Multiphoton ionization in superintense, high-frequency laser fields. I. General developments

Marcel Pont
Physics Department, University of Southern California, Los Angeles, California 90089-0484
(Received 2 November 1990; revised manuscript received 27 February 1991)

This is the first of two papers studying multiphoton ionization (MPI) in superintense, high-frequency laser fields. They are based on a general iteration scheme in increasing powers of the inverse frequency. To lowest order in the frequency, i.e., the high-frequency limit, the atom was shown to be stable against decay by MPI, though distorted. To next order in the iteration, an expression for the MPI amplitude was obtained. In the present paper, we present general developments from this expression, valid for arbitrary polarization, binding potential, intensity, and initial state. First we analyze the symmetry of the angular distributions of photoelectrons determined by this expression for the MPI amplitude. This expression can explain the asymmetries in the angular distributions of photoelectrons occurring in the case of elliptic polarization that were recently reported in experiments. In the radiation regime where our theory applies these asymmetries are, however, weak. In certain instances our theory yields asymmetries in cases where lowest-order perturbation theory (LOPT) fails to predict them. We prove that at low intensities our expression for the MPI amplitude yields results in agreement with LOPT evaluated at high frequencies. An important part of this paper consists, however, of the derivation of an alternative form for the MPI amplitude of atomic hydrogen, which is substantially simpler, though somewhat less accurate. We study the consequences of this simplified expression for the case of linearly polarized fields in the following paper [Phys. Rev. A 44, 2171 (1991)].

I. INTRODUCTION

Ambitious plans to construct laser systems, which are expected to attain intensities many orders of magnitude higher than the atomic unit, are now pursued by several laboratories throughout the world [1]. (The atomic unit of intensity, $I_0 = 3.51 \times 10^{16}$ W/cm^2, is defined as the intensity of a linearly polarized electromagnetic plane wave with an electric-field amplitude equal to the electrostatic field of the proton on the first Bohr orbit of atomic hydrogen.) With the possibility of experimental tests in sight, the theoretical description of atomic behavior in radiation fields well beyond the atomic unit of intensity becomes a fundamental problem of primary interest. This is the first of two papers that study multiphoton ionization of atoms under these extreme radiation conditions.

We will adopt the framework of a theory developed by Gavrila and Kaminski [2, 3]. The Gavrila-Kaminski theory applies to stationary decay in a monochromatic plane wave (Floquet description). It is based on an iteration scheme proceeding in increasing powers of the inverse frequency, with a certain parameter α_0, a combination of the intensity and frequency, kept fixed. This iteration scheme was shown to be convergent (in a pragmatic sense) under conditions that *the photon energy is high with respect to the ionization potential of the atom in the field.* To lowest order in the frequency, i.e., the high-frequency limit, the atom is stable against decay by multiphoton ionization. Its structure is then described by a Schrödinger equation containing a modified atomic binding potential, the "dressed" potential V_0, which depends on the frequency and intensity only as combined in the parameter α_0. So far, the theory has been applied to atomic hydrogen and its *radiative distortion and ac Stark shifts* have been analyzed in great detail for the ground state and its lower-lying excited states, in particular for the case of linear polarization [4] (for atomic hydrogen in circularly polarized fields, see Ref. [5]; for applications to scattering, see Ref. [6]). Among other things, we have found a drastic reduction of the ionization potential and a dramatic deformation of the electron cloud with α_0, i.e., with increasing intensity at fixed (high) frequency. For α_0=30, a value that can presently be realized in the laboratory, the drop of the ionization potential amounts to about a factor of 10. This reduction has favorable repercussions on the range of validity of the theory. For intermediate values of α_0, we find radiative stretching of the electron cloud in the direction of the polarization axis. (The corresponding increase in the atomic size can be two orders of magnitude for existing lasers.) It culminates in its *splitting into two disjoint parts* separated at high α_0 by a distance of $2\alpha_0$ ("dichotomy") [4].

To next order in the iteration, an expression for the multiphoton ionization amplitude was obtained in which the initial and final state are bound and scattering eigenstates of the dressed potential, respectively. In the present work we will be concerned with multiphoton ionization occurring in superintense, high-frequency laser fields. In contrast to our second paper we will here present general developments; we will assume arbitrary polarization, intensity, binding potential, and ini-

tial state, and we will take as a starting point this expression for the multiphoton ionization (MPI) amplitude of the high-frequency theory. In Sec. II we will recapitulate the basic equations of the Gavrila-Kaminski scheme.

Recent experiments on angular distributions of photoelectrons have attracted special attention because of an unexpected asymmetry occurring in the case of elliptic polarization. In Sec. III we analyze the symmetry of the angular distributions of photoelectrons for the case of arbitrary polarization and atomic binding potential, starting from the result for the multiphoton ionization amplitude obtained to lowest order in the iteration in ω^{-1}. Part of our findings agrees with well-known results obtained from lowest-order perturbation theory (see, e.g., [7]), but because of its *nonperturbative* nature, our theory leads, in general, to more complicated angular dependences. We find that the high-frequency theory of Gavrila and Kaminski does provide for the aforementioned asymmetries, although they will be weak in the radiation regime where it applies. In certain instances the present theory predicts asymmetries in cases where lowest-order perturbation theory fails to predict them.

Although our main interest lies in the nonperturbative radiation regime, we have deemed it worthwhile to show in Sec. IV that the high-frequency theory yields, at low intensities and high frequencies, angular-dependent decay rates which are in agreement with standard lowest-order perturbation theory. For simplicity, we will restrict ourselves, however, to the case of single-photon ionization.

For the remainder of this paper, we narrow the discussion down to atomic hydrogen. In Sec. V we analyze the behavior of the Gavrila-Kaminski expression for the multiphoton ionization amplitude for the case of high frequencies (irrespective of the value of the parameter α_0). We derive an expression in which only the values of the initial (deformed) bound state enter that pass over the proton during its oscillation (like a free electron) in the laboratory frame of reference. This could be done for the case of arbitrary polarization. This approximation represents a considerable simplification over the Gavrila-Kaminski expression. It enables us to calculate important quantities which can be inferred from experiment, such as the lifetime of the atom, the angular distributions of photoelectrons, and the peak pattern of the energy spectrum of the ejected electrons. These issues will be studied in detail for the case of atomic hydrogen, placed in a superintense, high-frequency field of linear polarization in the following paper.

II. THE HIGH-FREQUENCY THEORY OF ATOMIC BEHAVIOR IN INTENSE LASER FIELDS

The general formalism [2, 3] will be illustrated for a one-electron model atom with binding potential $V(\mathbf{r})$. We will assume that it is of a realistic but otherwise arbitrary form, i.e., Coulomb or with a Coulomb tail (e.g., a pseudopotential for the optically active electron of an alkali-metal atom). For the description of the laser field we take a classical, monochromatic plane wave in the dipole approximation [8]. With an appropriate choice of the initial phase (this can be done without affecting the generality of our results), the electric-field vector in the case of arbitrary polarization can be taken as

$$\mathbf{E}(t) = E_0[\mathbf{e}_1 \cos\omega t + \mathbf{e}_2 \tan(\chi/2)\sin\omega t]. \tag{1}$$

The system of unit vectors $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$ is chosen to be right-handed, with $\mathbf{e}_3$ in the propagation direction. If $\chi = 0°$ we have linear polarization, whereas if $\chi = \pm 90°$ we have circular polarization. The case $0° < |\chi| < 90°$ corresponds to the case of elliptic polarization. If χ is positive we call the polarization left-handed, whereas if χ is negative it is called right-handed [9].

The starting point of the theory is the so-called "space-translated" Schrödinger equation, originally proposed by Kramers [10] in the general context of quantum electrodynamics (see also Pauli and Fierz [11]) and rediscovered by Henneberger in the context of laser-atom interactions [12]:

$$\left(\frac{1}{2m}\mathbf{P}^2 + V(\mathbf{r} + \boldsymbol{\alpha}(t))\right)\Psi = i\hbar\frac{\partial\Psi}{\partial t}. \tag{2}$$

In this equation $\boldsymbol{\alpha}(t)$ denotes

$$\boldsymbol{\alpha}(t) = \alpha_0[\mathbf{e}_1 \cos\omega t + \mathbf{e}_2 \tan(\chi/2)\sin\omega t], \tag{3}$$

in which α_0 is a combination of the frequency and the intensity of the field:

$$\alpha_0 = -\frac{eE_0}{m\omega^2} = I^{1/2}\omega^{-2}\cos(\chi/2)\ \text{(a.u.)}, \tag{4}$$

where the atomic unit of intensity $I_0 = 3.51 \times 10^{16}\text{W cm}^{-2}$. Recall that we have defined the atomic unit of intensity by the intensity of a *linearly polarized* plane wave with an amplitude equal to the electrostatic field due to the proton on the first orbit of atomic hydrogen. Equation (2) describes in fact the dynamics in a frame of reference (henceforth called the "Kramers frame of reference") which oscillates along with a classical "free" electron driven by the electric field, whose position is pointed to by the tip of the vector $\boldsymbol{\alpha}(t)$. Consequently, in the Kramers reference frame the center of force appears to oscillate according to $-\boldsymbol{\alpha}(t)$ [13].

