

Supplementary Information for "Above-threshold ionization with X-ray free-electron lasers"

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Supplementary Note 1: Three-Dimensional Emission

The process of strong-field ionization can be described in a comprehensive three-dimensional scenario, closely mirroring the methodology employed in the one-dimensional analysis. The results presented here are consistent with the model outlined in Ref. [1]. However, it's important to note that these earlier results were derived for the general case of elliptical polarization. The current discussion simplifies these results to the case of linear polarization. This simplification allows for a more straightforward comparison with one-dimensional results presented in the main text. In the following discussion, we will walk through the steps and highlight any deviations from the idealized one-dimensional behavior as we go.

Our results are obtained directly from the first-order transition amplitude (Eqn. (30) of the main text):

$$\mathcal{M}^{(1)}(\mathbf{k}, t) \equiv \int_0^t dt_i e^{(i/\hbar)S(\mathbf{k}, t_i)} \left(-\frac{\partial}{\partial t_i} \right) \tilde{\phi}_0(\mathbf{k}, \mathbf{A}(t_i)). \quad (\text{S1})$$

This equation incorporates the bound-state contribution, denoted as $\tilde{\phi}_0(\mathbf{k}, \mathbf{A}(t_i))$, which is defined as:

$$\tilde{\phi}_0(\mathbf{k}, \mathbf{A}(t_i)) \equiv \int d\mathbf{r}_i \overline{\phi_{\mathbf{k}}^{(-)}(\mathbf{r}_i)} e^{-\frac{i|\mathbf{e}|}{\hbar c} \mathbf{A}(t_i) \cdot \mathbf{r}_i} \phi_0(\mathbf{r}_i) \quad (\text{S2})$$

The action, $S(\mathbf{k}, t_i)$, is given by:

$$S(\mathbf{k}, t_i) = \int_0^{t_i} d\tau \left[\frac{\mathbf{p}(\tau)^2}{2m_e} + I_p \right]. \quad (\text{S3})$$

In these equations, $\mathbf{A}(t) = -(c/\omega)\mathbf{E}\sin(\omega t)$ defines the vector potential, with $\mathbf{E}(t) \equiv -(1/c)\partial_t\mathbf{A}(t)$ and kinetic momentum $\mathbf{p}(t) \equiv \hbar\mathbf{k} + (|e|/c)\mathbf{A}(t)$. The initial bound state is represented as $\phi_0(x) = R_0(r)Y_{l_0}^{m_0}(\hat{\mathbf{r}})e^{(i/\hbar)I_{\text{p}}t}$, and we consider linearly polarized fields, $\mathbf{E}(t) \equiv \mathbf{E}\cos(\omega t)$, through the interaction $V_L(x) \equiv |e|\mathbf{E}(t) \cdot \mathbf{r}$. Here, $Y_l^m(\hat{\mathbf{r}})$ are the spherical harmonics, and the orbital angular momentum serves as the generalization of parity to three dimensions.

Supplementary Note 1.1: System-Dependent Properties

In order to describe the bound-state properties of the system, we evaluate the vector potential contribution, $\tilde{\phi}_0(\mathbf{k}, \mathbf{A}(t_i))$, using the following equation, which is similar to the one used previously:

$$e^{-\frac{i|e|}{\hbar c}\mathbf{A}(t_i) \cdot \mathbf{r}_i} = \sum_{n_A=-\infty}^{\infty} (-1)^{n_A} J_{n_A}(\mathbf{k}_A \cdot \mathbf{r}_i) e^{-in_A\omega t_i} \quad (\text{S4})$$

This equation includes an expansion, which describes emission in directions other than just along the field polarization [2]:

$$J_{n_A}(\mathbf{k}_A \cdot \mathbf{r}_i) = \sum_{N=|n_A|, |n_A|+2, \dots} \frac{\text{sign}(n_A)(-1)^{(N-|n_A|)/2}(\hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i)^N}{[(N-n_A)/2]![(N+n_A)/2]!} \left(\frac{k_A r_i}{2}\right)^N \quad (\text{S5})$$

Now, we have $-1 \leq \hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i \leq 1$ instead of $\hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i = \pm 1$ as before.

