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ATOMS, MOLECULES, OPTICS

THE EFFECT OF THE PHASE OF AN IONIZING ULTRASHORT LASER PULSE ON THE FORMATION OF QUANTUM VORTICES IN THE DENSITY DISTRIBUTION OF A PHOTOELECTRON

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Abstract. Quantum vortices formed by a photoelectron obtained as a result of over-barrier ionization of a two-dimensional hydrogen atom by an extremely short laser pulse are theoretically investigated. The sensitivity of quantum vortices to the initial phase of the ionizing field is analyzed. The interference effects responsible for the appearance of vortices are being clarified. For the model under consideration, the use of various gauges in describing the interaction of an electron with a field is discussed.

Keywords: *photoelectron*, *quantum vortex*, *momentum representation*, *probability flux*

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1. INTRODUCTION

The appearance of sources of electromagnetic pulses with an intensity and duration comparable to the corresponding atomic values has opened up opportunities for studying and controlling the states of single atoms and molecules, as well as their ensembles. Such capabilities are in demand both in quantum information applications and in the study of new control modes at atomic scales [1–10].

One of the nontrivial effects when a laser pulse with a duration of only a few atomic units of time is applied to an atom is the appearance of vortex structures in the electron density. The first theoretical prediction of such structures was made in 2010 in [11]. Later, in [12, 13], vortex structures were revealed in the momentum distribution for a photoelectron torn out during the ionization of a helium atom by a laser pulse. Experimental confirmation of vortex formations appeared in 2017 in [14]. In this experiment, the multiphoton ionization of potassium atoms by a sequence of femtosecond laser pulses was considered.

Currently, theoretical studies of such vortices are conducted in many scientific groups [15–31]. Approaches to the study of these formations are presented both by the development of original numerical methods for solving the nonstationary Schrodinger equation or equivalent equations of quantum hydrodynamics (see, for example, [32]) and by finding asymptotic solutions.

In [16–19], we investigated such vortex formations as quantum vortices [24] that arise during the superbarrier ionization of a two-dimensional hydrogen-like atom by an ultrashort laser pulse. These vortices manifest themselves as specific inhomogeneities in the spatial distribution of the photoelectron: the center of the vortex is the zero of the wave function, around which the velocity vector field circulates (note that the first one who connected the appearance of zeros of the wave function with the appearance of vortices was Dirac [33]).

Using the nonstationary perturbation theory, we obtained an analytical expression for the photoelectron wave function, which allowed us to see the interference nature of vortices and analyze their dependence on some pulse parameters. The results of the analytical approach were confirmed by numerical calculations.

In the mentioned works, the field of the laser pulse was modeled by a cosine dependence on a time interval $[0,T]$ ($1 < T < 10$ a.u.), containing only a few oscillation periods. At the same time, at the moments of switching on and off the pulse, the maximum amplitude value was always set, i.e. a variation of the limiting case of a sudden disturbance was realized [34]. Other variations of the effect of the laser pulse have not been investigated.

In this paper, remaining within the framework of the solutions obtained in [17- 19], the influence of the initial phase of the pulse field on the formation of quantum vortices will be investigated.

All calculations will be performed in the momentum space. In particular, the so-called «symmetric» probability flow [19, 35], sensitive to the phase of the wave function, will be used to identify quantum vortices in this space.

Attention will also be paid to the interference effects responsible for the formation of quantum vortices: in the momentum distribution of the photoelectron, we will highlight the corresponding interference term.

The issue of choosing a gauge when finding an approximate solution to the Schrodinger equation for the model under consideration will be considered separately.

The article is structured as follows. In section 2 the derivation of the photoelectron wave function is briefly presented and the approximations used are discussed. Expressions for probability flows are given. In section 3 the results of calculations and their analysis are shown. In section 4 the use of various gauges in describing the interaction of an electron with a field is discussed. The last section summarizes the work done.

The work uses the atomic system of units: $\hbar = 1$, $m_e = 1$, $e = 1$.

2. THEORETICAL APPROACH

The Hamiltonian $\hat{H}$ of a two-dimensional hydrogen atom interacting with a laser field has a standard form:

$$\hat{H} = \hat{H}_0 + \hat{V} = \hat{H}_0 - \hat{\mathbf{d}}\mathbf{F}(t), \tag{1}$$

where $\hat{H}_0$ is the unpertubed Hamiltonian of the atom, and the interaction $\hat{V}$ is written in the dipole approximation, where $\hat{\mathbf{d}} = -\hat{\mathbf{r}}$ is the operator of the dipole moment of the atom and $\mathbf{F}(t)$is the electric field of the laser.

