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Time-dependent Feshbach method to study resonant photoionization of He with ultrashort laser pulses.

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Abstract. A time-dependent Feshbach formalism is proposed to study the resonant photoionization of the helium atom using ultrashort laser pulses. This spectral method consist in solving the time-dependent Schrödinger equation by expanding the time-dependent wavepacket in terms of eigenfunctions defined in two orthogonal halfspaces: a bound-like resonant \mathcal{Q} and a non-resonant scattering-like \mathcal{P} . The latter eigenfunctions for the projected Hamiltonians $\mathcal{Q}H\mathcal{Q}$ and $\mathcal{P}H\mathcal{P}$ are not indeed eigenfunctions of the total Hamiltonian, so that the electrostatic coupling $\mathcal{Q}H\mathcal{P}$ acts as a *leaking* operator $\mathcal{Q}\rightarrow\mathcal{P}$ responsible for the temporal decay of resonances into the underlying continuum, keeping the physical insight of the Fano-Feshbach time independent formalisms. This method allows not only for accurate descriptions of the resonance parameters (energies, widths and Fano shape parameters) but also for the temporal evolution of the photodynamics involved in the resonant photoionization when using short laser pulses. We illustrate the performance of the method by analyzing the temporal formation of i) the one-photon ionization cross section below the second ionization threshold and the buildup of Fano profiles and ii) the *up-down* asymmetry of photoelectron angular distributions resulting from interferences of *S*-, *P*- and *D*-waves after simultaneous photoexcitation and decay of the lowest $^1S^e$, $^1P^o$ and $^1D^e$ resonant states, by using two sequential laser pulses with XUV harmonic frequencies separated by a time delay.

1. Introduction

In this contribution we describe a time-dependent Feshbach formalism as applied to resonant photoionization processes in helium subject to ultrashort laser pulses. The study of the resonant phenomena in atoms goes back to the pioneering theoretical work of U. Fano in 1935 [1] while under the tutorship of E. Fermi at the Institute of Physics of Rome University in Via Panisperna. Also, in 1935 Beutler reported an ultraviolet absorption spectrum of helium [2] in which a series of two-electron excitations were identified, showing anisotropic lineshapes in the absorption profile. The theoretical analysis in the pioneering paper by U. Fano in 1935 (see also [3]) was based on the interaction of a discrete state $|\varphi\rangle$ with a *flat structureless* discretized continuum described by states $|k\rangle$, all of them eigenfunctions of an unperturbed Hamiltonian H_0 , i.e., $\langle\varphi|H_0|\varphi\rangle=E_\varphi$ and $\langle k|H_0|k\rangle=E_k=k\delta$, where k is an integer number and δ is the constant energy spacing for the discretized continuum with a density of states $1/\delta$. A new Hamiltonian $H=H_0+V$ (more truly connected with the atomic realm) is able to couple the discrete state $|\varphi\rangle$ with the continuum $|k\rangle$ through the matrix element $v=\langle\varphi|V|k\rangle$.

The eigenfunctions $|\psi_\mu\rangle=(|\varphi\rangle\langle\varphi|+\sum_k|k\rangle\langle k|)|\psi_\mu\rangle$ of the new Hamiltonian H , i.e., $H|\psi_\mu\rangle=E_\mu|\psi_\mu\rangle$ [with energies given by the transcendental equation $\sum_k v^2/(E_\mu-E_k)=E_\mu$] contain



