \right)^2 \frac{\pi b^2 \xi^2}{3(2\ell+1)} \exp \left[-2\xi^2 \left(\frac{t_p^2}{a^2} + \frac{b^2 D^2}{4} \right) \right] \times \left| F_1(Ep, E\ell) I(E\ell, 1s) - i\pi F_2(E-p, E\ell) I(E-p, 1s) + \sum_k \frac{E_0 + \omega_1 + \xi^2 D - E}{E_0 + \omega_1 + \xi^2 D + E_k} I(E\ell, E_k p) I(E_k p) \right|^2, \quad (7)$$

where $E_- = E - \omega_2$. The first term under the absolute square in eq. (7) corresponds to the pole contribution, while the second and third terms correspond to the contributions from the continuum and from discrete excitations, respectively. In this first approximation, the intensity of the sideband as function of the time delay is Gaussian. The width of this Gaussian is independent of the transition amplitudes. Since the S_ℓ are positively definite, the intensity of the sideband as function of the angle, eq. (4), exhibits a maximum for parallel polarizations and a minimum for perpendicular polarizations. The depth of the signal modulation does not depend on the time delay t_p , but it generally depends on the two-photon detuning D and the pulse-duration parameter ξ .

Numerical calculations were performed for helium with the atomic potential of the form $rV(r) = -1 - (1 + 1.3313r) \exp(-3.0634r)$, which provides a good description of the excited helium states and the continuum. We took the time duration of the pulses as $a = 720$ fs, $b = 30$ fs and the frequencies $\hbar\omega_1 = 48.6$ eV (25.5 nm), $\hbar\omega_2 = 2.37$ eV (523 nm), which correspond to the first test measurements at the free-electron laser FLASH. Nine discrete intermediate p states of He were accounted for in the sum over k in eq. (7). The contribution from the discrete states turned out to be much smaller than from the other two terms in (7). In agreement with the present theory, the experimental data show a maximum of the ionization signal at parallel polarizations and a minimum at perpendicular polarizations. The relative

contribution of the outgoing s-wave of the photoelectron is larger than of the d-wave. The experimental ratio $S_0 : S_2 \approx 2.5$ and its calculated value at the detuning $D = 0$, $S_0 : S_2 \approx 2.93$, are in a good agreement.

In conclusion, we considered two non-stationary pump-probe photoionization processes in lithium and helium atoms, accessible with modern experimental facilities, and applied two different theoretical approaches: a direct numerical solution of the TDSE and second-order perturbation theory. In both cases one of the radiation pulses is due to the

VUV ionizing field, while the optical laser dressing field couples the ionization continuum to other channels. For lithium we showed that the angular distribution of the photoelectrons can be controlled by manipulating the time parameters of the femto-second pulses. Using the example of helium, where first test experimental results are available from the FLASH facility, we showed that second-order time-dependent perturbation theory is a practical method to consider the polarization dependence of the first ionization sidebands.

References

1. P.L.Knight, M.A.Lauder, B.J.Dalton, Phys. Rep. 190, 1 (1990).
2. N.V.Vitanov, T.Halfmann, B.W.Shore, K.Bergmann, Annu. Rev. Phys. Chem. 52, 763 (2001).
3. A.N.Grüm-Grzhimailo, A.D.Kondorskiy, K.Bartschat, J. Phys. B 39, 4659 (2006).
4. M.Meyer, D.Cubaynes, P.O'Keeffe et al, Phys. Rev. A 74, 011401 (2006).
5. R.D.Hudson, V.L.Carter, Phys. Rev. 137, A1648 (1965); J. Opt. Soc. Am. 57, 651 (1967).
6. Th.Mercouris, Y.Komninos, S.Dionissopoulou, C.A.Nicolaides, Phys. Rev. A 50, 4109 (1994); J. Phys. B 29, L13 (1996).

ДВОКОЛІРНА ІОНІЗАЦІЯ АТОМІВ ФЕМТОСЕКУНДНИМИ ІМПУЛЬСАМИ

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Розглянуто процеси накачки і зондування у фемтосекундному інтервалі з використанням зондуєчого ВУФ і взаємодієчого оптичного імпульсів: іонізація атома Li атом з континуумом, зв'язаним зі збудженням 3d-станом і іонізація атома He зі зв'язаними плоскими континуумами. У першому випадку розраховано спектри і кутові розподіли фотоелектронів і проаналізовано їх для двох лінійно поляризованих колінеарних полів за допомогою чисельного розв'язку нестационарного рівняння Шредінгера. Спектр і кутові розподіли контролюються часовою затримкою між імпульсами і дво-фотонним розстроюванням. У другому випадку увагу сконцентровано на модуляції сигналу іонізації He як функцією кута між векторами поля поляризації. У теорії залежних від часу збурень другого порядку для гауссівських згорток поля отримано і використано частково аналітичний розв'язок.