Resonant above-threshold ionization peaks at quantized intensities

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We suggest that electron-laser interactions can give rise to resonance phenomena as the intensity varies. A new QED perturbation theory is developed, in which the coupling between an electron and the second quantized laser mode is treated nonperturbatively. We predict, for example, the above-threshold ionization rate shows peaks at intensities with integer ponderomotive parameter. Such quantum resonance effects may be exploited to calibrate laser intensities in appropriate range.

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\textbf{Introduction} For a radiation field, the correspondence principle limit requires a large number of photons in unit volume. In accordance to this limit, as is widely accepted, the light field in a high-intensity laser beam can be treated classically or semi-classically. Indeed many aspects of the stimulated electron-laser interactions have been derived in this way [1]. However, according to quantum electrodynamics (QED), the fundamental theory for electron-photon interactions, even in this limit the radiation field still has particle-like structure in terms of photons, and stimulated emission and absorption give rise to strong fluctuations in photon number at high light intensities. The fluctuations are extremely tiny compared to the total photon number, thus are ignored in the classical treatments. But their absolute magnitude become appreciable at available high intensities and may give rise to novel, observable quantum effects.

A physical quantity that can be used to measure the strength of stimulated electron-laser-photon interactions is the dimensionless ponderomotive parameter:

$$z \equiv U_p / \hbar \omega_0 = 2 \pi e^2 I / m_e \hbar \omega_0^3,$$

where \(I\) is the intensity, and \(\hbar \omega_0\) the laser photon energy. It is essentially the ponderomotive energy \(U_p\) for an electron in the light field in units of the photon energy. Classically, \(U_p\) is the time-averaged kinetic energy of the wiggling motion of the electron in response to the radiation field. In quantum theory, Eberly first showed [2] the emergence of the parameter \(z\) in stimulated Compton scattering. He interpreted it as the number of photons in the possibly smallest interaction volume for the electron, and argued that when \(z\) becomes large, many photons at a time are interacting with the electron, and stimulated multi-photon coherent emissions and absorptions become frequent and significant. We will argue that it is more natural to interpret \(z\) as the average number of laser-mode photons in the photon cloud dressing the electron, whose formation is due to stimulated interactions. Thus, when the ponderomotive parameter \(z\) is close to an integer, the dressing photon cloud becomes resonant with the laser mode, which has a discrete energy spectrum. In view of this argument, we suggest that various processes in the electron-laser system should show resonance-like structures in their intensity dependence, as the ponderomotive parameter \(z\) becomes close to an integer.

Previously, Guo, Åberg and Crasemann [3] have given a nonperturbative approach based on almost exact eigenstates for the system of an electron coupled to a second quantized laser mode. It was then used to derive the above-threshold ionization (ATI) rate, leading to nonvanishing results only at \(z = \text{integer}\). This was a sort of precursor of the resonance effects, but it could not produce nonzero ATI rate at \(z \neq \text{integer}\). To remedy, in this paper we develop a new theory for QED at high laser photon density, by including non-laser radiation modes and treat their couplings to the electron as perturbation. In this way we are able to demonstrate the resonant ATI peaks at \(z = \text{integer}\) superposed on a smooth background at non-integer \(z\). This and similar intensity-dependent, resonating quantum effects of the light field in other multiphoton processes may be experimentally exploited to calibrate appropriately high laser intensities.

\textbf{New Perturbation Theory for QED} To properly deal with photon number fluctuations, we need to quantize the electromagnetic field, but still treat the electron quantum mechanically, ignoring pair production, vacuum polarization and other relativistic corrections for the electron as well, if the laser intensity is not too high.

In the Schrödinger picture, the Hamiltonian of the electron-radiation system is (with \(\hbar = c = 1\))

$$H = \frac{1}{2m_e} [-i \nabla - e \mathbf{A}(\mathbf{r})]^2 + \sum_k \omega_k N_k,$$

with \(N_k = a_k^\dagger a_k + 1/2\). Here the photon field operator is given by the time-independent vector potential in the radiation gauge (\(\nabla \cdot \mathbf{A} = 0\)):

$$\mathbf{A}(\mathbf{r}) = \sum_k \mathbf{A}_k(\mathbf{r}) = \sum_k g_k (e_k a_k e^{i \mathbf{k} \cdot \mathbf{r}} + \text{h.c.}),$$

with \(k\) labeling the photon modes, including the wave vector \(\mathbf{k}\) and transverse polarizations described by \(e\):
Here \( g_k = (2\omega_k V_\nu)^{-1/2} \), with \( \omega_k = |k| \) and \( V_\nu \) the normalization volume of the radiation field. \( a_k \) and \( a_k^\dagger \) are photon annihilation and creation operators.

