

Photoionization of hydrogenlike ions surrounded by a charged spherical shell

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Within the framework of a model of a hollow atom as a hydrogenlike multicharged ion located at the center of a spherical shell formed by highly excited electrons, the photoionization cross sections of the inner $1s$ level of the ion have been calculated. The results show that the existence of the outer electronic shell of the hollow atom results in oscillations in the energy dependence of the photoionization cross section. It has been demonstrated also that the photoionization cross section as a function of photon energy is extremely sensitive to the magnitude of the discontinuity of the electric field at the surface of the outer electronic shell.

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I. INTRODUCTION

A theoretical investigation of the photoionization of hydrogenlike multicharged ions surrounded by a spherically symmetrical shell formed by excited electrons of a hollow atom has been performed. Consideration of such exotic atomiclike systems is of great interest, originally scrutinized in the classic paper of Sommerfeld and Welker [1] which studied the behavior of the hydrogen atom in an impenetrable potential well. Actually, the investigation of this seemingly rather abstract problem proved to be useful for the study of the squeezed atom [2,3], atoms in spherical cavities [4,5], etc. A wide range of theoretical studies of the spectral features of spatially entrapped atoms have been reported in recent years (see, for example, [6] and references therein) since the discovery and experimental investigation of the doped fullerenes, $A@C_{60}$, spherical molecules consisting of C_{60} with an A atom inside the fullerene cage [7]. The fullerene shell significantly affects the spectral characteristics of the entrapped atom, including the processes of the photoionization of this atom. The calculations show that in the ionization cross section of inner shells of the entrapped atom A in such molecules there are the so-called confinement resonances [8]. The physical reason for these resonances is as follows: in photoionization of an atom which is inside the fullerene cage, the C_{60} shell plays the role of a resonator. Photoelectron wave distortion due to the passing of an electron over the spherical potential well formed by the C_{60} shell results in oscillations in the energy dependence of the cross section. The amplitude and period of these oscillations depend both on the characteristics of potential well of C_{60} and on the photoelectron kinetic energy.

The physical origin and nature of the confinement resonances is the same as the cause of the extended x-ray absorption fine structure (EXAFS) in condensed matter or molecules as described previously [9]. Kronig's short-range order theory [10–12] explains EXAFS by the modulations of the final state wave function of the photoelectron caused by the backscattering from the surrounding atoms. The difference in the two cases is that the source of the reflected waves in the EXAFS phenomenon are the nearby atoms in the crystalline solid, while in the $A@C_{60}$ structures, the source of

backscattered waves is the atoms of the C_{60} cage, considered individually [13–16] or as a spherical average.

It is evident that a similar effect can be expected in the case of deep inner-shell photoionization of *hollow* atoms (HA's) [17–20]—short-lived exotic atomic systems in which only a small number of electrons are localized at the deep inner-shell atomic levels; the remaining, larger number of electrons, are in high-excited atomic states. These atomiclike systems are formed, for example, in the process of ultrafast inner-shell ionization of normal atoms, stimulated by high-intensity short pulse x rays, when multiple inner-shell ionization predominates, leading to the formation of hollow atoms [21,22]. HA's are also created in the processes of glancing scattering of multi-charged ions and their neutralization near a metallic surface [23], or during ion transport through the nanocapillaries [24,25]. In these processes the electrons extracted from the metal surface are captured into excited atomic states, forming a spherically charged shell around the inner-shell electrons. The common characteristic feature of hollow atoms and doped fullerenes $A@C_{60}$ or their ions [26] is that the photoionization of the deep subshell of the HA and the A atom in the molecule $A@C_{60}$ can be considered as a process of ionizing the atomiclike system surrounded by a spherical shell. The partial reflection of the electron wave from a shell of high-excited electrons, as in the $A@C_{60}$ case, will have an impact on the energy dependence of the photoeffect cross section of the core levels of a HA.

The analysis of this problem is of importance because, despite the short lifetime, HA's play a vital role in high brightness x-ray measurements of ultrafast dynamics in chemistry [27], biochemistry, biology, and materials [28]. Therefore, the theoretical description of the processes with participation of these latest most exotic creations of atomic physics is of great interest.

