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Photon-statistics force in ultrafast electron dynamics

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This supplementary information file contains additional detail about the derivations and numerical calculations presented in the main text. In section I we solve the time-dependent Schrodinger equation (TDSE) for an electron interacting with a quantum state of light, to obtain the density matrix of the electron (Eq. (3) in the main text). In section II, we obtain a formula for the electric dipole of an atom driven by a quantum state of light, derived under the SFA (Eq. (6) in the main text). In section III, we derive the $P(\alpha)$ and $P(E)$ distributions for squeezed coherent light (Eq. (7.a) in the main text). In section IV we derive the coupled algebraic equations for the electronic trajectories, in case the driver is squeezed coherent. In section V we show that within the qSFA theory, squeezing is formally equivalent to an effective photon statistics force, presented in the main text (Eq. (10)). In section VI we derive Eq. (11) in the main text.

I. Solution of the time-dependent Schrodinger equation in a quantum-optical setting

Here we develop a general theory of the interaction between strong non-perturbative quantum light and an initially bound electron. To do this we write general Schrodinger equation for the joint density matrix of light and electron:

$$i\hbar\frac{\partial\rho}{\partial t}=H\rho-\rho H \tag{I.1}$$

where the Hamiltonian is:

$$H=H_{\mathrm{A}}+H_{\mathrm{F}}+\boldsymbol{d}\cdot\widehat{\boldsymbol{E}} \tag{I.2}$$

and $H_{\mathrm{F}}=\sum_{k\sigma}\hbar\omega\, a_{k\sigma}^{\dagger}a_{k\sigma}$. We aim to solve this equation to describe the emission of an electron in an external electromagnetic field $\boldsymbol{A}$ and static potential U. We employ the interaction picture: $\rho\to e^{-\frac{iH_F}{\hbar}t}\rho e^{\frac{iH_F}{\hbar}t}, H\to e^{-\frac{iH_F}{\hbar}t}He^{\frac{iH_F}{\hbar}t}$, to obtain the following Schrodinger equation:

$$i\hbar\frac{\partial\rho}{\partial t}=[H_0,\rho] \tag{I.3}$$

where $H_0=H_{\mathrm{A}}+\boldsymbol{d}\cdot\boldsymbol{E}$ and $\boldsymbol{E}(t)=i\sum_{\boldsymbol{k},\sigma}\sqrt{\frac{\hbar\omega}{2V\varepsilon_0}}\boldsymbol{\varepsilon}\left(a_{k\sigma}e^{-i\omega t}-a_{k\sigma}^{\dagger}e^{i\omega t}\right)$. We consider the following initial conditions for the density matrix:

$$\begin{cases}\rho(0)=\rho_A(0)\otimes\rho_F(0)\\ \rho_A(0)=|\psi_0\rangle\langle\psi_0|\\ \rho_F(0)=\int d^2\alpha d^2\beta\cdot P(\alpha,\beta^*)\dfrac{|\alpha\rangle\langle\beta|}{\langle\beta|\alpha\rangle}\end{cases} \tag{I.4}$$

Using the linearity of density matrix equation, we write:

$$\rho(t)\equiv\int d^2\alpha d^2\beta\cdot P(\alpha,\beta^*)\,\rho_{\alpha\beta}(t) \tag{I.5}$$

$$i\hbar\frac{\partial\rho_{\alpha\beta}(t)}{\partial t}=\left[H_0,\rho_{\alpha\beta}(t)\right]$$

$$\rho_{\alpha\beta}(0)=|\psi_0\rangle\langle\psi_0|\otimes\frac{|\alpha\rangle\langle\beta|}{\langle\beta|\alpha\rangle}$$

We now employ coherent shift operators with coherent parameters α and β (denoted by $D(\alpha)$ and $D(\beta)$) to define

$$\tilde{\rho}_{\alpha\beta} = D^{\dagger}(\alpha)\rho_{\alpha\beta}D(\beta) \tag{I.6}$$

$$\tilde{\rho}_{\alpha\beta}(0) = |\psi_0\rangle\langle\psi_0| \otimes \frac{|0\rangle\langle 0|}{\langle\beta|\alpha\rangle}$$

$$i\hbar\frac{\partial\tilde{\rho}_{\alpha\beta}}{\partial t} = D^{\dagger}(\alpha)H_0D(\alpha)\tilde{\rho}_{\alpha\beta} - \tilde{\rho}_{\alpha\beta}D^{\dagger}(\beta)H_0D(\beta),$$

Using $D^{\dagger}\hat{a}D = \alpha + \hat{a}$ (a property of coherent shift operators), we define:

$$i\hbar\frac{\partial\tilde{\rho}_{\alpha\beta}}{\partial t} = H_{\alpha}\tilde{\rho}_{\alpha\beta} - \tilde{\rho}_{\alpha\beta}H_{\beta} + \left[\boldsymbol{d}\cdot\widehat{\boldsymbol{E}}, \tilde{\rho}_{\alpha\beta}\right]$$

$$H_{\alpha} = H_{\mathrm{A}} + \boldsymbol{d}\cdot\boldsymbol{E}_{\boldsymbol{\alpha}}(t) \tag{I.7}$$

$$H_{\beta} = H_{\mathrm{A}} + \boldsymbol{d}\cdot\boldsymbol{E}_{\boldsymbol{\beta}}(t)$$

where $\boldsymbol{E}_{\alpha}(t) = \langle\alpha|\widehat{\boldsymbol{E}}|\alpha\rangle$, $\boldsymbol{E}_{\beta}(t) = \langle\beta|\widehat{\boldsymbol{E}}|\beta\rangle$ and H_A is the field free atomic Hamiltonian. Assuming the density matrices of light and matter are separable throughout the process (i.e., light and matter are not entangled):

$$\tilde{\rho}_{\alpha\beta}(t) = |\phi_{\alpha}(t)\rangle\langle\phi_{\beta}(t)| \otimes \tilde{\rho}_{\mathrm{light}}(t) \tag{I.8}$$

Here, $|\phi_{\alpha}(t)\rangle$ and $|\phi_{\beta}(t)\rangle$ are solutions of the semi-classical Schrodinger's equation:

$$i\hbar\frac{\partial}{\partial t}|\phi_a\rangle = H_{\alpha}|\phi_a\rangle \tag{I.9}$$

Plugging equations (I.8) and (I.9) the above definitions into equation (I.7):

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}\left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right) \\ &= H_\alpha\left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right) \\ &- \left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right)H_\beta \\ &+ \left[\boldsymbol{d}\cdot\widehat{\boldsymbol{E}},\left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right)\right] \end{aligned} \tag{I.10}$$

$$\begin{aligned} |\phi_\alpha(t)\rangle\langle\phi_\beta(t)|i\hbar\frac{\partial}{\partial t}\left(\tilde{\rho}_{\text{light}}(t)\right) + H_\alpha|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t) \\ &- |\phi_\alpha(t)\rangle\langle\phi_\beta(t)|H_\beta\otimes\tilde{\rho}_{\text{light}}(t) \\ &= H_\alpha\left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right) \\ &- \left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right)H_\beta \\ &+ \left[\boldsymbol{d}\cdot\widehat{\boldsymbol{E}},\left(|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|\otimes\tilde{\rho}_{\text{light}}(t)\right)\right] \end{aligned}$$

$$\begin{aligned} |\phi_\alpha(t)\rangle\langle\phi_\beta(t)|i\hbar\frac{\partial}{\partial t}\left(\tilde{\rho}_{\text{light}}(t)\right) \\ &= \boldsymbol{d}|\phi_\alpha(t)\rangle\cdot\widehat{\boldsymbol{E}}\left(\otimes\tilde{\rho}_{\text{light}}(t)\right)\langle\phi_\beta(t)| \\ &- |\phi_\alpha(t)\rangle\left(\tilde{\rho}_{\text{light}}(t)\right)\langle\phi_\beta(t)|\boldsymbol{d}\cdot\widehat{\boldsymbol{E}} \end{aligned}$$

