Physical Reality of Light-Induced Atomic States

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The physical reality of light-induced states in atoms has remained uncertain, ever since their discovery by Floquet theory. We now show that their existence is confirmed by time-dependent wave packet theory, and should manifest itself experimentally. By applying a realistic pulse to the atomic system, and calculating the energy spectrum of the ionized electrons, we find signals at the energies predicted by Floquet theory for the light-induced states, sometimes with towering intensity. Choosing the initial states such as to connect to the light-induced states via diabatic Floquet paths substantially enhances the yield. [S0031-9007(98)05747-0]

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The possibility that intense laser fields induce new states in atomic systems had been predicted some time ago for 1D models for both the high-frequency Floquet approximation [Bhatt *et al.* [1(a)]] and the full theory [Bardsley *et al.* [1(b)]]. However, it was the work of Potvliege, Shakeshaft, and collaborators on 3D models [2] that has clarified the circumstances of materialization of the light-induced Floquet states (LIS) from unphysical shadow states at energy thresholds $n\omega$ ($n =$ integer). Concerning physical systems, LIS were found for atomic hydrogen from full Floquet theory by Dörr *et al.* [3] and for H^- from the high-frequency approximation (including electron correlation) by Muller and Gavrila [4].

The class of Floquet solutions, to which the LIS belong, satisfies Gamow-Siegert boundary conditions [5]. Such solutions give an idealized description of the physics, as they have exponential asymptotic growth and, hence, do not belong to the quantum-mechanical Hilbert space. A number of conditions need to be satisfied for these solutions to represent physical situations [e.g., see [5(b),5(c)]]. Besides, they are calculated at constant field amplitude, and adiabaticity must be invoked in order to apply the results to laser pulses. With all of its limitations, Floquet theory has achieved impressive success over the years.

The physical significance of the LIS has remained, nevertheless, uncertain. This may be due to the fact that the general difficulties in interpretation of Floquet theory are now compounded with new ones, such as the evolution of the LIS cannot be followed from a field-free limit. Furthermore, they can manifest bizarre intensity dependence; for example, for the short-range potential models studied, the LIS can have transient existence, with materialization followed by disappearance, only to reappear after an intensity gap [2(c)]. Such behavior would be hard to accept as physical on the basis of Floquet theory only, unless firmly substantiated otherwise.

Yet, the physical reality of the LIS is an essential issue for the structure of the atom in the field, and the credibility of Floquet theory. The confirmation of the physical reality of the LIS can come only from the study of wave packets obeying the time-dependent Schrödinger equation, which is the proper framework for the application of quantum-mechanical laws, and directly reflects experiment. The simplest way of searching for them is to analyze the excess-photon ionization/above-threshold ionization (EPI/ATI) spectra of the ejected electrons, as the peaks in the spectra are located at multiples of ω from the energy level of origin. Whereas considerable information on LIS has been derived from Floquet theory, none has been extracted from the wave-packet approach. We show here, for the first time, that wave-packet evolution confirms the physical reality of the LIS, by detecting their signature in the EPI/ATI spectra of the ejected electrons.

Our analysis is carried out on a 1D atomic model. Such models have proven to be valuable tools for understanding and predicting intense laser-physics phenomena (e.g., the case of "dynamic" stabilization [6], or high-harmonic generation with ultrashort pulses [7]). The atomic potential we use has the "soft-core" Coulomb form:

$$V(x) = -[a^2 e^{-(x/a)^2} + x^2]^{-1/2}. \qquad (1)$$

It is finite at the origin, and its Coulomb tail supports a Rydberg series of states. We have chosen $a = 1.6$, such that the ground state energy is $W_0 = -0.500$ [8]; atomic units are used.