The detailed quantum-mechanical calculation of the process of populating of these atomic states is an extremely complex problem. Therefore, the detailed density distribution of the electrons forming the HA shell and the detailed behavior of its potential are unknown. But, whatever the density distribution is, the electric field of a HA has a jump while transiting through the charged electronic shell. The value of this jump is defined by the electric charge inside the shell and by the number of electrons in the shell. The HA potential

the photoelectron is moving in has no discontinuities at the inner and outer surfaces of the charged shell. This is the principal difference between the confined systems considered earlier and hollow atoms. If the reflected wave in the molecule $A@C_{60}$ results from the wave reflection from the potential well boundary, then in the hollow atoms this reflection is caused by the *discontinuity of potential derivative*, i.e., the potential jogs. Accordingly, the question then arises as to how the confinement resonances are modified in this case, i.e., how well can the confinement effects be seen. This paper is devoted to the analysis of these general questions using hollow atoms as examples.

To consider the process of the model HA photoionization we radically simplify the real physical picture by assuming that the HA consists of a single core $1s$ electron, and the remaining excited electrons of the HA shell are concentrated in an infinitesimally thin spherical layer of radius R that we consider as a parameter of the problem. Within the framework of this model we study the dependence of the photoionization cross section of the HA $1s$ level on electric field jump and HA ion charge, on electron shell radius and photoelectron orbital momentum, and on energy.

II. GENERAL FORMULAS

The neutralization of a positive ion near the metal surface, and the formation of the shell of highly excited electrons is due to, first, over-the-barrier transitions of conduction electrons to the Rydberg levels of the multicharged ion because only classically allowed over-the-barrier processes (but not tunneling) are sufficiently fast to be effective within the interaction time of the ion with surface [29–31]. According to this model, a HA shell is formed when the potential barrier between the metal and ion drops below the Fermi level and electrons from close to the Fermi edge of the metal surface can be transferred classically to the ion levels. This model predicts the principal quantum number n_c of the highly excited ion states into which the first over-the-barrier transitions will take place,

$$n_c \approx \frac{Z}{\sqrt{2W}} \left(1 + \frac{Z-0.5}{\sqrt{8Z}} \right)^{-1/2}, \quad (1)$$

where Z is the charge of the multicharged ion incident on the surface and W is the work function of the metal¹ the potential barrier, which is impenetrable for classical electron transitions, vanishes at a critical distance,

$$d_c \approx \frac{\sqrt{2Z}}{W}. \quad (2)$$

This critical distance should be of the same order of magnitude as the radius R of the hollow atom shell. According to Eqs. (1) and (2) for metallic surfaces with work function $W \approx 4-5$ eV and moderately high ion charges, $Z \approx 10$, the principal quantum numbers of excited electrons are $n_c \sim 11-13$ and the radius of the HA shell is about R

$\sim 25-30$ a.u.. These are the values of the radius R employed in our numerical calculations.

Now, assuming that Z_1 electrons of the shell are concentrated in an infinitesimally thin spherical layer of radius R , then the potential resulting from the electric field in which the electron moves in the process of the optical transition from the bound $1s$ state into a continuum of this model HA is

$$V(r) = \begin{cases} -\frac{Z}{r} + \frac{Z_1}{R} & \text{for } r \leq R, \\ -\frac{Z}{r} + \frac{Z_1}{r} & \text{for } r \geq R. \end{cases} \quad (3)$$

The Schrödinger equation for an electron moving in the potential field of Eq. (3) has the form

$$\Delta \psi_E + 2 \left(E + \frac{Z}{r} - \frac{Z_1}{R} \right) \psi_E = 0 \quad \text{for } r \leq R, \quad (4)$$

$$\Delta \psi_E + 2 \left(E + \frac{Z}{r} - \frac{Z_1}{r} \right) \psi_E = 0 \quad \text{for } r \geq R. \quad (5)$$

The first of these equations can be considered as the motion of an electron with the effective energy $E - Z_1/R$ in the attractive Coulomb field $V(r) = -Z/r$. The second equation (5) describes the motion of an electron with energy $E = k^2/2$ in the Coulomb field, $V(r) = -(Z - Z_1)/r$.