Tracing out the electronic degree of freedom, we obtain the final equation for time propagation of the density matrix of light:

$$i\hbar\frac{\partial\tilde{\rho}_{\text{light}}}{\partial t} = \boldsymbol{d}_\alpha\widehat{\boldsymbol{E}}\tilde{\rho}_{\text{light}} - \boldsymbol{d}_{\boldsymbol{\beta}}\tilde{\rho}_{\text{light}}\widehat{\boldsymbol{E}} \tag{I.11}$$

Perturbative solution of the time dependent Schrodinger's equation

The electric field operator is given by

$$\boldsymbol{E}(t) = i\sum_{\boldsymbol{k},\sigma}\sqrt{\frac{\hbar\omega}{2V\varepsilon_0}}\boldsymbol{\varepsilon}\left(a_{k\sigma}e^{-i\omega t} - a_{k\sigma}^\dagger e^{i\omega t}\right) \tag{I.12}$$

We decompose the density matrix of light into separate modes and employ equation (I.11):

$$\tilde{\rho}_{\text{light}} = \prod_{k\sigma} \tilde{\rho}_{k\sigma} \tag{I.13}$$

$$\tilde{\rho}_{k\sigma} = -\frac{i}{\hbar}\left[\int_0^t \left(\boldsymbol{d}_\alpha(\tau)\cdot\widehat{\boldsymbol{E}}_{k\sigma}\tilde{\rho}_{k\sigma}(\tau) - \boldsymbol{d}_\beta(\tau)\cdot\tilde{\rho}_{k\sigma}(\tau)\widehat{\boldsymbol{E}}_{k\sigma}\right)d\tau\right]$$

The initial condition is given by $\tilde{\rho}_{k\sigma}(0) = \frac{|0\rangle\langle 0|}{\langle\beta|\alpha\rangle}$. To first order in the light-matter coupling strength,

$$\tilde{\rho}_{k\sigma} = \tilde{\rho}_{k\sigma}(0) - \frac{i}{\hbar}\left[\int_0^t \left(\boldsymbol{d}_\alpha(\tau)\cdot\widehat{\boldsymbol{E}}_{k\sigma}\tilde{\rho}_{k\sigma}(0) - \boldsymbol{d}_\beta(\tau)\cdot\tilde{\rho}_{k\sigma}(0)\widehat{\boldsymbol{E}}_{k\sigma}\right)d\tau\right] \tag{I.14}$$

We employ the 1st order solution to obtain the 2nd order solution:

$$\begin{aligned}\tilde{\rho}_{k\sigma}(\tau) = \tilde{\rho}_{k\sigma}(0) &- \frac{i}{\hbar}\left[\int_0^t \left(\boldsymbol{d}_\alpha(\tau)\cdot\widehat{\boldsymbol{E}}_{k\sigma}\tilde{\rho}_{k\sigma}(0) - \boldsymbol{d}_\beta(\tau)\cdot\tilde{\rho}_{k\sigma}(0)\widehat{\boldsymbol{E}}_{k\sigma}\right)d\tau\right] \\ &+ \left(-\frac{i}{\hbar}\right)^2\Bigg[\int_0^t \Bigg(\boldsymbol{d}_\alpha(\tau_1) \\ &\cdot \widehat{\boldsymbol{E}}_{k\sigma}(\tau_1)\int_0^{\tau_1}\Big(\boldsymbol{d}_\alpha(\tau_2)\cdot\widehat{\boldsymbol{E}}_{k\sigma}(\tau_2)\tilde{\rho}_{k\sigma}(0) - \boldsymbol{d}_\beta(\tau_2) \\ &\cdot \tilde{\rho}_{k\sigma}(0)\widehat{\boldsymbol{E}}_{k\sigma}(\tau_2)\Big)\,d\tau_2 - \boldsymbol{d}_\beta(\tau_1) \\ &\cdot \int_0^{\tau_1}\Big(\boldsymbol{d}_\alpha(\tau_2)\cdot\widehat{\boldsymbol{E}}_{k\sigma}(\tau_2)\tilde{\rho}_{k\sigma}(0) - \boldsymbol{d}_\beta(\tau_2) \\ &\cdot \tilde{\rho}_{k\sigma}(0)\widehat{\boldsymbol{E}}_{k\sigma}(\tau_2)\Big)\,d\tau_2\,\widehat{\boldsymbol{E}}_{k\sigma}(\tau_1)\Bigg)\,d\tau_1\Bigg]\end{aligned} \tag{I.15}$$

The energy expectation value is given by:

$$\varepsilon = \sum_{k\sigma}\hbar\omega\mathrm{Tr}\left[\rho_{k\sigma}(\tau)a_{k\sigma}^{\dagger}a_{k\sigma}\right] = \sum_{k\sigma}\hbar\omega\mathrm{Tr}\left[D(\alpha)\tilde{\rho}_{k\sigma}(\tau)D^{\dagger}(\beta)a_{k\sigma}^{\dagger}a_{k\sigma}\right] \tag{I.16}$$

$$\varepsilon = \sum_{k\sigma} \frac{\omega^2}{2V\varepsilon_0} \left[\int_0^t \left((\boldsymbol{d}_\alpha(\tau_1) \cdot \boldsymbol{\varepsilon}_\sigma) e^{i\omega\tau_1} \int_0^{\tau_1} \left(\boldsymbol{d}_\beta(\tau_2) \cdot \boldsymbol{\varepsilon}_\sigma\right) e^{-i\omega\tau_2} d\tau_2 \right.\right.$$
$$\left.\left. + \left(\boldsymbol{d}_\beta(\tau_1) \cdot \boldsymbol{\varepsilon}_\sigma\right) e^{-i\omega\tau_1} \int_0^{\tau_1} (\boldsymbol{d}_\alpha(\tau_2) \cdot \boldsymbol{\varepsilon}_\sigma) e^{i\omega\tau_2} d\tau_2 \right) d\tau_1 \right]$$

Finally, the spectrum is given by

$$\frac{d\varepsilon}{d\omega} = \sum_{\sigma} \int d\Omega \frac{\omega^4}{2(2\pi)^3 c^3 \varepsilon_0} \left[\int_0^t \left((\boldsymbol{d}_\alpha(\tau_1) \cdot \boldsymbol{\varepsilon}_\sigma) e^{i\omega\tau_1} \int_0^{\tau_1} \left(\boldsymbol{d}_\beta(\tau_2) \cdot \boldsymbol{\varepsilon}_\sigma\right) e^{-i\omega\tau_2} d\tau_2 \right.\right.$$
$$\left.\left. + \left(\boldsymbol{d}_\beta(\tau_1) \cdot \boldsymbol{\varepsilon}_\sigma\right) e^{-i\omega\tau_1} \int_0^{\tau_1} (\boldsymbol{d}_\alpha(\tau_2) \cdot \boldsymbol{\varepsilon}_\sigma) e^{i\omega\tau_2} d\tau_2 \right) d\tau_1 \right] \tag{I.17}$$

$$\frac{d\varepsilon}{d\omega} = \frac{\omega^4}{6\pi^2 c^3 \varepsilon_0} \left[\int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \left(e^{i\omega\tau_1} \boldsymbol{d}_\alpha(\tau_1) \cdot e^{-i\omega\tau_2} \boldsymbol{d}_\beta(\tau_2) + e^{-i\omega\tau_1} \boldsymbol{d}_\beta(\tau_1) \right.\right.$$
$$\left.\left. \cdot\, e^{i\omega\tau_2} \boldsymbol{d}_\alpha(\tau_2) \right) \right]$$

Employing the mathematical identity

$$\int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \left(f(\tau_1) g(\tau_2) + f(\tau_2) g(\tau_1) \right) = \left(\int_0^t d\tau\, f(\tau) \right) \left(\int_0^t d\tau\, g(\tau) \right) \tag{I.18}$$