Now let us separate the laser modes, say a single mode labeled by \( k_0 \), from other photon modes: \( A = A_{k_0} + A' \), and try to first treat the electron-laser-mode interactions nonperturbatively, then add the coupling of the electron to non-laser modes as perturbation. Thus, we are led to split \( H = H_0 + V + V' \), with

\[
H_0 = \frac{(-i\nabla)^2}{2m_e} + \omega_0 N_0 + \sum_{k' \neq k_0} \omega' N',
\]

\[
V = -\frac{e}{m_e} A_{k_0}(r) \cdot (-i\nabla) + \frac{e^2 A_{k_0}(r)^2}{2m_e},
\]

\[
V' = -\frac{e}{m_e} A'(r) \cdot (-i\nabla) + \frac{e^2 A_{k_0}(r) \cdot A'(r)}{m_e},
\]

with the \( A'^2 \) term neglected.

For an electron in the laser field, we choose \( H_0 + V \) as the unperturbed Hamiltonian. The eigenstates of \( N' \) are simply the Fock states for the non-laser mode. For the electron-laser-mode subsystem, almost exact eigenstates has been obtained before [1], [3], which are labeled by a momentum \( p \) and an integer \( n \), denoted as \( \Psi_{p n} \). They are the nonrelativistic limit of the exact solutions [3] to the Dirac equation coupled to the quantized laser-mode. They form a complete, orthogonal set of states, called quantized field Volkov states (QFVS), which are the QED analog of the classical Volkov states [3]. Their non-relativistic limit is verified [3] to satisfy the Schrödinger equation up to errors of the same order of magnitude as relativistic corrections. In practice, we need only to consider their large photon-number limit, \( n \to \infty \), \( g_{k_0} \to 0 \) and \( \sqrt{n}g_{k_0} \to \Lambda \), with the QFVS simplified to

\[
\Psi_{p n} = V^{-1/2} \sum_{j \geq -n} \exp\{i(p + (z - j)k_0) \cdot r\} J_j(\eta, \zeta_p, \phi_p)^* \exp\{-ij\phi_p\} \quad n + j > .
\]

Here \( z \equiv e^2\Lambda^2/m_e\omega_0 \) is the ponderomotive parameter, \( \{n\} \) a laser-mode Fock state and

\[
\eta = \frac{1}{2} e \cos \xi, \quad \zeta_p = \frac{2|\epsilon|\Lambda}{m_e\omega_0} |p| \cdot \epsilon, \quad \phi_p = \tan^{-1}(p_y/p_x \tan \xi/2).
\]

The \( J_j \) is compounded of Bessel functions \( J_m \):

\[
J_j(\eta, \zeta_p, \phi_p) = \sum_{m=-\infty}^{\infty} J_m(\eta) J_{j-2m}(\zeta_p) e^{2im\phi_p},
\]

The energy and momentum \( \{P_0 = -i\nabla + N_{k_0}k_0\} \) eigenvalues of the QFVS are given by, respectively,

\[
E_0(p, n) = p^2/2m_e + (n + 1/2)\omega_0 + z\omega_0,
\]

\[
P_0(p, n) = p + (n + 1/2)k_0 + zk_0.
\]

The QFVS is a coherent superposition of Fock states in the laser mode with different photon number; this implies that the electron in the laser field is dressed by a coherent photon cloud which has a component in each Fock state with photon surplus (or deficit) \( j \), generated by stimulated emission and absorption. We interpret both the total energy and momentum in eq. (8) as consisting of contributions from the electron, the background photons, and the photon cloud, each being on shell. Therefore, it is natural to interpret the contributions from the photon cloud as the ponderomotive energy and momentum [3],

\[
U_p = z\omega_0, \quad P_p = zk_0,
\]

and identify the average number of laser photons in the dressing cloud with the ponderomotive parameter \( z \) defined in eq. (9). We emphasize that this interpretation of the ponderomotive parameter is the distinctive consequence of the exact QFVS solutions, not shared by any other existing perturbative approaches.