The radial parts of the electron wave functions are defined by [32]

$$\frac{d^2 P_{kl}}{dr^2} + \left(k^2 + \frac{2Z}{r} - \frac{2Z_1}{R} - \frac{l(l+1)}{r^2} \right) P_{kl} = 0 \quad \text{for } r \leq R, \quad (6)$$

$$\frac{d^2 P_{kl}}{dr^2} + \left(k^2 + \frac{2(Z - Z_1)}{r} - \frac{l(l+1)}{r^2} \right) P_{kl} = 0 \quad \text{for } r \geq R. \quad (7)$$

To reduce these equations to the standard form of differential equations for the continuum Coulomb functions [33], in Eq. (6) we introduce a new wave vector for the electron $k_1^2 = k^2 - 2Z_1/R$ (for $k^2 \geq 2Z_1/R$) and replace the variable r with $\rho_1 = k_1 r$. After these replacements the Eq. (6) reduces to the standard form

$$\frac{d^2 P_{kl}}{d\rho_1^2} + \left(1 + \frac{2\eta_1}{\rho_1} - \frac{l(l+1)}{\rho_1^2} \right) P_{kl} = 0 \quad \text{for } r \leq R, \quad (8)$$

where the parameter $\eta_1 = Z/k_1$. The regular solution of Eq. (8), the solution that is finite at the origin, coincides up to an arbitrary multiplicative constant with the regular Coulomb function $u_{kl}(\eta_1, \rho_1)$ [33] and has the form

$$P_{kl}(\rho) = D_l(k) u_{kl}(\eta_1, \rho_1). \quad (9)$$

The r -coordinate-independent amplitude factor $D_l(k)$ is defined by the matching conditions for the wave function, Eq. (7), with the wave function outside the shell.

In Eq. (7) we introduce the variable $\rho = kr$ and the parameter $\eta = (Z - Z_1)/k$, reducing it to the form

¹Atomic units are used throughout this paper.

$$\frac{d^2 P_{kl}}{d\rho^2} + \left(1 + \frac{2\eta}{\rho} - \frac{l(l+1)}{\rho^2}\right) P_{kl} = 0 \quad \text{for } r \geq R. \quad (10)$$

Outside the shell, $r \geq R$, the solution of Eq. (10) is a linear combination of the regular $u_{kl}(\eta, \rho)$ and irregular $v_{kl}(\eta, \rho)$ Coulomb functions

$$P_{kl}(\rho) = u_{kl}(\eta, \rho) \cos \Delta_l - v_{kl}(\eta, \rho) \sin \Delta_l. \quad (11)$$

Here the additional phase shift $\Delta_l(k)$ is due the influence of the potential of the HA shell. The regular and irregular Coulomb functions are characterized by the asymptotic behavior

$$u_{kl}(\eta, \rho \rightarrow \infty) \approx \sin\left(\rho + \eta \ln 2\rho - \frac{\pi l}{2} + \delta_l\right), \quad (12)$$

$$v_{kl}(\eta, \rho \rightarrow \infty) \approx -\cos\left(\rho + \eta \ln 2\rho - \frac{\pi l}{2} + \delta_l\right), \quad (13)$$

where $\delta_l(k) = \arg \Gamma(l+1-i\eta)$ is the Coulomb phase shift. In comparing the formulas, Eqs. (10)–(13), with those from Ref. [33], it should be noted that in this paper we deal with an attractive Coulomb field rather than a repulsive one.

The conditions of equality of the logarithmic derivative for the function defined by Eqs. (9) and (11) at $r=R$, result in the following expressions for the phase $\Delta_l(k)$ and the amplitude factor $D_l(k)$:

$$\tan \Delta_l = \frac{k_1 u'_{kl}(\eta_1, x_1) u_{kl}(\eta, x) - k u'_{kl}(\eta, x) u_{kl}(\eta_1, x_1)}{k_1 u'_{kl}(\eta_1, x_1) v_{kl}(\eta, x) - k v'_{kl}(\eta, x) u_{kl}(\eta_1, x_1)}, \quad (14)$$

$$D_l(k) = \frac{1}{u_{kl}(\eta_1, x_1)} [u_{kl}(\eta, x) \cos \Delta_l - v_{kl}(\eta, x) \sin \Delta_l]. \quad (15)$$

Here the variables $x=kR$, $x_1=k_1R$ and the wave function derivatives are $u'_{kl}(\eta, \rho) = du_{kl}(\eta, \rho)/d\rho$ and $v'_{kl}(\eta, \rho) = dv_{kl}(\eta, \rho)/d\rho$.

