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Theory of laser-assisted autoionization by attosecond light pulses

Z. X. Zhao[1,*] and C. D. Lin[1,2]
[1]*J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, Kansas 66506-2604, USA*
[2]*Physics Division, National Center for Theoretical Sciences, P. O. Box 2-131, Hsinchu, 30013, Taiwan*
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We present a quantum theory of the decay of an autoionizing state created in the attosecond xuv (extreme ultraviolet) pump and laser probe measurements within the strong field approximation employing resonance parameters from Fano's theory. From the electron spectra versus the pump-probe time delay, we show how the lifetimes of the resonances can be extracted directly from the time domain measurements.

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Many-electron atomic and molecular systems with excess internal energy relax to states of lower energy by rearranging their electronic structure. With the recently developed attosecond xuv (extreme ultraviolet) pulses [1,2], it has been demonstrated that it is possible to obtain time-resolved images of an Auger decay [3], akin to the time-resolved spectroscopy for tracking atomic motion in molecules with femtosecond laser pulses [4]. Limited by the pulse intensity, currently experimentalists employ an xuv pump-ir (infrared) laser probe technique where electrons generated by the xuv pulses are "steered" in the few-cycle ir laser field at varying pump-probe time delays [5,6]. Although the basic principle of such measurements can be qualitatively understood in terms of classical mechanics, quantitatively the electron spectra have to be calculated quantum mechanically. For simple laser-assisted photoionization processes, such a quantum theory is readily available by solving the time-dependent Schrödinger equation directly within the one-electron model. However, such calculations are quite tedious and a simpler approximate theory based on the strong field approximation (SFA) [7] has been found [8,9] to be more useful for the analyses of experiments.

In a time-resolved Auger measurement [3], the Auger electron spectrum in the laser field was analyzed based on an *ad hoc* model that combined a rate equation description of core-hole formation and subsequent quantum theory of Auger emission in the laser field. The theory has been improved recently by Smirnova *et al.* [10], and by Wickenhauser and Burgdörfer [11], where they treated the hole formation and the decay process coherently, albeit in a less transparent form to be useful for the experimentalists.

In this paper we generalized the strong field approximation to calculate the time-resolved electron spectra of an autoionizing state created in a typical xuv pump-ir laser probe (xuv+ir) experiment. The validity of the SFA thus developed is checked by solving the time-dependent Schrödinger equation using a model Hamiltonian that simulates an isolated autoionizing state populated in such xuv+ir experiments. From the simpler SFA theory we then illustrate how to extract the lifetime of an isolated resonance or two nearby resonances from the time-domain measurements. We note that in typical xuv+ir experiments, the attosecond xuv pulse is weak such that the initial autoionizing state is created perturbatively. Thus resonance parameters for characterizing this step are already available from measurements or from many-body calculations in the energy domain. Our starting point is thus to cast the photoexcitation amplitude in the energy domain into the time domain and then propagate it in the streaking ir laser field.

An isolated resonance in the energy domain is described by the Fano profile [12], $(q+\epsilon)^2/(1+\epsilon^2)$, where $\epsilon=(E-E_r)/(\Gamma/2)$ is the reduced energy measured from the resonance energy E_r in units of half width $\Gamma/2$. The parameter q measures the relative strength of the formation of the "bound" state and the direct continuum. If the interference between the bound and continuum states is small and is neglected, we call it a Lorentz resonance. In the energy domain such a resonance is described by the amplitude $1/[i(E-E_r)-\Gamma/2]$. In the time domain, after the Fourier transform, it becomes a decaying amplitude $F(t)=\exp[-iE_rt-(\Gamma/2)t]$. Under the strong field approximation the probability amplitude of finding a continuum electron with momentum $\vec{p}$ at time t after the laser pulse is over is given by

$$b_L(\vec{p},t) = i\int_{-\infty}^{t} dt_1 \int_{t_1}^{t} dt_2 \vec{E}_x(t_1)\cdot\vec{d}(\vec{p}')e^{iI_pt_1} \times \exp\left[\left(-iE_r-\frac{\Gamma}{2}\right)(t_2-t_1)\right] \times \exp\left\{-i\int_{t_2}^{t}\frac{[\vec{p}+\vec{A}(t)-\vec{A}(t_3)]^2}{2}dt_3\right\}, \quad (1)$$

where I_p is the ionization energy and $\vec{d}$ is the dipole transition moment.