The angular contributions can be expressed in terms of the Legendre polynomials, $P_{l_A}(\hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i)$:

$$(\hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i)^N = \sum_{l_A=N, N-2, \dots, \text{mod}(N,2)} \frac{N!(2l_A+1)P_{l_A}(\hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i)}{2^{(N-l_A)/2}[(N-l_A)/2]!(N+l_A+1)!!} \quad (\text{S6})$$

obtained from Ref. [3].

This gives us:

$$J_{n_A}(\mathbf{k}_A \cdot \mathbf{r}_i) = \sum_{l_A=q_A, q_A+2, \dots} (-1)^{(M_A-n_A)/2} f_{l_A}^{(n_A)}(k_A r_i) \sum_{m_A=-l_A}^{l_A} \overline{Y_{l_A}^{m_A}(\hat{\mathbf{k}}_A)} Y_{l_A}^{m_A}(\hat{\mathbf{r}}_i) \quad (\text{S7})$$

This equation is resolved in terms of the angular-momentum quantum numbers, l_A and m_A , with radial contributions given by:

$$f_{l_A}^{(n_A)}(k_A r_i) \equiv f_{l_A, M_A}^{(n_A)}(k_A r_i) {}_3F_4\left(\frac{M_A+2}{2}, \frac{M_A+1}{2}, 1\right)$$

$$; \frac{M_A - n_A + 2}{2}, \frac{M_A + n_A + 2}{2}, \frac{M_A - l_A + 2}{2}, \frac{M_A + l_A + 3}{2}; - \left(\frac{k_A r_i}{2} \right)^2 \Big) \quad (\text{S8})$$

Here, we have:

$$f_{l_A, M_A}^{(n_A)}(k_A r_i) \equiv \frac{(4\pi)M_A!(\sqrt{2})^{l_A}}{[(M_A - n_A)/2]![(M_A + n_A)/2]![(M_A - l_A)/2]!(M_A + l_A + 1)!!} \left(\frac{k_A r_i}{2^{3/2}} \right)^{M_A} \quad (\text{S9})$$

And:

$$P_{l_A}(\hat{\mathbf{k}}_A \cdot \hat{\mathbf{r}}_i) = \frac{4\pi}{2l_A + 1} \sum_{m_A = -l_A}^{l_A} \overline{Y_{l_A}^{m_A}(\hat{\mathbf{k}}_A)} Y_{l_A}^{m_A}(\hat{\mathbf{r}}_i) \quad (\text{S10})$$

from Ref. [4].

This expansion has been applied to the previous equations. This comprehensive set of equations allows us to describe the bound-state properties of the system in detail.

In addition, the vector potential momentum is defined as $\hbar \mathbf{k}_A \equiv |e| \mathbf{E} / \omega$, with $M_A \equiv \max(l_A, |n_A|)$ and $q_A \equiv \text{mod}(n_A, 2)$. We also introduce the generalized hypergeometric function [2]

$${}_pF_q(a_1, a_2, \dots, a_p; b_1, b_2, \dots, b_q; z) \equiv \sum_{m=0}^{\infty} \frac{(a_1)_m (a_2)_m \dots (a_p)_m}{(b_1)_m (b_2)_m \dots (b_q)_m} \frac{z^m}{m!} \quad (\text{S11})$$

where $(a)_m \equiv \Gamma(a + m) / \Gamma(a)$ is the Pochhammer symbol and $\Gamma(a)$ is the Euler integral.