The initial $1s$ state of the HA is localized in the region of space with radius $r \sim 1/Z \ll R$. This allows the neglect of the effect of the shell on this state, to an excellent approximation, and to consider its wave function to be hydrogenic, $\psi_{1s} = (P_{1s}/r) Y_{00}(\mathbf{r})$ where

$$P_{1s}(r) = 2Z^{3/2} r e^{-Zr}. \quad (16)$$

The dipole matrix element of the $1s$ photoionization of an ion is formed at some distances $\sim 1/Z$, i.e., in the region of space where the wave functions of the initial state, Eq. (16) and final state, Eq. (9), are hydrogenic. Therefore, the photoionization cross section of the HA, $\sigma^{\text{HA}}(k)$, up to the factor $D_1^2(k)$, coincides with the photoionization cross section of a hydrogenic ion with charge Z , $\sigma^{\text{HA}}(\omega) = D_1^2(k) \sigma(\omega)$, where [34]

$$\sigma(\omega) = \frac{2^9 \pi^2}{3cZ^2} \left(\frac{I_{1s}}{\omega}\right)^4 \frac{e^{-4\eta \operatorname{arccot} \eta}}{1 - e^{-2\pi\eta}}, \quad (17)$$

where c is the light speed and $\omega = I_{1s} + k^2/2$ is the photon energy. Thus, the calculation of the effect of the HA shell on

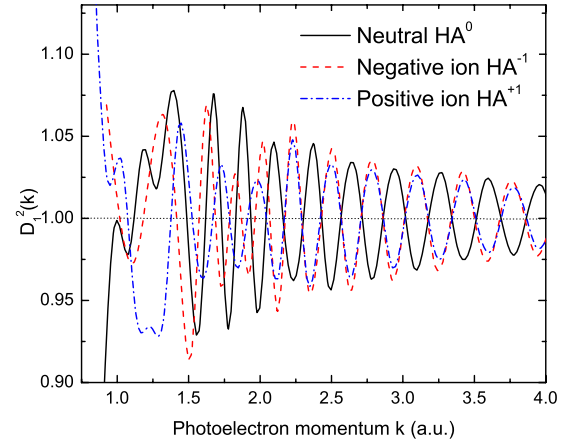


FIG. 1. (Color online) Amplitude factors $D_1^2(k)$ as a function of photoelectron momentum k for the three different charge states of the hollow atom, HA.

the cross section for $1s$ photoionization reduces to calculating the amplitude factor, Eq. (15), for photoelectron orbital momentum, $l=1$. Since in our model the hydrogenic ion in fact is located in the center of the spherical shell with charge $-Z_1$, the ionization potential of the $1s$ level of HA is equal to $I_{1s} = Z^2/2 - Z_1/R$.

III. RESULTS AND DISCUSSION

To obtain the numerical solutions of Eqs. (8) and (10), each of them is represented as a system of two differential equations of the first order,

$$\frac{dP_{kl}}{d\rho} = P'_{kl},$$

$$\frac{dP'_{kl}}{d\rho} = \left(-1 - \frac{2\eta}{\rho} + \frac{l(l+1)}{\rho^2}\right) P_{kl}. \quad (18)$$

This system of equations is solved by the Runge-Kutta method [35], which allows simultaneous calculation of the wave function and its derivative. The correctness of the numerical procedures is controlled by the calculation of the Wronskian of these equations, which must be equal to unity at all the points,

$$W_{kl}(\rho) = u_{kl}(\eta, \rho) v'_{kl}(\eta, \rho) - u'_{kl}(\eta, \rho) v_{kl}(\eta, \rho) = 1. \quad (19)$$

We performed the calculation of the $1s$ photoionization of a hydrogenic ion with the nuclear charge $Z=10$ for three systems: (i) the neutral hollow atom (HA^0) with one $1s$ electron and $Z_1=9$ electrons in the shell; (ii) the negative ion of the hollow atom (HA^{-1}) with one $1s$ electron and $Z_1=10$ electrons in the shell; and (iii) the singly charged positive ion HA^{+1} with $Z_1=8$ excited electrons in the shell. The calculated results for the amplitude factors, with shell radius $R=25$, are presented in Fig. 1. The functions $D_1^2(k)$ in all three cases have the form of damped oscillations relative to the asymptotic value, $D_1^2(k \rightarrow \infty) = 1$. The deviations from almost ideal damped sinusoid are observed only for small photo-

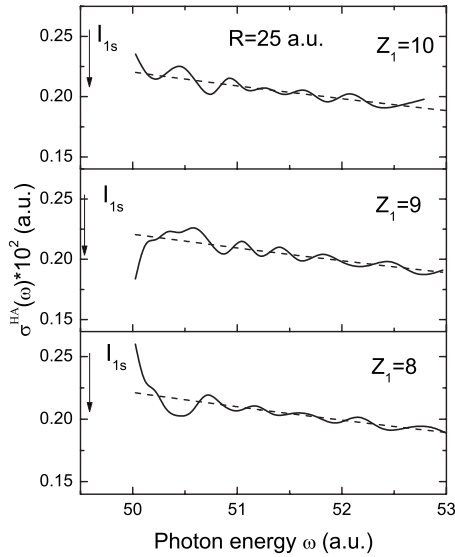


FIG. 2. Photoionization cross sections of the hollow atom HA^0 and its ions HA^{-1} , HA^{+1} as a function of photon energy ω . Ionization potentials I_{1s} are equal to 49.64, 49.60, and 49.68 a.u. for the shell charge Z_1 that is equal to 9, 8, and 10, respectively.