The solution of the nonstationary Schrodinger equation is sought in the form of the following superposition:

$$\left|\Psi(t)\right\rangle = \left|\Psi_{1,0}^{(0)}\right\rangle e^{-iE_1 t} + \sum_m \int_0^\infty b_{k,m}(t)\left|\Psi_{k,m}^{(0)}\right\rangle e^{-iE_k t} k dk. \tag{2}$$

Here, the first term corresponds to the ground (initial) state of an atom with energy $E_1 = -1/2$ [36, 37] (we assume the charge number $Z = 1/2$). The subscripts «1,0» of the vector $|\Psi_{1,0}^{(0)}\rangle$ indicate the main quantum number $n = 1$and the projection of the moment $m = 0$onto the axis z. The second term describes the state of a photoelectron and is represented by a superposition of cylindrical waves. The indices of the corresponding vectors $|\Psi_{k,m}^{(0)}\rangle$ characterize the energy of the photoelectron

$$E_k = k^2/2 = (k_x^2 + k_y^2)/2$$

and the projection of the moment $m = 0, \pm1, \pm2, \ldots$. Unknown amplitudes $b_{k,m}(t)$such that $b_{k,m}(0) = 0$, $\forall k,m$.

The desired state in the form (2) is similar to the one used in Keldysh's theory [38, 39], except that instead of the Volkov functions there are cylindrical waves. Such an entry implies that the intensity of the laser field should be less than the atomic one, so that the probability of ionization of an atom is small. It is also assumed that the effect of the Coulomb potential of the residual ion on the photoelectron is negligible.

The study of quantum vortices in the work will be carried out in the momentum space. To do this, we project the desired state $|\Psi(t)\rangle$ (2) onto the eigenvectors of the momentum operator $\hat{\mathbf{k}}$. Then, using the explicit form of the necessary wave functions [40]

$$\Psi_{1,0}^{(0)}(\mathbf{k}) = \frac{2\Phi_0(\varphi_k)}{(k^2+1)^{3/2}},$$
$$\Psi_{k',m}^{(0)}(\mathbf{k}) = (-i)^{|m|}\frac{\delta(k'-k)}{k'}\Phi_m(\varphi_k), \tag{3}$$