components of the discrete state $\langle\varphi|\psi_\mu\rangle = [1 + \sum_k v^2/(E_\mu - E_k)^2]^{-1/2}$ as well as of each state in the discretized pseudocontinuum $\langle k|\psi_\mu\rangle = v\langle\varphi|\psi_\mu\rangle/(E_\mu - E_k)$, so that the information of the discrete state $|\varphi\rangle$ is now diluted within the set of new eigenstates $\{|\psi_\mu\rangle\}$. More precisely, the presence of the discrete state $|\varphi\rangle$ strongly modifies the new quasicontinuum eigenspectrum $\{E_\mu, |\psi_\mu\rangle\}$ with respect to the original *flat* continuum $\{E_k, |k\rangle\}$ specially in the energy region around the energy position of the discrete state, $E=E_\varphi \pm \hbar\Gamma/2$, where Γ is the resonance width given by the Fermi's golden rule $\Gamma=\frac{2\pi}{\hbar}|\langle\varphi|V|k\rangle|^2\frac{1}{\delta}$. This simple model by Fano contains much of the relevant physics of the resonant phenomena in atoms. Firstly, time evolution of a state initially prepared to be $|\varphi\rangle$ will decay exponentially, as shown by a simple time propagation $|\Psi(t)\rangle = \exp(-iHt/\hbar)|\varphi\rangle = \sum_\mu |\psi_\mu\rangle \exp(-iE_\mu t/\hbar)\langle\psi_\mu|\varphi\rangle$ using the complete set of eigenstates $\{|\psi_\mu\rangle\}$. By projecting $|\Psi(t)\rangle$ with $\langle\varphi|$ (autocorrelation function) and after a simple integration by residues one arrives to the exponential decay law for the discrete state $P(t) = |\langle\varphi|\Psi(t)\rangle|^2 = \exp(-\Gamma t)$. Incidentally this simple model also allows to roughly explain the presence of asymmetric profiles in the photoabsorption spectrum [with the well-known Fano parametrization $\sigma \sim (q+\epsilon)^2/(1+\epsilon^2)$] by adding a second discrete state $|\chi\rangle$ to the previous model, coupled with a new interaction W to both the discrete state $|\varphi\rangle$ and the continuum $|k\rangle$ [1, 3]. Fano extended this model in 1961 after new experimental evidence of asymmetric peaks in the He spectrum, in one of the most cited papers in physics [4], then generalizing his 1935 result for a realistic electronic continuum, and explained that autoionization is due to configuration interaction between the discrete and continuum spectrum and the asymmetric peaks in the excitation spectra are to be attributed to the interference between the direct ionization to the continuum and the autoionization of doubly excited states. An extension of the Fano model (1961) to the time domain, motivated to better understand the ultrafast autoionization dynamics involved with short laser pulses, is discussed for instance in [5] and [6].

H. Feshbach also developed a unified theory for multichannel resonance phenomena in nuclear reactions [7], involving \mathcal{Q} and \mathcal{P} projection operators for the closed and open channels, which was later applied in atomic physics [8], where explicit forms for the projection operators were proposed (see also [9]). Accordingly, it is common to use the name of Fano-Feshbach formalism for resonances. All these developments were performed in the energy domain, mostly to explain spectra recorded with continuous radiation. The advent in the last decade of high frequency ultrashort laser pulses down to the attosecond duration [10] has driven theoreticians to develop approaches in the time domain to deal with new emergent nonlinear phenomena in these atomic time scales, which ultimately require the solution of the time dependent Schrödinger equation. Among fast evolving phenomena in atoms, the temporal formation and irreversible decay of transient metastable states play a fundamental role. For instance, the electron dynamics in helium autoionization has recently been monitored and controlled experimentally using isolated attosecond pulses [11], and showing control over the two interfering paths: direct photoionization and delayed autoionization, which provides the feasibility of observing the time resolved buildup of a Fano resonance in the time domain [12].

We present some details of a recently published work [13], where a general non-perturbative time-dependent Feshbach method is proposed (see also [14] for applications in molecules) to study the resonant photoionization dynamics in two-electron atoms subject to short laser pulses.

2. Theoretical description and computational details

While under the influence of short intense laser fields, we are bound to directly solve the time-dependent Schrödinger equation (TDSE) for He, $(H + V_L(t) - i\hbar\partial/\partial t)\Psi(\mathbf{x}_1, \mathbf{x}_2, t)=0$, which contains the laser-atom interaction given by $V_L(t)=(\mathbf{p}_1 + \mathbf{p}_2) \cdot \mathbf{A}(t)$ in the velocity gauge form of the dipolar approximation (equivalence with results in the length gauge yields a consistency proof for completeness). The vector potential for a linearly z -polarized laser pulse with duration T and central frequency ω reads $\mathbf{A}(t)=\hat{\mathbf{e}}_z f(t) \cos[\omega(t - T/2)]$, where $f(t)$ is the pulse envelope,