Since the Schrödinger equation Eq. (2) has time-periodic coefficients, as usual, a "quasienergy" solution of the Floquet-type [14] was sought:

$$\Psi(\mathbf{r}, t) = e^{-iEt/\hbar}\sum_{n=-\infty}^{+\infty} e^{-in\omega t}\Psi_n(\mathbf{r}). \tag{5}$$

We shall associate the $n = 0$ case with the initial channel of energy E ($\text{Re}E < 0$). Assuming that n_0 is the smallest (positive) integer for which $\text{Re}E_n > 0$, the channels for which $n < n_0$ are closed, whereas the channels for which $n \geq n_0$ are open.

Thus the following conditions on the asymptotic behavior of the Floquet components Ψ_n appearing in Eq. (5) were set:

$$\Psi_n(\mathbf{r}) \underset{\sim}{|\,\mathbf{r}\,|\to\infty} \begin{cases} f_n(\alpha_0,\omega,\hat{r})\frac{e^{i(k_n r-\gamma_n \ln 2k_n r)}}{r} & \text{(open channels)} \\ 0 \ \text{(exponentially)} & \text{(closed channels)} \end{cases} \quad (6)$$

where f_n is the n-photon ionization amplitude. Here we have considered the case of an ionic potential $V(\mathbf{r})$ with asymptotic charge Z (in the case of a short-range potential $Z = 0$) and we have defined the dimensionless quantity $\gamma_n = -Ze^2m/\hbar^2k_n$. The momenta $p_n = \hbar k_n$ are related to the quasienergy E of the initial state (associated with the channel $n = 0$) by the generalized Einstein-equation

$$\frac{p_n^2}{2m} = E_n = E + n\hbar\omega, \quad (7)$$

representing conservation of energy. Note that the boundary conditions as given in Eq. (6) are of the Siegert type [15].

The angular decay rate for n-photon ionization corresponding to Eq. (6) is given by

$$\frac{d\Gamma_n}{d\Omega} = \frac{p_n}{m}|f_n(\alpha_0;\omega;\hat{\mathbf{r}})|^2 \quad \text{(open channels)}, \quad (8)$$

if the solution of Eq. (2) is properly normalized. One may also define the angle-integrated n-photon ionization rate by

$$\Gamma_n = \frac{p_n}{m}\int |f_n(\alpha_0;\omega;\hat{\mathbf{r}})|^2 d\hat{\mathbf{r}} \quad \text{(open channels)} \quad (9)$$

and the total decay rate Γ, which is obtained by summing the partial decay rates Γ_n over all open channels.

Having set the boundary conditions, we now proceed with the solution of Eq. (2) (see Refs. [2, 3]). Following the Floquet analysis of the wave function Eq. (5), we Fourier analyze the potential $V(\mathbf{r}+\boldsymbol{\alpha}(t))$:

$$V(\mathbf{r}+\boldsymbol{\alpha}(t)) = \sum_{n=-\infty}^{+\infty} V_n(\alpha_0;\mathbf{r})e^{-in\omega t}. \quad (10)$$

The (time-independent) coefficients $V_n(\alpha_0,\mathbf{r})$ can be expressed as

$$V_n(\alpha_0;\mathbf{r}) = \frac{1}{2\pi}\int_{-\pi}^{+\pi} e^{in\phi}V(\mathbf{r}+\boldsymbol{\alpha}(\phi))d\phi, \quad (11)$$

where we have loosely written $\boldsymbol{\alpha}(\phi)$, since it does not depend on the frequency, instead of $\boldsymbol{\alpha}(\phi/\omega)$ [see Eq. (3)]. The insertion of Eqs. (5) and (10) into the space-translated Schrödinger equation Eq. (2) leads to an (infinite) set of coupled differential equations for the Floquet components Ψ_n.

It was shown by Gavrila and Kaminski that this system can be solved by successive approximations. To lowest order in the iteration, they obtain

$$\Psi_{\lambda,n} \simeq \Phi_\lambda\delta_{n,0}, \quad E_\lambda \simeq W_\lambda, \quad (12)$$

in which Φ_λ is a bound state of the "radiation dressed potential" V_0, with energy W_λ:

$$\left(\frac{1}{2m}\mathbf{P}^2 + V_0\right)\Phi_\lambda = W_\lambda\Phi_\lambda. \quad (13)$$

Thus, in the limit of high frequencies, the atom becomes stable against decay by multiphoton ionization. However, it may be strongly deformed, since at high α_0 the eigenvalues W_λ and the eigenfunctions Φ_λ may differ appreciably from their unperturbed versions (at $\alpha_0 = 0$).

To next order in the iteration, they found

$$\Psi_{\lambda,n} \simeq \Phi_\lambda\delta_{n,0} + G^{(+)}(W_\lambda + n\hbar\omega)V_n\Phi_\lambda(1-\delta_{n,0}). \quad (14)$$

Here $G^{(+)}$ denotes the Green's function associated with the Hamiltonian $H = \frac{1}{2}\mathbf{P}^2 + V_0$. By analyzing in coordinate representation the large $|\mathbf{r}|$ behavior of Eq. (14) and comparing this with Eq. (6), the following expression for the multiphoton ionization amplitudes in terms of solutions of Eq. (13) was obtained:

$$f_{\lambda,n}(\alpha_0,\omega,\hat{\mathbf{r}}) = -\frac{m}{2\pi\hbar^2}\left\langle \Phi_{\mathbf{k}_n}^{(-)}|V_n|\Phi_\lambda\right\rangle. \quad (15)$$

Here $\Phi_{\mathbf{k}_n}^{(\pm)}$ is the scattering solution of Eq. (13) for the energy $E_n = W_\lambda + n\hbar\omega$, behaving for large distances as a (modified) plane wave with momentum $\hbar\mathbf{k}_n$ plus an incoming or outgoing spherical wave for the superscripts $(-)$ and $(+)$, respectively. We assume it to be normalized to unit asymptotic amplitude. Note that the $f_{\lambda,n}$ — and therefore also the n-photon decay rates defined by Eq. (15) — are in fact ω dependent, because of the energy conservation relation Eq. (7) relating the initial and final states.

It was argued that the iteration procedure of the high-frequency theory converges (in a pragmatic sense) under the condition

$$\hbar\omega \gg |W_\lambda^0(\alpha_0)|, \quad (16)$$

where $W_\lambda^0(\alpha_0)$ is the lowest eigenvalue of Eq. (13) within the manifold of states which are radiatively coupled to the initial state Φ_λ. (For a more specific statement, see Sec. III.) Under condition Eq. (16), Eqs. (13) and (15) represent good approximations of the exact values. The convergence of the high-frequency iteration scheme was tested on one-dimensional model atoms, which allow an exact numerical solution, by Bardsley and Comella [16] (for the case of scattering see Bhatt, Piraux, and Burnett [17]). There are no restrictions on α_0 (except for being kept finite), i.e., the theory applies as well for $\alpha_0 \ll 1$, which is the realm of perturbation theory, as for $\alpha_0 > 1$ where the theory is nonperturbative. It is in this latter case that we shall be primarily interested.

Equation (13) was obtained earlier via other methods by Henneberger [12], and Gersten and Mittleman [18] (see also Mittleman [19]). Gersten and Mittleman were the first to realize its high-frequency character [20].

In the following we will use atomic units in which $\hbar = m = |e| = 1$.

III. THE SYMMETRY OF THE ANGULAR DISTRIBUTIONS OF PHOTOELECTRONS

As is well known, the angular distributions of electrons produced in multiphoton and excess-photon ionization

exhibit certain definite symmetries, depending on the polarization of the laser light used [7]. Recently Bashkansky, Bucksbaum, and Shuhmacher [21] reported on an (unexpected) asymmetry in the case of elliptically polarized light (see below). It has even been stated that theories that do not provide for this asymmetry are in a certain sense incomplete [22]. This stimulated us to analyze the symmetry of the angular distributions of photoelectrons on the basis of the expression for the n-photon ionization amplitude, Eq. (15), where the coupling potentials are given by Eq. (11).