where

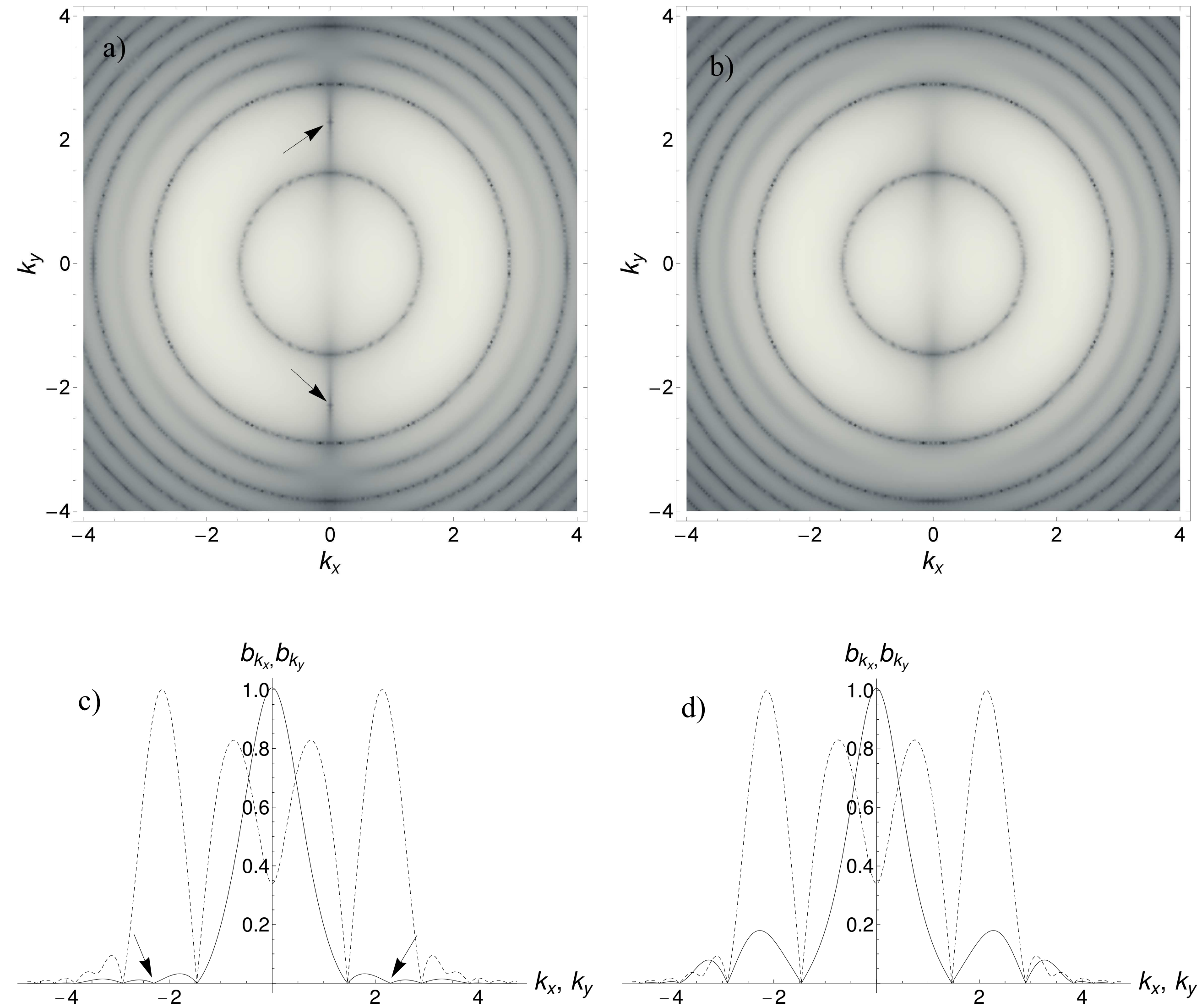

Fig.1. *a* and *b* are the density of the momentum distribution of the photoelectron $\ln(\rho)$. *c* and *d* are the dependences of the absolute values of the wave function $b_{k_x} b_{k_y}$ on the components k_x, k_y respectively. Pulse parameters: $F_0 = 0.4$, $\omega = \pi$, $\alpha = 0$, $T = 4$

$$\Phi_m(\varphi_k) = \frac{e^{im\varphi_k}}{\sqrt{2\pi}},$$

(k, φ_k) are the polar coordinates of the momentum $\mathbf{k}$, we get

$$\Psi(\mathbf{k},t) \equiv \langle \mathbf{k} | \Psi(t) \rangle = \frac{2\Phi_0(\varphi_k)}{(k^2+1)^{3/2}} e^{-iE_1 t} + + \sum_m b_{k,m}(t)(-i)^{|m|} \Phi_m(\varphi_k) e^{-iE_k t}. \quad (4)$$

Further, substituting decomposition (4) into the Schrodinger equation, we obtain a system of equations for finding unknown amplitudes $b_{k,m}(t)$:

$$\frac{\partial b_{k,m}(t)}{\partial t} = \frac{-i}{2}\Big(F_-(t)\delta_{m,+1} + F_+(t)\delta_{m,-1}\Big)\frac{6ke^{i\omega_{k1}t}}{(k^2+1)^{5/2}} + + \frac{(-i)^{|m-1|-|m|}}{2} F_-(t)\left(\frac{\partial}{\partial k} - ikt - \frac{m-1}{k}\right) b_{k,m-1}(t) + + \frac{(-i)^{|m+1|-|m|}}{2} F_+(t)\left(\frac{\partial}{\partial k} - ikt + \frac{m+1}{k}\right) b_{k,m+1}(t), \quad (5)$$

where $\omega_{k1} = (k^2+1)/2$ is the transition frequency $E_1 \to E_k$ and the designation is entered