By using the QFVS as unperturbed states, we can develop a new perturbation theory for the electron-radiation system, in which the electron-non-laser-mode coupling \( V' \) is treated as perturbation. Then the eigenstate for an electron in the laser field is the perturbed QFVS, \( \Psi_{p n', \epsilon} = \{p n', \epsilon\} + \{p n', \epsilon\}' \), with

\[
|p n', \epsilon\}' = \sum_{\tilde{p}, \tilde{\eta}, \tilde{\xi}} \langle \tilde{p} \tilde{n}, \tilde{\eta} | V' | p n', \epsilon\rangle \frac{E(\tilde{p} \tilde{n}, \tilde{\eta})}{E(\tilde{p} \tilde{n}, \tilde{\eta}) - E(p n', \epsilon)}.
\]

where \( |p n', \epsilon\}' = \Psi_{p n', \epsilon}^0 |p n', \epsilon\} \), with \( |p n', \epsilon\} \) a Fock state in a non-laser mode: \( E(\tilde{p} \tilde{n}, \tilde{\eta}) = E_0(\tilde{p}, \tilde{n}) + (n' + 1/2)\omega_0 \). Note that there is no energy shift up to first order.

**Calculation of the ATI Rate** For definiteness, let us consider the above-threshold ionization (ATI): A beam of neutral atoms with tightly bound electrons is injected into a monochromatic, elliptically polarized single-mode laser beam. Even if the photon energy is much less than the ionization energy, a bound electron can absorb simultaneously quite a number of, say ten to twenty, photons to become ionized with appreciable kinetic energy. Of course, before the ionized electron gets out of the laser beam, it has very strong stimulated interactions with the laser mode. During the ATI process, to balance the total energy and momentum, the electron may emit a photon not in the laser mode (spontaneous emission). We want to calculate the rate and angular distributions etc., and study their intensity dependence.

Let us start with the following initial state for the electron-radiation system: the electron in a bound state \( \Phi_i \), the laser mode in the Fock state \(|n_i\rangle \), and the non-laser modes in the vacuum state (with \( n'_i = 0 \) photons), denoted by \( |\Phi_i, n_i, 0\rangle \). In the final state of the ATI, denoted...
as $| P_f, n_f, n'_f \rangle$, the electron is in a free state with momentum $P_f$ outside the laser beam, the laser mode in the state $| n_f \rangle$ and at most one, say $k'$, of the non-laser modes in $| n'_f = 1 \rangle$ (to first order). Physically the ATI can be viewed as a two-step process $\{[3]:1\}$ The electron is first ionized into the laser field, so the intermediate state of the system is described by the perturbed QFVS given by eqs. $\{[3]:2\}$. The electron exits out of the laser beam and becomes a free electron.

With this physical picture in mind, we apply the standard formal theory for scattering $\{[3]:3\}$ and its adaption to the present situation $\{[3]:4\}$, up to the first order in perturbation theory, resulting in the transition amplitude

$$T_{fi} = \sum_{p,n,n'} \langle P_f, n_f, n'_f | \Psi_{pn,n'} \rangle \langle \Psi_{pn,n'} | V + V' | \Phi_i, n_i, 0 \rangle,$$

where the summation of intermediate states is subject to

$$E(p,n') = E_i = -E_b + (n_i + 1/2)ω_0 + 1/2ω',$$

$$= E_f = P_f^2/2m_e + (n_f + 1/2)ω_0 + (n'_f + 1/2)ω',$$

with $E_b$ the binding energy in the initial state $\Phi_i$, while both $n'$ and $n'_f$ are either 0 or 1, up to first order.