We arrive at the expression $\frac{d\varepsilon}{d\omega} = \frac{\omega^4}{6\pi^2 c^3 \varepsilon_0} \boldsymbol{d}_\alpha(\omega) \cdot \boldsymbol{d}_\beta^*(\omega)$ where $\boldsymbol{d}_\alpha(\omega) = \int \boldsymbol{d}_\alpha(\tau_1) e^{i\omega\tau_1} d\omega$ for the emission of $\tilde{\rho}_{\alpha\beta}$. The total emission is given by the integration:

$$\frac{d\epsilon}{d\omega} = \frac{\omega^4}{6\pi^2 c^3 \epsilon_0} \int d^2\alpha d^2\beta P(\alpha, \beta^*) \left(d_\alpha(\omega) \cdot d_\beta^*(\omega) \right) \tag{I.19}$$

To 1st order in light-matter coupling (i.e., neglecting any radiation reaction effect), the density matrix of the electron is given by:

$$\rho_{total} = \int d^2\alpha\, d^2\beta\, P_{\alpha\beta} |\phi_\alpha(t)\rangle\langle\phi_\beta(t)| \otimes \frac{|\alpha\rangle\langle\beta|}{\langle\beta|\alpha\rangle} \tag{I.20}$$

$$\rho_{electron} = \mathrm{Tr}_{\mathrm{F}} \left[\int d^2\alpha\, d^2\beta\, P_{\alpha\beta} |\phi_\alpha(t)\rangle\langle\phi_\beta(t)| \otimes \frac{|\alpha\rangle\langle\beta|}{\langle\beta|\alpha\rangle} \right]$$

$$\rho_{electron} = \int d^2\alpha d^2\beta \, P(\alpha,\beta^*)|\phi_\alpha(t)\rangle\langle\phi_\beta(t)|$$

II. Quantum-optical dipole moment under strong-field approximation

In this section, we derive the formula for the dipole moment expectation value for an electron driven by a quantum state of light with arbitrary photon statistics.

Interaction with a coherent state $|\alpha\rangle$

We consider the interaction of an atomic system with a single bound state with a coherent state of light. The ionization potential of this atomic system is denoted by I_p. The driving light occupies a coherent state $|\alpha\rangle = |\alpha_x + i\alpha_y\rangle$ (where α_x and α_y are real valued parameters), corresponding to a classical electromagnetic wave, whose vector potential is given by

$$\boldsymbol{A}_\alpha(t) = \frac{\epsilon^{(1)}}{\omega}\left(\alpha e^{-i\omega t} + \alpha^* e^{i\omega t}\right)\hat{z} = \frac{2\epsilon^{(1)}}{\omega}\left[\alpha_x \cos(\omega t) + \alpha_y \sin(\omega t)\right]\hat{z} \qquad \text{(II.1)}$$

Here, $\hat{z}$ is a unit vector parallel to the z axis. The electric field of this coherent state is denoted $E_\alpha(t) = -\partial \boldsymbol{A}_\alpha/\partial t \cdot \hat{z}$, and the time-dependent Schrodinger's equation (TDSE) of this atomic system is given by

$$i\hbar\frac{\partial|\phi_\alpha(t)\rangle}{\partial t} = \left[-\frac{1}{2m}\nabla^2 + U(z) - ezE_\alpha(t)\right]|\phi_\alpha(t)\rangle \qquad \text{(II.2)}$$

Here, $|\phi_\alpha(t)\rangle$ is the time dependent wavefunction of the system, and $U(z)$ is the atomic potential. Initially, the system is in its atomic ground state, denoted by $|0\rangle$. Under the standard approximations of the SFA theory [1], $|\phi_\alpha(t)\rangle$ can be written as

$$|\phi_\alpha(t)\rangle = e^{\frac{i}{\hbar}I_p t}\left(a(t)|0\rangle + \int b_\alpha(\boldsymbol{v},t)d^3\boldsymbol{v}\cdot|\boldsymbol{v}\rangle\right) \qquad \text{(II.3)}$$

We substitute (II.3) into (II.2) and to reformulate the TDSE in terms of kinetic momentum state amplitudes:

$$\frac{\partial b_\alpha(\boldsymbol{v},t)}{\partial t} = -\frac{i}{\hbar}\left(\frac{\boldsymbol{v}^2}{2m} + I_p\right)b_\alpha(\boldsymbol{v},t) - eE_\alpha(t)\frac{\partial b_\alpha(\boldsymbol{v},t)}{\partial v_z} + \frac{i}{\hbar}ed_z(\boldsymbol{v})E_\alpha(t) \qquad \text{(II.4)}$$

Here, $d_z(\boldsymbol{v}) = \langle \boldsymbol{v}|z|0\rangle$ and we have already employed the fact that the light field is linearly polarized, in writing $E_\alpha(t)$ as a scalar function and deriving b_α with respect to v_z (the velocity component parallel to the polarization of the driving field). Assuming that the ground state is non-depleted ($a(t) = 1$), the state of the atom is completely determined by the $b_\alpha(\boldsymbol{v}, t)$ continuum state amplitudes. The exact solution for $b(\boldsymbol{v}, t)$ is

$$b(\boldsymbol{v}, t) = \frac{i}{\hbar}\int_0^t dt' eE_\alpha(t')d_z\big(\boldsymbol{v} + e\boldsymbol{A}_\alpha(t) - e\boldsymbol{A}_\alpha(t')\big) \cdot \exp\left(-\frac{i}{\hbar}\int_{t'}^{t} dt'' \left[\frac{\big(\boldsymbol{v} + e\boldsymbol{A}_\alpha(t) - e\boldsymbol{A}_\alpha(t'')\big)^2}{2} + I_p\right]\right) \tag{II.5}$$

Neglecting bound-bound and continuum-continuum transitions, the dipole moment expectation value $z_\alpha(t) = \langle \phi_\alpha(t)|z|\phi_\alpha(t)\rangle$ is given by

$$z_\alpha(t) = \int d^3\boldsymbol{v}\, d_z^*(\boldsymbol{v})b(\boldsymbol{v}, t) + c.c. \tag{II.6}$$

We define canonical momentum $\boldsymbol{p}$:

$$\boldsymbol{p} = \boldsymbol{v} + e\boldsymbol{A}_\alpha(t) \tag{II.7}$$

This results in the final semi-classical equation for the dipole moment of an electron driven by a coherent state $|\alpha\rangle$:

$$z_\alpha(t) = \frac{i}{\hbar}\int d^3\boldsymbol{p}\int_0^t dt' eE_\alpha(t)d_z\big(\boldsymbol{p} - e\boldsymbol{A}_\alpha(t')\big)d_z^*\big(\boldsymbol{p} - e\boldsymbol{A}_\alpha(t)\big) \cdot \exp\left(-\frac{i}{\hbar}S_\alpha(\boldsymbol{p}, t, t')\right) + \text{c.c.} \tag{II.8}$$

$$S_\alpha(\boldsymbol{p}, t, t') = \int_{t'}^{t} dt'' \left(\frac{[\boldsymbol{p} - e\boldsymbol{A}(t'')]^2}{2} + I_p\right) \tag{II.9}$$

The function $S_\alpha(\boldsymbol{p}, t, t')$ is the semi-classical action of an electron driven by a coherent state $|\alpha\rangle$. The Ω frequency component of $z_\alpha(t)$ can be evaluated semi-analytically, by employing the saddle point approximation:

$$\begin{cases} \frac{\partial S_\alpha}{\partial \boldsymbol{p}} = \boldsymbol{0} \\ \frac{\partial S_\alpha}{\partial t'} = 0 \\ \frac{\partial S_\alpha}{\partial t} = \hbar\Omega \end{cases} \tag{II.10}$$

The system of equations (II.10) defines the three physically meaningful (and experimentally observable) stationary parameters $[\boldsymbol{p} = \boldsymbol{p}_\Omega, t' = t_{0_\Omega}, t = t_{1_\Omega}]$, associated with the emission of the frequency Ω. Here, $\boldsymbol{p}$ is inferred as the canonical momentum of a three-step electronic trajectory, t_0 is the time of ionization, and t_1 is time of recombination. We emphasize that this solution is completely semi-classical and does not include the effect of vacuum fluctuations (i.e., noise associated with the quantum-mechanical uncertainty principle).