$$F_\pm(t) = F_x(t) \pm iF_y(t).$$

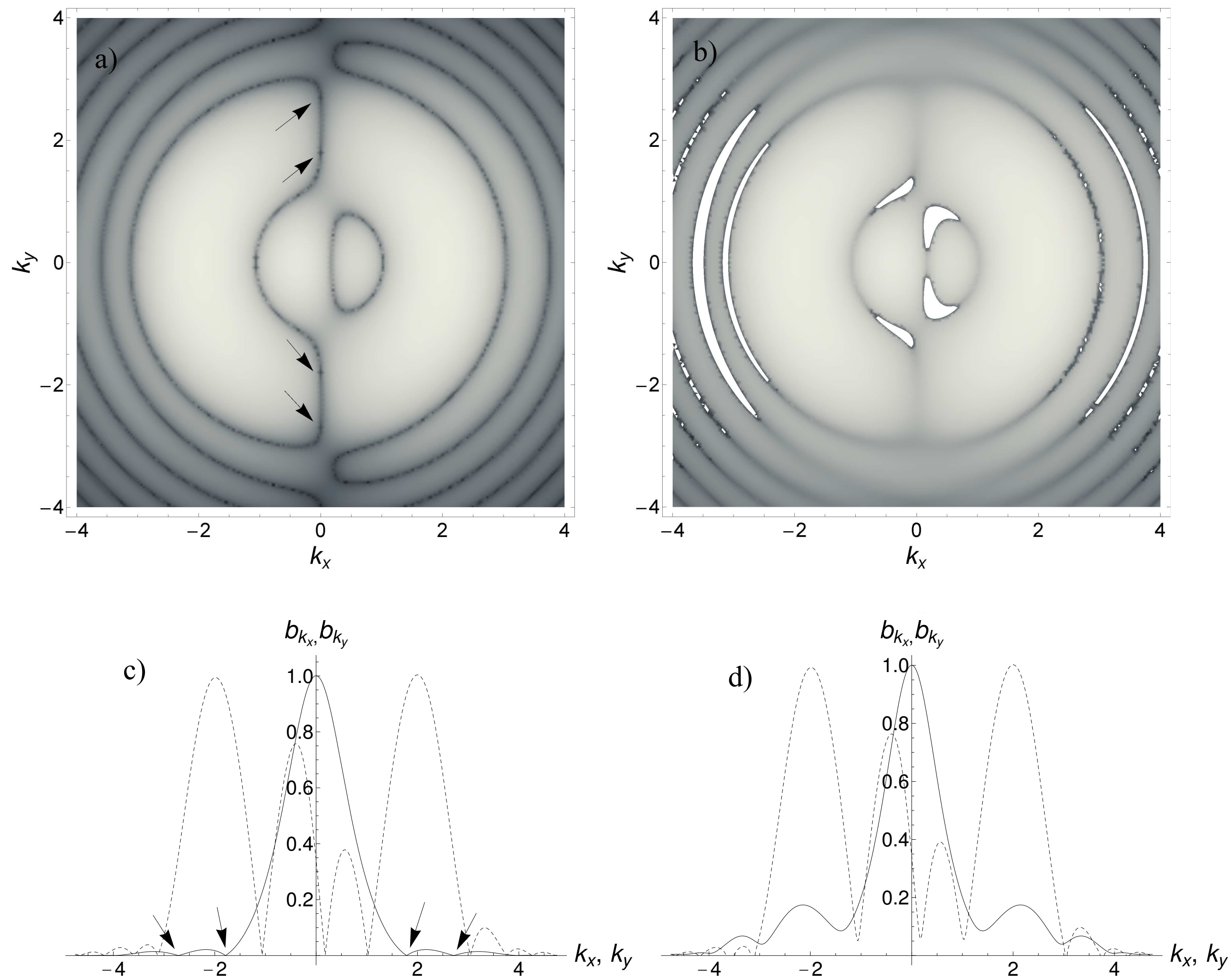

Fig.2. *a* and *b* are the density of the momentum distribution of the photoelectron $\ln(\rho)$. *c* and *d* are the dependences of the absolute values of the wave function $b_{k_x} b_{k_y}$ on the components k_x, k_y respectively. Pulse parameters: $F_0 = 0.4$, $\omega = \pi$, $\alpha = 0$, $T = 3$

In the derivation (5), an explicit expression was used for the interaction operator in the momentum representation:

$$\hat{V} = \mathbf{F}(t) i \nabla_k, \quad \nabla_k = \partial / \partial \mathbf{k}.$$

The difference between system (5) and the corresponding system of equations used by us earlier (see [17–19]) consists in taking into account the arbitrary polarization of the field $\mathbf{F}(t)$.

We are looking for a solution to system (5) in the form of an perturbation series

$$b_{k,m}(t) = \sum_{s=0}^{\infty} b_{km,10}^{(s)}(t),$$

where $b_{km,10}^{(s)} \sim F^s$, and the added subscript "10" indicates the initial bound state of the electron. Hence, the wave function of a photoelectron in the second order of perturbation theory is equal to

$$\tilde{\Psi}(\mathbf{k},t) = -i\sqrt{\frac{2}{\pi}} b_{k1,10}^{(1)}(t)\cos(\varphi_k) e^{-iE_k t} + \\ + \frac{1}{\sqrt{2\pi}} b_{k0,10}^{(2)}(t) e^{-iE_k t} - \sqrt{\frac{2}{\pi}} b_{k2,10}^{(2)}(t)\cos(2\varphi_k) e^{-iE_k t}, \tag{6}$$

where the tilde over Ψ indicates that the bound state is omitted, i.e. the interference between the initial and final states of the electron is neglected. Note that in our previous works [17, 18] the notation was used