chosen here to be $f(t)=\sin^2(\pi t/T)$ for the sake of computational simplicity and defined in the time interval $t \in [0, T]$ and zero elsewhere. The vector potential amplitude A_0 is related to the laser intensity I through $A_0=\sqrt{I[W/cm^2]/3.5095 \cdot 10^{16}\omega^2}$. Time-dependent non-perturbative computations for processes involving the electronic continuum for two-electron atoms and strong fields were firstly addressed by Zhang and Lambropoulos [15, 16] with \mathcal{L}^2 discretized methods based on expansions in terms of B-splines. Following their procedure, it is usual to expand the time-dependent wavefunction on the basis set of two-electron eigenstates of the total Hamiltonian H , $\Psi(\mathbf{r}_1, \mathbf{r}_2, t)=\sum_{L,n} C_{L,n(E)}(t)\Phi_{n(E)}^{LS}(\mathbf{r}_1, \mathbf{r}_2) \exp[-iE_{L,n(E)}t/\hbar]$, which is equivalent to use the $|\psi_\mu\rangle$ eigenstates in the model by Fano described in the introduction, i.e., they already have the effect of resonant states but diluted among the H eigenstates. Although the photoelectron energy spectrum can be retrieved after the laser pulse of duration T through the expansion coefficients, $dP/dE(E_e, t \rightarrow \infty)=\sum_{L,n(E=E_e)} |C_{L,n(E=E_e)}(t)|^2 \rho(E = E_e)$, if the laser pulse has a duration shorter than the lifetime of the Fano resonant state, the information on the autoionization decay dynamics is not given straightforwardly at any time t by the probabilities $|C_{L,n(E=E_e)}(t)|^2$ since these are expansion coefficients for stationary eigenstates of the field-unperturbed Hamiltonian H , i.e., these expansion coefficients for the pseudocontinuum eigenstates carry unresolved information of the autoionization decay of the discrete resonant state into the continuum and the time resolution of the autoionization process is *hidden* within the dynamics. Instead it is better to expand the time dependent wavepacket in the Fano configuration basis of discrete $|\varphi\rangle$ and *flat* pseudocontinuum $|k\rangle$ states, which are not eigenstates of the total Hamiltonian H but of an uncoupled Hamiltonian H_0 .

Consequently, we make use of an spectral method by expanding the time-dependent wavefunction with the stationary Feshbach eigenstates $|\mathcal{Q}\Phi_r\rangle$ (resonant) and $|\mathcal{P}\Psi_E^0\rangle$ (non resonant continuum) of the $\mathcal{Q}H\mathcal{Q}$ and $\mathcal{P}H\mathcal{P}$ projected Hamiltonians, respectively, which play a similar role to those of the discrete $|\varphi\rangle$ and the *flat* pseudocontinuum $|k\rangle$ states in the Fano model. In fact, since $\mathcal{Q} + \mathcal{P}=1$ (completeness), $\mathcal{P}^2=\mathcal{P}$ and $\mathcal{Q}^2=\mathcal{Q}$ (idempotency), and $\mathcal{Q}\mathcal{P}=0$ (orthogonality), the total Hamiltonian H and its eigenstates $|\Psi\rangle$ can be written *a la Feshbach* in the form $H=H_0+V$ and $|\Psi\rangle=|\mathcal{Q}\Phi_r\rangle+|\mathcal{P}\Psi_E^0\rangle$, respectively, where $H_0=\mathcal{Q}H\mathcal{Q}+\mathcal{P}H\mathcal{P}$ and $V=\mathcal{Q}H\mathcal{P}+\mathcal{P}H\mathcal{Q}$, and both $|\mathcal{Q}\Phi_r\rangle$ and $|\mathcal{P}\Psi_E^0\rangle$ are eigenfunctions of H_0 through the following equations

$$(\mathcal{Q}H\mathcal{Q} - \mathcal{E}_r)|\mathcal{Q}\Phi_r\rangle = 0; \quad (\mathcal{P}H\mathcal{P} - E)|\mathcal{P}\Psi_E^0\rangle = 0. \quad (1)$$