Let us now define our choice of coordinate system, i.e., our choice for the (right-handed) system of unit vectors $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$ of Eqs. (1) and (3). Recall that $\mathbf{e}_3$ was chosen in the propagation direction. In the case of elliptic or circular polarization, we will choose our coordinate system with the x, y, and z axes along $\mathbf{e}_1$, $\mathbf{e}_2$, and $\mathbf{e}_3$, respectively. In the case of linear polarization we choose the z axis along $\mathbf{e}_1$ and the x and y axes along $\mathbf{e}_2$ and $\mathbf{e}_3$, respectively.

In order to investigate the symmetry of the angular dependences of photoelectrons we need to know the behavior of the n-photon ionization amplitude under spatial transformations. In our case [in view of Eq. (15)] we have to examine the transformation properties of $\Phi_{\mathbf{k}}^{(-)}$, V_n, and Φ_λ. Since Φ_λ and $\Phi_{\mathbf{k}}^{(-)}$ are eigenstates of the dressed potential, the transformations pertaining to them are those which leave V_0 invariant.

Henceforth we will assume the atomic binding potential to be spherically symmetric. It is easily seen from Eq. (11) that the full *symmetry group of* V_0 is then in fact identical to the one which carries the figure (line segment, ellipse, or circle) described by the tip of the electric vector, over into itself [23]. As one can easily check, the full symmetry group (consisting of rotations, the inversion, reflections, and rotary reflections) of V_0 can be generated by combining (a) rotations about the z axis, (b) reflection in the origin (inversion), and (c) reflection in the xz plane.

The transformation properties of the scattering eigenfunction $\Phi_{\mathbf{k}}^{(-)}$, specified uniquely by its boundary conditions, are the simplest. If $\mathcal{O}$ is a symmetry mapping of V_0 we have

$$\Phi_{\mathbf{k}}^{(-)}(\mathcal{O}^{-1}\mathbf{r}) = \Phi_{\mathcal{O}\mathbf{k}}^{(-)}(\mathbf{r}), \tag{17}$$

i.e., the wave vector of the new state is simply the transformed old wave vector.

Let us first consider rotations about the z axis [case (a)], denoting rotation by the angle γ about the z axis as $\mathcal{R}_\gamma$. For *linear* and *circular* polarization the dressed potential is axially symmetric about the z axis and hence we can associate a magnetic quantum number m with the initial state

$$\Phi_\lambda(\mathcal{R}_\gamma^{-1}\mathbf{r}) = e^{-im\gamma}\Phi_\lambda(\mathbf{r}). \tag{18}$$

Also, we have from Eq. (17)

$$\Phi_{\mathbf{k}}^{(-)}(\mathcal{R}_\gamma^{-1}\mathbf{r}) = \Phi_{\mathcal{R}_\gamma\mathbf{k}}^{(-)}(\mathbf{r}). \tag{19}$$

It is easily shown from Eqs. (11) and (3) that

$$V_n(\mathcal{R}_\gamma^{-1}\mathbf{r}) = \begin{cases} V_n(\mathbf{r}) & \text{(linear polarization)} \quad (20) \\ e^{\mp in\gamma}V_n(\mathbf{r}) & \text{(circular polarization),} \quad (21) \end{cases}$$

where the upper (lower) sign corresponds to left (right) circular polarization. Using these transformation relations in Eq. (15), we obtain

$$f_{\lambda,n}(\mathcal{R}_\gamma^{-1}\hat{\mathbf{r}}) = \begin{cases} e^{-im\gamma}f_{\lambda,n}(\hat{\mathbf{r}}) & \\ \qquad \text{(linear polarization)} & (22) \\ e^{-i(m\pm n)\gamma}f_{\lambda,n}(\hat{\mathbf{r}}) & \\ \qquad \text{(circular polarization).} & (23) \end{cases}$$

Equations (22) and (23) imply rotational symmetry about the z axis of $d\Gamma_{\lambda,n}/d\Omega$.

For the case of *elliptic polarization* we may examine the transformation properties for rotation by 180° about the z axis. From Eqs. (11) and (3) we have

$$V_n(\mathcal{R}_{180°}\mathbf{r}) = (-1)^n V_n(\mathbf{r}). \tag{24}$$

Thus we see in particular that V_0 is left invariant by a rotation about the z axis by 180°. Consequently, the initial state may be chosen such that it is symmetric or antisymmetric under this operation, i.e.,

$$\Phi_\lambda(\mathcal{R}_{180°}\mathbf{r}) = (-1)^r\Phi_\lambda(\mathbf{r}), \tag{25}$$

and we also have

$$\Phi_{\mathbf{k}}^{(-)}(\mathcal{R}_{180°}\mathbf{r}) = \Phi_{\mathcal{R}_{180°}\mathbf{k}}^{(-)}(\mathbf{r}). \tag{26}$$

From these relations it is easily shown that

$$f_{\lambda,n}(\mathcal{R}_{180°}\hat{\mathbf{r}}) = (-1)^{r+n}f_{\lambda,n}(\hat{\mathbf{r}}). \tag{27}$$

Consequently, $d\Gamma_{\lambda,n}/d\Omega$ is symmetric with respect to a rotation through 180° about the z axis.

Returning to the case of *arbitrary* polarization, let us now consider reflection in the origin [case (b)]. From Eqs. (11) and (3) we have

$$V_n(-\mathbf{r}) = (-1)^n V_n(\mathbf{r}). \tag{28}$$

Furthermore, from Eq. (17) we have

$$\Phi_{\mathbf{k}}^{(-)}(-\mathbf{r}) = \Phi_{-\mathbf{k}}^{(-)}(\mathbf{r}) \tag{29}$$

and for bound states we have

$$\Phi_\lambda(-\mathbf{r}) = (-1)^P\Phi_\lambda(\mathbf{r}), \tag{30}$$

with P the parity quantum number. From these relations it is easy to show that

$$f_{\lambda,n}(-\hat{\mathbf{r}}) = (-1)^{n+P}f_{\lambda,n}(\hat{\mathbf{r}}). \tag{31}$$

Equation (31) implies that the angular distribution of ejected electrons is invariant with respect to reflection in the origin, independent of the polarization.

The above-discussed transformation properties of $f_{\lambda,n}$ have some interesting side results concerning the vanishing of the electron flux in the polar and equatorial directions. For example, combining Eq. (22) for $\gamma = 180°$ with Eq. (31) in the xy plane, we find that for linear polarization

$$\frac{d\Gamma_{\lambda,n}}{d\Omega}(\theta = 90°) = 0 \quad \text{for } m+n+P \text{ odd.} \tag{32}$$

This issue is discussed further in Ref. [24].

Let us finally examine case (c). Denoting the reflection in the xz plane by $\mathcal{S}$, we have from Eqs. (11) and (3)

$$V_n(\mathcal{S}\mathbf{r}) = [V_n(\mathbf{r})]^* . \tag{33}$$

From this relation we see that the dressed potential is invariant with respect to a reflection in the xz plane if it is complex conjugated at the same time. This invariance property of V_0 implies [25]

$$\Phi_{\mathbf{k}}^{(-)}(\mathcal{S}\mathbf{r}) = \left[\Phi_{-\mathcal{S}\mathbf{k}}^{(+)}(\mathbf{r})\right]^* . \tag{34}$$

Compatible with the transformation properties under rotations about the z axis and under reflection in the origin (see above), we may impose on the bound states the following transformation behavior under reflection in the xz plane:

$$\Phi_\lambda(\mathcal{S}\mathbf{r}) = [\Phi_\lambda(\mathbf{r})]^* . \tag{35}$$

Consequently

$$f_{\lambda,n}(\hat{\mathbf{r}}) = \left(-\frac{1}{2\pi}\left\langle \Phi_{-\mathcal{S}\mathbf{k}_n}^{(+)} \,|V_n|\, \Phi_\lambda \right\rangle\right)^* \neq [f_{\lambda,n}(-\mathcal{S}\hat{\mathbf{r}})]^* , \tag{36}$$

the inequality being due to the difference in ingoing or outgoing wave boundary conditions on the final state. By combining this with Eq. (31), it follows that $d\Gamma_{\lambda,n}/d\Omega$ for elliptic polarization is in general *not* symmetric with respect to a reflection in a plane which passes through the major (or minor) axis of the ellipse described by the tip of the electric-field vector (perpendicular to the plane of polarization).