It's important to note that $Y_{l_A}^{m_A}(\hat{\mathbf{r}}_i)$ signifies the angular momentum transfer and $e^{-in_A \omega t}$ represents the total energy transfer $n_A \hbar \omega$. We further expand the continuum state as

$$\phi_{\mathbf{k}}^{(-)}(\mathbf{r}_i) = \frac{1}{k} \sum_{l_k=0}^{\infty} (i)^{l_k} e^{-i\eta_{l_k}(k)} R_{k, l_k}(r_i) \sum_{m_k=-l_k}^{l_k} \overline{Y_{l_k}^{m_k}(\hat{\mathbf{k}})} Y_{l_k}^{m_k}(\hat{\mathbf{r}}_i) \quad (\text{S12})$$

In this equation, $R_{k, l_k}(r_i)$ is the radial continuum state with phase shift $\eta_{l_k}(k)$. Here, $\phi_{\mathbf{k}}^{(-)}(\mathbf{r}_i)$ describes scattering states which are asymptotically described at $r \rightarrow \infty$ by a plane wave plus an ingoing spherical wave [5, 6]. The normalization condition

$$\int_0^{\infty} dr r^2 R_{k, l}(r) R_{k', l}(r) = \delta(k - k') \quad (\text{S13})$$

is enforced and scattering phase-shifts are determined from

$$R_{k,l_k}(r_i) \xrightarrow{r_i \rightarrow \infty} \frac{1}{r_i} \sqrt{\frac{\pi}{2}} \sin(kr_i - \eta \log(2kr_i) - l_k \pi/2 + \eta_{l_k}(k)) \quad (\text{S14})$$

with Sommerfeld parameter $\eta \equiv -Z_{\text{ion}}|e|^2/\hbar v$ as before.
Therefore, we can evaluate

$$\begin{aligned} \left(-\frac{\partial}{\partial t}\right) \tilde{\phi}_0(\mathbf{k}, \mathbf{A}(t)) &= (i/\hbar) \sum_{n_A=-\infty}^{\infty} (n_A \hbar \omega) e^{-in_A \omega t} \sum_{l_k, m_k} K_{l_k}(k) Y_{l_k}^{m_k}(\hat{\mathbf{k}}) \\ &\quad \times \sum_{l_A}^I A_{l_A}^{m_A(n_A)} (Y_{l_A}^{m_A})_{l_0, m_0}^{l_k, m_k} I_{l_k, l_A, l_0}^{(n_A)}(k) \end{aligned} \quad (\text{S15})$$

with

$$\begin{aligned} A_{l_A}^{m_A(n_A)} &\equiv (-1)^{(M_A+n_A)/2} \overline{Y_{l_A}^{m_A}(\hat{\mathbf{k}}_A)}, \quad K_{l_k}(k) \equiv (-i)^{l_k} e^{i\eta_{l_k}(k)}, \\ m_A &\equiv m_k - m_0 \quad \text{and} \quad \sum_{l_A}^I \equiv \sum_{l_A \in \{q_A, q_A+2, \dots\} \cap \{|m_A|, |m_A|+1, \dots\}} \end{aligned} \quad (\text{S16})$$

where the sum is over all $l_A = q_A, q_A + 2, \dots$ satisfying $l_A \geq |m_A|$.
The angular integrals

$$(Y_{l_A}^{m_A})_{l_0, m_0}^{l_k, m_k} \equiv \int d\Omega_{\mathbf{r}_i} \overline{Y_{l_k}^{m_k}(\hat{\mathbf{r}}_i)} Y_{l_A}^{m_A}(\hat{\mathbf{r}}_i) Y_{l_0}^{m_0}(\hat{\mathbf{r}}_i) \quad (\text{S17})$$

describe the corresponding angular momentum selection rules which are evaluated as Wigner-3j symbols.

The radial integral

$$I_{l_k, l_A, l_0}^{(n_A)}(k) \equiv \frac{1}{k} \int_0^\infty dr_i r_i^2 R_{k, l_k}(r_i) f_{l_A}^{(n_A)}(k r_i) R_0(r_i) \quad (\text{S18})$$

determines how the initial state influences the photo-electron distribution and is numerically evaluated in the current work. Additional dimensions results in additional ionization channels.

