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# Screening Effects in Multielectron Ionization of Heavy Atoms in Intense Laser Fields

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Multielectron ionization of Xe is described in terms of multistep processes, driven by a laser field which is screened by the motion of the outer 5*p* shell. In stepwise multiple ionization of the 5*p* shell, screening is successively reduced. The *effective, local intensity* will therefore *increase* during the stripping of the outer shell. In the 4*d* inner-shell region the effective intensity is very low. Finally we point out difficulties connected with the tentative identification of recently observed 4*d*-Auger spectra.

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With use of intense lasers, it is now possible to test the dynamics of outer and inner shells in heavy atoms in radically new ways. The *intensity* can be used as an independent parameter, in addition to the frequency $\omega$ and the polarization, in order to study the nonlinear response of the system.

In this Letter, we shall focus attention on a few important problems that have been investigated in a number of recent experiments on Xe performed at various wavelengths (532 and 1064 nm,[1] 193 nm,[2,3] and even 10 $\mu$m[4]), as well as in recent theoretical studies,[5-10] namely, (i) multielectron ionization of the outer 5*p* and 5*s* shells,[1-4] (ii) single-electron ionization of the inner 4*d* shell,[3] and (iii) *N*-*OO* Auger transitions as a probe of 4*d* inner-shell ionization.[3]

One of the major points of the present paper is the introduction of the concept of *effective intensity* $I_{\text{eff}}$. As a result of *screening of the external laser field* (intensity $I$) by the outermost 5*p* shell, the atomic electrons will experience a frequency- and space-dependent *effective local field* (intensity $I_{\text{eff}}$). As long as the frequency $\omega$ of the external field is low (lower than the excitation energy gap), and the intensity is not too high, the effective local field at frequency $\omega$ will be weaker, or much weaker, inside than outside the atom. Moreover, the higher harmonics of the induced field will be negligible. Therefore, this *lowering of the effective intensity* due to screening will make it more difficult to ionize singly the outer 5*p* shell and practically impossible to ionize the inner 4*d* shell.[11]

However, if the effective intensity nevertheless is sufficient for multiple ionization, the stripping of the 5*p* shell will successively reduce the effects of screening. We therefore suggest that the effective local intensity may increase in a significant way during stripping of the outer shell, leading to an enhanced yield of highly charged ions compared with singly charged ions.

The basis for the present[9,10] treatment is diagrammatic many-body theory, which provides systematic perturbative solutions of the time-dependent Schrödinger equation. This type of approach has been very successful in the description of many-electron dynamics in one-photon ionization processes.[12,13] In the present work we use diagrammatic perturbation theory to finite and infinite order for describing multiphoton, multielectron excitation and ionization amplitudes; i.e., we consider nonlinear response. The infinite-order aspects involve renormalization of electron-photon coupling and energy due to many-electron screening, ac-Stark shift, and multielectron ionization. The finite-order aspects involve low-intensity expansion of all physical quantities and absorption of a finite number of photons. The way that screening is treated in the present work has much in common with the recent work by Zangwill[14] and by Szöke and Rhodes[15] using the time-dependent local-density approximation (see Zangwill and Soven[16]).

We shall first discuss *N*-photon, one-electron ionization in order to establish a simple form of the effective electron-photon interaction. We then study the effects of decreasing screening during stepwise ionization.

In the present formulation, modifications of the external field and of the photoelectron wave functions appear through an effective electron-photon interaction. Moreover, the core-hole level is influenced by the ac-Stark effect and by double ionization (and higher) processes.