Recently Bashkansky, Bucksbaum, and Shuhmacher [21] reported on the appearance of this asymmetry, which occurs only in the case of elliptically polarized light, and noted that it could not be explained within the standard Keldysh-Faisal-Reiss (KFR) model [26–28]. Note that our theory does not have this deficiency. However, if we replace $\Phi_{\mathbf{k}}^{(-)}$ by a *plane wave*, it then follows from Eqs. (31) and (36) that the asymmetry does *not* occur. [In this case the inequality sign in Eq. (36) becomes an equal sign.] This situation is similar to the KFR theory, in which the final state interaction of the ejected electron is disregarded [29]. However, our theory does not apply to the experiments carried out by Bashkansky, Bucksbaum, and Shuhmacher since the frequency condition, Eq. (16), is not fulfilled. At high frequencies the final-state interaction is weak and may be neglected because the electron is ejected with large momentum (see Sec. V of this paper). Thus we predict that in our radiation regime the above-mentioned asymmetry may occur, but will be weak. For a discussion of the findings of the experiment of Ref. [21] in the framework of lowest-order perturbation theory (LOPT), see Refs. [22] and [30].

In the case of linear polarization, the orbital angular momentum component along the polarization axis is conserved and therefore we can assign a definite magnetic quantum number to the *decaying* state, Eq. (5). [This is an alternative route to prove Eq. (22) from Eq. (6).] In the case of elliptic or circular polarization, the Floquet component $\Psi_{\lambda,n}(\mathbf{r})$ of the decaying state gets multiplied by $(-1)^{r+n}$ ($r = m$ in the case of circular polarization) when the decaying state is rotated by 180° about the z axis [see Eqs. (6) and (27)], while it gets multiplied by $(-1)^{P+n}$ when the decaying state is reflected in the origin [see Eqs. (6) and (31)]. Consequently, the full decaying state has a definite parity equal to $r+P$ when both operations are combined. Since a rotation about the z axis by 180° combined with a reflection in the origin yields a reflection in the xy plane, we see that for elliptic or circular polarization the parity associated with reflection in the xy plane is conserved for the atom decaying in the field [31]. Knowing now which manifolds of states are left uncoupled by the radiation field, we can be somewhat more specific about the condition of validity, Eq. (16), namely about the question of which manifold of states — with lowest energy eigenvalue $W_\lambda^0(\alpha_0)$ — should be considered in Eq. (16). It will now be clear that, in the case of linear polarization, this is the manifold of states with the same magnetic quantum number as the initial state, whereas for circularly or elliptically polarized fields, it is the manifold of states with the same parity with respect to a reflection in the polarization plane as the initial state.

Let us finally note that the well-known selection rule in LOPT, which states that each absorbed photon is associated with a change of the orbital angular momentum of the atom by one unit, does not apply to the present situation, since (unlike the dipole operator in LOPT) in the Gavrila-Kaminski theory the transition operator V_n is not a spherical tensor operator of rank 1. The fact that this rule does not apply to the present theory is a direct consequence of its nonperturbative nature. As a consequence, the distributions of photoelectrons predicted by our theory generally possess a more complicated angular dependence than that obtained in LOPT. For example, according to LOPT, single-photon ionization from an s state leads to outgoing electron waves of p-type only. In our case, ionization from the (radiatively deformed) initial state which (adiabatically) develops from it leads to outgoing electron waves consisting of p, f, h, etc. partial waves. In fact, in this particular example of single-photon ionization, LOPT predicts a "fourfold" symmetry for the angular dependence of photoelectrons created by an elliptically polarized light source of low intensity (see Sec. IV). However, in more intense fields this fourfold symmetry will be broken. This feature (an example of a typical nonperturbative feature, i.e., which LOPT fails to predict) is contained in the Gavrila-Kaminski theory.

IV. THE CONNECTION WITH PERTURBATION THEORY

In this section we demonstrate that for low intensities, the Gavrila-Kaminski theory yields angular-dependent decay rates that are in agreement with LOPT, evaluated at high frequencies, in accordance with what was said in Sec. II of this paper. We will demonstrate this in the

following for the simpler case of single-photon ionization. The more general case will be considered in Ref. [32].

In LOPT (choosing the velocity gauge) the single-photon ionization amplitude $f_\lambda(\hat{\mathbf{r}})$ is given by

$$f_\lambda^{\mathrm{LOPT}}(\hat{\mathbf{r}}) = \frac{i\sqrt{I}}{4\pi\omega}\left\langle u_{\mathbf{k}}^{(-)}\left|\epsilon\cdot\mathbf{P}\right|u_\lambda\right\rangle . \tag{37}$$

Here u_λ and $u_{\mathbf{k}}^{(-)}$ represent eigenstates of the (unperturbed) Hamiltonian $H_0 = \mathbf{P}^2/2 + V$ at energies $W_\lambda(0)$ and $k^2/2 = W_\lambda(0)+\omega$, respectively. In the general case of arbitrary polarization, the *polarization vector* ϵ is complex and given by

$$\epsilon = \mathbf{e}_1\cos(\chi/2) + i\mathbf{e}_2\sin(\chi/2). \tag{38}$$

[Note that it follows from Eq. (38) that ϵ is of unit length in the generalized sense, i.e., $\epsilon\cdot\epsilon^* = 1$.]

In order to make the connection with the Gavrila-Kaminski expression, Eq. (15), we cast Eq. (37) in the so-called acceleration form [33]

$$f_\lambda^{\mathrm{LOPT}}(\hat{\mathbf{r}}) = -\frac{\sqrt{I}}{4\pi\omega^2}\left\langle u_{\mathbf{k}}^{(-)}\left|(\epsilon\cdot\boldsymbol{\nabla})V\right|u_\lambda\right\rangle . \tag{39}$$

Let us now compare Eq. (39) with the Gavrila-Kaminski expression, Eq. (15). In order to make the connection we first replace Φ_λ and $\Phi_{\mathbf{k}}^{(-)}$ by their versions at $\alpha_0 = 0$, u_λ, and $u_{\mathbf{k}}^{(-)}$, respectively. (Note that at low enough intensity, we have $\alpha_0 \ll 1$.) Next we replace the transition operator V_1 by its low-α_0 form [34]

$$V_1 \simeq \frac{\sqrt{I}}{2\omega^2}(\epsilon\cdot\boldsymbol{\nabla})V . \tag{40}$$

With these substitutions Eq. (15) becomes identical to Eq. (39). Thus we conclude that for *single-photon* ionization, the Gavrila-Kaminski theory yields angular-dependent ionization decay rates in *exact* agreement with LOPT, irrespective of the frequency, provided $\omega > |W_\lambda^0(0)|$. This is related to the absence of Green's functions in the general LOPT expression for the MPI amplitude if $n = 1$. However, in the case of *absorption of more than one photon*, Green's functions do occur in this expression and the Gavrila-Kaminski theory agrees with results obtained by LOPT *only for high enough frequency* [see Eq. (16)].

Let us recall that for the case of atomic hydrogen, the eigenfunctions u_λ and $u_{\mathbf{k}}^{(-)}$ are known in closed form and Eq. (37) can then be evaluated analytically [35]. The high-frequency behavior of the decay rate can be obtained directly from Eq. (37) [36]. For high frequencies, the Coulomb final state in Eq. (37) can be replaced by a plane wave (Born approximation), since in that case the photoelectron is ejected with high speed (neglect of "final-state interaction"). An elementary calculation from Eq. (37) then yields

$$\frac{d\Gamma}{d\Omega} = \frac{Ik}{2\pi\omega^5}|\epsilon\cdot\mathbf{e}_k|^2 \simeq \frac{I}{\sqrt{2}\pi\omega^{9/2}}|\epsilon\cdot\mathbf{e}_k|^2 \tag{41}$$

and the corresponding angular-integrated decay rate yields

$$\Gamma \simeq \frac{2^{3/2}I}{3\omega^{9/2}}, \tag{42}$$

in agreement with the high-frequency form of the LOPT expression [35]. In Sec. V we will *generalize* this well-known result to the case of (arbitrarily) *strong fields*.

Let us finally remark that, in LOPT, single-photon ionization from an s state leads to an angular dependence given by $|\epsilon\cdot\mathbf{e}_k|^2$ for *any* spherically symmetric atomic binding potential. [This notion can be easily checked by making a partial wave analysis of Eq. (37).] For elliptic polarization we find that the angular dependence of photoelectrons is given by $d\Gamma/d\Omega \propto \sin^2\theta[\cos^2(\chi/2)\cos^2\phi + \sin^2(\chi/2)\sin^2\phi]$. Note that the angular distribution of photoelectrons created by elliptically polarized light has a *fourfold* symmetry (reflection symmetry with respect to a plane perpendicular to the polarization plane and passing through the major or minor axis of the ellipse described by the tip of the electric-field vector).

V. THE BEHAVIOR OF THE MULTIPHOTON IONIZATION AMPLITUDE AT HIGH FREQUENCIES (ATOMIC HYDROGEN)

In this section we will derive an asymptotic expression for the angular-dependent multiphoton ionization amplitude $f_{\lambda,n}$ of Eq. (15), valid in the limit of high frequencies (say $\omega\to\infty$), *irrespective of the value of* α_0. Henceforth we will assume that the atomic binding potential is of Coulomb form, i.e., $V(\mathbf{r}) = -1/|\mathbf{r}|$. On the other hand, no assumptions about the state of polarization are made.

