



Time dependent aspects in the ionizations following atomic photoexcitations

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ABSTRACT

The time dependent aspects of the probability amplitudes in atomic excitations and their subsequent decay processes are discussed. The spin-orbit precession in an atomic ion core gives us a good clock to measure the characteristics of the autoionizing states. The lowest $3s$ sub-valence shell excitation of potassium atoms is attributed to an optically forbidden quartet state resonance, which are realized by the spin-orbit migration of the residual ion cores in the course of indirect photoionization processes. There is a class of multiple-electron ionization processes which provide us with the post-collision interaction effects. An electron that slowly leaves the ion behaves as a clock in the atomic system. The role of these atomic clocks are studied by tracing the time dependent state vector of the total system with the aid of accurate relativistic calculations of atomic structures and ionization dynamics. The post-collision interaction effects in an artificial many step Auger cascade and in a multi-step Auger cascade following the argon $1s$ vacancy creation by photon impact are studied extensively.

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1. Introduction

In the excitations or ionizations of atomic systems, we sometimes have coherent excitations of multiple-electronic-states that have slightly different energies. If we observe those energy differences in a time domain, we may find quantum beats [1]. If autoionization or Auger process take place during these quantum beats, electrons will carry out the information of the beats [2,3]. When atomic or atomic ionic autoionization or Auger process take place under the presence of a slowly receding photoelectron, the autoionizing or Auger electron will carry out the information of the receding electrons. We have, then, the so called post-collision interaction effect in the energy spectrum of emitted electrons or photoions [4,5]. The post-collision interaction effect is a time stamp placed on the energy spectra of emitted or scattered electrons [6–8]. We can, furthermore, observe the enhancement of the post-collision effects in the multiple-electron escape from the Auger cascade of the deep inner-shell photoionized atomic ions [9–11].

Generally speaking, we can find a clock in an atom if we virtually divide the atomic system into two or more parts and observe some of the sub-systems; we may observe time dependent effects in such atoms. In the present paper, we discuss the time dependent aspects in the processes of electron emissions from atoms after their electronic-state excitations or ionizations by photon impact. In Section 2.1, we consider the time correlations between the atomic autoionization processes and atomic internal motions. Autoionization of photon-impact induced doubly or inner-shell excited atomic states may sometimes create multiplet states in residual ions. In atomic

species with moderate atomic numbers, the multiplet nature is typically caused by spin-orbit interactions in ionic open-shell electrons [2,3]. In these systems, the open-shell undergoes precessions between different total angular momentum states of ions, and its cycle time gives us a good clock to measure the lifetime of the autoionizing states. If the lifetime is much longer than the cycle time, the autoionizing states may carry the information of the spin-orbit migration. In Section 2.2, we consider the time correlations between multiple electrons which start to leave atoms at different instances. Due to the correlations among the escaping electrons, we sometimes observe the effects that are so called post-collision interaction effects [7,10,11]. An electron that is emitted from an atom at an instance will propagate forming a wavepacket. When we have two or more electrons that are emitted from the same atom and observe the energy spectra of one electron of them, we may find information of such wavepacket in the energy spectra due to its de-coherence caused by the presence of other electrons. The post-collision interaction can be formulated theoretically by introducing wavepackets adjacent to the emitted electrons.

In this paper, we investigate the role of this type of atomic clocks in detail by tracing the time dependent state vector of the total system with the aid of accurate relativistic calculations of atomic structures and ionization dynamics [12–14].

2. The time development of the atomic system in photoionization or autoionization processes

We investigate firstly the time propagation of electrons which are emitted from atoms by photoionization or non-radiative decay.