We consider Floquet solutions $\Psi^{(F)}$ of the time-dependent Schrödinger equation in the space-translated version [see [5(c)], Eq. (18)], which is unitarily equivalent to its more familiar forms in the velocity or length gauges. For monochromatic radiation, the electric field can be taken $\mathcal{E}(t) = \mathcal{E}_0 \sin \omega t$, where $\mathcal{E}_0$ is constant. Inserting in the Schrödinger equation

$$\Psi^{(F)}(x,t) = e^{-iEt} \sum_{n=-\infty}^{+\infty} \Phi_n(x) e^{-in\omega t} \qquad (2)$$

yields a system of coupled equations for $\Phi_n(x)$ [see [5(c)], Eqs. (28), (29)]. The natural parameters entering the

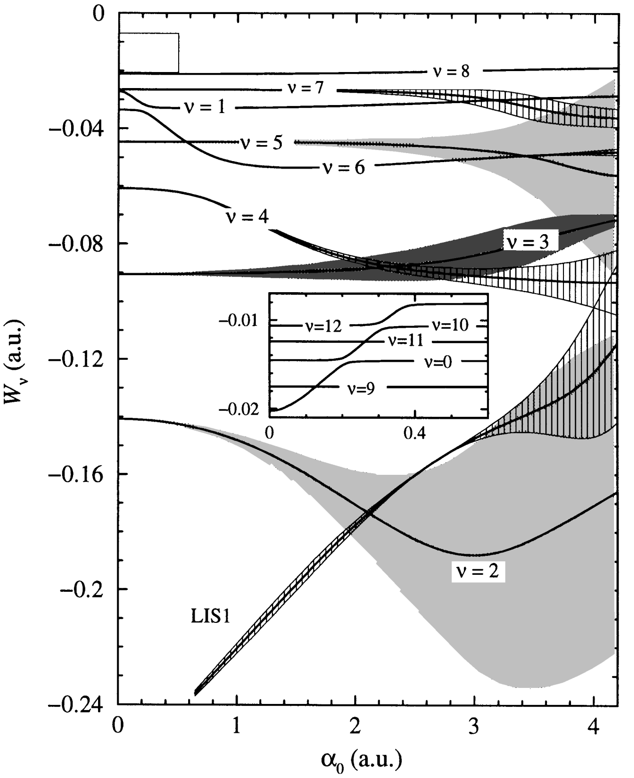

FIG. 1. Floquet quasienergies $E_\nu \equiv W_\nu - (i/2)\Gamma_\nu$, at $\omega = 0.24$ a.u.; Γ_ν is represented by the length of the vertical segment (shading) associated with W_ν. The states $\nu = 0$ and 9–12 are given in the inset.

theory are now ω and $\alpha_0 \equiv \mathcal{E}_0 \omega^{-2}$ (α_0 is the classical quiver amplitude of the electron in the field). We impose Gamow-Siegert boundary conditions on the $\Phi_n(x)$ [see [5(c)], Sect. III.A, or [9], Eqs. (6), (7)]:

$$\Phi_n(x) \to \begin{cases} f_n \exp(i\theta_n), & \text{for } x \to \infty, \\ (-1)^{P+n} f_n \exp(-i\theta_n), & \text{for } x \to -\infty, \end{cases} \tag{3}$$

where $\theta_n \equiv k_n x - (k_n)^{-1} \ln 2k_n x$, with $k_n = \pm[2(E + n\omega)]^{1/2}$, and P is the parity of the Floquet solution. This leads to a complex eigenvalue problem yielding the "quasienergies" $E \equiv W - i(\Gamma/2)$. If the signs of the square roots of k_n are chosen such that there are outgoing fluxes in the "open channels" [i.e., $(\mathrm{Re}\, E + n\omega) > 0$], but none in the "closed channels" [i.e., $(\mathrm{Re}\, E + n\omega) < 0$], the Floquet solution is physical. Otherwise, the Floquet solution is unphysical, and is called a "shadow" [2]. In the physical case, W is interpreted as the average energy (modulo ω) of the decaying state in the field, and Γ as its total ionization rate. By varying α_0 (or $\mathcal{E}_0$) continuously, at some critical value a physical solution can go over into a shadow; the reverse can also occur, which is the materialization of a LIS. We recall that, if a solution $\Psi^{(F)}$ is represented by the set $\{E, \Phi_n(x)\}$, it can be represented just as well by the family of sets $\{E' \equiv E + p\omega, \Phi'_n \equiv \Phi_{n+p}(x)\}$, where p is an integer [see Eq. (2)]. Nevertheless, observable results, such as the values of the EPI/ATI energies, are not affected by this ambiguity.