In order to obtain the asymptotic behavior of Eq. (15) for high frequencies, we first replace $\Phi_{\mathbf{k}_n}^{(-)}$ by a plane wave. Thus

$$f_{\lambda,n} = -\frac{1}{2\pi}\langle\mathbf{k}_n|V_n|\Phi_\lambda\rangle . \tag{43}$$

The replacement of $\Phi_{\mathbf{k}_n}^{(-)}$ by a plane wave requires some comments. In the first place, we would like to remind the reader that our calculations are carried out in the Kramers frame of reference, oscillating along with the classical free electron in the radiation field. In fact, in the laboratory frame of reference our plane wave corresponds to the exact solution of the Schrödinger equation for a free electron driven by the oscillating electric field, commonly referred to as the "Volkov solution" [37]. Therefore the replacement of the final state by a plane wave fully accounts for the interaction of the electron ejected from the atom with the radiation field. This approximation for $\Phi_{\mathbf{k}_n}^{(-)}$ represents in fact the Born approximation to the scattering wave function. The physical reason for making this approximation is that, because of the high frequency, the electron is ejected with large momentum [see the energy conservation equation Eq. (7)]. Due to the short interaction time during which the electron leaves the atom, the attraction by the dressed potential on its way out (the final-state interaction) can be neglected. Our expression Eq. (43) is therefore exact in the limit of high frequencies, irrespective of the value of α_0.

A *practical* range of the validity of our approximation Eq. (43) is difficult to assess. Taking the ionization po-

tential as a measure of the strength of the dressed potential, we will assume its applicability under circumstances where the photoelectron energies are large with respect to a characteristic binding energy of the atom *in the field.* (The fact that for frequencies high with respect to the ionization potential the final-state interaction can be neglected was illustrated in Sec. IV for the case of single-photon ionization from the ground state of atomic hydrogen in weak fields, i.e., for which $\alpha_0 \ll 1$.) However, our starting point Eq. (15) is already subject to the condition of validity, Eq. (16). We will assume the applicability of Eq. (43) whenever condition Eq. (16) is satisfied. As α_0 increases, the dressed potential gets progressively weaker. This is clearly reflected by the steep decrease of the ionization potential with α_0 that we have seen earlier (see Ref. [4] for a discussion of this phenomenon). Note that, because of this decrease, the high-frequency condition Eq. (16) is much easier to satisfy at high α_0 than at low values of α_0.

In view of Eq. (11) we may write Eq. (43) in the form

$$f_{\lambda,n} = \frac{1}{2\pi}\int_{-\pi}^{+\pi} e^{in\phi} F_\lambda(\mathbf{k}_n, -\boldsymbol{\alpha}(\phi))d\phi, \tag{44}$$

where we have introduced the "generating function" $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ by

$$F_\lambda(\mathbf{k},\boldsymbol{\zeta}) = \frac{1}{2\pi}\int \frac{e^{-i\mathbf{k}\cdot\mathbf{r}}}{|\mathbf{r}-\boldsymbol{\zeta}|}\Phi_\lambda(\mathbf{r})d\mathbf{r}. \tag{45}$$

[In Eq. (11) we have set $V(\mathbf{r}) = -1/|\mathbf{r}|$.] In order to obtain the high-frequency behavior of the multiphoton ionization amplitude we need the high-k behavior of $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ in Eq. (45). To extract this limit, we cast $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ in the form

$$F_\lambda(\mathbf{k},\boldsymbol{\zeta}) = \frac{e^{-i\mathbf{k}\cdot\boldsymbol{\zeta}}}{2\pi}\int \frac{e^{-i\mathbf{k}\cdot\mathbf{r}}}{|\mathbf{r}|}\Phi_\lambda(\mathbf{r}+\boldsymbol{\zeta})d\mathbf{r}. \tag{46}$$

The high-k behavior of $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ can now be obtained by scaling the integration variable $\mathbf{r}$ with k. Thus setting $\mathbf{r} = \boldsymbol{\xi}\, k^{-1}$ in Eq. (46) yields

$$F_\lambda(\mathbf{k},\boldsymbol{\zeta}) = \frac{e^{-i\mathbf{k}\cdot\boldsymbol{\zeta}}}{2\pi k^2}\Phi_\lambda(\boldsymbol{\zeta})\int \frac{e^{-i\mathbf{e}_k\cdot\boldsymbol{\xi}}}{|\boldsymbol{\xi}|}\frac{\Phi_\lambda(\boldsymbol{\xi}\, k^{-1}+\boldsymbol{\zeta})}{\Phi_\lambda(\boldsymbol{\zeta})}d\boldsymbol{\xi}\,, \tag{47}$$

where $\mathbf{e}_k$ denotes the unit vector in the direction of $\mathbf{k}$. Note that if $k \to \infty$ the ratio of $\Phi_\lambda(\boldsymbol{\xi}\, k^{-1}+\boldsymbol{\zeta})$ and $\Phi_\lambda(\boldsymbol{\zeta})$ simply plays the role of a factor of convergence in the above integral, which in this limit yields 4π. With this value $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ becomes

$$F_\lambda(\mathbf{k},\boldsymbol{\zeta}) \simeq \frac{2e^{-i\mathbf{k}\cdot\boldsymbol{\zeta}}}{k^2}\Phi_\lambda(\boldsymbol{\zeta}). \tag{48}$$

This expression is exact in the limit of high k.

Let us now derive a practical estimate of the range of validity of the above relation. In order to replace the integral in Eq. (47) by its high-k limit, the ratio of $\Phi_\lambda(\boldsymbol{\xi}\, k^{-1}+\boldsymbol{\zeta})$ and $\Phi_\lambda(\boldsymbol{\zeta})$ should not differ too much from unity as ξ increases from zero to values for which the integrand still contributes significantly to the integral, say for values of ξ of a few times unity. Since in Eq. (44) $\boldsymbol{\zeta}$ is in fact restricted to the orbit described by the proton in the Kramers reference frame, we may express this condition in more physical terms as follows: The wave function of the initial state should practically stay unaltered if excursions of the order of the wavelength of the ejected electron from this orbit are made. We will be interested mainly in ionization from the ground state. We may estimate for this case the characteristic distance of variation of the wave function by $|W_0^0(\alpha_0)|^{-1/2}$. If we require Eq. (48) to hold for all open channels, we find that the present estimate again coincides with the high-frequency condition Eq. (16) [38].

By substituting Eq. (48) into Eq. (44), we finally obtain the following expression for the high-frequency behavior of the n-photon ionization amplitude $f_{\lambda,n}$:

$$f_{\lambda,n} = \frac{1}{\pi k_n^2}\int_{-\pi}^{+\pi} e^{in\phi}e^{i\boldsymbol{\alpha}(\phi)\cdot\mathbf{k}_n}\Phi_\lambda(-\boldsymbol{\alpha}(\phi))d\phi, \tag{49}$$

in the form of a one-dimensional integral. It is interesting to note that the prefactor in Eq. (49) is in fact the Fourier transform of the Coulomb potential. This indicates that it is possible to generalize the expression of Eq. (49) to a class of atomic binding potentials (singular at $\mathbf{r} = \mathbf{0}$) by replacing the prefactor in Eq. (49) by the Fourier transform of the atomic binding potential under consideration. We will not explore this issue any further here and focus our interest on the case of atomic hydrogen.

Let us briefly comment here on the relation of Eq. (49) to LOPT. Since Eq. (49) is valid for any value of α_0, provided that the frequency satisfies $\omega \gg |W_\lambda^0(\alpha_0)|$, it should be possible to recover Eq. (41) in the limit of vanishingly small intensities. Let us demonstrate this here. For low enough intensities, α_0 is small and we have

$$\Phi_\lambda(-\boldsymbol{\alpha}(\phi)) \simeq \Phi_\lambda(0), \tag{50}$$

where $\Phi_\lambda(\mathbf{0})$ denotes the value of the *unperturbed* initial state (which we assume here to be of s symmetry) at the origin. Since also $\alpha_0 k_n \ll 1$, we may expand the exponential in Eq. (49) in powers of $\boldsymbol{\alpha}(\phi)\cdot\mathbf{k}_n$. Making these replacements, Eq. (49) with $\boldsymbol{\alpha}(\phi)$ given by Eq. (3) yields (retaining only the lowest-order term in powers of the intensity, which corresponds to single-photon ionization)

$$\frac{d\Gamma}{d\Omega} = \frac{\alpha_0^2}{k\cos^2(\chi/2)}|\boldsymbol{\epsilon}\cdot\mathbf{e}_k|^2|\Phi(0)|^2$$
$$\simeq \frac{I}{\sqrt{2}\omega^{9/2}}|\boldsymbol{\epsilon}\cdot\mathbf{e}_k|^2|\Phi(0)|^2. \tag{51}$$

In the last step we have made the replacements $\alpha_0 = I^{1/2}\omega^{-2}\cos(\chi/2)$ [see Eq. (4)] and $k \simeq \sqrt{2\omega}$ [we used Eq. (7), i.e., $k_n = \sqrt{2[E(\alpha_0)+n\omega]}$ for the case of single-photon ionization, and invoked the condition of high-frequency $\omega \gg |W_\lambda^0(\alpha_0)|$]. For s states of atomic hydrogen, we have the well-known relation $|\Phi(0)|^2 = 1/\pi n^3$, where n denotes the principal quantum number. Note that for principal quantum number $n = 1$, Eq. (51) is identical to the high-frequency form of the LOPT result, Eq. (41). This demonstrates that if $\alpha_0 k_n \ll 1$ and $\alpha_0 \ll 1$, Eq. (49) yields results that are in agreement with LOPT.