In the weak field limit, where γ is much greater than 1, we can approximate the function $f_{l_A}^{(n_A)}(r_i/\gamma a_0)$ with its lowest-order polynomial contribution. This approximation is similar to previous methods used. However, as the fields intensify and γ approaches or becomes less than 1, additional higher-order contributions become significant. Consequently, the emission exhibits a qualitatively distinct behavior.

Supplementary Note 1.2: System-Independent Properties

The action factor, given by

$$e^{(i/\hbar)S(\mathbf{k}, t_i)} = e^{-i\mathbf{k} \cdot \boldsymbol{\xi}(0)} e^{iN(k)\omega t_i} e^{i(Z/2) \sin(-2\omega t_i)} e^{i\mathbf{k} \cdot \boldsymbol{\xi}(t_i)} \quad (\text{S19})$$

is evaluated using the following equation, similar to the one used previously:

$$e^{i\mathbf{k}\cdot\boldsymbol{\xi}(t_i)} = \sum_{n_S=-\infty}^{\infty} (i)^{n_S} J_{n_S}(\mathbf{k}\cdot\boldsymbol{\xi}) e^{-in_S\omega t_i} \quad (\text{S20})$$

In this equation, $\hbar\mathbf{k}$ represents the momentum of the liberated electrons, and the action, $S(\mathbf{k}, t_i)$, is given by:

$$S(\mathbf{k}, t_i) = N(k)\hbar\omega t_i + (\hbar\mathcal{Z}/2)\sin(-2\omega t_i) + \hbar\mathbf{k}\cdot[\boldsymbol{\xi}(t_i) - \boldsymbol{\xi}(0)] \quad (\text{S21})$$

The phase factor is expanded as follows:

$$e^{i(\mathcal{Z}/2)\sin(-2\omega t_i)} = \sum_{a=-\infty}^{\infty} J_a(\mathcal{Z}/2) e^{-2ia\omega t_i} \quad (\text{S22})$$

And the Bessel functions are expanded as:

$$J_{n_S}(\mathbf{k}\cdot\boldsymbol{\xi}) = \sum_{l_S=q_S, q_S+2, \dots} (-1)^{(M_S-n_S)/2} f_{l_S}^{(n_S)}(k\xi) \sum_{m_S=-l_S}^{l_S} \overline{Y_{l_S}^{m_S}(\hat{\boldsymbol{\xi}})} Y_{l_S}^{m_S}(\hat{\mathbf{k}}) \quad (\text{S23})$$

Here, $f_{l_S}^{(n_S)}(k\xi)$ is evaluated as in Eqn. (S8), but with $q_S \equiv \text{mod}(n_S, 2)$ and $M_S \equiv \max(|n_S|, l_S)$.

$N(k) \equiv (\varepsilon_k + \tilde{I}_p)/\hbar\omega$ describes the number of photons of energy $\hbar\omega$ required for an electron to overcome the field-shifted ionization potential and enter the continuum at energy $\varepsilon_k \equiv \hbar^2 k^2/2m_e$ as before. $\mathcal{Z}$ is again the continuum-state intensity parameter and $\boldsymbol{\xi}(t) \equiv \boldsymbol{\xi} \cos(\omega t)$, with $\boldsymbol{\xi} \equiv |e|\mathbf{E}/m_e\omega^2$, describes the quiver motion of a free electron.

Given a certain number of quanta n_A from the vector potential term, we can express the integral as follows:

$$\begin{aligned} \int_0^t dt_i e^{(i/\hbar)S(\mathbf{k}, t_i) - in_A\omega t_i} = \\ e^{-i\mathbf{k}\cdot\boldsymbol{\xi}(0)} \sum_{n_S=-\infty}^{\infty} \sum_{l_S=q_S, q_S+2, \dots} \sum_{m_S=-l_S}^{l_S} X_{l_S}^{m_S(n_S)}(k) Y_{l_S}^{m_S}(\hat{\mathbf{k}}) \\ \times \sum_{a=-\infty}^{\infty} J_a(\mathcal{Z}/2) \delta_t([N(k) - n]\omega/2) \end{aligned} \quad (\text{S24})$$

where n is defined as $n \equiv n_A + n_S + 2a$, and the coefficients $X_{l_S}^{m_S(n_S)}(k)$ are given by:

$$X_{l_S}^{m_S(n_S)}(k) \equiv (i)^{M_S} f_{l_S}^{(n_S)}(k\xi) \overline{Y_{l_S}^{m_S}(\hat{\boldsymbol{\xi}})}. \quad (\text{S25})$$