Further, to substantiate the existence of the LIS, we propagate solutions of the time-dependent Schrödinger equation in the length gauge, evolving from a field-free initial state (for $t < 0$), under the influence of a laser pulse $\mathcal{E}(t) = \mathcal{E}_0(t) \sin \omega t$, where $\mathcal{E}_0(t) = \mathcal{E}_0 \sin^2(\pi t/\tau)$, where τ is the length of the pulse. For $t > \tau$, the atom is again free, and the probability density for the EPI/ATI spectrum is given by

$$dP/dW = \textstyle\sum_\Pi |(u_W^{(\Pi)}, \Psi_L)|^2, \tag{4}$$

where Ψ_L is the length-gauge wave packet, and $u_W^{(\Pi)}$ is a continuum energy eigenfunction of the field-free Hamiltonian, with parity Π. As wave packets can be expanded in Floquet states [characterized in the present case by a time-dependent $\alpha_0(t)$] (e.g., see [10]), the calculated EPI/ATI spectrum has sharp peaks located at $(\mathrm{Re}\, E_\nu + n\omega)$, where E_ν are the discrete quasienergies of the states present. Several Floquet states are populated in general ("shakeup"), because the variation of the pulse is never fully adiabatic. The coefficients of these states evolve rather smoothly (for our pulse choice) until an avoided crossing of two $\mathrm{Re}\, E_\nu(\alpha_0(t))$ curves is reached, where the corresponding coefficients can have

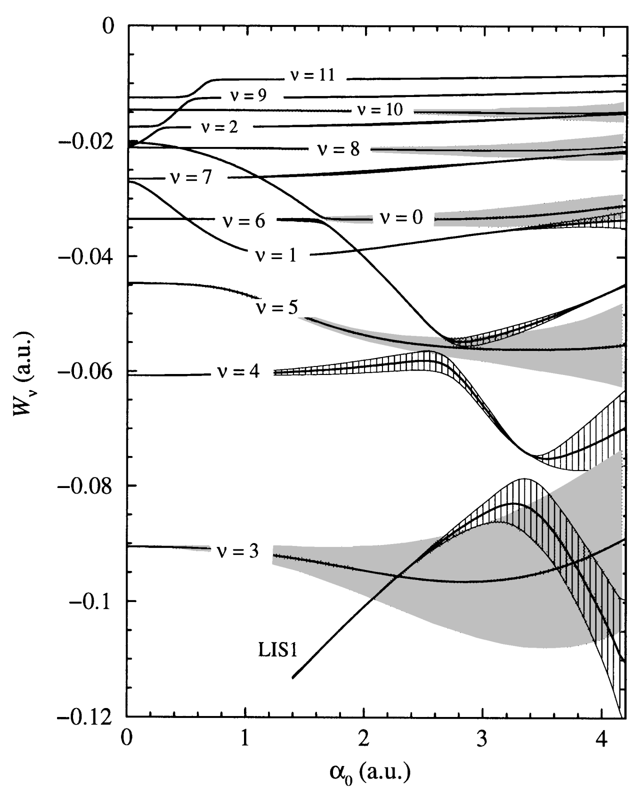

FIG. 2. Floquet quasienergies E_ν at $\omega = 0.12$ a.u. Γ_ν is represented as in Fig. 1.