For further analysis of Eq. (49) (as well as for numerical evaluations) it is preferable to express it as an (infinite) sum of Bessel functions. This can be done by substituting into Eq. (49) the Fourier series of the periodic $\Phi_\lambda(-\alpha(\phi))$:

$$\Phi_\lambda(-\alpha(\phi)) = \sum_{m=-\infty}^{+\infty} e^{-im\phi}\Phi_{\lambda,m}. \tag{52}$$

This yields

$$f_{\lambda,n} = \frac{2}{k_n^2}\sum_{m=-\infty}^{+\infty} \mathcal{I}_{n-m}\Phi_{\lambda,m}, \tag{53}$$

where we have defined the integrals $\mathcal{I}_m$ by

$$\mathcal{I}_m = \frac{1}{2\pi}\int_{-\pi}^{+\pi} e^{im\phi}e^{i\boldsymbol{\alpha}(\phi)\cdot\mathbf{k}_n}d\phi. \tag{54}$$

Note that the n-photon ionization amplitude $f_{\lambda,n}$ given in Eq. (53) depends on the polarization through the $\Phi_{\lambda,m}$'s and $\mathcal{I}_m$'s.

By making use of the well-known integral representation of the Bessel functions of integer order [39]

$$J_m(x) = \frac{1}{2\pi i^m}\int_{-\pi}^{+\pi} e^{im\phi}e^{ix\cos\phi}d\phi, \tag{55}$$

we may express the integral $\mathcal{I}_m$ – with $\boldsymbol{\alpha}(\phi)$ given by Eq. (3) — into a Bessel function of order m. In the general case of elliptic polarization ($0° < |\chi| < 90°$) $\mathcal{I}_m$ becomes

$$\mathcal{I}_m = i^m e^{im\beta}J_m(\alpha_0 k_n \sin\theta\sqrt{\cos^2\varphi + \sin^2\varphi\tan^2(\chi/2)}), \tag{56}$$

where we have defined the angle β by

$$\beta = \arg\left(\frac{\cos\varphi + i\sin\varphi\tan(\chi/2)}{\sqrt{\cos^2\varphi + \sin^2\varphi\tan^2(\chi/2)}}\right). \tag{57}$$

In the case of circularly polarized light ($\chi = \pm 90°$) this reduces to

$$\mathcal{I}_m = i^m e^{\pm im\varphi}J_m(\alpha_0 k_n \sin\theta), \tag{58}$$

where the upper (lower) sign corresponds to left- (right-) handed polarization. In the case of linear polarization ($\chi = 0°$) Eq. (54) becomes

$$\mathcal{I}_m = i^m J_m(\alpha_0 k_n \cos\theta). \tag{59}$$

Note that Eq. (59) does not connect continuously with Eq. (56) as the angle χ goes to zero, because of our choice of coordinate system.

Among the possible states of polarization, the case of *circular polarization* takes a special position. Indeed, we see that for an initial (deformed) bound state of definite magnetic quantum number m only one term in the expansion of Eq. (52) survives: $\Phi_{\lambda,s} = \delta_{s,\mp m}\Phi_\lambda(-\alpha_0\mathbf{e}_x)$. Thus we find from Eqs. (53) and (58) that the n-photon ionization amplitude $f_{\lambda,n}$ takes the particularly simple form

$$f_{\lambda,n} = \frac{2}{k_n^2}i^{n\pm m}J_{n\pm m}(\alpha_0 k_n \sin\theta)\Phi_\lambda(-\alpha_0\mathbf{e}_x)e^{\pm i(n\pm m)\varphi} \tag{60}$$

and the corresponding angular-dependent ionization decay rate becomes

$$\frac{d\Gamma_{\lambda,n}}{d\Omega} = \frac{4}{k_n^3}J_{n\pm m}^2(\alpha_0 k_n \sin\theta)|\Phi_\lambda(-\alpha_0\mathbf{e}_x)|^2. \tag{61}$$

From Eq. (60) we see that in the case of circularly polarized light, the ionization amplitude factorizes into a product of a term which derives from the atomic binding potential, the probability amplitude to find the electron on the circle of charge (with radius α_0) generating the dressed potential and a Bessel function. It is interesting to note that a similar factorization occurs in the celebrated Kroll and Watson theory connected to a different but to some extent related physical situation, namely that of free-free scattering in intense, low-frequency radiation fields [40].

Equation (53) is an important result and forms a considerable simplification over the original Gavrila-Kaminski expression Eq. (15). [For example, in Eq. (61) only the amplitude to find the electron at the circle of charge and the binding energy (namely via the final momentum k_n) enter as unknown.] At the same time, it has essentially the same range of validity, but it is obviously less accurate at high, but finite frequencies. It does not, for example, yield the (weak) asymmetries in the angular distributions of photoelectrons which occur in the case of elliptically polarized light, as discussed in Sec. III. Once the initial-state wave function has been calculated, the extraction of the Fourier components $\Phi_{\lambda,n}$ defined in Eq. (52) followed by the evaluation of Eq. (53) is a straightforward numerical exercise. Moreover, it is a very useful starting point for studying analytically the general behavior of angular dependences of photoelectrons, branching ratios for decay by multiphoton ionization of the atom as a function of the number of photons absorbed, etc., under certain special radiation conditions, in particular when the interaction of the light field with the atom becomes highly nonperturbative (see the following paper) [41].

From Eq. (54) we have the relation $\mathcal{I}_m = \delta_{m,0}$, valid for $\mathbf{k}_n$ orthogonal to $\boldsymbol{\alpha}(\phi)$. From this notion and Eq. (53) we find a particularly simple relation between the angular-dependent rate for electrons ejected in a direction perpendicular to the plane of polarization for elliptically or circularly polarized light (or in a direction perpendicular to the axis of polarization for the case of linearly polarized light), and the nth Fourier component of $\Phi_\lambda(-\alpha(\phi))$ of Eq. (52). Thus we find for elliptic or circular polarization

$$\frac{d\Gamma_{\lambda,n}(\theta = 0°, 180°)}{d\Omega} = \frac{4}{k_n^3}|\Phi_{\lambda,n}|^2, \tag{62}$$

whereas for linear polarization this relation becomes

$$\frac{d\Gamma_{\lambda,n}(\theta = 90°)}{d\Omega} = \frac{4}{k_n^3}|\Phi_{\lambda,n}|^2. \tag{63}$$

Here we have used the expression for the angular-dependent rate in terms of the n-photon ionization amplitude, Eq. (8). By inspecting the behavior of $\Phi_\lambda(-\boldsymbol{\alpha}(\phi))$ in Eq. (52) under the increase of ϕ by π, we find that $\Phi_{\lambda,n}$ is identically zero for gerade states if n is odd, whereas it vanishes in the case of ungerade states if n is even. Consequently, we find that for elliptically or circularly polarized light, $d\Gamma_{\lambda,n}(\theta = 0°, 180°)/d\Omega$ vanishes for n odd in the case of gerade states and for n even in the case of ungerade states. The same applies to $d\Gamma_{\lambda,n}(\theta = 90°)/d\Omega$ in the case of linearly polarized light [42].