H_0 eigenstates are coupled by the V term, which in the Feshbach formalism corresponds, for instance, to the *leaking* operator $\mathcal{Q}H\mathcal{P}$, responsible for the resonance decay from the bound-like resonant part $|\mathcal{Q}\Phi_r\rangle$ to the nonresonant scattering-like part $|\mathcal{P}\Psi_E^0\rangle$ of the total wavefunction. Because of the spatially localized ($|\mathcal{Q}\Psi\rangle \rightarrow 0$) and temporal transient nature of resonant states ($\mathcal{Q}H\mathcal{P}$ couplings are not effective for $t \rightarrow \infty$ since the whole resonant population in \mathcal{Q} halfspace eventually vanishes), then $\lim_{r,t \rightarrow \infty} H=H_0$ and $\lim_{r,t \rightarrow \infty} |\Psi\rangle=|\mathcal{P}\Psi_E^0\rangle$. For practical computations, our expansion reads $|\Psi(t)\rangle=\sum_b C_b(t)|\Xi_b\rangle \exp(-iE_b t/\hbar) + \sum_r C_r(t)|\mathcal{Q}\Psi_r\rangle \exp(-iE_r t/\hbar) + \int dE C_E(t)|\mathcal{P}\Psi_E^0\rangle \exp(-iEt/\hbar)$, where $\{|\Xi_b\rangle, E_b\}_{b=1}^{N_b}$ corresponds to the set of N_b bound states, $\{|\mathcal{Q}\Psi_r, E_r\}_{r=1}^{N_r}$ to the set of N_r resonant states, with $E_r=\mathcal{E}_r+\Delta_r$, and Δ_r is the perturbative second-order energy correction (energy shift) to the resonance energy \mathcal{E}_r due to the surrounding continuum states coupled by the V term, and $\{|\mathcal{P}\Psi_E^0, E\}_E$ refers to the *flat* non-resonant continuum states. Now, by inserting this ansatz into the TDSE one arrives to a large set of coupled differential equations which can be expressed in packed block form, with $\{n, m\}=\{b, r, E\}$, as follows

$$i\hbar \begin{pmatrix} \dot{\mathbf{C}}_b \\ \dot{\mathbf{C}}_r \\ \dot{\mathbf{C}}_E \end{pmatrix} = e^{iE_n t/\hbar} \begin{pmatrix} 0 & V_L(t)_{b,r} & V_L(t)_{b,E} \\ V_L(t)_{r,b} & 0 & \mathcal{Q}H\mathcal{P}_{r,E} \\ V_L(t)_{E',g} & \mathcal{P}H\mathcal{Q}_{E',r} & V_L(t)_{E',E} \end{pmatrix} e^{-iE_m t/\hbar} \begin{pmatrix} \mathbf{C}_b \\ \mathbf{C}_r \\ \mathbf{C}_E \end{pmatrix}, \quad (2)$$

where $V_L(t)_{n,m}$ are the dipolar coupling matrix elements and QHP are the V electrostatic couplings in the Fano model. These QHP couplings are active during and after the laser pulse until the full depletion of the $|\mathcal{Q}\Psi\rangle$ resonant halfspace. Eigenproblems associated to the asymptotic basis expansion $H_0=QH\mathcal{Q}+\mathcal{P}H\mathcal{P}$ are solved using a configuration interaction (CI) method based on expansions in terms of antisymmetrized products of atomic orbitals, the latter expanded in terms of 200 B-splines with polynomial order $k=7$ enclosed within a box of size $L=150$ a.u., and using an exponential-linear sequence for the knot-points to guarantee a reasonable good description of both the inner radial localized part of resonances and the outer radial part of the continuum wavefunctions.

The $QH\mathcal{Q}$ stationary eigenproblem is solved for the lowest 19 $1S^e$, 26 $1P^o$ and 25 $1D^e$ resonance states located below the $\text{He}^+(N=2)$ ionization threshold using around 10000 two-electron configurations which, for instance, produce the following resonant parameters (E_r, Γ_r) for the lowest and fastest decaying resonances: (-0.777533 a.u., 0.005051 a.u.) for the $1S^e$ $2(1,0)_2^+$ resonance with a lifetime $\tau=4.79$ fs; (-0.692642 a.u., 0.001392 a.u.) for the $1P^o$ $2(0,1)_2^+$ resonance with $\tau=17.4$ fs; and (-0.701512 a.u., 0.002528 a.u.) for the $1D^e$ $2(1,0)_2^+$ resonance with a lifetime 9.56 fs. To keep simplicity in the application of this method we firstly choose to deal with resonant photoionization to final states of energy E in the case of one open continuum channel, $\text{He}^+(1s)+e^-(\varepsilon\ell)$, with $\varepsilon=E-E_{\text{He}^+(1s)}$. Accordingly the *flat* non-resonant continuum states are also computed with a CI method, in terms of antisymmetrized configurations $\langle \mathbf{x} | \mathcal{P}\Psi_{E_i}^0 \rangle = \sum_{k=1}^{N_P} C_k^i \mathcal{A} \{ \phi_{1s}(\mathbf{x}_1) \cdot \phi_{k,\ell=L}(\mathbf{x}_2) \}$ where one of the electrons is fixed to the $1s$ orbital and for the other its index k runs up to $N_P=170$ orbitals built with B-splines basis for each angular momentum ℓ (*static exchange approximation*), to obtain at least 75 continuum states lying between the first $\text{He}^+(N=1)$ and the second $\text{He}^+(N=2)$ ionization thresholds. With this set, we interpolate 2000 points for both dipolar and QHP couplings to improve the density of states and to *smooth* the time propagation. Thus the solution of the coupled differential equations in Eq. (2) involves tens of bound b and resonant r states, but thousands of discretized continuum states E_i , properly normalized to the Dirac delta. Nevertheless, all results shown here can be easily calculated in desktop computers.