Interaction with an arbitrary quantum state of light under the strong-field approximation

Next, we consider the interaction of the same atomic system with a single mode of light with arbitrary quantum statistics. The density matrix (quantum state) of the driving light is specified by *generalized Glauber representation* $P(\alpha, \beta)$,

$$\hat{\rho}_{drive} = \int d^2\alpha \, d^2\beta \, P(\alpha, \beta) \frac{|\alpha\rangle\langle\gamma^*|}{\langle\beta^*|\alpha\rangle} \tag{II.11}$$

In the previous supplementary section, we have obtained that the density matrix of an electron driven by $\hat{\rho}_{drive}$ is given (to lowest order in light matter coupling) by

$$\rho_e = \int d^2\alpha \, d^2\beta \, P(\alpha, \beta^*) |\phi_\alpha(t)\rangle\langle\phi_\beta(t)| \tag{II.12}$$

where $|\phi_\alpha(t)\rangle$ solves equation (II.2). In this case the dipole moment is given by

$$z(t) = \mathrm{Tr}[\rho_e(t)x] = \int d^2\alpha \, d^2\beta \, P(\alpha, \beta^*) \langle\phi_\beta(t)|z|\phi_\alpha(t)\rangle \tag{II.13}$$

Employing equation (II.3) in the main text, and again neglecting bound-bound and continuum-continuum transitions (a standard assumption of the SFA theory) we have

$$\langle\phi_\beta(t)|z|\phi_\alpha(t)\rangle = \int b_\alpha(\boldsymbol{v}, t) d^3\boldsymbol{v} \cdot \langle 0|z|\boldsymbol{v}\rangle + \int b_\beta^*(\boldsymbol{v}, t) d^3\boldsymbol{v} \cdot \langle\boldsymbol{v}|z|0\rangle \tag{II.14}$$

Notably, by keeping only bound-continuum transitions, we can decompose $\langle\phi_\beta(t)|z|\phi_\alpha(t)\rangle$ into terms that depends separately on α and β. This separation is exploited to simplify supplementary equation (II.3):

$$z(t) = \int d^2\alpha \left(\int d^2\beta\, P(\alpha,\beta^*)\right)\int b_\alpha(\boldsymbol{v},t)d^3\boldsymbol{v}\cdot\langle 0|z|\boldsymbol{v}\rangle + \int d^2\beta \left(\int d^2\alpha\, P(\alpha,\beta^*)\right)\int b_\beta^*(\boldsymbol{v},t)d^3\boldsymbol{v}\cdot\langle\boldsymbol{v}|z|0\rangle \tag{II.15}$$

We employ the general property of the generalized Glauber representation $P(\alpha,\beta^*) = P(\beta,\alpha^*)$. This property stems from the connection between the generalized Glauber representation and the Husimi quasi-probability distribution $Q(\alpha) = \frac{1}{\pi}\langle\alpha|\rho|\alpha\rangle$:

$$P(\alpha,\beta) = \frac{1}{4\pi}\exp\left[-\frac{|\alpha-\beta^*|^2}{4}\right]Q\left(\frac{\alpha+\beta^*}{2}\right) \tag{II.16}$$

Through this relation, we have $\int d^2\beta\, P(\beta,\alpha^*) = \int d^2\beta\, P(\alpha,\beta^*)$, hence,

$$z(t) = \int d^2\alpha \left(\int d^2\beta\, P(\alpha,\beta^*)\right)\int b_\alpha(\boldsymbol{v},t)d^3\boldsymbol{v}\cdot\langle 0|z|\boldsymbol{v}\rangle + c.c. \tag{II.17}$$

By equation (II.6) in the main text, this reduces to

$$z(t) = \int d^2\alpha P(\alpha) z_\alpha(t) \tag{II.18}$$

Where $z_\alpha(t)$ is the semi-classical dipole moment expectation value for a coherent drive $|\alpha\rangle$, and $P(\alpha) \equiv \int d^2\beta\, P(\alpha,\beta^*)$ and is not to be confused with the standard Glauber representation. $z_\alpha(t)$ can be obtained either by solving the time-dependent Schrodinger's equation, or directly from the semi-classical SFA formula (II.8), to arrive at the final expression:

$$z(t) = \frac{i}{\hbar}\int d^2\alpha \int d^3\boldsymbol{p}\int_0^t dt' eE_\alpha(t) d_z\big(\boldsymbol{p}-e\boldsymbol{A}_\alpha(t')\big)d_z^*\big(\boldsymbol{p}-e\boldsymbol{A}_{\boldsymbol{\alpha}}(t)\big) \cdot P(\alpha)\exp\left(-\frac{i}{\hbar}S_\alpha(\boldsymbol{p},t,t')\right) + c.c. \tag{II.19}$$

III. Derivation of $P(\alpha)$ for squeezed coherent light

III. A. Integration of $P(\alpha,\beta^*)$

In this section, we derive the distribution function $P(\alpha)=\int d^2\beta P(\alpha,\beta^*)$ for the case of squeezed coherent states of light. For a squeezed coherent state of light $|\gamma,r\rangle$ where γ is the coherent parameter and r is the squeezing parameter, the Husimi $Q(\alpha)$ distribution is given by [2]

$$Q(\alpha)=\frac{1}{\pi\cosh(r)}\exp\left(-2\frac{\left(\alpha_y-\gamma_y\right)^2}{1+e^{2r}}-2\frac{(\alpha_x-\gamma_x)^2}{1+e^{-2r}}\right) \tag{III.1}$$

The generalized Glauber representation of this state is constructed through

$$P(\alpha,\beta)=\frac{1}{4\pi}e^{-|\alpha-\beta^*|^2/4}Q\left(\frac{\alpha+\beta^*}{2}\right) \tag{III.2}$$

The resulting distribution is:

$$P(\alpha,\beta^*)=\exp\left[-\frac{(\alpha_x-\beta_x)^2}{4}-\frac{(\alpha_x+\beta_x-2\gamma_x)^2}{2+2e^{-2r}}-\frac{\left(\alpha_y-\beta_y\right)^2}{4}\right. \\ \left.-\frac{\left(\alpha_y+\beta_y-2\gamma_y\right)^2}{2+2e^{2r}}\right] \tag{III.3}$$

Here, $\alpha=\alpha_x+i\alpha_y$ (and similarly β and γ). We explicitly calculate $P(\alpha)=\int d^2\beta P(\alpha,\beta^*)$:

$$P(\alpha)=\frac{1}{4\pi^2\cosh(r)}\int d\beta_x\exp\left[-\frac{(\alpha_x-\beta_x)^2}{4}\right. \\ \left.-\frac{(\alpha_x+\beta_x-2\gamma_x)^2}{2+2e^{-2r}}\right]\int d\beta_y\exp\left[-\frac{\left(\alpha_y-\beta_y\right)^2}{4}\right. \\ \left.-\frac{\left(\alpha_y+\beta_y-2\gamma_y\right)^2}{2+2e^{2r}}\right]=\frac{1}{4\pi^2\cosh(r)}\ I_1I_2 \tag{III.4}$$

$$I_1=\int d\beta_x\exp\left[-\frac{(\alpha_x-\beta_x)^2}{4}-\frac{(\alpha_x+\beta_x-2\gamma_x)^2}{2+2e^{-2r}}\right]$$

$$I_2 = \int d\beta_y \exp\left[-\frac{(\alpha_y - \beta_y)^2}{4} - \frac{(\alpha_y + \beta_y - 2\gamma_y)^2}{2 + 2e^{2r}}\right]$$

These final expression for $P(\alpha)$ is

$$P(\alpha) = \frac{1}{\pi \cosh(r)} \frac{e^{-\frac{2(\alpha_y - \gamma_y)^2}{3+e^{2r}} - \frac{2(\alpha_x - \gamma_x)^2}{3+e^{-2r}}}(1 + e^{2r})}{\sqrt{3 + 10e^{2r} + 3e^{4r}}} \quad \text{(III.5)}$$

III. B. Infinite volume of quantization

Let us take the limit $\epsilon^{(1)} \to 0$ and $V \to \infty$ (these are the single-photon amplitude and quantization volume, respectively). Within this limit, the $P(\alpha)$ distribution of a coherent state $|\gamma\rangle$ approaches a Dirac delta function $\delta(\alpha - \gamma)$, and so, $|\gamma\rangle$ becomes equivalent to a classical electromagnetic field $E_\gamma(t)$. Additionally, taking this limit lifts the ambiguity in the relation between dimensionless coherent parameters and electric field amplitudes $E = 2\epsilon^{(1)}\alpha$, which in principle depends on the frequency and volume of quantization ($\epsilon^{(1)} = \sqrt{\hbar\omega/2\epsilon_0 V}$). Hence, we also exploit this limit to reformulate the distribution $P(\alpha)$ with a distribution $P(E)$ that does not depend on the quantization volume.

