



LETTER

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Quantum fluctuations in strong field ionization

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Abstract – Based on the field quantization, the angular distribution of the ionization rate of hydrogen atoms with the effect of a finite number of photons is computed in this letter. We find that the angular distribution of the ionization rate will fluctuate with the initial photon number of the field, and have a peak value higher than that predicted by Keldysh-Faisal-Reiss theory. This ionization peak is the embodiment of quantum effect. Furthermore, our calculations show that the phenomenon of ionization peak also exists when the initial field state is a coherent state.

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Introduction. – The interaction between the intense laser and matter [1] has always been a hot topic. Meanwhile, the leap-forward development of high-power laser technology in the past four decades [2] has enabled many significant theoretical predictions to be realized successively, such as the famous Kapitza-Dirac (KD) effect [3–5] and high-order multiphoton Thomson scattering (HMTS) [6-8], etc. We notice that the number of photons in these processes is actually assumed large enough, such that the light field strictly described by the quantum state [9] will not be perturbed during the mutual process. However, for the laser produced in the laboratory, its coherence is strongly restricted [10–12]. Therefore, for some physical processes involving lasers, it is necessary to investigate the influence of the initial photon number l of the field on the physical results. This paper will take the strong field ionization of hydrogen atoms that we are interested in as an example, and give some corresponding results.

Keldysh made a pioneering work on describing the ionization of atoms induced by intense electromagnetic fields in 1965 [13]. Thereafter, Faisal and Reiss [14,15] carried forward Keldysh's work. Their work is commonly known as Keldysh-Faisal-Reiss (KFR) theory, which has been widely accepted as the mainstream theory of atomic ionization in the strong laser field. KFR theory can not only successfully explain the spectral line characteristics [16] of above-threshold ionization (ATI) [17], but also describe the phenomenon of tunneling ionization well [13,18]. However, the electromagnetic field $A^{\mu}(x)$ in KFR theory is regarded as a classical field, and it may cause a part of quantum effects to be concealed. Therefore, from the perspective of field quantization, KFR theory is still semiclassical, not a full quantum theory. The strong field ionization theory based on quantum electrodynamics (QED) has been established by Guo *et al.* [19–22]. With the field quantization, they obtained the same results as the semiclassical theory [15,23,24] by setting $l \to \infty$.

In this letter, we consider such a situation where l is not infinite, but finite. Then, the angular distribution $\frac{dW}{d\Omega}$ (see fig. 1) of the ionization rate of ground-state hydrogen atoms with the effect of infrared circularly polarized light ($\lambda = 912.44 \text{ nm}$) can be obtained numerically. Our results reveal how $\frac{dW}{d\Omega}$ will fluctuate with l and the fluctuation is very obvious when l is small.

Model and method. – The motion equation (using units $\hbar = c = 1$) for a single electron in the light field is

$$[i\partial \!\!\!/ + eA\!\!\!/(x) - m]\psi(x) = 0, \tag{1}$$

where -e is the electron charge and e > 0. m is the electron rest mass. \not{a} is the Feynman slash notation, representing $g_{\mu\nu}\gamma^{\mu}a^{\nu}(\mu,\nu=0,1,2,3)$; γ^{μ} stands for 4×4 Dirac matrices and the space-time metric $g^{\mu\nu} =$ diag(1, -1, -1, -1). The single-mode circularly polarized light field is used in this letter and it can be expanded as $A_{\mu}(x) = g(\varepsilon_{\mu}a e^{-ikx} + \varepsilon_{\mu}^*a^{\dagger} e^{ikx})$. Here, $g = (2\omega V)^{-1/2}$, is the normalization factor of the light field. $k = (\omega, 0, 0, \omega)$. ω is the angular frequency of photons and V is the space volume occupied by l initial-state photons. $\varepsilon = (0, \varepsilon)$, where $\varepsilon = e_x \cos(\xi/2) + ie_y \sin(\xi/2)$ and $\xi = \pi/2$ corresponds to circularly polarized light. a^{\dagger} and a are one pair of creation and annihilation operators for photons.