In Eq. (49) only these values of Φ_λ enter that during the oscillatory motion of the (distorted) electron cloud in the laboratory frame of reference pass over the position of the proton. This is in itself a remarkable result. Equation (49) can be given an interesting *dynamical physical interpretation*. To this end consider the following expression:

$$\tilde{\Psi}_\lambda(\mathbf{r},t) = \int d\mathbf{r}' \int dt' \mathcal{G}_0^{(+)}(\mathbf{r},\mathbf{r}',t-t')\rho_\lambda(\mathbf{r}',t'), \tag{64}$$

with

$$\rho_\lambda(\mathbf{r},t) = -\frac{\Phi_\lambda(-\boldsymbol{\alpha}(t))\exp(-iW_\lambda t)}{|\mathbf{r}+\boldsymbol{\alpha}(t)|}. \tag{65}$$

In Eq. (64) we have denoted by $\mathcal{G}_0^{(+)}(\mathbf{r},\mathbf{r}',t)$ the time-dependent (retarded) Green's function for a free electron. Our interest in Eq. (64) lies in the fact that it represents an approximation for the *open-channel part* of the decaying state of Eq. (5) [43]. Indeed, one can easily convince oneself [by Floquet analyzing the approximation Eq. (64) and determining the asymptotic behavior of its Floquet components for large $|\mathbf{r}|$, following Eq. (6)], that the multiphoton ionization amplitudes associated with Eq. (64) coincide with those given in Eq. (49).

From Eq. (64) we see that $\tilde{\Psi}_\lambda(\mathbf{r},t)$ represents a superposition of outgoing waves, created (at any time in the past) by the source density $\rho_\lambda(\mathbf{r},t)$ given by Eq. (65). Thus the amplitude of the outgoing waves is proportional to the strength of the electrostatic interaction between the electron and the proton [$= -1/|\mathbf{r}+\boldsymbol{\alpha}(t)|$] and the amplitude for finding the electron at the momentary position of the proton in the Kramers reference frame [$= e^{-iW_\lambda t}\Phi_\lambda(-\boldsymbol{\alpha}(t))$]. This suggests that, under conditions of high frequency, outgoing electron waves, corresponding to ionization of the atom, are created in the near vicinity of the proton.

It is interesting to note here that such an outcome is also obtained from a classical consideration. [We leave here aside under what conditions a (semi-) classical approach to our problem is valid.] As we have seen, in the Kramers frame of reference the electron is subject to the electrostatic force exerted by the (harmonically oscillating) proton. Suppose that *during one cycle of the oscillation*, the electron position vector $\mathbf{r}(t)$ appearing in Newton's dynamical equation of motion $d^2\mathbf{r}(t)/dt^2 = -\nabla V(\mathbf{r}(t)+\boldsymbol{\alpha}(t))$ can be replaced by a constant on its right-hand side. It is then easily shown that, within this approximation, the electron motion is dictated by the time-averaged interaction potential (the dressed potential) with a periodic oscillatory motion superposed on it (not to be confused with the jitter motion of a free electron in the laboratory frame of reference) and consequently the atom is *stable*. (The energies of the electron before and after one cycle are the same.) Deviations from this description lead to instability of the electron trajectory, i.e., to ionization. Of course, the validity of the picture sketched above relies on the following assumptions: (a) the frequency is sufficiently high, (b) the electron moves sufficiently slowly, and (c) the atomic binding potential is sufficiently smooth. It thus follows from the above discussion that, for a given high frequency, it is only during times when the electron passes close to the proton that it can pick up significant amounts of energy with which it can eventually leave the atomic system (*ionization*).

One should be aware that our result that only the behavior of the initial-state wave function near the proton is relevant for the ionization process is typical for the high-frequency regime that we are studying and can by no means be generalized to the case of arbitrary frequency [except for the obvious case of an atomic binding potential of a (very) short range].

Equation (49) obviously yields zero if the initial-state wave function Φ_λ vanishes on the distribution of charge creating the dressed potential. In the linear case this happens for states with $m \neq 0$, while in the elliptic or circular case this happens for states which are antisymmetric with respect to a reflection in the plane of polarization. It is self-evident that to these cases Eq. (49) [or Eq. (53)] does not apply; the leading term in ω^{-1} of $f_{\lambda,n}$ is apparently of higher order. Equation (49) can be generalized to these cases. The high-k behavior of $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ in the more general case can be obtained by setting $\mathbf{r} = \boldsymbol{\xi}\, k^{-1}$ in Eq. (46), as above. The expansion of $\Phi_\lambda(\boldsymbol{\xi}\, k^{-1}+\boldsymbol{\zeta})$ in a Taylor series about the point $\boldsymbol{\zeta}$ then yields $F_\lambda(\mathbf{k},\boldsymbol{\zeta})$ in the form of an asymptotic series in k^{-1}. The high-ω behavior of $f_{\lambda,n}$ can be obtained by retaining only the lowest nonvanishing term of this series and by the insertion of this into Eq. (44). For the case that $\nabla\Phi_\lambda$ is nonvanishing on the distribution of charge, this leads to

$$\begin{aligned} F_\lambda(\mathbf{k},\boldsymbol{\zeta}) &= \frac{e^{-i\mathbf{k}\cdot\boldsymbol{\zeta}}}{2\pi k^3}\nabla\Phi_\lambda(\boldsymbol{\zeta})\cdot\int e^{-i\mathbf{e}_k\cdot\boldsymbol{\xi}}\frac{\boldsymbol{\xi}}{|\boldsymbol{\xi}|}d\boldsymbol{\xi} \\ &= \frac{4e^{-i\mathbf{k}\cdot\boldsymbol{\zeta}}}{ik^3}\mathbf{e}_k\cdot\nabla\Phi_\lambda(\boldsymbol{\zeta}). \end{aligned} \tag{66}$$

Thus the n-photon ionization amplitude becomes

$$f_{\lambda,n} = \frac{2}{i\pi k_n^3}\int_{-\pi}^{+\pi} e^{in\phi}e^{i\boldsymbol{\alpha}(\phi)\cdot\mathbf{k}_n}\mathbf{e}_k\cdot\nabla\Phi_\lambda(-\boldsymbol{\alpha}(\phi))d\phi. \tag{67}$$

In the following paper, we will analyze the consequences of Eqs. (52),(53), and (59) for the case of atomic hydrogen in a superintense, high-frequency laser field of linear polarization (see also Ref. [24]).

ACKNOWLEDGMENTS

The work presented here was carried out at the FOM—Institute for Atomic and Molecular Physics,

Kruislaan 407, 1098 SJ Amsterdam, the Netherlands. It was part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (Foundation for the Fundamental Research on Matter) and was made possible by financial support from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organization for the Advancement of Research). The author is greatly indebted to Dr. M. Gavrila, with whom he had many interesting and stimulating discussions. This work was finalized for publication by support of the National Science Foundation under Grant No. PHY-9017079.

[1] *Ultrafast Phenomena VI*, edited by T. Yajima, K. Yoshihara, C. B. Harris, and S. Shionoya (Springer, Berlin, 1988), Pts. I and II.

[2] M. Gavrila and J.Z. Kaminski, Phys. Rev. Lett. **52**, 614 (1984), and unpublished.

[3] M. Gavrila, in *Atoms in Unusual Situations*, Vol.143 of *NATO Advanced Study Institute Series B: Physics*, edited by J.P. Briand (Plenum, New York, 1987), p.225.

[4] For applications to atomic deformation in linearly polarized fields, see M. Pont, N. Walet, M. Gavrila, and C.W. McCurdy, Phys. Rev. Lett. **61**, 939 (1988); M. Pont, N. Walet, and M. Gavrila, Phys. Rev. A **41**, 477 (1990).

[5] For applications to atomic deformation in circularly polarized fields, see M. Pont, M.J. Offerhaus, and M. Gavrila, Z. Phys. D **9**, 297 (1988); M. Pont, Phys. Rev. A **40**, 5659 (1989).

[6] The following references concern applications of the Gavrila-Kaminski theory to scattering. The Coulomb case was treated by M.J. Offerhaus, J.Z. Kaminski, and M. Gavrila, Phys. Lett. A **112**, 151 (1985); J. van de Ree, J.Z. Kaminski, and M. Gavrila, Phys. Rev. A **37**, 4536 (1988); the Yukawa case by M. Gavrila, M.J. Offerhaus, and J.Z. Kaminski, Phys. Lett. A **118**, 331 (1986).

[7] G. Leuchs and H. Walther, in *Multiphoton Ionization of Atoms*, edited by S.L. Chin and P. Lambropoulos (Academic, New York, 1984), Chap. 5.

[8] This is clearly an idealization of the pulsed beams which are used in experiments. These pulses have a certain inhomogeneity in space and time. A comparison of theory with experiment requires special care with regard to this point. It is possible to obtain excellent agreement with experiment by incorporating these effects [in connection with this, see in particular, M. Dörr, R.M. Potvliege, and R. Shakeshaft, Phys. Rev. A **41**, 558 (1990)].

[9] We have adopted here the convention of the book by Hecht and Zajac [see E. Hecht and A. Zajac, *Optics* (Addison Wesley, Reading, MA, 1974), Chap. 8], which is also the one adopted in Ref. [21]. The sense of polarization is then defined as the direction in which the electric vector rotates as seen by an observer towards whom the wave is moving.