We may write the *N*-photon, single-electron ($i \rightarrow \epsilon$) ionization current according to[10]

$$J_{\epsilon}(\omega,N) \sim |\langle \epsilon | \tilde{U}^{N}_{\text{eff}} | i \rangle|^2 \frac{\Gamma_i/2\pi}{(\epsilon - \epsilon_i - N\omega)^2 + (\Gamma_i/2)^2}. \tag{1}$$

$i$ and $\epsilon$ represent one-electron levels of the isolated atom. $\tilde{U}^N_{\text{eff}}$ is the effective $N$-photon, one-electron interaction which incorporates screening of the field, as well as the ac-Stark effect on the photoelectron. $\epsilon_i$ is the core-hole energy dressed by the ac-Stark effect and multiple ionization, and $\Gamma_i$ is the associated damping. The effective amplitude $\langle \tilde{U}^N_{\text{eff}} \rangle$ may be broken up into contributions from various radiative and nonradiative harmonics $(\omega, 2\omega, \ldots)$ of the effective field. Here, we shall only consider the effective field at frequency $\omega$, which should dominate at not too high intensities. We then have ($\omega_{ki} = \epsilon_k - \epsilon_i$, etc.)

$$\langle \epsilon | \tilde{U}^N_{\text{eff}} | i \rangle = \sum_{kmn} \frac{\langle \epsilon | \tilde{U}^1_{\text{eff}} | k \rangle \cdots \langle m | \tilde{U}^1_{\text{eff}} | n \rangle \langle n | \tilde{U}^1_{\text{eff}} | i \rangle}{[\omega_{ki} - (N-1)\omega] \cdots (\omega_{mi} - 2\omega)(\omega_{ni} - \omega)} \tag{2}$$

with

$$\langle m | \tilde{U}^1_{\text{eff}} | n \rangle = \langle m | U^1_{\text{eff}} | n \rangle / [1 - \beta(\omega; I)]. \tag{3}$$

In Eq. (3), $U^1_{\text{eff}}$ represents the screened electron-photon interaction, and $\beta(\omega, I)$ describes the ac-Stark effect on the photoelectron. The form of Eq. (3) is obtained by approximate summation of an infinite series of Stark shifts.

The matrix element of the screened electron-photon coupling may be written as

$$-\langle m | U^1_{\text{eff}} | n \rangle = \langle m | \mathbf{E}_{\text{eff}}(\mathbf{r}, \omega) \cdot \mathbf{r} | n \rangle \tag{4a}$$

$$= E \langle m | \mathbf{e} \cdot \mathbf{r} \epsilon_k^{-1}(\mathbf{r}, \omega) | n \rangle \tag{4b}$$

$$= I^{1/2} \langle \epsilon_k^{-1}(\omega) \rangle_{mn} \langle m | \mathbf{e} \cdot \mathbf{r} | n \rangle \tag{4c}$$

$$= \langle I^{1/2}_{\text{eff}}(\omega) \rangle_{mn} \langle m | \mathbf{e} \cdot \mathbf{r} | n \rangle, \tag{4d}$$

where the space-dependent inverse dielectric function $\epsilon_k^{-1}(\mathbf{r}, \omega)$ may be approximated by[12,13] ($k$ denotes the initial-state ion charge)

$$\epsilon_k^{-1}(\mathbf{r}, \omega) = 1 - \sum_{nj} c^k_{nj} \frac{\langle j | 1/r_{12} | n \rangle_k \langle n | r \epsilon_k^{-1}(\mathbf{r}, \omega) | j \rangle_k r^{-1}}{[(\omega^k_{nj})^2 - \omega^2]/2\omega^k_{nj}}, \tag{5a}$$

$$c^k_{nj} = \tfrac{1}{3}(n^k_j - \delta_{ij})(2l_n + 1) \begin{pmatrix} l_j & 1 & l_n \\ 0 & 0 & 0 \end{pmatrix}^2. \tag{5b}$$

$n^k_j$ denotes the number of electrons in the $j$th shell, and $\delta_{ij}$ corrects for the self-interaction: $\delta_{ij} = 1$ if $i$ and $j$ belong to the same subshell; otherwise $\delta_{ij} = 0$.

Since the screened electron-photon interaction also appears in the Stark shift, Eq. (3) may be further simplified according to

$$-\langle m | \tilde{U}^1_{\text{eff}} | n \rangle = \frac{\langle I^{1/2}_{\text{eff}}(\omega) \rangle}{1 - \beta_0(p\omega) \langle I_{\text{eff}}(\omega) \rangle} \langle m | \mathbf{e} \cdot \mathbf{r} | n \rangle. \tag{6}$$

$\beta_0(p\omega)$ describes the ac-Stark effect for a system of noninteracting electrons through the polarizability of the excited level reached after absorption of $p$ photons. $\langle I_{\text{eff}}(\omega) \rangle$ is an average effective intensity due to many-electron screening.

