Atomic Inner-Shell Excitation Induced by Coherent Motion of Outer-Shell Electrons

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Outer-shell electrons coherently driven by intense radiation can transfer energy in a direct intra-atomic process to inner-shell excitations. Provided that the effective momentum transfer Δq is sufficiently low ($\Delta q \lesssim \hbar/a_0$), the amplitudes governing the coupling of the outer electrons to the atomic core constructively sum. The effective cross section, which can be related to fast atom-atom collisions ($\gtrsim 10$ MeV/u), is evaluated in a limiting form closely resembling the Bethe result for inelastic electron scattering from atoms.

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Recent experiments[1,2] examining the nature of multiquantum ionization of atoms in intense ultraviolet fields have exhibited several anomalous characteristics. Among them are (1) reactions of unexpectedly high order, involving as many as 99 photons, and (2) a clear statement, from the atomic-number dependence, that the shell structure of the atom is the main determinant of the strength of the interaction. All the conspicuous characteristics of the experimental findings could be consolidated by that single principle. From the standpoint of this Letter, the main implication of these results[2] is that, at a sufficiently high intensity, the electrons in the outer atomic shells can be *coherently* driven by the incident wave to produce extremely high localized current densities j on the order of $10^{14} \leqslant j \leqslant 10^{15}$ A/cm^2. A multielectron atom undergoing a nonlinear interaction of this type responds in a *fundamentally different fashion* from that of a single-electron atom. It is expected that this ordered motion, which represents a very high level of atomic excitation corresponding quantum mechanically to a multiply excited configuration in which all the electrons in a shell are in excited orbitals, would have a lifetime τ given approximately by that characteristic of autoionization. This would place the lifetime in the range of $10^{-14} \geqslant \tau \geqslant 10^{-15}$ sec, a time scale approximately comparable to the period of the ultraviolet frequencies used in the studies of ionization.[1,2] In consideration of the outermost shells, an ultraviolet electric field strength E on the order of an atomic unit $E_0 = e/a_0^2$ is the regime in which the envisaged motion is expected to become an important factor in the dynamics. This corresponds to an electromagnetic intensity $I_0 \sim 7 \times 10^{16}$ W/cm^2.

The coherent oscillation of the electrons in outer atomic shells induced by irradiation at ultraviolet frequencies at intensities $I \gtrsim I_0$ has important consequences for the coupling of energy to atomic inner shells. Moreover, as described below, the influence of this type of atomic motion can be related to certain characteristics[3] of high-energy ($\gtrsim 10$ MeV/u) atom-atom collisions. At sufficiently high intensity, a relatively simple physical model can be envisaged which illustrates these effects. For simplicity, consider an intensity above $\sim 10^{19}$ W/cm^2, for which the peak electric field is more than ten times e/a_0^2, so that loosely bound outer electrons can be approximately modeled as completely free particles. Therefore, for those electrons, we can represent their motion as that of free electrons accelerating in intense coherent fields.[4,5] In this limiting case, we imagine that the atom is composed of two parts: (a) an outer shell of electrons driven in coherent oscillation by the radiative field, and (b) a remaining atomic core for which direct coupling to the radiation field is neglected. Coupling between these two systems can occur, since the outer electrons can, through inelastic "collisions," lead to the production of electronically excited core states. Indeed, since the outer electrons could acquire relativistic velocities at intensities on the order of 10^{21} W/cm^2, the production of electron-positron pairs by an *intra-atomic* process analogous to the well-known trident graph[6,7] becomes possible.

The role of coherence in the motion of the outer electrons in the excitation of the core is readily described by appeal to descriptions of energetic atom-atom (A/B) collisions. In this comparison, a correspondence is established between the scattering of the coherently radiatively driven outer electrons from the atomic core and the respective interaction of the electrons in the projectile atom A with the target atom B. Consider the process

$$A + B(0) \xrightarrow{\sigma_{n0}} A + B^*(n) \qquad (1)$$

in which A is a ground-state neutral atom with atomic number Z_A and $B^*(n)$ represents an electronically excited configuration of the target system with quantum numbers collectively represented by (n). In the plane-wave Born approximation, the cross section σ_{n0} can be written in the form presented by Briggs and Taulbjerg[3] as

$$\sigma_{n0}=(8\pi e^4/v^2)\int_{K_{\min}}^{K_{\max}}|\epsilon_{n0}^B(\mathbf{K})|^2[|Z_A-\sum_j\omega_j\langle\phi_j^A|\exp(i\mathbf{K}\cdot\mathbf{s}_A)|\phi_j^A\rangle|]^2 dK/K^3 \tag{2}$$

in which

$$\epsilon_{n0}^B(\mathbf{K})=\int dr_B^3\psi_{nB}^*(\mathbf{r}_B)\exp(i\mathbf{K}\cdot\mathbf{r}_B)\psi_{0B}(\mathbf{r}_B). \tag{3}$$