This equation describes the formation and the decay of the resonance in three steps. First, the xuv attosecond pulse creates the resonance at time t_1 with momentum $\vec{p}'$, where the formation amplitude is evaluated by the electric dipole approximation. Second, this resonance propagates from time t_1 to t_2 and decays at t_2 with an amplitude $\exp[(-iE_r-\Gamma/2)(t_2-t_1)]$. Third, at t_2, the continuum electron emerges in the laser field and propagates from t_2 to the final time t. In this model, the ir laser does not participate in the creation of the resonance and the xuv pulse does not influence the

*Email address: zxzhao@phys.ksu.edu

propagation of the continuum electron after its creation. Recall that the vector potential and the electric field vector are related by $\vec{E}(t)=-\partial\vec{A}/\partial t$. We also assume that the decay amplitude of the resonant state is not modified by the laser. The continuum electron is born at time t_2 with momentum $\vec{p}'$, which is related to the momentum $\vec{p}$ at time t when the external field vanishes by $\vec{p}'=\vec{p}+\vec{A}(t)-\vec{A}(t_2)$.

Equation (1) can be generalized to an isolated Fano resonance directly. For the Fano resonance the scattering amplitude in the energy domain is given by $f_F(E)=(q+\epsilon)/(1-i\epsilon)$. Upon transforming to the time domain, this would give the amplitude

$$F_F(t)=\frac{\Gamma}{2}(q-i)e^{-iE_rt-\Gamma t/2}+i\delta(t-0). \tag{2}$$

This expression would replace the middle term in Eq. (1) in the calculation of electron spectra for a Fano resonance. Note that the first term of Eq. (2) describes the decay of the "discrete" resonance and the second term describes the prompt photoemission, both in the time-varying electric field of the ir laser. Clearly this theory reduces to the Fano theory for an isolated resonance in the energy domain in the absence of the ir laser. It also reduces to the SFA theory for laser-assisted photoionization given by Kitzler *et al.* [8] in the absence of the discrete resonance. Although the derivation is for an autoionizing state, the theory can be applied to describe Auger electron as well if the prompt photoelectron and the Auger electron do not interact. We emphasize again that within the SFA, only the resonance parameters in the energy domain are used and there is no need to calculate electron spectrum starting from the formidable time-dependent many-electron Schrödinger equation.

The theory presented so far is based on the strong field approximation. The validity of the SFA has been confirmed for laser-assisted photoionization [8] when the photoelectron energy is large compared to the Ponderomotive energy of the ir laser field. To check if the present formulation using SFA is indeed valid, we solved the time-dependent Schrödinger equation for a many-electron atom in the xuv+ir field within the two-state approximation and with the same ansatz as in the SFA. For simplicity, consider a generic two-electron atom in an external time-dependent electric field and assume that the time-dependent wave function can be solved by the expansion $\Psi(\vec{r}_1,\vec{r}_2,t)=\Sigma_\mu F_\mu(\vec{r}_1,t)\Phi_\mu(\vec{r}_2)$, where the summation is truncated to two target states only. By projecting out the second electron we obtained the coupled equations

$$i\frac{\partial}{\partial t}\begin{pmatrix}F_1\\F_2\end{pmatrix}=\begin{bmatrix}H_1 & V_c\\V_c & H_2\end{bmatrix}\begin{pmatrix}F_1\\F_2\end{pmatrix}+\begin{bmatrix}\vec{r}\cdot\vec{E}_{l+x}(t) & \vec{r}\cdot\vec{E}_x(t)\\\vec{r}\cdot\vec{E}_x(t) & 0\end{bmatrix}\begin{pmatrix}F_1\\F_2\end{pmatrix} \tag{3}$$

for the first electron. Note we use subscripts ℓ and x to indicate electric fields from the laser and the xuv, respectively. This model two-state Hamiltonian is constructed to include the essential ingredients of exciting an autoionizing state in the xuv+ir fields. Without the upper "core" state $\Phi_2(\vec{r}_2)$, it reduces to a description of laser-assisted photoionization. The coupled equations are to be solved using partial wave expansion. For definiteness, assume initially $\Phi_1(\vec{r}_1)$ is an s state. The weak xuv pulse will lead it to p states only while the ir laser will couple it to many other angular momentum states via multiphoton processes. In the numerical calculation, following the spirit of SFA, the basis functions for the lower channel include the ground state and the continuum states only. For the upper channel, only one bound eigenstate is included in the basis expansion. This will be a bound p state that is coupled to the ground s state in the lower channel by the xuv pulse. Once this bound p state is formed, it can decay to the continuum p states of the lower channel through the V_c term, resulting in autoionization as described by Fano [12]. We have made a model atom with potentials V_1 and V_2 and their coupling V_c. The asymptotic energy separation of the two potentials was chosen to be 27.21 eV. With the chosen potentials the resonance parameters were calculated: $E_r=22.9$ eV (measured from the first threshold), $\Gamma=0.055$ eV (lifetime of 12 fs), and $q=-4.2$. These resonance parameters are used to generate the photoelectron spectra in the strong field approximation using Eqs. (1) and (2). In the meanwhile the coupled time-dependent Schrödinger equations for the model atom in the xuv+ir fields are solved numerically to obtain the photoelectron spectra. We assume the time dependence of both the xuv and the ir laser pulses are Gaussian and the few-cycle ir laser has a carrier envelope phase ϕ.