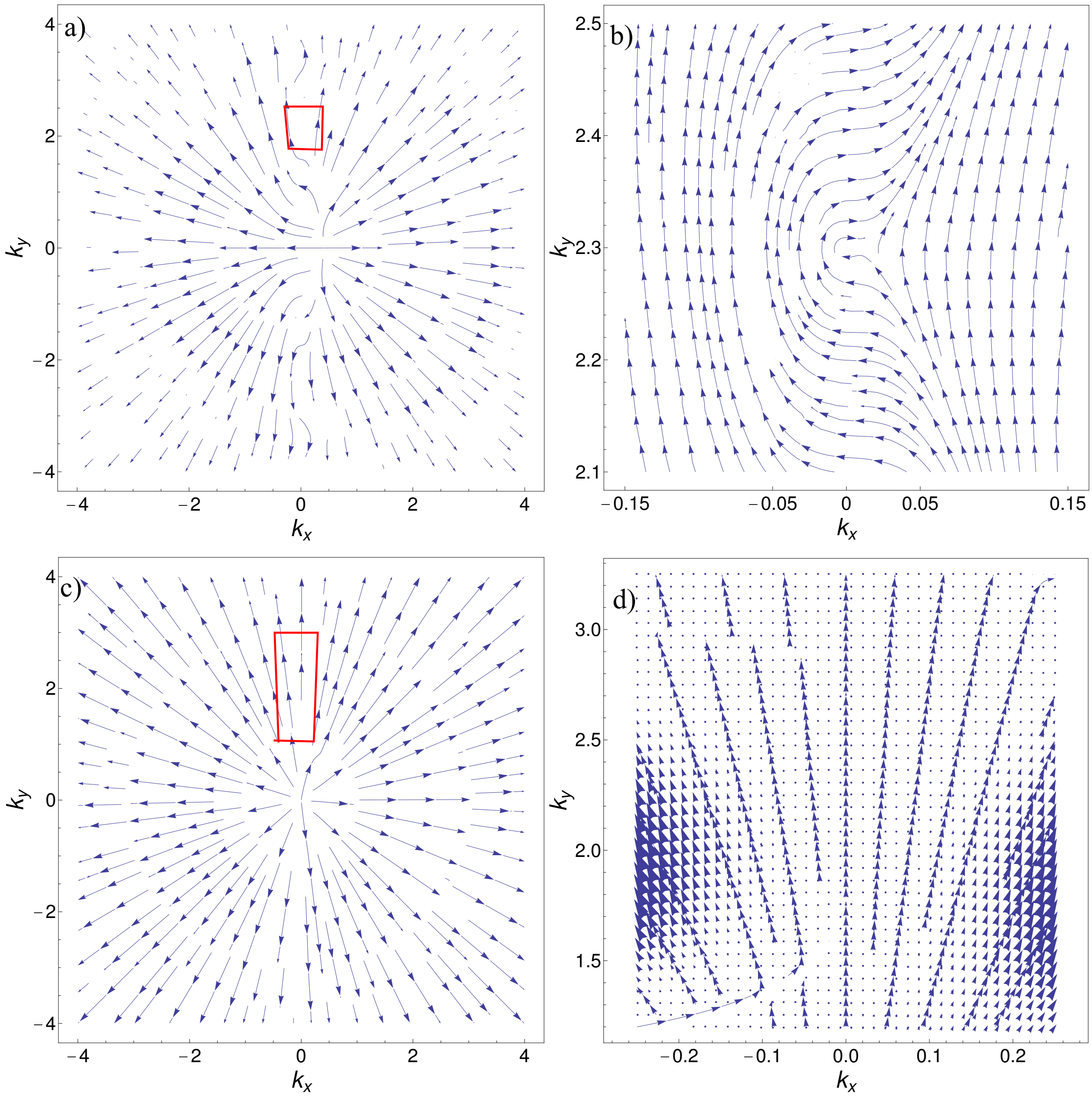

Fig. 3. A vector field for “symmetric” flow $\bar{\mathbf{j}}$. $T = 4$ (*a*, *b*), $T = 3$ (*c*, *d*). *b*, *d* are Enlarged regions of the vector field (red rectangles) containing quantum vortices

$$\tilde{\Psi}(\mathbf{k},t) = b(\mathbf{k},t)e^{-iE_k t}.$$

Also, when writing (6), it was taken into account that

$$b^{(1)}_{k1,10}(t) = b^{(1)}_{k-1,10}(t),$$
$$b^{(2)}_{k2,10}(t) = b^{(2)}_{k-2,10}(t).$$

For clarity, we will write out an explicit expression for the density of the pulse distribution of a photoelectron, which we will need when discussing interference effects.

$$\rho(\mathbf{k},t) \equiv \left|\tilde{\Psi}(\mathbf{k},t)\right|^2 = \frac{1}{\pi}\left\{\begin{array}{c} 2\cos^2(\varphi_k)\left|b^{(1)}_{k1,10}(t)\right|^2 + \\ +\frac{1}{2}\left|b^{(2)}_{k0,10}(t)\right|^2 + 2\cos^2(2\varphi_k)\left|b^{(2)}_{k2,10}(t)\right|^2 - \\ -2Re\left[ib^{(1)}_{k1,10}(t)b^{(2)*}_{k0,10}(t)\right]\cos(\varphi_k) + \\ +4Re\left[ib^{(1)}_{k1,10}(t)b^{(2)*}_{k2,10}(t)\right]\cos(\varphi_k)\cos(2\varphi_k) - \\ -2Re\left[b^{(2)}_{k0,10}(t)b^{(2)*}_{k2,10}(t)\right]\cos(2\varphi_k) \end{array}\right\}, \quad (7)$$

where $Re[z]$ means the real part z.