Throughout this section, the squeezing parameter r is assumed to be real and positive. If one wishes to change the relative squeezing phase, it can be done by changing the phase of the pump.

We consider the $P(\alpha)$ distribution of the state $|\gamma, r\rangle$ (equation (III.5)). The number of photons in such a state is given by

$$N = \langle\gamma, r|\hat{n}|\gamma, r\rangle = |\gamma|^2 + \sinh^2(r) \quad \text{(III.6)}$$

The intensity of this beam is given by

$$I \equiv \frac{c\hbar\omega}{V} N \equiv \frac{c\hbar\omega}{V}(|\gamma|^2 + \sinh^2(r)) \equiv I_{coh} + I_{vac} \quad \text{(III.7)}$$

Where we have defined $I_{coh} = \frac{c\hbar\omega}{V}|\gamma|^2$ and $I_{vac} = \frac{c\hbar\omega}{V}\sinh^2(r)$, and V is the quantization volume. The squeezing parameter r can be reformulated as $r = \operatorname{asinh}\left(\pm\sqrt{\frac{V I_{vac}}{c\hbar\omega}}\right)$, where the sign of r determines the type of squeezing (plus/minus for phase/amplitude, respectively). The electric field of the squeezed coherent state $|\gamma, r\rangle$ is given by $E(t) = 2\epsilon^{(1)}[-\gamma_x \sin(\omega t) + \gamma_y \cos(\omega t)]$, hence, we preform a substitution of integration variables in from dimensionless coherent parameters to electric field quadratures, $E_{\alpha_x} = 2\epsilon^{(1)}\alpha_x$ and $E_{\alpha_y} = 2\epsilon^{(1)}\alpha_y$.

Let us perform the substitution:

$$e^{-\frac{2(\alpha_y-\gamma_y)^2}{3+e^{2r}}-\frac{2(\alpha_x-\gamma_x)^2}{3+e^{-2r}}} = e^{-\frac{2\left(E_{\alpha_y}-E_{\gamma_y}\right)^2}{\left(2\epsilon^{(1)}\right)^2(3+e^{2r})}-\frac{2\left(E_{\alpha_x}-E_{\gamma_x}\right)^2}{\left(2\epsilon^{(1)}\right)^2(3+e^{-2r})}} = \tag{III.8}$$

$$e^{2r} = e^{2\,\mathrm{asinh}\left(\sqrt{\frac{VI_{vac}}{c\hbar\omega}}\right)} = 1 + 2\frac{VI_{vac}}{c\hbar\omega} + 2\sqrt{\left(\frac{VI_{vac}}{c\hbar\omega}\right)^2 + \frac{VI_{vac}}{c\hbar\omega}}$$

$$e^{-2r} = e^{-2\,\mathrm{asinh}\sqrt{\frac{VI_{vac}}{c\hbar\omega}}} = 1 + 2\frac{VI_{vac}}{c\hbar\omega} - 2\sqrt{\left(\frac{VI_{vac}}{c\hbar\omega}\right)^2 + \frac{VI_{vac}}{c\hbar\omega}}$$

Plugging the single photon amplitude $\epsilon^{(1)} = \sqrt{\frac{\hbar\omega}{2\epsilon_0 V}}$, and taking the limit $V \to \infty$, we have:

$$\left(2\epsilon^{(1)}\right)^2(3 + e^{-2r}) \to 0 \tag{III.9}$$

$$\frac{2}{(2\epsilon^{(1)})^2(3 + e^{2r})} \to \frac{1}{2|E_{vac}|^2}$$

where E_{vac} fulfills $I_{vac} = \frac{1}{2}\epsilon_0 c|E_{vac}|^2$. Hence, $P(E_\alpha) = P\left(E_{\alpha_x} + iE_{\alpha_y}\right)$ becomes

$$P(E_\alpha) = \frac{1}{norm}\, e^{-\frac{\left(E_{\alpha_y}-E_{\gamma_y}\right)^2}{2|E_{vac}|^2}}\, \delta\left(E_{\alpha_x} - E_{\gamma_x}\right) \tag{III.10}$$

Thus, the final equation for $z(t)$, the SFA dipole moment of an atom driven by a squeezed coherent state $|\gamma, r\rangle$ is given by

$$z(t) = \frac{1}{\sqrt{2\pi}|E_{vac}|}\int dE_{\alpha_y}\, e^{-\frac{\left(E_{\alpha_y}-E_{\gamma_y}\right)^2}{2|E_{vac}|^2}}\, z_{E_{\alpha_y},E_{\gamma_x}}(t) \tag{III.11}$$

IV. Derivation of the coupled algebraic equations

I.V.A. Saddle point approximation

In this section, we derive the coupled algebraic equations for the dipole moment. The quantum-optical analogue of the semi-classical action is given by

$$\underbrace{S_q(\boldsymbol{p},t',t,\alpha)}_{\text{quantum-optical action}} = \underbrace{\int_{t'}^{t} dt'' \left(\frac{[\boldsymbol{p}-e\boldsymbol{A}_\alpha(t'')]^2}{2}+I_p\right)}_{\text{semi-classical action}} + \underbrace{i\log\big(P(\alpha)\big)}_{\text{photon statistics}} \tag{IV.1}$$

The emission of high-order harmonics at frequencies $\Omega = n\omega$ will mainly originate from the stationary points of $S_q(\boldsymbol{\kappa}_q) - \hbar\Omega t$ with respect to all variables $\boldsymbol{\kappa}_q = [p,t',t,\alpha]$, which satisfy the condition $\nabla_{\boldsymbol{\kappa}_q}\big(S_q(\boldsymbol{\kappa}_q) - \hbar\Omega t\big) = 0$. For the case of squeezed coherent light $|\gamma, r\rangle$, we have

$$S_q(\boldsymbol{p},t',t,\alpha) = -2i\frac{(\alpha_x-\gamma_x)^2}{3+e^{-2r}} - 2i\frac{\left(\alpha_y-\gamma_y\right)^2}{3+e^{2r}} + \int_{t'}^{t} dt''\left(\frac{[p-eA_\alpha(t'')]^2}{2}+I_p\right) - \hbar\Omega t \tag{IV.2}$$