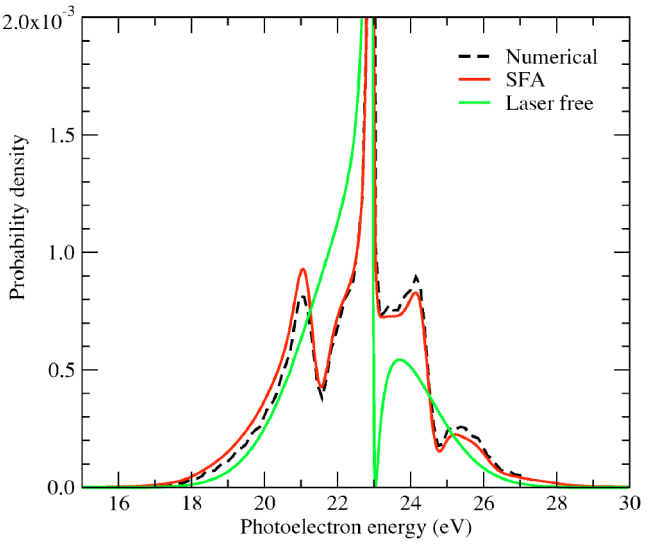

FIG. 1. (Color online) Laser-dressed total photoelectron spectrum for an autoionizing state created in xuv+ir laser fields. The spectra from the strong field approximation and from the numerical calculations are shown to agree with each other. The electron spectrum in the absence of lasers is also shown. See text for the parameters used in the calculation.

In Fig. 1 we compare the angle-integrated photoelectron spectra obtained with the following parameters: xuv pulse duration 500 as, central photon energy 39 eV, and peak intensity 10^{12} W/cm^2, while the laser has a duration of 5 fs, the central photon energy is 1.65 eV, peak intensity is 10^{12} W/cm^2, and the carrier envelope phase is $\phi=0$. The spectra are obtained with no time delay. The comparison shows that the numerical result from the model calculation

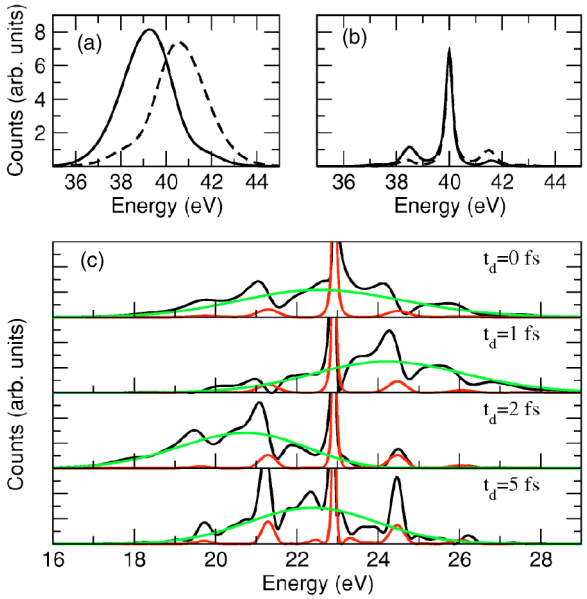

FIG. 2. (Color online) Streaked electron spectra in the forward direction for two Lorentz resonances of lifetime of (a) 0.2 fs and (b) 2 fs, excited by a 0.5-fs xuv pulse. The xuv-laser delay time is zero and two carrier envelope phases are shown: 0 (solid) and π (dashed). For other parameters used, see text. (c) Streaked electron spectra (black) for a Fano resonance at different time delays. Also shown are the electron spectra of the Lorentz resonance alone [in red (dark gray)] and the photoionization alone [in green (gray)]. Parameters for the resonance, the xuv, and the laser are as in Fig. 1.