According to the spirit of strong field approximation (SFA) [25] and formal scattering theory [26], the transition

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Fig. 1: The diagram of interaction between the ground-state hydrogen atom and the photon flux containing l photons. W is the ionization rate and θ represents the angle between the momentum of photoelectron and the laser propagation direction.

matrix element S_{fi} of the ground-state electron in the hydrogen atom is written as

$$S_{fi} = -i \int_{-\infty}^{+\infty} \mathrm{d}t \langle \psi_{pns}(x) | - e\gamma^0 \mathcal{A}(x) | \Psi(x), l \rangle.$$
 (2)

In eq. (2), $|\psi_{pns}(x)\rangle$ is the solution [19–22] of eq. (1), namely the Volkov state [27], and its expression is given in detail in the Supplementary Material Supplementarymaterial.pdf (SM). p is the fourdimensional momentum of the photoelectron; n is a natural number and $s = \pm \sin \xi$. $|\Psi(x), l\rangle = \Psi(x) \otimes |l\rangle$ represents the direct product of the ground state of one atom $\Psi(x)$ and the photon-number state $|l\rangle$. The relativistic normalized ground-state hydrogenic wave functions for spin-up and spin-down states in spherical coordinates are given by Bethe and Salpeter [28] as

$$\Psi^{\uparrow}(r,\theta',\phi,t) = Mr^{\tau-1} \begin{pmatrix} 1\\ 0\\ \frac{i(1-\tau)}{\alpha}\cos\theta'\\ \frac{i(1-\tau)}{\alpha}\sin\theta'e^{i\phi} \end{pmatrix} e^{-iE_{0}t},$$

$$\Psi^{\downarrow}(r,\theta',\phi,t) = Mr^{\tau-1} \begin{pmatrix} 1\\ \frac{i(1-\tau)}{\alpha}\sin\theta'e^{-i\phi}\\ -\frac{i(1-\tau)}{\alpha}\cos\theta' \end{pmatrix} e^{-iE_{0}t},$$
(3)

where

$$M = \frac{(2m\alpha)^{\tau+1/2}}{(4\pi)^{1/2}} e^{-m\alpha r} \left(\frac{1+\tau}{2\Gamma(1+2\tau)}\right)^{1/2}, \qquad (4)$$
$$\tau = (1-\alpha^2)^{1/2},$$

The above α is the fine-structure constant; E_0 is the ground-state energy of the hydrogen atom.

If the initial hydrogen atom is not polarized, it should be described by the mixed state. So when calculating the ionization rate W, we should average the initial spin, that is,

$$W = \frac{1}{2} \lim_{T \to \infty} \frac{\mathrm{d}}{\mathrm{d}T} \sum_{\boldsymbol{p}} \sum_{n} \sum_{s_f} \sum_{s_i} |S_{fi}|^2, \qquad (5)$$

where s_i and s_f represent the initial and final spin of the electron, respectively; $\frac{1}{2}$ comes from averaging the initial spin; p is the momentum of the photoelectron.

spin; p is the momentum of the photoelectron. So far, the precise result of $\frac{dW}{d\Omega}$ can be obtained numerically through eq. (5) and the detailed calculation process of eq. (5) is shown in the SM. In the calculations hereafter, atomic units will be adopted.

Results. – First, we define the parameter as follows

$$I = 4\pi \frac{l\omega}{V}.$$
 (6)

We note that the corresponding realistic laser intensity $I_r = 137 l \omega / V$ [29], since $I_r = 137 I / (4\pi)$. For the given parameters l, I, θ and ω , we can numerically calculate $\frac{dW}{d\Omega}$ in the non-relativistic limit and long wavelength approximation. Since we are using circularly polarized light, $\frac{dW}{d\Omega}$ only depends on θ . Besides, $\frac{dW}{d\Omega}$ is symmetric about the XY-plane (the direction of laser propagation is agreed to be the Z-axis), that is, $\frac{dW}{d\Omega}|_{\theta} = \frac{dW}{d\Omega}|_{\pi-\theta}$. As shown in fig. 2(a), the change curve of $\frac{dW}{d\Omega}$ with θ