3. Illustrative results

(i) *Time resolved Fano profiles and one-photon ionization cross section.*

The time dependent formation of the Fano profiles can be analyzed from the photoelectron spectrum, i.e., photoionization probabilities differential in energy computed from the continuum expansion coefficients, $dP(E,t)/dE=|C_E(t)|^2\rho(E)$, where $\rho(E)$ corresponds to the density of states of our discretized pseudocontinuum. We have solved the set of coupled equations (2) with the initial condition set for the ground state of He and using a laser pulse with central frequency $\omega=2.211$ a.u., intensity $I=10^{10}$ W/cm² and duration $T=20$ fs. The photoelectron spectra for continuum energies close to the lowest $1P^o$ resonance is plotted in figure 1. The results from our time-dependent Feshbach method eventually converge to the asymptotic *energy domain* asymmetric profile (for a Fano shape parameter $q=-2.8$) for times much longer than the pulse duration, and show transient oscillations due to interferences associated to the still active resonance decay (the resonance lifetime here is $\tau\sim 18$ fs). Incidentally, it is shown in [13] that within this method the resonance population closely follows an exponential decay law after the pulse, i.e., $P_r(t)=P_r(T)e^{-\Gamma(t-T)}$ with $\Gamma=0.001392$ a.u., and for a propagation time $t=50$ fs in figure 1, the population of the lowest $1P^o$ resonance is not yet fully depleted.

The one-photon ionization cross section can also be obtained from the total ionization probability $P_I(t)$ extracted from our solution of the TDSE, by normalizing with the photon flux of the laser pulse, then using the expression $\sigma[\text{cm}^2]=\omega[\text{Joules}]P_I(t)/I[\text{W}/\text{cm}^2]C[s]$, where $C[s] = \int_0^T dt [f(t)]^2 = 3T[s]/8$. Figure 2 shows the temporal formation of the one-

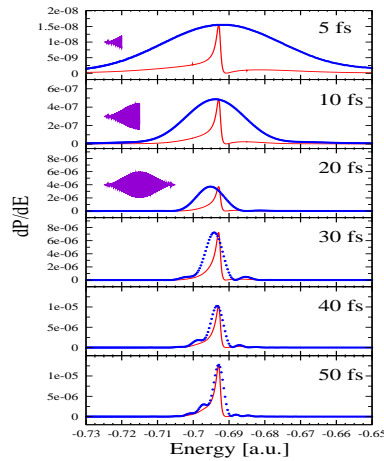


Figure 1. Time resolved buildup of the Fano profile in the photoelectron energy spectrum in the neighbourhood of the lowest He $1P^o$ resonance, obtained with a laser pulse (inset) of duration $T=20$ fs, frequency $\omega=2.210$ a.u. and intensity $I=10^{10}$ W/cm 2 . Blue dots: time-dependent Feshbach formalism; red solid line: asymptotic stationary result according to Fano theory [4].

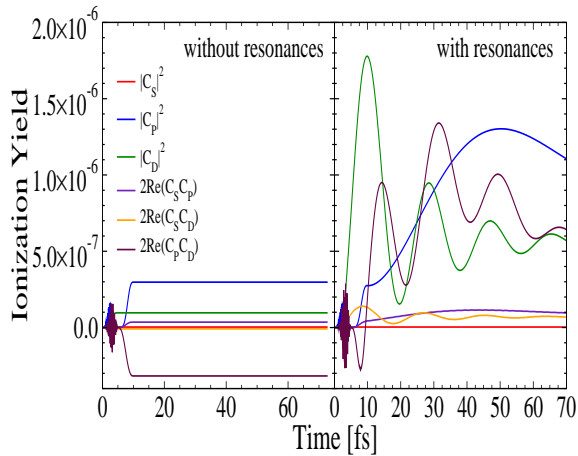


Figure 3. Time evolution of the photoelectron spectrum for the continuum components with fixed energy E (degenerated with that of the lowest $1P^o$ resonance), using two sequential laser pulses with durations $T_1=T_2=5$ fs, frequencies $\omega_1=\omega_2/2=1.105$ a.u. and intensities $I_1=10^3 \times I_2=10^{13}$ W/cm 2 separated by a time delay $\tau=5$ fs. S - and D -partial waves excited by the first laser pulse interfere with the P -partial wave excited by the second pulse.