Inspection shows only the following terms are nonzero:

$$T_0 = \sum_{p,n} \langle P_f, n_f, 0 | p, n \rangle \langle p, n | 0 \rangle \langle 0 | V | \Phi_i, n_i, 0 \rangle,$$

$$T_1 = \sum_{p,n} \langle P_f, n_f, 1 | p, n \rangle \langle p, n | 1 \rangle \langle 1 | V | \Phi_i, n_i, 0 \rangle,$$

$$T_2 = \sum_{p,n} \langle P_f, n_f, 1 | p, n \rangle \langle p, n | 1 \rangle \langle 1 | V | \Phi_i, n_i, 0 \rangle,$$

$$T_3 = \sum_{p,n} \langle P_f, n_f, 1 | p, n \rangle \langle p, n | 0 \rangle \langle 0 | V | \Phi_i, n_i, 0 \rangle.$$

The zeroth order term $T_0$ has been calculated before $\{[3]:5\}$. $T_1$ and $T_2$, as well as $T_0$, contribute only at $z = \text{integer}$, while $T_3$ contributes both at $z = \text{integer}$ and $z \neq \text{integer}$. We are interested in $z \neq \text{integer}$, so we focus on $T_3$.

After a lengthy calculation, introducing $j = n_i - n$, $j' = n_f - n$ and $q = n_i - n_f$, we finally obtain

$$T_3 = \frac{e^2\omega_0}{m_e} V e^{-1/2} \Phi_i | P_f - qk + k' \rangle e^{ij\theta/2} \frac{1}{\omega_0} \sum_{j',j''} \delta_{z-j} \delta_{z-j'} \delta_{z-j''}$$

$$\mathcal{J}_{j,j'}(\zeta, \eta, \phi P_f) e^{-ij\phi P_f} \mathcal{J}_{j'}(\zeta, \eta, \phi P_f + k') e^{ij\phi P_f + k'}$$

$$- e^{ij\phi P_f} \mathcal{J}_{j}(\zeta, \eta, \phi P_f + k') e^{ij\phi P_f + k'}$$

$$\frac{e}{\omega_0} \sum_{j',j''} \delta_{z-j} \delta_{z-j'} \delta_{z-j''}$$

$$+ e\Lambda e^* \mathcal{J}_{j} e^{ij\phi P_f} \mathcal{J}_{j'}(\zeta, \eta, \phi P_f + k') e^{ij\phi P_f + k'}$$

$$e^* \mathcal{J}_{j}(\zeta, \eta, \phi P_f) e^{ij\phi P_f + k'} + e\Lambda e^* \mathcal{J}_{j} e^{ij\phi P_f} \mathcal{J}_{j'}(\zeta, \eta, \phi P_f + k') e^{ij\phi P_f + k'}.$$

A careful study shows that the kinetic energy difference for the photoelectron before and after exiting out of the light field is of the order of relativistic corrections. Thus, energy conservation implies a discrete spectrum for both the photoelectron and the non-laser photon:

$$\omega' \approx [z - (j - q)]\omega_0,$$

$$P_f^2/2m_e \approx j\omega_0 - E_b - z\omega_0 \geq 0.$$

The physical interpretation is clear: the electron is ionized by absorbing $j$ photons simultaneously and, upon exiting out of the laser field, completely shakes off its ponderomotive energy (or the dressing photon cloud), by emitting $j - q$ laser photons and a non-laser photon with the remaining ponderomotive energy.

We express the energy delta function $\delta(E_f - E_i)$ as

$$\left(\frac{m_e}{2\omega_0}\right)^{1/2} \frac{e^2\omega_0}{(2\pi)^3} \left(\frac{m_e}{2\omega_0}\right)^{1/2} (q - \epsilon_b - \nu)^{1/2},$$

where $\epsilon_b \equiv E_b/\omega_0$ and $\nu \equiv \omega'/\omega_0$ and $P_f \equiv | P_f |$. Then the total ATI rate is given by

$$W = \int_{\epsilon_b} \frac{dW}{d\epsilon_b d\nu} | T_3 |^2 2\pi \delta(E_i - E_f) d^3P_f d^3k',$$

where angular distribution is given by

$$\frac{dW}{d\epsilon_b d\nu} = e^2\omega_0 (2\pi)^3 \left(\frac{m_e}{2\omega_0}\right)^{1/2} \left(\frac{2m_e\omega_0^3}{(2\pi)^3}\right)^{1/2} (q - \epsilon_b - \nu)^{1/2},$$

$$\mathcal{J}_{j}(\zeta, \eta, \phi P_f) e^{ij\phi P_f} \mathcal{J}_{j'}(\zeta, \eta, \phi P_f + k') e^{ij\phi P_f + k'}.$$