relies on l. From it, we can see that as l increases, color curves gradually approach the black curve predicted by KFR theory. This phenomenon is not surprising, because the fundamental principles of quantum mechanics suggest that quantum theory tends to be classical in the large quantum number limit $l \to \infty$. However, the black curve is not always higher than the color ones. For example, when $\theta \lesssim 57.3$ degrees, the red curve is higher than the black one. Corresponding to $\theta = 0.9$ rad (51.6 degrees), we give the variation curve of $\frac{dW}{d\Omega}$ as a function of l (see fig. 2(b)). What is interesting is that $\frac{dW}{d\Omega}$ first grows to a peak higher than the value predicted by KFR theory in the range of small photon number and then it decays, which is a novel quantum fluctuation effect. Furthermore, our calculations show that when θ is very small, the peak value of the curve is much larger than $\frac{dW}{d\Omega}$ predicted by KFR theory, even several orders of magnitude higher (see fig. 4 for details), which seems incredible. Nevertheless, laser pulses produced in modern physics laboratory generally contain a large number of photons $(l > 10^5)$ [29], which makes it difficult for us to detect the fluctuation of $\frac{dW}{d\Omega}$.

Also, with I and θ as variables, we calculated the photon number N_p at the peak of the curve. The results are shown in fig. 3. We can clearly see that N_p increases with θ when I is constant. However, it is a pity that the data in the blank parameter area in fig. 3(a) could not be obtained due to computing resource constraints. This makes it impossible for us to directly judge whether ionization



Fig. 2: The numerical results of $\frac{dW}{d\Omega}$. The parameters ω and I are 0.05 a.u. and 0.0029 a.u., respectively. (a) The change curve of $\frac{dW}{d\Omega}$ with θ , wherein the black and color curves represent the prediction values of $\frac{dW}{d\Omega}$ by KFR theory and our model, respectively. (b) Here $\theta = 0.9$ rad. The blue short-dashed line refers to the prediction result of our model, and it will fluctuate with the initial photon number l.



Fig. 3: Panel (a): the contour map corresponding to $\omega = 0.05$ a.u.. The parameters $I \in (0.0005, 0.0555]$ and $\theta \in (0, \pi/2]$. Here, $\Delta I = 0.0005$ a.u. and $\Delta \theta = \pi/200$ rad. As shown in fig. 2(b), N_p represents the photon number at the peak. Panel (b): N_p as a function of θ . It seems that N_p has an exponential growth when I is constant. This reflects the feature that the contours in (a) become denser as θ increases ($0 < \theta \le \pi/2$). In addition, this drawing also shows that the smaller the I, N_p the faster the growth.

peaks will also appear in the blank area. To this end, we can take

V

$$V_r = \left(\frac{\mathrm{d}W}{\mathrm{d}\Omega}\right)_{peak} \left/ \left(\frac{\mathrm{d}W}{\mathrm{d}\Omega}\right)_{KFR},\tag{7}$$

where $(\frac{dW}{d\Omega})_{peak}$ is the peak value of $\frac{dW}{d\Omega}$ (see fig. 2(b)), and $(\frac{dW}{d\Omega})_{KFR}$ represents the $\frac{dW}{d\Omega}$ predicted by KFR theory.



Fig. 4: $W_r - 1$ as a function of θ . Our purpose is to explore the behavior of W_r while $\theta \to \pi/2$, so only part of the curve of $W_r - 1$ are given here. The black dashed line is the fitting curve, corresponding to the analytic function $f(\theta) = b/\theta^c$.

Table 1: Here, we list the fitting parameters corresponding to the five curves in fig. 4.

I (a.u.)	b	С
0.01	34.649 ± 0.072	27.823 ± 0.081
0.02	1619.699 ± 26.838	36.464 ± 0.143
0.03	18029.060 ± 526.876	41.878 ± 0.186
0.04	118735.243 ± 4722.964	46.422 ± 0.220
0.05	566157.025 ± 27882.483	50.238 ± 0.251

In fig. 4, it is shown that $W_r - 1$ decreases with θ and gradually tends to 0 in the area close to $\pi/2$. This reflects the fact that the ionization peak phenomenon is very weak when θ ($\theta \leq \pi/2$) is large; on the contrary, it is fairly obvious. Through calculation, we find that $W_r(\theta) - 1$ can be well fitted by function

$$f(\theta) = \frac{b}{\theta^c}.$$
(8)

Here, b and c are the parameters of the fitting function. The relevant data is given in table 1.