Now let's write out and discuss the advantages of the so-called [19, 35] "symmetric" probability flow

$$\bar{\mathbf{j}}(\mathbf{k},t) = -\frac{1}{2i}\left[\tilde{\Psi}^*(\mathbf{k},t)\nabla_k\tilde{\Psi}(\mathbf{k},t) - \tilde{\Psi}(\mathbf{k},t)\nabla_k\tilde{\Psi}^*(\mathbf{k},t)\right]. \quad (8)$$

Let's write the found wave function (6) in the form

$$\tilde{\Psi}(\mathbf{k},t) = \sqrt{\rho(\mathbf{k},t)}e^{-i\chi(\mathbf{k},t)},$$

where $\chi(\mathbf{k},t)$ is the phase. Then, substituting $\tilde{\Psi}(\mathbf{k},t)$ in (8), we get

$$\bar{\mathbf{j}}(\mathbf{k},t) = -\rho(\mathbf{k},t)\nabla_k\chi(\mathbf{k},t).$$

Thus, the flow is $\bar{\mathbf{j}}(\mathbf{k},t)$ sensitive to the phase of the wave function $\tilde{\Psi}(\mathbf{k},t)$, while the standard flow in the pulse space is given by the expression

$$\mathbf{j}(\mathbf{k},t) = \mathbf{k}\rho(\mathbf{k},t).$$

We will also use a normalized «symmetric» flow

$$\bar{\mathbf{v}}(\mathbf{k},t) = \bar{\mathbf{j}}(\mathbf{k},t) / \rho(\mathbf{k},t) = -\nabla_k\chi(\mathbf{k},t),$$

which we will call the velocity field.

3. RESULTS AND DISCUSSIONS

A two-dimensional hydrogen atom is irradiated by a linearly polarized laser pulse, the electric field strength of which has the form

$$\mathbf{F}(t) = \mathbf{e}_x F_0\cos(\omega t - \alpha)\left[\theta(T-t) - \theta(-t)\right], \quad (9)$$

where $\mathbf{e}_x$ is the unit vector in the direction of the axis x, F_0— constant amplitude, ω— frequency, α— initial phase, $\theta(t)$— Heaviside step function, T— pulse duration.

The density of the distribution (7) and the «symmetric» flow (8) (hereinafter simply the flow) will be considered at times $t > T$:

$$\rho = \rho(k_x,k_y,t > T), \quad \bar{\mathbf{j}} = (\bar{j}_x,\bar{j}_y),$$

where

$$\bar{j}_i = \bar{j}_i(k_x,k_y,t > T), \quad i = x,y.$$

3.1 Interference contribution

First, let's check which of the interference terms in expression (7) is responsible for the formation of quantum vortices. Let's choose the pulse parameters close to those for which vortices have already been identified [16, 17]: $F_0 = 0.4$, $\omega = \pi$, $\alpha = 0$, $T = 4$.

Figure 1 shows graphs of the photoelectron momentum distribution ρ (for a clearer display, graphs are plotted for $\ln(\rho)$), as well as the following dependences of the absolute value of the wave function normalized to their maxima:

$$b_{k_x} \equiv \left|\tilde{\Psi}(k_x,0,t > T)\right| / \left|\tilde{\Psi}_{max}\right|,$$

$$b_{k_y} \equiv \left|\tilde{\Psi}(0,k_y,t > T)\right| / \left|\tilde{\Psi}_{max}\right|.$$

For the selected pulse parameters, two symmetrical vortices are formed with centers at points $k_{x_0} = 0$, $k_{y_0} = \pm\sqrt{2\pi - 1} \approx \pm 2.3$ (indicated by arrows, Fig. 1*a*,*c*). Now, comparing density (7) with