In expressions (2) and (3), v is the relative atom-atom velocity, the ϕ_j^A are orthonormal spin orbitals representing the electrons on the projectile atom, ω_j is the statistical weight of the shell, $\mathbf{K}$ is the momentum transfer in the collision, and ψ_{0B} and ψ_{nB} represent the electron wave functions of the target system. The summation over the index j appearing in Eq. (2) extends over all occupied orbitals so that in the limit $\mathbf{K}\rightarrow 0$, the summation tends to the number of electrons N_A associated with the projectile atom.[3] Since $N_A=Z_A$ for a neutral atom, complete screening[3] occurs in the low–momentum-transfer limit and the nuclear and electronic contributions cancel exactly. Therefore, in this limit, the amplitudes of the electrons combine coherently and the contribution to the cross section σ_{0n} arising from the motion of the *electrons* in atom A is increased by a factor of N_A^2 over that of a single electron at the same collision velocity v. Alternatively, for sufficiently low momentum transfer such that $Ka_0 << \hbar$, the electron cloud acts as a coherent scattering center with a mass $N_A m_e$, a charge $N_A e = Z_A e$, a velocity v, and a kinetic energy $N_A(\frac{1}{2}m_e v^2)$. Significantly, on account of the coherence, the single-particle energies $(\frac{1}{2}m_e v^2)$ add so that, in principle, this value could be *below* the magnitude required to produce the excitation of the target atom *B*.

In sufficiently high-field strengths, coherently accelerated electrons in outer atomic shells can interact with the remaining atomic core system in a manner closely analogous to the atom-atom scattering described above. If a plane-wave Born-approximation description is used, the cross section representing energy transfer can be derived directly from expression (2) with $Z_A=0$. We now describe an example illustrating the circumstances under which this may occur. Since the basic physical concepts can be very simply represented in the high-field limit $(E >> E_0)$, we consider a peak electric field strength $E\sim 0.5\times 10^{12}$ V/cm so that an electron acquires an energy of ~ 10 keV in a distance comparable to an atomic dimension (~ 2 Å). At this field strength, which corresponds to an intensity of $\sim 3\times 10^{20}$ W/cm^2, the electron accelerates to the 10-keV energy in a time which is approximately 1% of an optical cycle for an ultraviolet wave length of ~ 200 nm, a condition consistent with the validity of the assumption of a constant field strength for accelerations on the scale of atomic dimensions. The resulting velocity of $\sim 8\times 10^9$ cm/sec corresponds, for atom-atom collisions, to a collision energy of ~ 20 MeV/u. Therefore, the motion of these electrons simulates the *electronic* collisional environment that would occur in fast atom-atom encounters, but with the important *absence* of the nuclear contribution arising from the Z_A term in expression (2). In this case, no shielding is present in the low–momentum-transfer limit.

It is now possible to estimate the contribution to σ_{0n} for an inner-shell excitation arising from coherently excited atomic shells. For this we take expression (2) with $Z_A=0$ and restrict $K_{\max}$ to $\lesssim \hbar/a_0$, to fulfill the condition for full shielding which, for this situation, corresponds to totally *constructive* interference of the electron amplitudes. We further take Z_1 to denote the number of electrons in the outer shells and expand Eq. (2) for $\epsilon_{n0}^B(\mathbf{K})$ in the customary fashion so that only the leading dipole term x_{0n} is retained. Finally, for a core excitation energy ΔE we put $K_{\min}\simeq\Delta E/v$, the condition that holds for ΔE much less than the collision energy. With these modifications, the coherent piece σ_{0n}^c can be written as

$$\sigma_{0n}^c\simeq\frac{8\pi e^4 x_{0n}^2 Z_1^2}{v^2\hbar^2}\int_{\Delta E/v}^{\hbar/a_0}\frac{dK}{K}, \tag{4}$$

a result which, with the exception of the restriction on $K_{\max}$ and the Z_1^2 factor, is exactly the form of the well-known result for inelastic scattering of electrons on atoms developed by Bethe.[8] The final result, valid for

$$\alpha\left(\frac{v}{c}\right)\left(\frac{m_e c^2}{\Delta E}\right)>1, \tag{5}$$

is

$$\sigma_{0n}^c\simeq 8\pi\alpha^2\left(\frac{c}{v}\right)^2 Z_1^2 x_{0n}^2\ln\left[\alpha\left(\frac{v}{c}\right)\left(\frac{m_e c^2}{\Delta E}\right)\right], \tag{6}$$