The shape term, defined as:

$$\delta_t(x) \equiv e^{ixt} \text{sinc}(xt)t \quad (\text{S26})$$

describes the distribution of final energy states after the absorption of $n_A + n_S + 2a$ photons by a finite flat-top pulse.

In the scenario of a weak field limit, where $\mathcal{Z}$ is significantly less than 1, we can substitute the functions $J_a(\mathcal{Z}/2)$ and $f_{l_S}^{(n_S)}(k_n \xi)$ with their respective lowest-order polynomial contributions, akin to our previous methods. However, as the fields strengthen and $\mathcal{Z}$ nears 1, the impact of additional higher-order contributions cannot be ignored. This is analogous to the findings from our one-dimensional analysis.

Supplementary Note 1.3: Amplitude and Yields

When we combine all contributions, we obtain the ionization amplitude:

$$\begin{aligned} \mathcal{M}^{(1)}(\mathbf{k}, t) = & e^{-i\mathbf{k} \cdot \boldsymbol{\xi}(0)} (i/\hbar) \sum_{l_k, m_k} K_{l_k}(k) \sum_{n_A=-\infty}^{\infty} (n_A \hbar \omega) \\ & \times \sum_{l_A}^l A_{l_A}^{m_A(n_A)} (Y_{l_A}^{m_A})_{l_0, m_0}^{l_k, m_k} I_{l_k, l_A, l_0}^{(n_A)}(k) \sum_{n_S=-\infty}^{\infty} \sum_{l_S=q_S, q_S+2, \dots} \sum_{m_S=-l_S}^{l_S} X_{l_S}^{m_S(n_S)}(k) \\ & \times \sum_l (Y_{l_S}^{m_S})_{l_k, m_k}^{l, m} Y_l^m(\hat{\mathbf{k}}) \sum_{a=-\infty}^{\infty} J_a(\mathcal{Z}/2) \delta_t([N(k) - n]\omega/2) \quad (\text{S27}) \end{aligned}$$

where we have applied:

$$Y_{l_S}^{m_S}(\hat{\mathbf{k}}) Y_{l_k}^{m_k}(\hat{\mathbf{k}}) = \sum_{l=\max(|l_k-l_S|, |m|)}^{l_k+l_S} (Y_{l_S}^{m_S})_{l_k, m_k}^{l, m} Y_l^m(\hat{\mathbf{k}}) \quad (\text{S28})$$

with

$$\sum_l \equiv \sum_{l=\max(|l_k-l_S|, |m|)}^{l_k+l_S}, \quad (\text{S29})$$

and $m \equiv m_k + m_S$. The yield is given by:

$$P^{(\text{ion})}(t) = \int d\mathbf{k} |\mathcal{M}(\mathbf{k}, t)|^2. \quad (\text{S30})$$

In the multi-dimensional theory, a broader scope of possibilities unfolds compared to the one-dimensional theory. Each channel (l, m) corresponds to the absorption of n photons, resulting in channels with definite parity $\text{mod}(l_0 + n, 2)$, mirroring patterns established in previous analyses.

In the one-dimensional scenario, a two-photon process typically leads to the emission of an s -state ($l = 0$). Conversely, in the three-dimensional theory, both s -states ($l = 0$) and d -states ($l = 2$) populate the continuum.