Let us now consider multielectron ($q+$) ionization.[5-10] In the present treatment, the dominating process will be *stepwise ionization,* leading to successive creation of $A^{1+}, A^{2+}, \ldots, A^{q+}$ ions. Single-electron excitation in any intermediate $A^{k+}$ ion stage is driven by an effective interaction $\tilde{U}^{N_k}_{\text{eff}}$ representing total absorption of $N_k$ photons of frequency $\omega$. To simplify matters, we shall only consider the minimum number $N_k$ of photons necessary for ionization of the $A^{k+}$ ions.

At sufficiently low intensities, and in the absence of saturation effects, the number of detected $A^{q+}$ ions can be written as a product of the different steps according to

$$N^{q+} = \prod_{k=0}^{q-1} \left( \frac{I}{\omega} \right)^{N_k} \sigma_k \tau_k, \tag{7}$$

where

$$\sigma_k = \omega^{N_k} | \langle \epsilon | \tilde{U}^{N_k}_{\text{eff}} | i \rangle |^2_{\epsilon = \epsilon^k_i + N_k \omega} \tag{8}$$

is the $N_k$-photon cross section. $\tau_k$ is the $N_k$th-order pulse duration, and $\epsilon^k_i$ is the dressed ionization energy ($A^{k+} - A^{(k+1)+}$).

Finally, by inserting the effective interaction in Eq. (6) into Eq. (7), we obtain

$$N^{q+} = \prod_{k=0}^{q-1} (\tilde{I}_k / \omega)^{N_k} \sigma_{k0} \tau_k. \tag{9}$$

$\sigma_{k0}$ is the cross section for noninteracting electrons, given by Eq. (8) with $\tilde{U}_{\text{eff}}^{N_k}$ replaced by the external interaction $U^{N_k}$. All effects of screening and ac-Stark shift have been transferred to an *effective average intensity* $\tilde{I}_k$, which we *symbolically* write as

$$\tilde{I}_k = \frac{I |\langle \epsilon_k^{-1}(\omega) \rangle|^2}{|1 - \langle \beta_{k0}(\omega) \rangle |\langle \epsilon_k^{-1}(\omega) \rangle|^2 I|^2}, \tag{10}$$

where $\langle \epsilon_k^{-1}(\omega) \rangle$ and $\langle \beta_{k0}(\omega) \rangle$ denote averages arising from proper treatment of the sums in Eq. (2).

We have evaluated the dielectric function $\epsilon_k^{-1}(\omega)$ in Eq. (5) for neutral Xe ($k = 0$) in the frequency range $0 \text{ eV} < \omega < 6 \text{ eV}$, using a local-density one-electron basis. For the matrix element $\langle nd | U_{\text{eff}}^1(\omega) | 5p \rangle$, we find typical average values $\langle \epsilon_0^{-1}(\omega = 0) \rangle = 0.7$ and $\langle \epsilon_0^{-1}(\omega = 6) \rangle = 0.5$, and for the matrix element $\langle nf | U_{\text{eff}}^1(\omega) | 4d \rangle$, we find average values $\langle \epsilon_0^{-1}(\omega = 0) \rangle = +0.2$ and $\langle \epsilon_0^{-1}(\omega = 6) \rangle = -0.2$. It follows that the external intensity is reduced because of (mainly) $5p$ screening by a factor of 2–4 in the $5p$ region and by a factor of 25 (or more) in the $4d$ region. These results agree quite well with the static results of Zangwill and Soven.[16]

During successive steps of outer-shell $5p$ ionization of Xe, the average dielectric function $\langle \epsilon_k^{-1}(\omega) \rangle$ in the $5p$ region will be increased from a value around 0.5–0.7 for the complete $5p$ shell to unity when there remains only a single $5p$ electron to be ionized. Therefore, for a given external laser intensity, the effective intensity [Eq. (9)] should increase for each step of the stripping process, increasing the probability for creation of the higher charge states.