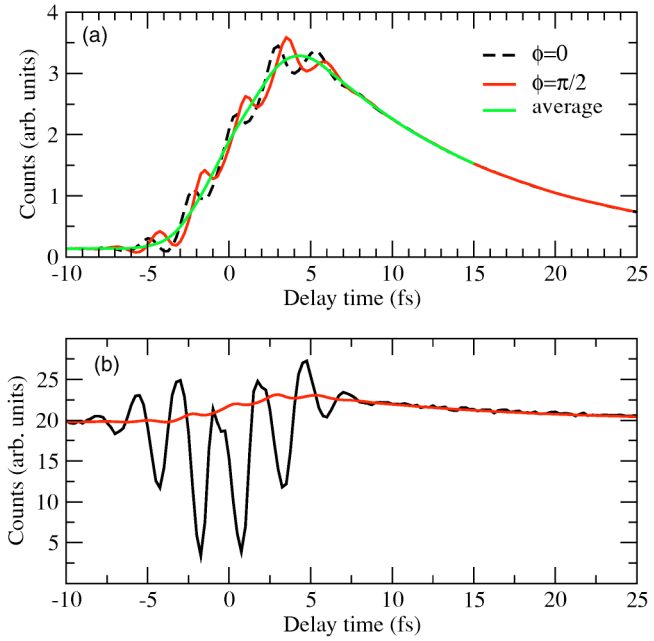

FIG. 3. (Color online) (a) Electron counts in the forward direction for a Lorentz resonance within the first sideband (20.4, 22.1) eV vs time delay, shown for two carrier envelope phases of the laser and the counts after phase average. (b) Similar electron counts for a Fano resonance for electrons within the energy of (20.4, 22.1) eV. The smooth curve gives the counts for the corresponding Lorentz resonance part, except vertically shifted by a constant.

and from the SFA are in good agreement. The Fano profile of the resonance is significantly modified by the laser field and evidences of possible sidebands appear (see later) at 1.65 eV from the resonance on both sides. Such general agreement has been found in other tests. Thus we will use the SFA model developed here to calculate the electron spectra in xuv+ir experiments.

We next compare the present SFA prediction for the photoelectron spectra in the forward direction with the Auger electron spectra from the recent calculations of Smirnova *et al.* [10], using the same resonance parameters. The xuv has a pulse length of 0.5 fs, a mean photon energy of 60 eV, and a peak intensity of 10^{12} W/cm^2, while the ir laser has a duration of 6.5 fs, a photon energy of 1.5 eV, and a peak intensity of 10^{11} W/cm^2, and ϕ=0. Two Lorentz resonances with lifetimes of (a) 0.2 fs and (b) 2.0 fs, are considered. Figures 2(a) and 2(b) shown here are to be compared to their Figs. 2(a) and 2(c), respectively. Although the formation of the resonance is different in the two cases, the dependence of the electron spectra on the laser carrier phase and on the lifetime of the resonance are similar. For the short lifetime resonance, the electron spectrum is broadened significantly. For the longer lifetime state, the sidebands are clearly resolved. In Fig. 2(c) we show the electron spectra in the forward direction for a Fano resonance, with the parameters used in Fig. 1 but at different time delays, calculated from the SFA. Electron spectra from the isolated Lorentz resonance part and from the direct photoionization part are shown for each time delay as well. For the Lorentz resonance alone the sidebands are clearly seen. For direct ionization the electron spectrum is broadened and its center shifts with the time delay. The shift stops when the xuv pulse and the ir laser do not overlap in time any more, as shown for the delay of 5 fs. The total electron spectrum, as the result of interference, appears to be highly "irregular" with respect to electron energy and to the time delay.

Using the present theory, let us analyze the procedure used by Drescher *et al.* [3] for deducing the Auger (or Lorentz resonance) lifetime based on the area of the first sideband for varying time delays between the xuv pulse and the ir laser. In Fig. 3(a) we show the area of the first sideband for electron energy between 20.4 and 22.1 eV vs the time delay from −10 to 25 fs for a Lorentz resonance represented by the parameters in Fig. 1 [examples of these sidebands can be seen in Fig. 2(c)]. Calculations were shown for two carrier envelope phases: 0 and π/2. Note that for the small time delay, there is a dependence on the carrier envelope phase. For the larger time delay where the xuv and ir pulses do not overlap in time, there is no such phase dependence. On the other hand, by averaging over the carrier envelope phase, a smooth curve is recovered and the result is similar to what was observed by Drescher *et al.* [3]. Note that the modulation in Fig. 3(a) at smaller time delay is separated by 2.5 fs—the period of the ir laser.