Momentum derivative $\frac{\partial S_q}{\partial p} = 0$*:*

$$\int_{t'}^{t} dt''[eA_\alpha(t'')] = (t-t')p \tag{IV.3}$$

Ionization moment derivative $\frac{\partial S_q}{\partial t'} = 0$*:*

$$\frac{[p-eA_\alpha(t')]^2}{2} = -I_p \tag{IV.4}$$

Recombination moment derivative $\frac{\partial S_q}{\partial t} = 0$*:*

$$\frac{[p-eA_\alpha(t)]^2}{2} = \hbar\Omega - I_p \tag{IV.5}$$

Coherent parameter derivative – x quadrature:

$$A_\alpha(t) = \frac{\epsilon^{(1)}}{\omega}\left(\alpha e^{-i\omega t} + \alpha^* e^{i\omega t}\right) = \frac{2\epsilon^{(1)}}{\omega}[\alpha_x\cos(\omega t) + \alpha_y\sin(\omega t)] \tag{IV.6}$$

$$\frac{1}{i}\frac{\partial S_q}{\partial \alpha_x} = -4\frac{(\alpha_x - \gamma_x)}{3+e^{-2r}}$$
$$+\frac{2i\epsilon^{(1)}}{\omega}\left\{\frac{1}{\omega}p\,[\sin(\omega t) - \sin(\omega t')]\right.$$
$$-\frac{\epsilon^{(1)}}{\omega}e\alpha_x\left\{t - t' + \frac{1}{2\omega}(\sin(2\omega t) - \sin(2\omega t'))\right\}$$
$$\left.+\frac{\epsilon^{(1)}}{\omega}e\alpha_y\frac{1}{2\omega}[\cos(2\omega t) - \cos(2\omega' t')]\right\} = 0$$

Coherent parameter derivative – y quadrature:

$$A_\alpha(t) = \frac{\epsilon^{(1)}}{\omega}\left(\alpha e^{-i\omega t} + \alpha^* e^{i\omega t}\right) = \frac{2\epsilon^{(1)}}{\omega}[\alpha_x \cos(\omega t) + \alpha_y \sin(\omega t)] \qquad \text{(IV.7)}$$

$$\frac{1}{i}\frac{\partial \tilde{S}}{\partial \alpha_y} = -4\frac{\left(\alpha_y - \gamma_y\right)}{3+e^{2r}}$$
$$+\frac{2i\epsilon^{(1)}}{\omega}\left\{-\frac{1}{\omega}p[\cos(\omega t) - \cos(\omega t')]\right.$$
$$+\frac{\epsilon^{(1)}}{\omega}e\alpha_x\frac{1}{2\omega}[\cos(2\omega t) - \cos(2\omega t')]$$
$$\left.-\frac{\epsilon^{(1)}}{\omega}e\alpha_y\left\{t - t' - \frac{1}{2\omega}[\sin(2\omega t) - \sin(2\omega t')]\right\}\right\} = 0$$

Infinite volume of quantization

Upon taking the limit of an infinite volume of quantization $V \to \infty$ the following limits apply: $\epsilon^{(1)} \to 0$, $\frac{\hbar\omega}{2\epsilon_0 V}(3+e^{2r}) \to 2|E_{vac}|^2$, $\frac{\hbar\omega}{2\epsilon_0 V}(3+e^{-2r}) \to 0$. With these limits, the coupled algebraic equations take the form:

$$\int_{t'}^{t} dt''[eA_\alpha(t'')] = (t-t')p$$
$$\frac{[p - eA_\alpha(t')]^2}{2} = -I_p$$

$$\frac{[p - eA_\alpha(t)]^2}{2} = \hbar\Omega - I_p \tag{IV.8}$$

$$\epsilon_0 c(E_{\alpha_y} - E_{\gamma_y}) = \frac{2i}{\omega}\, I_{vac} \left\{ \begin{matrix} -\frac{1}{\omega} p\,[\cos(\omega t) - \cos(\omega t')] \\ -e\frac{E_{\alpha y}}{2\omega}\left\{t - t' - \frac{1}{2\omega}[\sin(2\omega t) - \sin(2\omega t')]\right\} \\ +e\frac{E_{\alpha_x}}{4\omega^2}[\cos(2\omega t) - \cos(2\omega t')] \end{matrix} \right\}$$

$$E_{\alpha_x} = E_{\gamma_x}$$

I.V.B. Derivation directly from $P(E_\alpha)$ distribution

Here, we derive the coupled algebraic equations (IV.8) using the distribution

$$P\left(E_{\alpha_y}\right) = \frac{1}{\sqrt{2\pi}|E_{vac}|} e^{-\frac{\left(E_{\alpha_y} - E_{\gamma_y}\right)^2}{2|E_{vac}|^2}} \tag{IV.9}$$

which corresponds to squeezing of the x quadrature, so that the amplitude of electric field fluctuations in the anti-squeezed quadrature E_{α_y} is E_{vac}. The corrected action is given by

$$S\left(\boldsymbol{p}, t', t, E_{\alpha_y}\right) = S_\alpha(\boldsymbol{p}, t', t) + i\log\left(P\left(E_{\alpha_y}\right)\right) \tag{IV.10}$$

$$S\left(\boldsymbol{p}, t', t, E_{\alpha_y}\right) = \int_{t'}^{t} dt'' \left(\frac{[\boldsymbol{p} - e\boldsymbol{A}_\alpha(t'')]^2}{2} + I_p\right) - i\,\frac{\left(E_{\alpha_y} - E_{\gamma_y}\right)^2}{2|E_{vac}|^2}$$

Where the vector potential $A_\alpha(t'')$ is

$$A_\alpha(t'') = \frac{1}{\omega}\left(E_{\alpha_y}\sin(\omega t'') + E_{\gamma_x}\cos(\omega t'')\right) \tag{IV.11}$$

The first three coupled equations are trivially identical to the semi-classical coupled equations because the correction to the semi-classical action is independent of $\boldsymbol{p}, t', t$. The fourth equation is given by:

$$\frac{\partial S}{\partial E_{\alpha_y}} = \partial_{E_{\alpha_y}} \int_{t'}^{t} dt'' \left(\frac{\left[\boldsymbol{p} - e\frac{E_{\alpha_y}\sin(\omega t'') + E_{\gamma_x}\cos(\omega t'')}{\omega}\right]^2}{2} \right) - i\,\frac{E_{\alpha_y} - E_{\gamma_y}}{|E_{vac}|^2} = 0$$

Rearranging, it equates to:

$$E_{\alpha_y} = E_{\gamma_y} + i\frac{|E_{vac}|^2}{\omega}\left\{\begin{array}{c} -\frac{pe}{\omega}\left(\cos(\omega \mathrm{t}) - cos(\omega t')\right) \\ -e^2\frac{E_{\alpha_y}}{2\omega}\left[t - t' - \frac{\sin(2\omega t) - \sin(2\omega t')}{2\omega}\right] \\ +e^2\frac{E_{\gamma_x}[\cos(2\omega t) - \cos(2\omega t')]}{4\omega^2} \end{array}\right\}$$

This is precisely the fourth equation of (IV.8), as can be seen by substituting $I_{vac} = \frac{1}{2}\epsilon_0 c|E_{vac}|^2$.

<u>I.V.C. Yoctosecond time-delays induced by bare vacuum fluctuations.</u>

In Figure 4, we show that vacuum fluctuations at the frequency of the pump field induce yoctosecond time delays to the electronic trajectories in standard HHG experiments. This is found by solving the coupled algebraic equations (II.3)(II.7) for a squeezed coherent field $|\alpha, 0\rangle$, i.e., by taking $r = 0$. Notably, even for a coherent state, these equations are not identical to the semi-classical SFA equations, with deviations introduced by a nonzero value for $\epsilon^{(1)} = \sqrt{\frac{\hbar\omega}{2\epsilon_0 V}}$.

For a coherent state driver, the electric field variance is given by $\Delta E^2 = {\epsilon^{(1)}}^2$. That is, $\epsilon^{(1)}$ is the amplitude of electromagnetic vacuum fluctuations at the frequency of the driver. In free space, this amplitude depends on the focusing of the beam, and was measured[44] to be of the order of $1 - 100\frac{V}{cm}$ for focal spot radii in the range $100\mu m - 1\mu m$. To obtain an upper bound for the time delays (i.e., the ys time delays presented in the main text), we plug in $\epsilon^{(1)} = 1500\frac{V}{cm}$ (3e-7 a.u.).