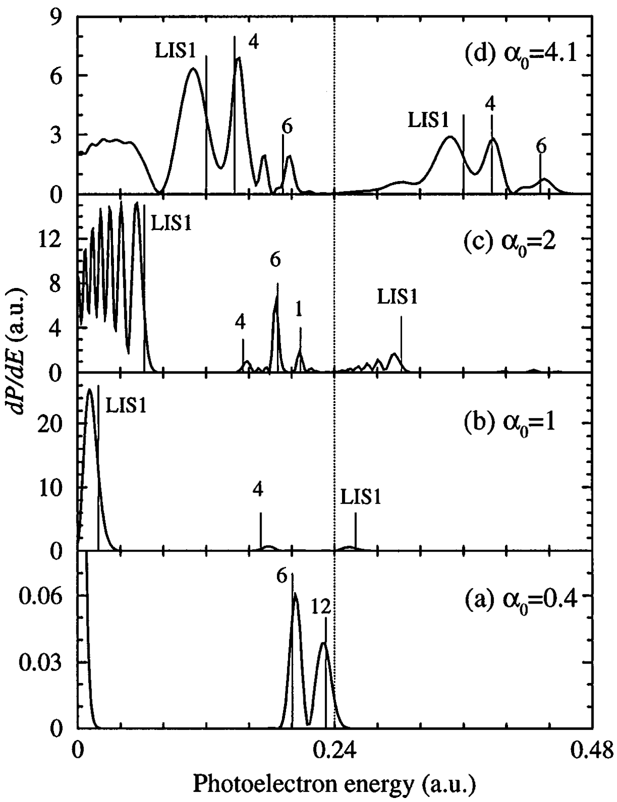

FIG. 3. EPI/ATI spectra at $\omega = 0.24$ a.u., with initial state $\nu = 0$, for the peak values of α_0 given. $\tau = 31$ cycles, except for (c), where $\tau = 61$ cycles. The markings give the energies of electrons originating from states ν labeled as in the corresponding Floquet diagram.

a rapid variation due to diabatic transitions; see the discussion by Potvliege and Shakeshaft [11]. Thus, during the pulse, the atomic population evolves mainly via Floquet states with $\mathrm{Re}\,E_\nu(\alpha_0(t))$ forming a (discontinuous) sequence of small-curvature segments ["diabatic paths" (DP)]. The energies and widths of the peaks appearing in the EPI/ATI spectrum will reflect these paths, with due weight given to the magnitude of $\Gamma(\alpha_0(t))$ along them.

The complex Floquet eigenvalue problem was solved by the method described in [9], extended here in the case of a Coulomb-tail potential, such as Eq. (1). The wave packets were propagated on a spatial grid by the method described in [10].

In Fig. 1, we present the quasienergy spectra for $\omega = 0.24$ (6.53 eV) and $\alpha_0 < 4.2$ ($\mathcal{E}_0 < 0.24$), and in Fig. 2, for $\omega = 0.12$ (3.26 eV) and $\alpha_0 < 4.2$ ($\mathcal{E}_0 < 0.06$). Using the Floquet ambiguity, all energies $W(\alpha_0) \equiv \mathrm{Re}\,E(\alpha_0)$ have been placed in the energy band $(-\omega, 0)$. Each of the spectra shown contains one LIS, denoted by LIS1 (there are more at higher α_0 values). Note that LIS1 in Fig. 2 has a transient existence.

LIS1 in Fig. 1 can be reached (modulo ω) by a DP starting from the ground state $\nu = 0$, and passing through the Rydberg states $\nu = 10$, 12, etc. (see inset of Fig. 1). It is remarkable that there are two DP leading to LIS1 in Fig. 2. One of them starts from $\nu = 0$, passes through $\nu = 6$, 4, and accesses the *descending* branch of the LIS1 curve. The other starts from $\nu = 2$ and passes through the Rydberg states $\nu = 9$, 11, etc., to access (modulo ω) the *ascending* branch of the LIS1 curve. Thus, the initial states considered should be best suited to access the LIS1 of Figs. 1 and 2 [12].

Figures 3–5 display the EPI/ATI spectra (for kinetic energy $<2\omega$ only), calculated with a pulse length $\tau = 31$ cycles, unless otherwise stated. Figure 3 corresponds to $\omega = 0.24$ and initial state $\nu = 0$. At $\alpha_0 = 0.4$, for which $W(\alpha_0)$ is located on the $\nu = 12$ segment of the DP identified in Fig. 1, we see, indeed, the signal from $\nu = 12$, along with the signature of $\nu = 6$, populated by shakeup. At $\alpha_0 = 1$, the signal from the just-materialized LIS1 dominates that from $\nu = 4$, populated by shakeup [13]. The spectrum at $\alpha_0 = 2$ (recorded with $\tau = 61$ cycles) shows interference fringes associated with the LIS signal; this interference effect was discussed (for regular states) by Bardsley *et al.* [1b], Reed and Burnett [14], Telnov and Chu [15]; [16]. The peaks $\nu = 6$, 4, 1 are present due to shakeup [13]. At $\alpha_0 = 4.1$, there is displacement of LIS1 and a distortion of its interference pattern. Figure 4, at $\omega = 0.12$, with $\nu = 2$ as the initial condition, illustrates the materialization of LIS1 (ascending branch) of Fig. 2.