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The state vector of an atom $\Psi_I(t_0)$ at time t_0 under an interaction Hamiltonian H_I may satisfy the following equation of motion,

$$i\hbar \frac{d\Psi_I(t_0)}{dt_0} = H_I \Psi_I(t_0). \quad (1)$$

We assume here that H_I is weak; we can expect that $\Psi_I(t_0) \simeq \Psi_I(0) \equiv |i\rangle$ for arbitrary t_0 . The fraction of the transition amplitude for an electron in the ionization continuum $|f_k\rangle$ is represented by

$$d\langle f_k | \Psi_I(t_0) \rangle = \frac{i}{\hbar} \langle f_k | H_I | i \rangle \times \exp \left[\frac{i}{\hbar} \int_0^{t_0} (\epsilon_f - \omega - \epsilon_0) dt' \right] dt_0, \quad (2)$$

where ϵ_f and ϵ_0 are the energies of initial and final states, respectively, and ω is the incident photon energy or free electron energy depending on the ionization processes under consideration. Then the fraction of outgoing electron wavefunction may be represented in the asymptotic region by

$$\begin{aligned} dF(t-t_0) &\equiv d|f_k\rangle_S \int \rho_k dk \langle f_k | \Psi_I(t_0) \rangle \\ &= \int \frac{\rho_k}{(2\pi)^{\frac{3}{2}}} \frac{\exp[i(kr - \frac{\ell}{2}\pi + \delta_\ell)]}{kr} dk \exp \left[-\frac{i}{\hbar} \epsilon_f(t-t_0) \right] \\ &\quad \times \frac{i}{\hbar} \langle f_k | H_I | i \rangle \exp \left[\frac{i}{\hbar} \int_0^{t_0} (\epsilon_f - \omega - \epsilon_0) dt' \right] dt_0, \end{aligned} \quad (3)$$

where k and ρ_k is the wavenumber and the state density of the continuum, respectively, and r , ℓ and δ_ℓ are the radial coordinate, angular momentum, and phase shift of the emitted electron, respectively. The integration over the wavenumber in Eq. (3) would give a wavepacket G with the electron velocity v ; we have

$$dF(t-t_0) = G_{k_\omega}(r-v(t-t_0)) \exp[i(k_\omega r - \epsilon_{k_\omega}(t-t_0))] dt_0, \quad (4)$$

where $\epsilon_{k_\omega} = \epsilon_f - \epsilon_0$ is the emitted electron energy and k_ω is its adjacent wavenumber. Normally the wavepacket vanishes after the t_0 integration to $t_0 \rightarrow \infty$, and there are no wavepackets in the system, because, in the isolated system, the Hamiltonian is invariant with respect to the displacement of time and so is the adjacent wavefunction. We may find wavepackets if the system (1) loses the coherence with respect to the time t_0 , (2) has a variable ϵ_{k_ω} and consequently k_ω , or (3) has a finite range in t_0 to be integrated. And, further on, if the ionization takes place into multiple channels with slightly different resonance energies, the inter-channel interference may occur despite the energy difference. We illustrate an example on this case in Section 2.1, and also illustrate another example from case (2) in Section 2.2.

2.1. The effect of spin-orbit migration in sub-valence ion core observed in atomic autoionization spectra

Suppose we have an excited atomic or an ionic state that can be represented by direct products of its sub-system wavefunctions, say, ψ_{j,E_j} and φ_{k,E_k} , where j and k are the quantum numbers of the sub-systems and E_j and E_k are their adjacent energies. We can represent the electronic-state wavefunction of the system $\Phi(t-t_0)$ by

$$\Phi(t-t_0) = \sum_{j,k} c_{j,k} \psi_{j,E_j} \varphi_{k,E_k} \exp[-i(E_j + E_k)(t-t_0)], \quad (5)$$

where $c_{j,k}$ is the amplitude of the sub-state (j,k) , and t and t_0 stand for the actual time and the time the state is created, respectively. If the excitation is instantaneous, we may observe the quantum beat due to the superposition of the states $\psi_{j,E_j} \varphi_{k,E_k}$ with different (j,k) that are coherently excited simultaneously. On the other hand, if the excitation process lasts long compared to the lifetime of the excited states, the wavefunction of the states will be represented by the superposition of the wavefunctions $\Phi(t-t_0)$ at different t_0 . In Fig. 1, we illustrate this feature for the case of atomic autoionization

processes as an example. We may then observe the excitations individually to the excitation energies. It is worthy to study these features by investigating the time development of the sub-states; the time given by the inverse of the energy difference $1/(E_j - E_f)$ gives the characteristic clock time of the atomic sub-system.