When restricting ourselves to linear polarization along the quantization axis ($\mathbf{E} \propto \mathbf{k}_A \propto \boldsymbol{\xi} \propto \hat{\mathbf{z}}$), we find:

$$\overline{Y_{l_A}^{m_A}(\hat{\mathbf{k}}_A)} = \sqrt{\frac{2l_A + 1}{4\pi}} \delta_{m_A,0} \quad (\text{S31})$$

and similarly:

$$\overline{Y_{l_S}^{m_S}(\hat{\boldsymbol{\xi}})} = \sqrt{\frac{2l_S + 1}{4\pi}} \delta_{m_S,0} \quad (\text{S32})$$

These conditions lead to the final simplified amplitude:

$$\begin{aligned} \mathcal{M}^{(1)}(\mathbf{k}, t) = & e^{-i\mathbf{k} \cdot \boldsymbol{\xi}(0)} (i/\hbar) \sum_{n_A} (n_A \hbar \omega) \sum_{l_A}^l A_{l_A}^{0(n_A)} \sum_{l_k} K_{l_k}(k) (Y_{l_A}^0)^{l_k, m_0} I_{l_k, l_A, l_0}^{(n_A)}(k) \\ & \times \sum_{n_S} \sum_{l_S}^l X_{l_S}^{0(n_S)}(k) \sum_l (Y_{l_S}^0)^{l, m_0} Y_l^{m_0}(\hat{\mathbf{k}}) \sum_a J_a(Z/2) \delta_t([N(k) - n]\omega/2). \end{aligned} \quad (\text{S33})$$

Transitioning to the long-pulse limit $t \rightarrow \infty$, we derive the photo-electron momentum distribution:

$$W(\mathbf{k}) \approx \lim_{t \rightarrow \infty} t^{-1} |\mathcal{M}^{(1)}(\mathbf{k}, t)|^2 \equiv \sum_{n=n_{\text{th}}}^{\infty} |\mathcal{M}_n^{(1)}(\hat{\mathbf{k}})|^2 \delta(k - k_n) \quad (\text{S34})$$

with angular contributions:

$$\begin{aligned} \mathcal{M}_n^{(1)}(\hat{\mathbf{k}}) = & e^{-i\mathbf{k}_n \cdot \boldsymbol{\xi}(0)} (i/\hbar) \sqrt{\frac{2\pi}{v_n}} \sum_{n_A} (n_A \hbar \omega) \sum_{l_A}^l A_{l_A}^{0(n_A)} \\ & \times \sum_{l_k} K_{l_k}(k_n) (Y_{l_A}^0)^{l_k, m_0} I_{l_k, l_A, l_0}^{(n_A)}(k_n) \sum_a J_a(Z/2) \sum_{l_S}^l X_{l_S}^{0(n_S)}(k_n) \sum_l (Y_{l_S}^0)^{l, m_0} Y_l^{m_0}(\hat{\mathbf{k}}) \\ & \equiv e^{-i\mathbf{k}_n \cdot \boldsymbol{\xi}(0)} \sum_l C_l^{m_0}(k_n) Y_l^{m_0}(\hat{\mathbf{k}}) \end{aligned} \quad (\text{S35})$$

pertaining to each photon process $\hbar k_n = \sqrt{2m_e(n\hbar\omega - \tilde{I}_p)}$, with electron mass m_e and $n_S \equiv n - n_A - 2a$.

Integrating over $\mathbf{k}$ yields the total ionization rate:

$$w = \int d\mathbf{k} W(\mathbf{k}) = \sum_{n=n_{\text{th}}}^{\infty} \sum_l w_l^{m_0}(k_n) = \sum_{n=n_{\text{th}}}^{\infty} w_n \quad (\text{S36})$$

with partial contributions:

$$w_l^{m_0}(k_n) \equiv k_n^2 |C_l^{m_0}(k_n)|^2. \quad (\text{S37})$$

These findings closely mirror the steps undertaken in the one-dimensional analysis at each stage. The intuitive understanding is preserved, and the increased dimensionality leads to a greater number of ionization channels. In practical calculations, the evaluation of ${}_3F_4(\cdot)$ may present difficulties. The earlier representation of these results, as detailed in Ref. [1], might provide enhanced efficiency, contingent upon the specific application and the software package used for evaluating special functions.

Supplementary References

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