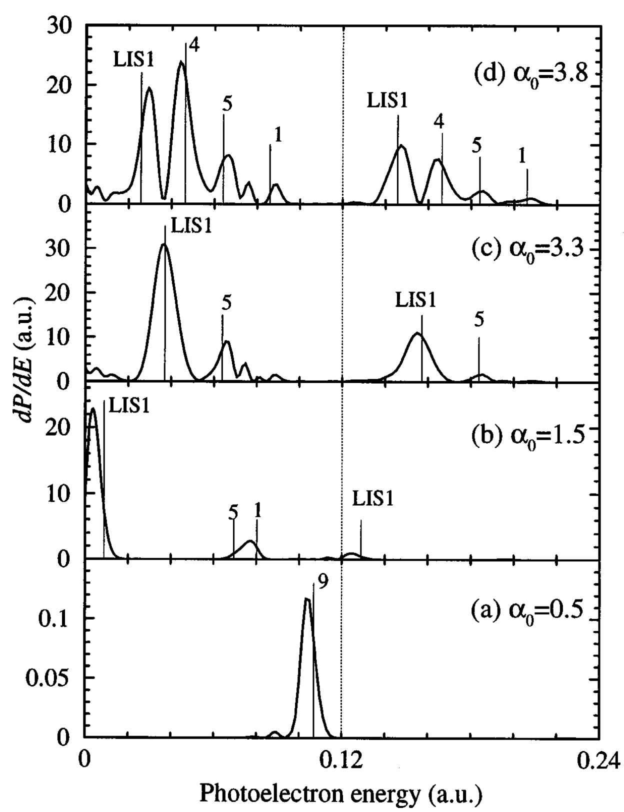

FIG. 4. EPI/ATI spectra at $\omega = 0.12$ a.u., initial state $\nu = 2$, $\tau = 31$ cycles.

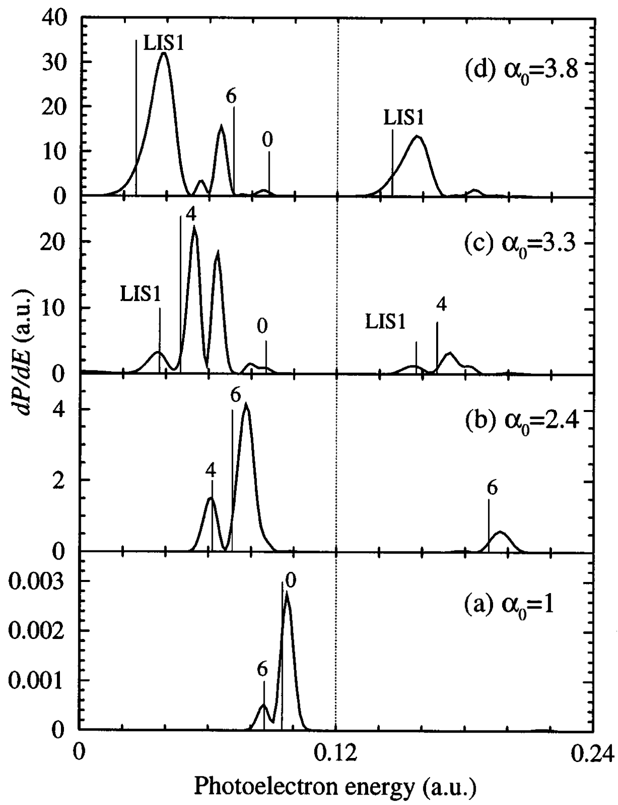

FIG. 5. EPI/ATI spectra at $\omega = 0.12$ a.u., initial state $\nu = 0$, $\tau = 31$ cycles.

Figure 5 displays the EPI/ATI spectrum for $\omega = 0.12$ and initial state $\nu = 0$. At $\alpha_0 = 1$, $W(\alpha_0)$ is on the $\nu = 0$ fragment of the DP; besides the signature of $\nu = 0$, there is a smaller signal from $\nu = 6$, excited by shakeup. At $\alpha_0 = 2.4$, $W(\alpha_0)$ is on the $\nu = 6$ segment of the DP; the large signature from $\nu = 6$ is accompanied by one of $\nu = 4$, excited by shakeup (note that $\nu = 4$ has a relatively large rate). At $\alpha_0 = 3.3$, we are at the diabatic passage from $\nu = 4$ to the descending branch of LIS1; see Fig. 2. The presence of LIS1 is clear, along with the signals from $\nu = 4$ (with interference fringes [16]) and $\nu = 0$. At $\alpha_0 = 3.8$, besides the dominant presence of LIS1, we have signals from $\nu = 6$ (shakeup from $\nu = 4$, with diabatic transfer to $\nu = 6$) and $\nu = 0$ (shakeup from $\nu = 6$, with diabatic transfer to $\nu = 0$).