Since the function $f(\theta) > 0$, $W_r(\theta) > 1$ when $0 < \theta \le \pi/2$. On these grounds, we could conclude that the ionization peak phenomenon will appear in all parameter areas in fig. 3(a). It means that, for the hydrogen atom, the fluctuation effect of $\frac{\mathrm{d}W}{\mathrm{d}\Omega}$ caused by QED is universal when $\omega = 0.05$ a.u..

The preceding discussion results are based on the initial state of the light field being the photon-number state $|l\rangle$, which is difficult to be achieved in the current laboratory [29,30]. Strictly speaking, the quantum state of the radiation field should be described by the so-called coherent state [9]

$$|\beta\rangle = e^{-\frac{1}{2}|\beta|^2} \sum_{n=0}^{\infty} \frac{\beta^n}{\sqrt{n!}} |n\rangle, \qquad (9)$$



Fig. 5: Here I = 0.0029 a.u. and $\omega = 0.05$ a.u.: (a) $\theta = 0.7$, (b) $\theta = 0.8$, (c) $\theta = 0.9$, and (d) $\theta = 1.0$ (rad).

where $\beta = \sqrt{l_{ave}}$ and l_{ave} is the mean photon number in the initial light field. The $|n\rangle$ state component in $|\beta\rangle$ follows the Poisson distribution

$$P_n = \frac{\beta^{2n}}{n!} e^{-\beta^2}.$$
 (10)

Let the transition matrix element corresponding to the coherent state be $S_{fi}(\beta)$, and it satisfies

$$S_{fi}(\beta) = -i \int_{-\infty}^{+\infty} dt \langle \psi_{pns}(x) | -e\gamma^0 \mathcal{A}(x) | \Psi(x), \beta \rangle$$
$$= \sum_{l=0}^{\infty} \sqrt{P_l} S_{fi}, \tag{11}$$

where $|\Psi(x),\beta\rangle = \Psi(x) \otimes |\beta\rangle$; S_{fi} is shown in eq. (2).

Due to the fact that S_{fi} contains delta function, we have

$$|S_{fi}(\beta)|^2 = \sum_{l=0}^{\infty} P_l |S_{fi}|^2.$$
 (12)

The above eq. (12) indicates that when the initial state of the light field is the coherent state $|\beta\rangle$, the ionization rate should be the expected value of the ionization rate of all photon-number states, and the corresponding probability distribution satisfies the Poisson distribution. In order to calculate $\frac{dW}{d\Omega}$ numerically, we need to truncate *l* in eq. (12) according to the characteristics of the Poisson distribution. Accordingly, we give a related case, and the results are shown in fig. 5. Here, we find that the results of the photon-number state and coherent state are almost the same. So it tells us that even if the initial state of the light field is the coherent state, there will still be a peak in the change curve of $\frac{dW}{d\Omega}$. Of course, there are some subtle differences. For example, compared with the photonnumber state, the ionization peak value corresponding to the coherent state will decrease.

Conclusions. – In this paper, the angular distribution of the ionization rate of hydrogen atoms under the action of a finite number of photons is obtained numerically with the field quantization. We find that with the curve of $\frac{dW}{dQ}$ varying with l there appears the phenomenon of ionization peak, that is, $\frac{dW}{d\Omega}$ suddenly increases to a peak, then slowly decreases and shows an asymptotic behavior, where the asymptotic value is close to the predicted value of KFR theory. This is obviously a quantum effect. Through data fitting, we prove that the phenomenon of ionization peak is general for hydrogen atoms when $\omega = 0.05$ a.u. In addition, this phenomenon also exists when the initial field state is a coherent state, which gives us the opportunity to verify it experimentally. Finally, it should be pointed out that our work in this letter is carried out in the nonrelativistic limit and with the effect of circularly polarized light. The angular distribution of the ionization rate of hydrogen atoms in other cases is worth exploring, and we look forward to more novel results.

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