V. Derivation of the effective photon statistics forces

In this section, we solve the coupled algebraic equations (IV.8) to show that within the qSFA, photon statistics are formally equivalent to a *time-dependent force*. That is, **within the quantum-optical qSFA theory, the motion of the electron is formally governed by an effective semi-classical theory that includes an effective photon-statistics force, in addition to the classical electric field of the driving pulse.**

According to equations (IV.8), the effective vector potential exerted by squeezed-coherent light on the electron is given by $A_\alpha(t) = \frac{1}{\omega}\left(E_{\alpha_x}\cos(\omega t) + E_{\alpha_y}\sin(\omega t)\right)$. We note that in the absence of squeezing, $A_\alpha(t) = A_\gamma(t)$ where $A_\gamma(t)$ is the classical vector potential carried by the driving light. The last two equations of (IV.8) can be solved exactly, resulting in

$$A_\alpha(t) = \frac{1}{\omega}\left(E_{\gamma_x}\cos(\omega t) + E_{\alpha_y}\sin(\omega t)\right) \tag{V.1}$$

$$E_{\alpha_y} = \frac{E_{\gamma_y} + \frac{i}{\omega}\, E_{vac}^2\left\{-\frac{1}{\omega}pe\,[\cos(\omega t) - \cos(\omega t')] + e^2\frac{E_{\gamma_x}}{4\omega^2}[\cos(2\omega t) - \cos(2\omega t')]\right\}}{\left(1 + ie^2\frac{E_{vac}^2}{2\omega^2}\left\{t - t' - \frac{1}{2\omega}[\sin(2\omega t) - \sin(2\omega t')]\right\}\right)}$$

E_{α_y} can be rearranged as

$$E_{\alpha_y} = \frac{E_{\gamma_y} + \frac{ie}{\omega}\, E_{vac}^2 \int_{t'}^{t} dt''\sin(\omega t'')\left(p - e\frac{E_{\gamma_x}}{\omega}\cos(\omega t'')\right)}{1 + ie^2\frac{E_{vac}^2}{2\omega^2}\int_{t'}^{t} dt''\,(1 - \cos(2\omega t''))} \tag{V.2}$$

Taking $E_{vac}^2/2\omega^2 \ll 1$ and neglecting high-order terms in E_{vac}, we have

$$E_{\alpha_y} \approx E_{\gamma_y} + \frac{ie}{\omega}\, E_{vac}^2 \int_{t'}^{t} dt''\sin(\omega t'')\left(p - e\frac{E_{\gamma_x}}{\omega}\cos(\omega t'')\right) - ie^2\frac{E_{\gamma_y}E_{vac}^2}{2\omega^2}\int_{t'}^{t} dt''\,(1 - \cos(2\omega t'')) \tag{V.3}$$

Using $p = v(t) + eA_\alpha(t) = v(t) + \frac{e}{\omega}\left(E_{\gamma_x}\cos(\omega t) + E_{\alpha_y}\sin(\omega t)\right)$ where $v(t)$ is the velocity of the electron at time t, we have $p - e\frac{E_{\gamma_x}}{\omega}\cos(\omega t'') = v(t) + e\frac{1}{\omega}E_{\alpha_y}\sin(\omega t)$. Hence

$$E_{\alpha_y} - E_{\gamma_y} \approx \int_{t'}^{t} dt''\left(\frac{ie}{\omega}\, E_{vac}^2 v(t'')\sin(\omega t'') + \frac{i}{\omega^2}e^2\, E_{vac}^2(E_{\alpha_y} - E_{\gamma_y})\sin^2(\omega t'')\right) \tag{V.4}$$

Rearranging,

$$E_{\alpha_y} - E_{\gamma_y} \approx \frac{\int_{t'}^{t} dt''\frac{ie}{\omega}\, E_{vac}^2 v(t'')\sin(\omega t'')}{\left(1 - \frac{i}{\omega^2}\, e^2 E_{vac}^2\int_{t'}^{t} dt''(\sin^2(\omega t''))\right)} \approx \frac{ie}{\omega}\, E_{vac}^2\int_{t'}^{t} dt'' v(t'')\sin(\omega t'') \tag{V.5}$$

We plug this into the first equation of (IV.8), which describes the displacement of the electron as a function of time:

(V.6)

$$\int_{t'}^{t} dt'' \frac{e}{\omega}\left(E_{\gamma_x}\cos(\omega t'') + E_{\gamma_y}\sin(\omega t'') + \frac{ie}{\omega}\ E_{vac}^2 \sin(\omega t'') \int_{t'}^{t} dt''' v(t''') \sin(\omega t''')\right) = (t - t')p$$

Now, we make use of the formula

(V.7)

$$\int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \left(f(\tau_1)g(\tau_2) + f(\tau_2)g(\tau_1)\right) = \left(\int_0^t d\tau\, f(\tau)\right)\left(\int_0^t d\tau\, g(\tau)\right)$$

To write the equality:

(V.8)

$$\frac{ie}{\omega^2}\ E_{vac}^2 \left(\int_{t'}^{t} dt'' \sin(\omega t'')\right)\left(\int_{t'}^{t} dt'''\ v(t''') \sin(\omega t''')\right) =$$

$$\int_{t'}^{t} dt'' \frac{ie}{\omega^2}\ E_{vac}^2 \int_{t'}^{t''} dt''' \left(\left(v(t'') + v(t''')\right) \sin(\omega t'') \sin(\omega t''')\right)$$

Hence,

(V.9)

$$\int_{t'}^{t} dt'' e\left[A_\gamma(t'') + \frac{ie}{\omega^2}\ E_{vac}^2 \int_{t'}^{t''} dt''' \left(\left(v(t'') + v(t''')\right) \sin(\omega t'') \sin(\omega t''')\right)\right] = (t - t')p$$

We now postulate the existence of an effective semi-classical action, S_{eff}, which solves equation (V.9).

$$\frac{dS_{eff}}{dp} = \int_{t'}^{t} dt'' \left\{p - e\left[A_\gamma(t'') + \frac{ie}{\omega^2}\ E_{vac}^2 \int_{t'}^{t''} dt''' \left(\left(v(t'') + v(t''')\right) \sin(\omega t'') \sin(\omega t''')\right)\right]\right\} = 0$$

This is solved by the effective semi-classical action:

$$S_{eff}(p,t',t) = \int_{t'}^{t} dt'' \; \frac{\left[p - eA_\gamma(t'') - \frac{ie^2}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau)\right]^2}{2} \tag{V.10}$$

That is, the strong-field dynamics of the electron in the squeezed coherent field are formally equivalent to dynamics in an effective semi-classical theory, where the electron is subject to the classical vector potential $A_\gamma(t'')$ and an effective photon statistics force

$$A_{sq}(t'') = \frac{ie}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau) \tag{V.11}$$

Consistency of the effective photon statistics force with the complete qSFA theory

Here, we derive the effective semi-classical coupled algebraic equations associated with the effective photon statistics force. We show that they precisely reproduce the coupled algebraic equations derived by the complete quantum-optical qSFA theory.

p derivative

Using the property $v(\tau) = p - eA_\gamma(\tau)$, $v(t'') = p - \boldsymbol{e}A_\gamma(t'')$ we write:

$$\frac{dS_{eff}}{dp} \tag{V.12}$$

$$= \frac{d}{dp}\int_{t'}^{t} dt'' \; \frac{\left[p - eA_\gamma(t'') - \frac{ie^2}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau \left(p - eA_\gamma(\tau)\right) \sin(\omega\tau)\right]^2}{2}$$

Explicitly, equates to (up to 2nd order in E_{vac}):

$$\frac{dS_{eff}}{dp} = \int_{t'}^{t} dt'' \left(p - eA_\gamma(t'') - \frac{ie^2}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau \left(v(\tau) + v(t'')\right) \sin(\omega\tau)\right) \tag{V.13}$$

This reproduces (V. 9), i.e., the 3rd equation of (IV.8).