In Fig. 2, we illustrate the experimental photoion spectra of potassium 3s-photoionization together with the result of theoretical calculation, which were performed by Koide et al. [2]. They observed a doubly dipped nature in the 3s \rightarrow 4p photoexcitation spectral profile. The similar feature has been observed for several other systems [15,16,3]. The window type structure at 36.7 eV is assigned as of 4P resonance, and the structure at 37.4 eV, of 2P resonance. These excited discrete state configurations undergoes the configuration interaction to the optically favored $3s^2 3p^5 4s + \epsilon_{j_e}$ configuration. The energy difference ΔE_j between the ionic $3s^2 3p^5 ({}^2P_{j=3/2}) 4s$ and $3s^2 3p^5 ({}^2P_{j=1/2}) 4s$ states is almost 0.5 eV whereas the lifetime widths Γ_S of the atomic excited $3s^1 3p^6 4s 4p ({}^4,2P)$ states are around 0.2 eV. Because $\Delta E_j > \Gamma_S$, we may say that the ionic $3s^2 3p^5 ({}^2P_j) 4s$ states are well defined and the ionic states can be labeled in terms of the quantum number j . The wavefunction for the $3s^2 3p^5 4s + k_{j_e}$ continuum states, $\psi_{j,E'}$, may be given by a direct product of the wavefunction of the ion, $\psi_{j,E_j}^{\text{ion}}$, and the wavefunction of emitted electron, ϕ_{ϵ,j_e} , as

$$\psi_{j,E'} = \psi_{j,E_j+\epsilon} = \psi_{j,E_j}^{\text{ion}} \phi_{\epsilon,j_e}. \quad (6)$$

We express the wavefunction of atomic excited $3s^1 3p^6 4s 4p ({}^4,2P)$ states $\varphi_{L,S}$, where L stands for the total orbital angular momentum and $L=1$ in the present case, and where S stands for the total spin angular momentum and $2S+1=4$ or 2 in the present case. Then, according to Fano's theory [17], the total wavefunctions after the photoexcitation, $\Phi_{L,S}$, are given by linear combinations of $\varphi_{L,S}$ and $\psi_{j,E'}$ as

$$\Phi_{L,S} = a_{L,S} \varphi_{L,S} + \sum_j \int d\epsilon b_{j,E_j}(\epsilon) \psi_{j,E_j}^{\text{ion}} \phi_{\epsilon,j_e}, \quad (7)$$

where $a_{L,S}$ and $b_{j,E_j}(\epsilon)$ are the expansion coefficients. From this equation we can see that the case of one discrete and two or more continua in Fano's theory [17] applies to the present photoionization processes.

Now, further on, we can point out, firstly, that the configuration interactions between $\varphi_{L,S}$ and all the $\psi_{j,E'}$ are finite for both cases $2S+1=4$ and 2 . Both the quartet and doublet $3s^1 3p^6 4s 4p$ states undergo the configuration interaction with the $3s^2 3p^5 ({}^4P_j) 4s + \epsilon_{j_e}$ continua. Therefore we may have Fano's spectral profiles both on

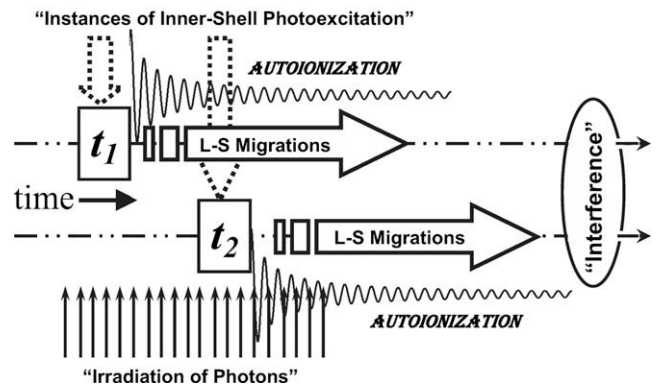


Fig. 1. Schematic drawing of the interference features in autoionization of atoms with LS multiplet ion cores. The fractions of wavefunction created at times t_1 and t_2 due to the irradiation of photons propagate in time with ion cores that undergo spin-orbit migration, and they interfere each other realizing the appearance of "optically forbidden" excitations.

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