Can the lifetime of a Fano resonance be extracted in a similar manner in view of the fact that the electron spectra with respect to the time delay, as seen in Fig. 2(c), do not show any clear sidebands? In Fig. 3(b), we show the electron counts within the interval (20.4, 22.1) eV, as in the isolated Lorentz resonance, versus the time delay. For small time de-

lays, there are large oscillations. But for long time delays, the oscillation becomes weaker. In Fig. 3(b), the smooth line is from the Lorentz resonance part alone, except shifted by a constant. Clearly this shows that interference from the direct ionization is small at large time delays and the smooth curve can be fitted by an exponential law $\exp(-\Gamma t_d)$ (t_d is the time delay), from which the lifetime of the resonance is extracted. In fact in such an analysis we have tested that the extracted lifetime is independent of the specific selection of the energy bins used for the total electron counts.

The present formulation can be easily extended to obtain electron spectra for two nearby resonances. Consider two Lorentz resonances at 24.8 and 25.0 eV, with lifetimes of 30 and 40 fs, respectively, created by a 0.5-fs xuv pulse at a peak intensity of 10^{12} W/cm^2 into an ir laser of 10 fs, a frequency at 1.65 eV, and a peak intensity of 10^{12} W/cm^2. The streaked electrons from the two resonances will interfere. In Fig. 4(a) we show the spectra near the first sideband versus the time delay. One can see clearly that the electron spectrum shows oscillations within the sideband. The total electron counts within (22.4, 24.0) eV vs the time delay are shown in Fig. 4(b). The damped oscillations can be fitted by $c_1 e^{-\Gamma_1 t_d} + c_2 e^{-\Gamma_2 t_d} + c_{12} e^{-(\Gamma_1+\Gamma_2)t_d/2} \cos(\Delta E t_d + \phi_0)$, where t_d is the time delay, from which one can extract the two lifetimes and the energy separation ΔE between the two resonances. (Note: The line in the figure was fitted with the two given lifetimes and the input energy separation.)

In summary, we have presented a quantum theory for describing the photoelectron spectra of an autoionizing state created by an attosecond xuv pulse in the streaked few-cycle ir laser field within the strong field approximation that is expected to form the basis of future time-resolved experiments on resonances with attosecond pulses. We illustrated the utility of the theory by extracting the lifetime of an isolated and two nearby resonances from the time-resolved electron spectra. Future work may involve the possibility of extracting the q parameter of a Fano resonance and applications to more resonances. We comment that no real strong field effects are included in the present theory. For higher ir laser powers, one may expect new resonances to appear, similar to new resonances induced by the static electric field [13]. In such cases, there is no clear alternative but to develop a real time-dependent many-electron theory.

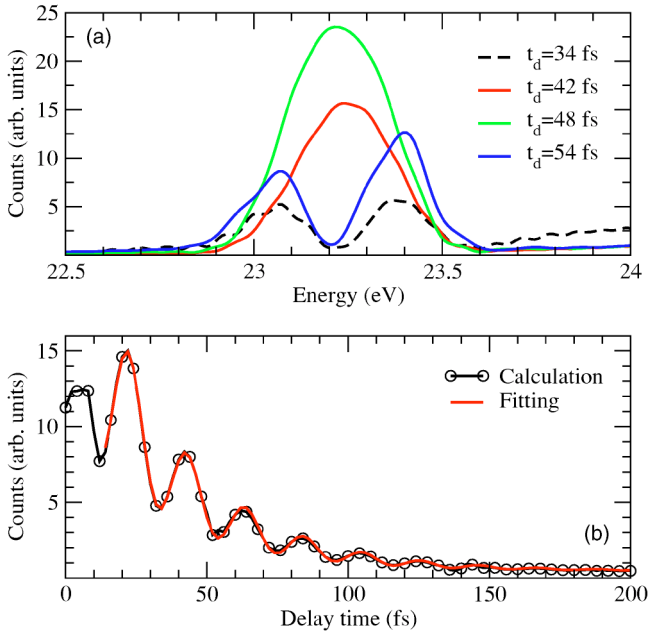

FIG. 4. (Color online) (a) Streaked electron spectra within the first sideband vs time delay for two nearby Lorentz resonances excited by an attosecond pulse. (b) The oscillatory total electron counts within the sideband vs time delay can be fitted to extract the lifetimes of the two resonances and their energy separation (see text).

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