electron momentum and near $k \sim 1.8$ a.u. It is interesting that the curves corresponding to the positive and negative ions practically coincide, while $D_1^2(k)$ for the neutral HA^0 appears to be 180° out of phase with them. Despite the fact that in the three cases the continuum wave functions outside the shell are quite different (spherical Bessel functions for HA^{-1} , Coulomb functions with $Z=1$ for HA^0 or the Coulomb functions with $Z=2$ for HA^{+1}), the three amplitude functions $D_1^2(k)$ decrease with increasing photoelectron momentum k in a similar manner.

The photoionization cross sections for the hollow atom and its ions, $\sigma^{HA}(\omega)$, are given in Fig. 2 by solid lines. The ionization potentials of the $1s$ levels for each case are shown as arrows. The cross sections have the form of curves oscillating around the background cross section of the free hydrogenic ion, shown in Fig. 2 as dashed lines. The amplitudes of these oscillations are small, $\sim 5\%$ – 8% of the background cross sections, similar to the typical amplitude of EXAFS oscillations [12]. However, the differences among the cross sections for HA^{-1} , HA^0 , and HA^{+1} are somewhat surprising considering the potentials, $V(r)$ of Eq. (3), for each of these cases, as shown in Fig. 3. The differences in the discontinuities in the derivatives of the potentials at $r=R$ for the three HA systems seem insignificant, but their presence dramatically changes the character of the HA cross sections $\sigma^{HA}(k)$ as compared with that for the free hydrogenic ion. Instead of the monotonic decreasing cross section $\sigma(k)$ that is characteristic of the free ion, we have pronounced oscillations in the cross sections $\sigma^{HA}(k)$. The oscillations are a consequence of the reflection of the photoelectron wave from the shell at $r=R$. The reflected photoelectron wave interferes with the “direct” one. The characteristics of the reflected wave are strongly dependent upon the details of the discontinuity in the derivative of the potential; as a result, the interference manifested is also dependent upon the discontinuity. Thus,

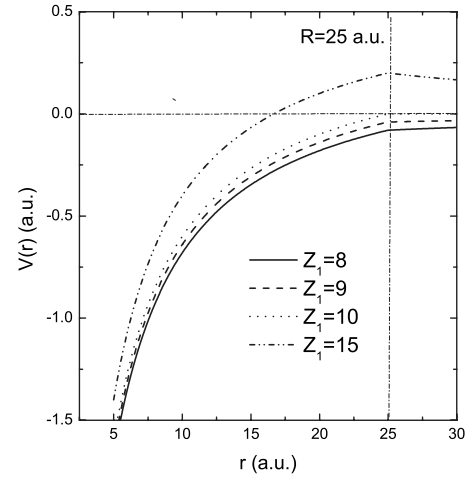


FIG. 3. Potentials $V(r)$ of the hollow atom for different values of shell charge Z_1 .

the energy dependence of the photoionization cross section of the inner shell of the HA is extremely sensitive to a potential gradient, i.e., to the jump of the electric field $\Delta E = Z_1/R^2$ at the shell radius, $r=R$.

For $Z_1 > Z$ the charged shell creates a potential barrier that can lead, as shown in [26], to the appearance of quasistationary states of the system. As an example, the potential in which the photoelectron moves when the number of electrons in the hollow atom shell is equal to $Z_1 = 15$ is presented in Fig. 3. In this case we deal with the fivefold negatively charged shell. However, because of the large shell radius R , as compared to that considered in [26], the barrier height in our case is insufficient for the formation of quasistationary states.