To conclude, we have shown that the LIS have all the properties that can be ascribed to the other Floquet states, e.g., can be constituents of diabatic paths, and can give dominant, well resolved peaks in EPI/ATI spectra obtained with single-pulse laser excitation. As the pulse parameters considered (frequency, peak field strength, duration) are realistic, the LIS should manifest themselves experimentally, and, in fact, this may already have been the case [17].

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[1] (a) R. Bhatt, B. Piraux, and K. Burnett, Phys. Rev. A **37**, 98 (1988); (b) J. N. Bardsley *et al.*, J. Phys. B **21**, 3899 (1988); (c) J. N. Bardsley and M. J. Comella, Phys. Rev. A **39**, 2252 (1989).

[2] (a) R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 6190 (1988); see also (b) M. Dörr and R. M. Potvliege, Phys. Rev. A **41**, 1472 (1990); (c) A. S. Fearnside *et al.*, Phys. Rev. A **51**, 1471 (1995); (d) R. M. Potvliege and R. Shakeshaft, in *Atoms in Intense Laser Fields*, edited by M. Gavrila (Academic, Boston, 1992), Sect. III, p. 373.

[3] M. Dörr *et al.*, Phys. Rev. A **43**, 3729 (1991).

[4] H. G. Muller and M. Gavrila, Phys. Rev. Lett. **71**, 1693 (1993).

[5] For general presentations of the strong-field Floquet method, see (a) Shih-I Chu, Adv. At. Mol. Phys. **21**, 197 (1985); Adv. Chem. Phys. **73**, 739 (1989); (b) R. Shakeshaft, J. Opt. Soc. Am. B **4**, 705 (1987); (c) M. Gavrila, in *Atoms in Intense Laser Fields* [Ref. [2(d)]], p. 435.

[6] Q. Su, J. H. Eberly, and J. Javanainen, Phys. Rev. Lett. **64**, 862 (1990); J. H. Eberly, R. Grobe, C. K. Law, and Q. Su, in *Atoms in Intense Laser Fields* [Ref. [2(d)]], p. 301.

[7] J. P. Christov *et al.*, Phys. Rev. Lett. **77**, 1743 (1996).

[8] The first energy levels are $W_0 = -0.500$, $W_1 = -0.267$, $W_2 = -0.141$, and $W_3 = -0.0906$.

[9] M. Marinescu and M. Gavrila, Phys. Rev. A **53**, 2513 (1996).

[10] For a discussion of this point and references, see J. C. Wells, I. Simbotin, and M. Gavrila, Laser Phys. **7**, 525 (1997).

[11] R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 4597 (1988); **40**, 3061 (1989).

[12] It is not clear if, for a Coulomb-tail potential, there are LIS that cannot be reached by a DP. This is certainly true for short range potentials with a finite number of field-free bound states.

[13] The shakeup may actually occur to $\nu = 1$, followed by diabatic transfer to $\nu = 6$ and $\nu = 4$.

[14] V. C. Reed and K. Burnett, Phys. Rev. A **43**, 6217 (1991).

[15] D. A. Telnov and Shih-I Chu, J. Phys. B **28**, 2409 (1995).

[16] This is the interference of the electron de Broglie waves of given kinetic energy, emitted during the turn-on, with those of the same energy, emitted during the turn-off. When the binding energy $|W(\alpha_0)|$ increases (decreases) with α_0, the interference peaks extend to the right (left) of the Floquet energy.

[17] In the experiments of H. Rottke *et al.*, J. Phys. B **26**, L15 (1993); G. G. Paulus *et al.*, J. Phys. B **29**, L249 (1996), series of EPI peaks were observed that could not be ascribed to known atomic levels, and could originate from LIS; see also [2d], Sect. VIII.