Resonant ATI Peaks From eq. $\{[3]:13\}$ one can infer that the amplitude $T_3$ becomes very large, if $z$ is sufficiently close to an integer, because then one of the terms in the sum can have a very small denominator $j' - z$. Thus, we predict that there are resonant ATI peaks at quantized intensities with $z = N$ or

$$I = NI_0 \equiv \frac{N\hbar m_e \omega_0^3}{2\pi e^2},$$

with $N \geq 1$ an integer. Note that $I_0$ is proportional to the cube of the laser frequency $\omega_0$.

As an example, in Fig. 1 we present the numerically calculated total photoelectron counts, collected in the direction of polarization, for ATI of xenon in a single-mode linearly polarized laser beam with wavelength 1064 nm. It indeed confirms the emergence of resonance peaks at quantized intensities, with $I_0 = 1.10 \times 10^{13}$ W-cm$^{-2}$. Note the smooth background away from the resonant peaks, grossly dictated by the classical description of the laser field. Because the widths of the resonance peaks in Fig. 1 are rather narrow, for the ATI measurements to test our theory the laser intensity should be very stable and adjustable almost continuously.

To compare, we recall that the Keldysh-Faisal-Reiss theory $\{[3]:15\}$ gives the ATI amplitude

$$T_{fi}^{\text{KFR}} = \langle P_f | V | \Phi_i \rangle,$$

(21)
the order of calibrating the intensity of laser beams in the range with the light field, if verified, would provide means for near an integer.

In the above, as in the usual treatments, we have ignored the effects of the ionic Coulomb interactions, since the Coulomb matrix elements are of the order $e^6$ while those of $V'$ are of the order $e$. We have also used the Fock states as the basis for the laser mode. If one uses Glauber coherent states to describe the initial and final states of the laser field, the ATI amplitude can be easily derived by superposing our amplitudes. This gives rise to a spread in photon number $n_i$ and $n_j$. But the corresponding spread in $z$ is expected to be very small. So our prediction of the resonant ATI peaks is unaffected.

Our ATI rate diverges at exactly integral $z$. This problem is easy to remedy, by including in eq. (10) an imaginary part (a finite width) for the QFVS energy $E(p, 0)$, due to its ability to decay through spontaneous emission via the coupling $V'$ to non-laser modes.

**Other Intensity-Dependent Quantum Effects**

Our argument for the resonance effects in the electron-laser system is very general, based only on the intensity dependent stimulated interactions and the discrete photon structure of the laser mode. So we expect to see them in other multiphoton processes, and our approach to QED at high laser photon density is applicable as well.

One example is a slow electron transversing a single-mode laser beam. Classically, the ponderomotive energy acts like an effective repulsive potential, so at high intensities the electron can hardly get into the laser beam. But according to our argument, the stimulated electron-laser interactions will give rise to a photon cloud dressing the electron, which can be resonant with the laser mode. So we predict that when the laser intensity is close to the quantized values $N\lambda$, there will be resonance peaks for the penetration probability for slow electrons transversing the laser beam. Our new perturbation theory is applicable to make quantitative predictions.

It is easy to generalize our approach to more than one laser modes, since the corresponding QFVS have been obtained before [10]. For example, one may consider electrons scattered by a standing wave formed by two laser modes. Previously, Bucksbaum *et al.* [1] have experimentally discovered a dramatic peak splitting in the angular distribution of the scattered electron. This has been theoretically explained in ref. [2] using the QFVS states, which could not deal with the angular region inside the splitting angle. Our new perturbation theory can be employed to deal with the angular region in between the peaks, and is expected to reveal a characteristic variation in the peak separation as $I/I_0$ changes continuously near an integer.

These and similar intensity-dependent quantum effects of the light field, if verified, would provide means for calibrating the intensity of laser beams in the range with the order of $I/I_0$ from unity to at least few tens.

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FIG. 1. Total ATI photoelectron count, collected in the laser polarization direction, vs. laser intensity for xenon in a single-mode, linearly polarized laser at 1064 nm.