It is of interest to inquire how the results depend upon the choice of shell radius R . To examine this point, amplitude factors were calculated for different radii of the HA^0 shell ($R=25$, 27, and 30) and are given in Fig. 4. All the results for the photoelectron momentum $k \geq 3$ have the form of damped oscillations with slowly changing periods. The oscillation amplitude decreases exponentially. The envelope curves de-

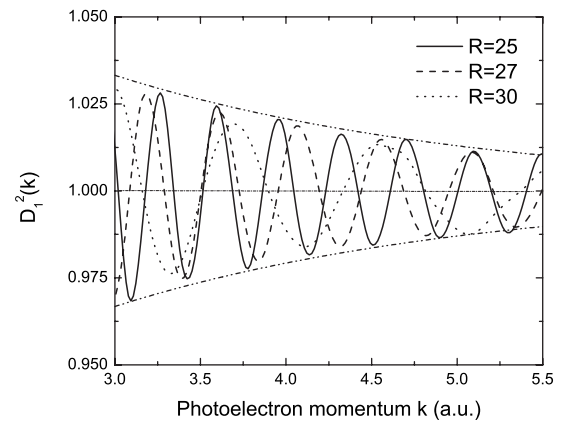


FIG. 4. Amplitude factors $D_1^2(k)$ as a function of photoelectron momentum k for three different radii R of the hollow atom HA^0 shell.

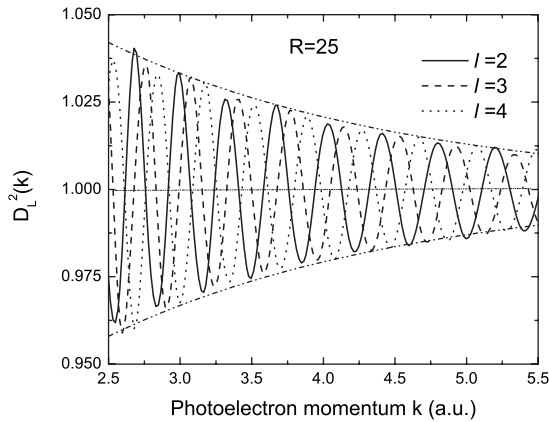


FIG. 5. Amplitude factors $D_l^2(k)$ for different orbital electron moments l at a fixed radius $R=25$ of the hollow atom HA^0 shell.

finied by the equation $Y(k)=1\pm A \exp(-k/\kappa)$ with the parameters $A\approx 0.136$ and $\kappa\approx 2.126$ are also given in this figure. In any case, although the choice of the shell radius changes the phases of the oscillations, as seen in Fig. 4, it makes no qualitative difference.

All of the above results concern only the $l=1$ continuum wave. However, for photoionization of initial states that are not ns states, other continua will come into play. Certainly, the positions of the resonances in the oscillations of the $D_l^2(k)$, as a function of photoelectron momentum k , depend on the electron orbital momentum l . To get a feeling for the dependence on l , the calculated results for the amplitude factors for different values of l are presented in Fig. 5 where it is seen that the curves of differing l are shifted relative to each other owing to the differing centrifugal repulsion added to the effective potential for each different l . This means that the parameters defining the photoelectron angular distribution, which are defined by combinations of the dipole, quadrupole, etc., matrix elements, have a more complex structure, as a function of energy, than the total cross section, $\sigma^{HA}(k)$, due to the interference of the oscillations of the matrix elements corresponding to the emission of photoelectrons with various orbital angular momenta.

IV. CONCLUSIONS

Ultrafast atomic processes resulting from x-ray interactions with atoms or the interaction of atomic ions with a metal surface can give rise to the formation of hollow atoms. In the present paper, we have considered a class of x-ray studies of these objects, specifically hollow atom photoionization. We used as examples the photoionization processes of HA^0 , HA^{+1} , and HA^{-1} atomiclike systems. Within the framework of the rather simple model of the HA it has been demonstrated that the existence of a charged shell of HA leads to an oscillatory energy dependence of the photoionization cross section of a deep inner shell. It has also been shown that the cross section $\sigma^{HA}(\omega)$ as a function of photon energy ω is extremely sensitive to the jump of the electric field due to the excited electrons of the HA shell.

Recent progress in the emission of high-intensity short-pulse x rays [36–38] leads to new sources of high-power x rays and creates new opportunities for x-ray measurements in biology, material science, and chemistry. The results of this paper, we believe, will stimulate and, to some extent, direct experimental studies in this rapidly developing field of x-ray spectroscopy.

Finally, we reiterate that the phenomenology found in this investigation, specifically the oscillations in the photoionization cross section, is similar to the phenomenon of confinement resonances found in the photoionization of fullerene-encapsulated atoms [8], including the photoionization of C_{60} itself (and its ions) [39]. In all these cases, the underlying cause is the same, *viz.* the continuum states of the atom inside the additional spherical potential are affected by the reflection of the photoelectron wave from that spherical potential.

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