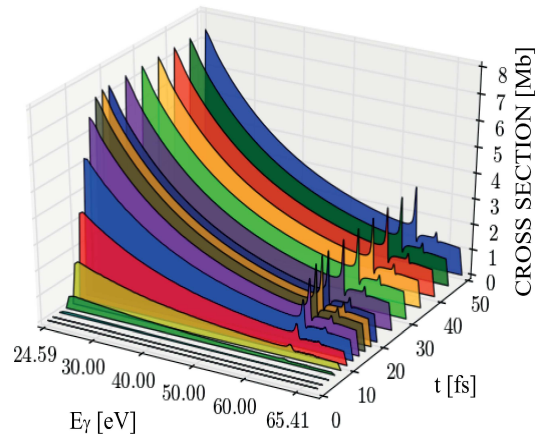


Figure 2. Temporal formation of the one-photon ionization cross section (in Mbarn) from the ground state to the $1P^o$ continuum of He, between the first He $^+$ ($N=1$) and the second He $^+$ ($N=2$) ionization thresholds, using the same laser pulse described in figure 1. The buildup of Fano profiles for the two fastest decaying resonances, $2(0,1)_2^+$ ($\tau \sim 18$ fs) and $2(1,0)_3^-$ ($\tau \sim 82$ fs) is a clearly visible feature.

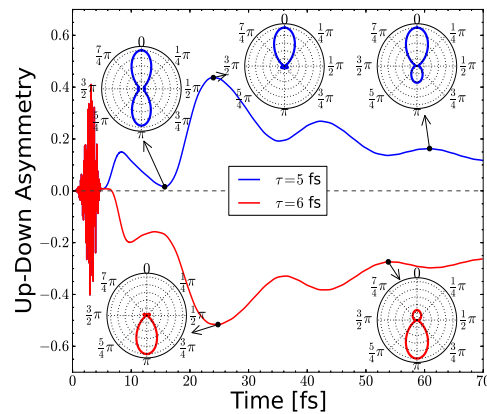


Figure 4. Time dependence for the *up-down* asymmetry of the photoelectron angular distributions produced by the two sequential pulses with the harmonic frequencies quoted in caption of figure 3, for two different time delays ($\tau=5$ fs and 6 fs) between the two laser pulses. The interfering term contributed by P - and D -waves [the line $2\text{Re}(C_S C_P)$ in figure 3] dominates for the abrupt change in the angular distribution asymmetry when the time delay τ is modified.

photon ionization cross section between the first and the second ionization thresholds for the same laser parameters described previously. Since only the first and the third lowest $^1P^o$ resonances in He have lifetimes under 100 fs, two noticeable Fano peaks associated to these states develop in the spectrum. In this work we have not improved our method in order to extract asymptotic amplitudes for the continuum $C_E(t \rightarrow \infty)$ from our wavepacket (which is propagated only up to 80 fs). Methods to extract spectra from the correlated wavepackets in He have been summarized and discussed recently [17], and some related contributions are also presented in this conference.

(ii) *Asymmetry in the photoelectron angular distributions.*

To play around with our Feshbach method we analyze the manipulation of the asymmetry in the photoelectron angular distributions by using two sequential laser pulses with XUV harmonic frequencies. The first pulse with ω_1 is able to ionize the He atom by two-photon absorption close to the resonant region of the lowest $^1S^e$ and $^1D^e$ doubly excited states, which generates partial S - and D - waves both by direct photoionization and by delayed autoionization. After a controlled time delay τ between the two laser pulses, defined as the time interval between the centers of the two pulses, the second pulse with lower intensity and $\omega_2=2\omega_1$ populates resonant and continuum $^1P^o$ states by one-photon absorption. Figure 3 shows the crucial importance of the resonance decay in the photoelectron spectrum and the complex interference of S - P - D continuum waves [here $\frac{dP}{dE}(E) \sim |C_E^S(t) + C_E^P(t) + C_E^D(t)|^2 \rho(E)$]. More specifically, the P - D interference is the dominant contribution (see figure 3) and it changes dramatically as a function of the time delay τ between the two harmonic pulses. Consequently, the preferred direction for the escaping photoelectron (*up or down* in figure 4) could be selected by appropriately choosing the time delay τ between the two consecutive pulses.

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