in which α is the fine-structure constant. For the example considered, the restriction on the logarithmic factor limits ΔE to a maximum value of approximately 1 keV, an energy corresponding to the region near the M edge of xenon,[9] a case which serves as a suitable numerical example. If we take the charge radius[10] of the M shell of xenon as the scale for x_{0n}, then $x_{0n}\sim 0.2a_0$, and if we assume that $Z_1=18$, accounting for the three outermost xenon shells $(5p^6 5s^2 4d^{10})$, then the resulting cross section is on the order of $\sigma_{0n}^c\sim 7\times 10^{-18}$ cm^2 with the weakly varying logarithmic term taken as a factor of $O(1)$. This value is somewhat greater than the total photon cross section[11,12] in the region near the M edge of xenon. Furthermore, since expression (4) respects dipole selection rules, considerable state selectivity is present as only odd-parity excited core levels would be produced.

The upper limit in the integral in expression (4) can be extended to $K_{\max}=2Z_A m_e v$ if appropriate projectile-atom wave functions ϕ_j^A are used. This procedure produces a final cross section σ_{0n} with a magnitude of the same scale as that represented by Eq. (6), but with a somewhat different detailed dependence on v and ΔE. This refinement leaves the principal conclusion unchanged.

The coherent interaction described above can be viewed as a form of dynamic configuration interaction in which the fields of the participating electrons sum constructively. Constructive addition naturally results if the scale of the momentum transfer Δq communicated in the interaction is sufficiently small so that the length $\hbar/\Delta q$ is greater than the spatial scale of the scattering system. The physical origin of this effect is the same as that which generates the coherent forward scattering[13] observed in nuclear collisions.[14]

Obviously, all types of possible excited configurations cannot fully benefit from this type of coherent motion regardless of the field strengths used. For example, the coherence is unimportant in the amplitude for intra-atomic electron-positron pair production by the trident diagram shown in Fig. 1, since the momentum transfer Δq associated with the propagator for pair production in this interaction is such that

$$\hbar/\Delta q \sim \lambda\!\!\!^{-}_c << a_0. \tag{7}$$

Indeed, from Eq. (5), at sufficiently high intensity in the limit $v \rightarrow c$, the maximum value of $\Delta E_{\max}$ is given by

$$\Delta E_{\max} \sim \alpha m_e c^2 = 3.73 \text{ keV}. \tag{8}$$

Therefore, the cross section[6] for pair production in the field of a nucleus is easily shown to be

$$\sigma_t = (28/27\pi) Z_1 (Z\alpha)^2 r_0^2 (\ln\gamma)^3 \tag{9}$$

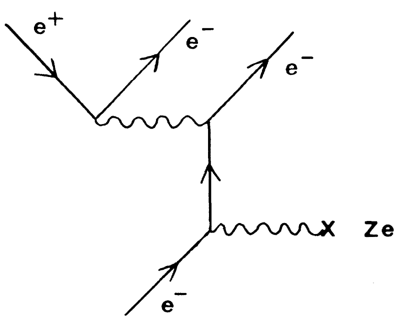

FIG. 1. Trident graph representing electron-positron pair production by collision on an energetic electron with a fixed center of charge Ze.

in which r_0 is the classical radius of the electron and γ is the customary relativistic factor

$$\gamma = [1-(v/c)^2]^{-1/2}. \tag{10}$$

At sufficiently high intensity ($\gtrsim 10^{21}$ W/cm^2), for $Z_1=50$ and $Z_2=90$, and with $\gamma \simeq 5$, $\sigma_t \simeq 2\times10^{-24}$ cm^2, a value that would, under reasonable experimental conditions with an ultraviolet laser of 1–10-J output and ~ 100-fs pulse length, make possible the generation of ~ 100 pairs/pulse by this mechanism.

Coherently driven motions in outer electron shells can generate an enhanced intra-atomic coupling for the excitation of inner shells. The interaction, which can be viewed alternatively as a form of configuration interaction or electron scattering, has, on account of the constructive addition of amplitudes, a cross section which scales as the square of the number (Z_1) of outer electrons participating in the motion. A strong and highly nonlinear coupling arises as a direct consequence. Coherent motions of this type should enable the selective excitation of atomic inner-shell states in the kiloelectronvolt energy range to be produced by intense irradiation of atoms at ultraviolet wave lengths. The physical nature of this process of intra-atomic energy transfer bears a direct relationship with energetic atom-atom collisions. Similar conclusions can be reached by alternative theoretical approaches, such as those involving the time dependent Hartree-Fock method.[15]

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