t' derivative

$$\frac{dS_{eff}}{dt'} = \frac{d}{dt'}\int_{t'}^{t} dt'' \; \frac{\left[p - eA_\gamma(t'') - \frac{ie^2}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau)\right]^2}{2} = \tag{V.14}$$

$$= \frac{ie^2}{\omega^2} E_{vac}^2 \int_{t'}^{t} dt''\, v(t'') \sin(\omega t'') v(t') \sin(\omega t') - \frac{\left[p - A_\gamma(t')\right]^2}{2}$$

This reproduces the 2nd equation of (IV.8), as can be seen by substituting (IV.5) into (IV.8):

$$E_{\alpha_y} - E_{\gamma_y} \approx \frac{ie}{\omega} E_{vac}^2 \int_{t'}^{t} dt'' v(t'') \sin(\omega t'') \tag{V.15}$$

$$-\frac{[p - eA_\alpha(t')]^2}{2} = -\frac{\left[p - eA_\gamma(t') - \sin(\omega t') \frac{ie^2}{\omega^2} E_{vac}^2 \int_{t'}^{t} dt'' v(t'') \sin(\omega t'')\right]^2}{2}$$

$$= -\frac{\left[p - eA_\gamma(t')\right]^2}{2} + \frac{ie^2}{\omega^2} E_{vac}^2 v(t') \sin(\omega t') \int_{t'}^{t} dt'' v(t'') \sin(\omega t'')$$

t derivative

$$\frac{dS}{dt} = \frac{d}{dt} \int_{t'}^{t} dt'' \frac{\left[p - eA_\gamma(t'') - \frac{ie^2}{\omega^2} E_{vac}^2 \sin(\omega t'') \int_{t_0}^{t''} d\tau\, v(\tau) \sin(\omega \tau)\right]^2}{2} \tag{V.16}$$

$$= \frac{\left[p - eA_\gamma(t) - \frac{ie^2}{\omega^2} E_{vac}^2 \sin(\omega t) \int_{t_0}^{t} d\tau\, v(\tau) \sin(\omega \tau)\right]^2}{2}$$

This precisely equates the third equation in (IV.8):

$$\frac{[p - A_\alpha(t)]^2}{2} = \frac{\left[p - eA_\gamma(t) - \frac{ie^2}{\omega^2} E_{vac}^2 \sin(\omega t) \int_{t_0}^{t} d\tau\, v(\tau) \sin(\omega \tau)\right]^2}{2} \tag{V.17}$$

VI. Newtonian trajectories in the effective photon statistics force

In this section, we attribute physical meaning to the effective photon statistics force, using the effective semi-classical SFA dipole moment:

$$S_{eff}(p, t', t) = \int_{t'}^{t} dt'' \frac{\left[p - eA_\gamma(t'') - \frac{ie^2}{\omega^2} E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega \tau)\right]^2}{2}$$

$$z_{eff}(t) = \frac{i}{\hbar} \int d^3\boldsymbol{p} \int_0^t dt' E_\gamma(t) d_z\left(p - A_\gamma(t')\right) d_z^*\left(p - A_\gamma(t)\right) \cdot \exp\left(-iS_{eff}(p, t, t')\right) + \text{c.c.} \tag{VI.1}$$

$$S_{\text{eff}}(\boldsymbol{p},t,t') = \int_{t'}^{t} dt'' \frac{\left[p - eA_\gamma(t'') - e\frac{i}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau)\right]^2}{2} + I_p \quad \text{(VI.2)}$$

Expanding the exponent:

$$\begin{aligned} &\exp\left(-iS_{eff}(p,t,t')\right) \\ &\qquad \approx \exp\left(-iS_\gamma(p,t,t')\right) \qquad \text{(VI.3)} \\ &\qquad \times \exp\left(-\frac{E_{vac}^2 e^2}{\omega^2} v(t'') \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau)\right) \end{aligned}$$

$$S_{eff}(p,t',t) = \int_{t'}^{t} dt'' \, \frac{\left[p - eA_\gamma(t'') - \frac{ie^2}{\omega^2}\, E_{vac}^2 \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau)\right]^2}{2}$$

$$\begin{aligned} &\exp\left(-iS_{eff}(p,t,t')\right) \\ &\qquad = \exp\left(-i\int_{t'}^{t} dt'' \, \frac{\left[p - eA_\gamma(t'')\right]^2}{2} - \frac{e^2}{\omega^2}\, E_{vac}^2\, v(t'') \sin(\omega t'') \int_{t'}^{t''} d\tau\, v(\tau) \sin(\omega\tau)\right) \end{aligned}$$

Therefore, the effective photon statistics force results in exponential decay/ growth of the dipole moment, with the rate of decay proportional to the velocity.

VII. Enhancement of multi-photon processes

Quantum states of light such as squeezed vacuum and stochastic light are known to enhance multiphoton processes, which compete with high harmonic generation. For example, stochastic and squeezed vacuum light enhance multiphoton ionization, which may result in bound state depletion[3,45]. Enhancement of multiphoton processes of order n scales as the n'th order coherence $g^{(n)}(0)$ of the driving light. Stochastic light enhances multiphoton ionization because its n'th order coherence is given by $g^{(n)} = n!$. Squeezed vacuum light enhances multiphoton ionization because it's n'th order coherence is given by $g^{(n)} = (2n-1)!!$.

In this paper, we explore HHG driven by squeezed coherent light, in a regime where only minute bunching occurs and $g^{(n)} \approx 1$, thus multi-photon processes are not enhanced. The n'th order coherence $g^{(n)}$ of squeezed coherent light $\left|\beta, re^{i\phi}\right\rangle$ is given by (reference [6], equation 3.27):

$$g^{(n)}(\tau = 0) = \frac{G_{nn}}{G_{11}^n}$$

$$G_{nn} = (n!)^2 \sum_{k=0}^{n} \left| H_k \left(\frac{iW}{\sqrt{2S}} \right) \right|^2 \frac{|S|^k M^{n-k}}{2^k (k!)^2 (n-k)!}$$

$$M = \frac{1}{2} \left[(4|S|^2 + 1)^{1/2} - 1 \right]$$

$$S = e^{i\phi} \cosh(r) \sinh(r) = e^{i\phi} (\sinh^2(r) + e^{-r} \sinh(r)) \approx e^{i\phi} \sinh^2(r)$$

$$W = \beta$$

in which $H_k(x)$ are Hermite polynomials, and W, S, M are coefficients related to $|\beta, r\rangle$. In our paper, we operate in a regime where $I_{coh} \gg I_{vac}$, i.e., the coherent component of the light is much brighter compared to the component originating from squeezing. In this limit $|\beta|^2 = |W|^2 \gg M, S$, the element G_{nn} becomes (ref[7] , equation 4.1):

$$G_{nn} \approx |W|^{2n} + \frac{n(n-1)}{2} |W|^{2n-4} \left(SW^{*2} + S^* W^2 \right) + n^2 |W|^{2n-2} M$$

For the cases discussed in the paper,

$$|W|^2 = N_{coh} \approx 10^{14}$$

$$S < 10^{12} = \frac{|W|}{100}$$

$$M \approx S < 10^{12} = \frac{|W|}{100}$$

Therefore,

$$G_{nn} \approx |W|^{2n} \left(1 + \frac{n(n-1)}{2} |W|^{-4} \left(SW^{*2} + S^* W^2 \right) + n^2 |W|^{-2} M \right) \approx$$

$$\approx |W|^{2n} \left(1 + \frac{n(n-1)}{2} \frac{|W|^{-1}}{100} + \frac{n^2 |W|^{-1}}{100} \right) \approx$$

$$\approx |W|^{2n} \left(1 + 10^{-14} \left(\frac{n(n-1)}{2 \times 100} + \frac{n^2}{100} \right) \right) \approx |W|^{2n}$$

We have $G_{nn} \approx |W|^{2n}$, and $g^{(n)} = \frac{G_{nn}}{G_{11}^n} = \frac{|W|^{2n}}{(|W|^2)^n} = 1$, i.e., no bunching occurs, and therefore enhancement of multiphoton ionization is negligible and the strong-field approximation is valid.

Supplementary References

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