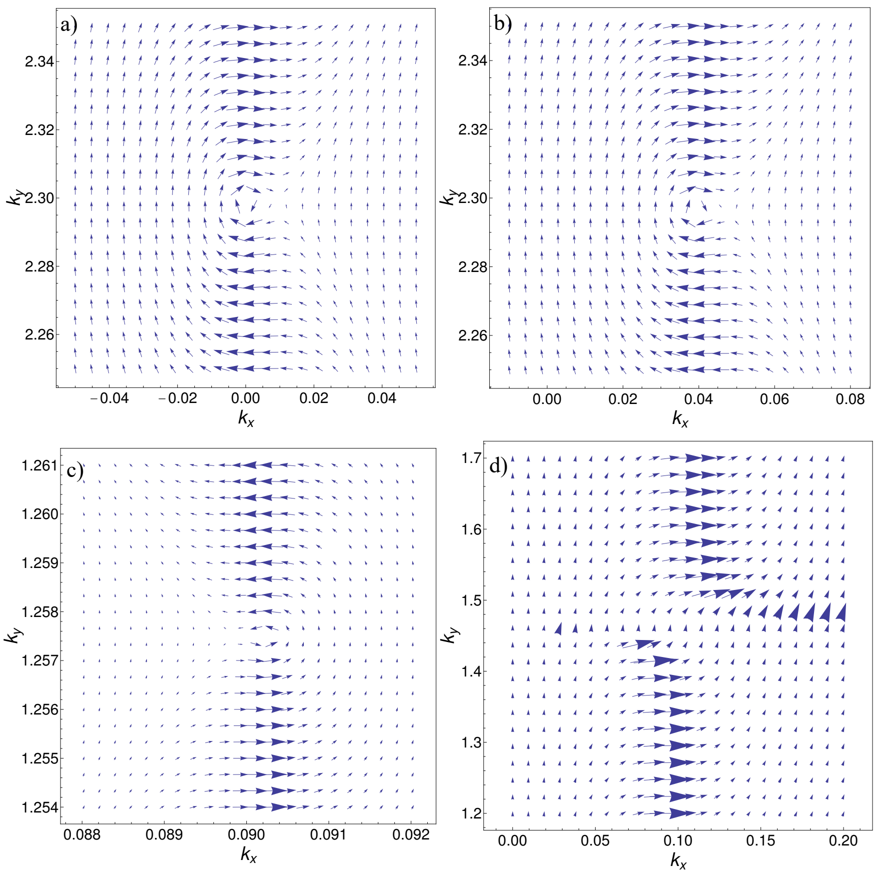

Fig. 4. A vector field for $\overline{\mathbf{v}}$. $\alpha = 0$ (*a*), $\pi / 10$ (*b*), $\pi / 3$ (*c*), $\pi / 2$ (*d*). For all graphs $F_0 = 0.4$, $\omega = \pi$, $T = 4$

the same density, but in which one of the three interference terms is discarded, we find that the last term in (7) is responsible for the formation of vortices. This term describes the interference of photoelectron states corresponding to cylindrical waves with quantum numbers $m = 0, \pm 2$. These states are the result of a two-photon transition through intermediate states of a continuous spectrum:

$$\left|\Psi^{(0)}_{1,m=0}\right\rangle \to \sum_k \left|\Psi^{(0)}_{k,m=\pm 1}\right\rangle \to \left|\Psi^{(0)}_{k,m=0,\pm 2}\right\rangle.$$

In Fig. 1 *b*, *d*, graphs are plotted without taking into account this interference term. It can be seen that there are no vortices.

A similar result on the influence of interference terms can be obtained for other values of the pulse duration, for example, when T is odd and two pairs of symmetric vortices are observed [16, 17].

In Fig. 2 for $T = 3$ (the other parameters are the same as in Fig. 1), the ρ and b_{k_x}, b_{k_y} are presented. The exclusion of the last interference term in (7) leads to the disappearance of vortices (Fig. 2 *b*, *d*), the centers of which are given by coordinates $k_{x_0} = 0$, $k_{y_0} = \pm\sqrt{4\pi / 3 - 1} \approx \pm 1.78$, $k_{x_0} = 0$, $k_{y_0} = \pm\sqrt{8\pi / 3 - 1} \approx \pm 2.71$ (fig. 2 *a*, *c*).

3.2 "Symmetric" flow $\overline{\mathbf{j}}$

As shown in [19], it is convenient to identify quantum vortices in momentum space using «symmetric» flow (8). In Fig. 3 vector field is presented for the two pulse durations considered above.

Fig. 3*a*, *c* shows that in both cases the field $\overline{\mathbf{j}}$ diverges from the center and on large scales this divergence is close to radial. However, in the area where quantum vortices are observed, the streamlines are curved (highlighted in red rectangles). In Fig. 3b,d, these areas are enlarged.

For a single isolated vortex, which occurs at $T = 4$, the presence of a rotational component of the field around the center of the vortex is clearly visible (see also [19]). At $T = 3$ there is no such beautiful pattern for the case, but the area of the broken streamlines is visible. It is along this narrow region (the light arc), as we showed earlier (see Fig. 4 in [16]), that two vortices are localized.

3.3 Sensitivity of quantum vortices to the initial phase of the field

Figure 4 shows the velocity vector field $\bar{\mathbf{v}} = (\bar{j}_x / \rho, \bar{j}_y / \rho)$ for four cases: $\alpha = 0$ (*a*), $\pi / 10$ (*b*), $\pi / 3$ (*c*), $\pi / 2$ (*d*). The other pulse parameters are the same as in Fig. 1.

It can be seen that as the phase increases, the vortex shifts. For α close to the $\frac{\pi}{2}$ characteristic vortex behavior of the field is observed in a significantly narrower range of values k_x than in the case when $\alpha = 0$. For sinusoidal dependence, i.e. when the switching on and off of the pulse ceases to be sudden, it is no longer possible to identify the vortex.

Figure 5 shows the dependence of the modulus of the wave function $b_{k_y,min} \equiv |\tilde{\Psi}(k_{x,min}; k_y, t > T)|$ on k_y at different projection values $k_{x,min}$ corresponding to the local minimum $\tilde{\Psi}$. The graph shows the displacement of the vortex center with increasing phase α.

It is important to note that the strict zero of the found wave function $\tilde{\Psi}$ (6), indicating the center of the vortex, takes place only at $\alpha = 0$. For the others considered $\alpha \neq 0$ corresponding values $k_{x,min}$ characterize local minima, while $b_{k_y,min} \neq 0$.