The essential point is then that in the present picture, the effective intensity is strongly *reduced* at the lowest ionization stages, making, e.g., single ionization *less probable* than expected from perturbation theory using the external intensity. Therefore, when comparing one-electron perturbation theory with experiment, one should in principle plot the measured number of $\text{Xe}^{k+}$ ions versus the corresponding average *effective intensity* $I_{\text{eff}}$, not the external laser intensity. This will displace the ions-vs-intensity curve for each ion species towards lower intensities (slopes will be conserved!). As a result, the intensity interval over which the $\text{Xe}^{k+}$ ions appear will be expanded. In particular, the $\text{Xe}^{1+}$ (and $\text{Xe}^{2+}$) ions will apear at substantially lower intensities [a factor of 2–4 at $\omega = 0$–$6$ eV)] than suggested by the nominal laser intensity, while $\text{Xe}^{6+}$ will not be shifted at all.

In the case of $4d$ inner-shell single ionization the interaction strength is very strongly reduced in the $4d$ region which, in combination with the high order of the process, makes direct $4d$ ionization highly improbable.[17]

The present discussion is incomplete in the case of very high intensities, where nonlinear components of the induced fields and the ac-Stark shifts become important. With increasing external intensity the amplitude of the $5p$ shell will increase, and so will intensity of the higher harmonics ($2\omega, 3\omega, \ldots$) of the induced field. In particular, there could be resonance enhancement of various higher harmonics due to multiple (possibly collective) excitation of the $5p$ shell. This view has been advocated by Rhodes and co-workers[2,3,15] and Chin, Yergeau, and Lavigne[4] in order to explain experiments at quite high intensities (above $10^{15}$ W/cm$^2$) where they have observed what they interpret to be Auger electrons arising from $4d$ ionization.

With regard to the problem of how $N$th-order $4d$ inner-shell ionization actually occurs, there are two extreme possibilities as we see it: (i) Stripping of the outer shell occurs during the first part of the pulse, which reduces the screening. The effective intensity in the inner-shell region then approaches the external intensity and leads to $N$th-order direct $4d$ ionization at frequency $\omega$. (ii) All the necessary energy $N\hbar\omega$ is first absorbed into a bound multiple (possibly collective) excitation of the outer shell. The stored energy $N\hbar\omega$ is then transferred to the inner shell, in the most extreme case via the $N$th harmonic of the interaction (Coulomb or radiative), and in the least extreme case by successive deexcitation of the "quanta" of the "collective" excitation via lower harmonics.

In case (i), the $4d$-Auger emission occurs in the presence of very few $5s, 5p$ outer-shell electrons; maybe only $4d^{-1}$-$5s^{-2}$ is possible. In case (ii), there is at least a possibility of having $4d^{-1}$-$5p^{-2}$ processes involving a complete $5p^6$ shell.

We emphasize that *the Auger spectrum provides a direct probe of the high-intensity dynamics:* The positions of the Auger lines identify the initial configuration of the Auger process ($4d^9 5s^x 5p^y$), and the intensity distribution over the various configurations provides information about the dynamics, i.e., the time development of the ionization process. For instance, we note that the most prominent Auger lines observed by Rhodes and co-workers[3] were tentatively identified as $4d^{-1}$-$5s^{-2}$ and to some extent $4d^{-1}$-$5s^{-1}5p^{-1}$. This could be consistent with the $5p$ shell being nearly stripped when the $4d$ ionization occurs: With a full $5p^6$ shell, the $4d^{-1}$-$5p^{-2}$ Auger processes would dominate, giving rise to Auger electrons around 35 eV. Such electrons were not observed.

On the other hand, there are serious problems with the picture of a nearly stripped $5p$ shell, since $4d$ ionization should then be a highly unlikely process.[17] Moreover, the Auger energies will shift towards higher energies; e.g., the $4d^{-1}$-$5s^{-2}$ transition will shift from about 10 eV in neutral Xe to around 40 eV in $\text{Xe}^{6+}$ (Hartree-Fock calculations; present work). Again, no electrons were observed at these energies.