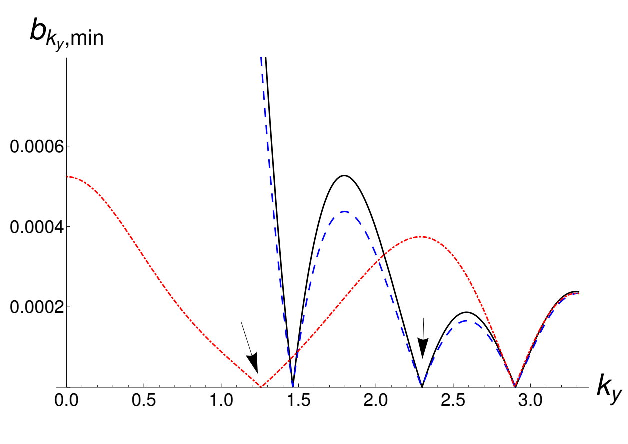

Fig. 5. Dependence of the modulus of the wave function $b_{k_y,min}$ on the projection k_y near the center of the vortex: $\alpha = 0$, $k_{x,min} = 0$ — solid line, $\alpha = \pi/10$, $k_{x,min} \approx 0.04$ — strokes, $\alpha = \pi/3$, $k_{x,min} \approx 0.09$ — dashpoint. For all graphs $F_0 = 0.4$, $\omega = \pi$, $T = 4$

4. INTERACTION OF AN ELECTRON WITH A FIELD IN "VELOCITY GAUGE"

As is known, the exact solutions of the Schrodinger equation obtained in two different gauges — in "length gauge" and in "velocity gauge", give the same predictions for the probabilities of transitions in atomic systems. Of course, this is the case only when a non-relativistic system interacts with that part of the electromagnetic field modes whose wavelength significantly exceeds the size of the system (see, for example, [41]).

In the case of approximate solutions found, for example, using perturbation theory, the results obtained in different gauges are not required to coincide [38]. Therefore, it is of interest to consider the problem solved here in «velocity gauge».

Let's write out the corresponding perturbation operator

$$\hat{V} = \frac{1}{c}\mathbf{A}(t)\hat{\mathbf{p}} + \frac{1}{2c^2}\mathbf{A}^2(t), \tag{10}$$

where the vector potential is

$$\mathbf{A}(t) = -c\mathbf{e}_x \int_0^t F_x(t')dt',$$

$$F_x(t) = \mathbf{e}_x \cdot \mathbf{F}(t),$$

(see (9)). Taking the desired wave function in the form (2) and performing the same manipulations as in the case of "length gauge", we obtain the following equations for unknown amplitudes:

$$\begin{aligned} \frac{\partial b_{k,m}(t)}{\partial t} &= \frac{A_x(t)}{2c}\left(\delta_{m,+1} + \delta_{m,-1}\right)\frac{2ke^{i\omega_{k1}t}}{(k^2+1)^{3/2}} - \\ &- (i)^{|m|+1-|m-1|}\frac{A_x(t)}{2c}k \cdot b_{k,m-1}(t) - \\ &- (i)^{|m|+1-|m+1|}\frac{A_x(t)}{2c}k \cdot b_{k,m+1}(t) - i\frac{A_x^2(t)}{2c^2}b_{k,m}(t) - \\ &- i\frac{A_x^2(t)}{c^2}\delta_{m,0}\frac{e^{i\omega_{k1}t}}{(k^2+1)^{3/2}}. \end{aligned} \tag{11}$$

It can be seen that the system (11) has a simpler appearance compared to the system (5) obtained in "length gauge": there are no derivatives of k, i.e.

these equations are relatively simple for numerical calculations.

As preliminary calculations have shown, the application of perturbation theory to the system (11), up to and including the second order, makes it possible to detect the quantum vortices studied in this work. The results obtained in two different calibrations are close to each other. The small discrepancies do not affect the main conclusions of the work and can be investigated separately.

Note that in obtaining (11), the same basic functions of the unperturbed problem were used as in deriving the system (5). This choice is justified by the approximation made in the work of the sudden switching on and off of the field [41].

5. CONCLUSION

In this article, the influence of the initial phase of an extremely short ionizing laser pulse on the formation of quantum vortices was theoretically investigated. It is shown that phase variation can lead to both a localized vortex with a well-defined center and a “smeared” vortex structure. At the same time, an important aspect here is the implementation of the limiting case of a sudden disturbance, which is a kind of “Jarring” type “turn-on” [34].

The interference contribution to the photoelectron distribution density responsible for the formation of vortices is extracted. This contribution is due to the interference of photoelectron states formed during a two-photon transition through intermediate states of a continuous spectrum.

The possibility of identifying quantum vortices in momentum space using “symmetric” flow (8) has been confirmed. Unlike the work [19], which considered the even case T, this work also investigated the odd case T, when two pairs of quantum vortices appear.

The choice of gauge in describing the interaction of an electron with a field is discussed.

The equations for the probability amplitudes in “velocity gauge” (11) have a simpler form than in the case of “length gauge” (5) and are relatively simple for numerical calculations. The small differences found between the results of preliminary calculations obtained in different calibrations do not affect the main conclusions of the article.

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