In an analogous manner, the *x-ray emission spectrum* (e.g., $4d^{-1}$-$5p^{-1}$ lines) will also serve as a probe of the dynamics. A direct probe could also be obtained by an actual measurement of the intensity of the radiative emission of the various harmonics.

The idea that at high intensities a collective outer-shell resonance may drive the inner-shell ionization[3,4,15] via higher harmonics is very attractive. It remains to be demonstrated, however, that damping of the resonance due to multielectron ionization of the outer shell is not so large that the energy transfer to the inner shell becomes negligible. After all, complete stripping of the 5*p* shell is a common phenomenon at those high laser intensities ($>10^{15}$ W/cm$^2$), where higher harmonics can be expected to become important according to Szöke and Rhodes.[15]

Finally, we note that at high intensities there should be a close connection between multiple excitation of the 5*p* shell on the one hand, and multiple ionization and generation of higher harmonics of the induced field on the other. If large-amplitude oscillation of the 5*p* shell is central to the problem, multiple ionization could proceed via highly excited intermediate states. This may not, we feel, be described in terms of stepwise ionization through a sequence of ionic ground states, and would require an extension of the present approach.

In summary, in stepwise multiple ionization of the outer shell, screening is successively reduced. The *effective, local intensity will therefore increase* during the stripping of the outer shell. This might partly explain the relative ease with which highly charged ions are produced in, e.g., the rare gases. Moreover, the effective intensity at frequency $\omega$ will be low in the inner-shell region, making inner-shell ionization highly improbable.

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[1] A. L'Huillier, L. A. Lompré, G. Mainfray, and C. Manus, Phys. Rev. A **27**, 2503 (1983), and J. Phys. B **16**, 1363 (1983).

[2] T. S. Luk, U. Johann, H. Egger, H. Pummer, and C. K. Rhodes, Phys. Rev. A **32**, 214 (1985).

[3] C. K. Rhodes, Science **229**, 1345 (1985), and references therein.

[4] S. L. Chin, F. Yergeau, and P. Lavigne, J. Phys. B **18**, L213 (1985).

[5] M. Crance, J. Phys. B **17**, 3503, 4333, L635 (1984), and **18**, 455 (1985); M. Crance and M. Aymar, J. Phys. (Paris) **46**, 887 (1985).

[6] T. Åberg, A. Blomberg, J. Tullki, and O. Goscinski, Phys. Rev. Lett. **52**, 1207 (1984).

[7] S. Geltman, Phys. Rev. Lett. **54**, 1909 (1985).

[8] P. Lambropoulos, Phys. Rev. Lett. **55**, 2141 (1985).

[9] A. L'Huillier, L. Jönsson, and G. Wendin, to be published.

[10] A. L'Huillier, These de Doctorat d'Etat, University of Paris, 1986 (unpublished).

[11] This is reminiscent of metallic screening, where light with frequency below the plasma frequency becomes reflected at the metal surface and cannot penetrate deeply into the bulk.

[12] G. Wendin, in *New Trends in Atomic Physics,* edited by G. Grynberg and A. Stora, Les Houches Summer School Proceedings Vol. 38 (Elsevier, New York, 1984), and references therein.

[13] G. Wendin, Phys. Rev. Lett. **53**, 724 (1984).

[14] A. Zangwill, J. Chem. Phys. **78**, 5926 (1983).

[15] A. Szöke and C. K. Rhodes, Phys. Rev. Lett. **56**, 720 (1986); A. Szöke, Lawrence Livermore National Laboratory Report No. UCRL-93156, Pt. 1, Rev. 1, 1985 (to be published).

[16] A Zangwill and P. Soven, Phys. Rev. A **21**, 1561 (1980).

[17] A likely situation is that an intense laser field will sequentially strip first the $5p^6$ shell and then the $5s^2$ shell [at around $10^{16}$ W/cm$^2$ (Ref. 2)]. The 4*d*-ionization energy then rises to about 180 eV in Xe$^{8+}$ (Hartree-Fock calculation, present work), which probably explains why Luk *et al.* (Ref. 2) did not observe Xe$^{9+}$ even at their highest intensities (around $10^{17}$ W/cm$^2$).