[10] H.A. Kramers, *Collected Scientific Papers* (North-Holland, Amsterdam, 1956), p. 866.

[11] W. Pauli and M. Fierz, Nuovo Cimento **15**, 167 (1938).

[12] W.C. Henneberger, Phys. Rev. Lett. **21**, 838 (1968).

[13] Note that the shift to the accelerated frame of reference is only an interpretation of the Kramers transformation and that no assumptions concerning the validity of the Schrödinger equation in the accelerated frame of reference are needed.

[14] J.H. Shirley, Phys. Rev. B **138**, 979 (1965).

[15] A.J. Siegert, Phys. Rev. **56**, 750 (1939).

[16] J.N. Bardsley and M.J. Comella, Phys. Rev. A **39**, 2252 (1989).

[17] R. Bhatt, B. Piraux, and K. Burnett, Phys. Rev. A **37**, 98 (1988).

[18] J.I. Gersten and M.H. Mittleman, J. Phys. B **9**, 2561 (1976).

[19] M.H. Mittleman, *Theory of Laser-Atom Interactions* (Plenum, New York, 1982), Chap. 3.

[20] They formulated, however, the validity condition for the purely plane-wave case (and for the case of circular polarization they were considering) too restrictively, by requiring that in Eq. (16) ω be large with respect to the field-free ($\alpha_0 = 0$) value of the ionization potential $|W_0^0(0)|$, instead of $|W_\lambda^0(\alpha_0)|$.

[21] M. Bashkansky, P.H. Bucksbaum, and D.W. Shuhmacher, Phys. Rev. Lett. **60**, 2458 (1988).

[22] P. Lambropoulos and X. Tang, Phys. Rev. Lett. **61**, 2506 (1988).

[23] In the general case of elliptic polarization this yields the group D_{2h}. In the special cases of linear and circular polarization the symmetry group is $D_{\infty h}$.

[24] M. Pont, Ph.D. thesis, University of Amsterdam, 1990 (unpublished).

[25] Invariance under complex conjugation implies that V_0 is real. When we also make use of this latter notion, we find that V_0 is also invariant with respect to a reflection in the xy plane. However, we need not make use of these properties separately. As one can easily check, invoking these invariance properties separately does not yield any new information about the invariance properties of the angular dependences of photoelectrons. The reason that reflection in the xz plane appears in combination with the operation of complex conjugation is the following one. The dynamics of the atom in the radiation field of Eq. (3) is invariant under "time reversal" only in conjunction with a reflection in the xz plane. This can be easily checked by carrying out the appropriate substitutions into the time-dependent Schrödinger equation, Eq. (2). [Time-reversal is defined by the transformation $\Psi(\mathbf{r}, t) \rightarrow \Psi^*(\mathbf{r}, -t)$.]

[26] L.V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys.—JETP **20**, 1307 (1965)].

[27] F. Faisal, J. Phys. B **6**, L312 (1973).

[28] H.R. Reiss, Phys. Rev. A **22**, 1786 (1980).

[29] We are referring here to the standard KFR theory. As others have shown [see, e.g., S. Basile, F. Trombetta, and G. Ferrante, Phys. Rev. Lett. **61**, 2453 (1988)], it is possible to modify the theory so as to take into account the final-state interaction. In that case there is allowance for the asymmetry.

[30] H.G. Muller, G. Petite, and P. Agostini, Phys. Rev. Lett. **61**, 2507 (1988).

[31] These conservation laws are immediately apparent from the symmetry of the dynamics of the atom in the radiation field. In the case of linear polarization, the (time-dependent) Schrödinger equation Eq. (2) is invariant with

respect to a rotation about the axis of polarization, while in the case of elliptic or circular polarization it is invariant with respect to a reflection in the plane of polarization.

[32] M. Gavrila and J.Z. Kaminski (unpublished).

[33] H.A. Bethe and E.E. Salpeter, in *Quantum Mechanics of One- and Two Electron Systems*, edited by S. Flügge, Encyclopedia of Physics XXXV Pt. I (Springer, Berlin, 1957) p. 338. This form is obtained from Eq. (37) by application of the identity $[H_0, \epsilon \cdot \mathbf{P}] = i(\epsilon \cdot \nabla)$ V and by noting that u_λ and $u_{\mathbf{k}_n}^{(-)}$ are eigenstates of H_0 at the aforementioned energies. [The terminology "acceleration form" is derived from the appearance of the force $\mathbf{F} = -\nabla V$ in the matrix element of Eq. (39).]

[34] We have, from Eqs. (3), (4), and (38),

$$\alpha(\phi) = \frac{\sqrt{I}e^{-i\phi}}{2\omega^2}\epsilon + \text{c.c.},$$

with $\epsilon = \mathbf{e}_1 \cos(\chi/2) + i\mathbf{e}_2 \sin(\chi/2)$ $(\epsilon \cdot \epsilon^* = 1)$ and c.c. denoting complex conjugate. By expanding in Eq. (11) $V(\mathbf{r} + \alpha(\phi))$ in powers of $\alpha(\phi)$ and retaining only the lowest nonvanishing term in α_0, we obtain

$$\begin{aligned} V_1(\alpha_0, \mathbf{r}) &\simeq \frac{1}{2\pi}\int_{-\pi}^{+\pi} e^{i\phi}\{1 + [\alpha(\phi)\cdot\nabla]\} \\ &\quad \times V(\mathbf{r})d\phi + \mathcal{O}(\alpha_0^2) \\ &= \left(\frac{1}{2\pi}\int_{-\pi}^{+\pi} e^{i\phi}\alpha(\phi)d\phi\right)\cdot\nabla V(\mathbf{r}) \\ &\quad + \mathcal{O}(\alpha_0^2). \end{aligned}$$

Insertion of the above expression for $\alpha(\phi)$ yields immediately the desired result of Eq. (40).

[35] A. Sommerfeld and G. Schur, Ann. Phys. (Leipzig) **4**, 409 (1920); see also W. Heitler, *The Quantum Theory of Radiation*, (Oxford University Press, Oxford, 1954), Sec. 21. When expressed in our quantities and units one obtains for ionization from the ground state the well-known result

$$\frac{d\Gamma^{\text{LOPT}}}{d\Omega} = \frac{I}{\omega^5}\mathcal{F}(k)|\epsilon \cdot \mathbf{e}_k|^2,$$

valid irrespective of the polarization. The factor $\mathcal{F}$ is given by

$$\mathcal{F}(k) = \frac{\exp(-4/k \arctan k)}{1 - \exp(-2\pi/k)}.$$

For high frequencies, we have $k \simeq \sqrt{2\omega} \gg 1$ and $\mathcal{F}$ may be approximated by $\mathcal{F}(k) \simeq k/2\pi$. We then find agreement with Eqs. (41) and (42).

[36] See, e.g., H.A. Bethe and R. Jackiw, *Intermediate Quantum Mechanics* (Benjamin Cummings, Reading, MA, 1968), Chap. 12.

[37] D.M. Volkov, Z. Phys. **94**, 250 (1935).

[38] From the generalized Einstein equation Eq. (7) we have $k_n = \sqrt{2[n\omega + E_0(\alpha_0)]}$. Combining this with the high-frequency condition Eq. (16), $\omega \gg |W_0^0(\alpha_0)|$, we find that $k_n \approx \sqrt{2n\omega}$. Therefore the largest wavelength of the ejected electrons equals $\lambda \approx \pi\sqrt{2/\omega}$. If we require this wavelength to be small with respect to the characteristic length of variation of the ground-state wave function $|W_0^0(\alpha_0)|^{-1/2}$, we obtain the condition $\omega \gg |W_0^0(\alpha_0)|$ as before.

[39] I.S. Gradsteyn and I.M. Ryzhik, *Table of Integrals, Series and Products* (Academic, New York, 1980), formula 8.411.1.

[40] N.M. Kroll and K.M. Watson, Phys. Rev. A **8**, 804 (1973).

[41] A study of the angular-dependent rate for multiphoton ionization from the ground state of atomic hydrogen in a circularly polarized laser field has recently been carried out [M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990)]. A detailed study of the case of linear polarization will be presented in the following paper.

[42] These findings are in accordance with general symmetry considerations (see Ref. [24]), if we take into account that the validity of Eq. (53) is restricted to initial states of $m = 0$ in the case of linear polarization and of $r + P$ even in the case of elliptic or circular polarization. If these conditions are not satisfied, $\Phi_\lambda(-\alpha(\phi))$ vanishes for all ϕ and Eq. (53) yields an angular decay rate identically zero (see below). For circular polarization we find from Eq. (52) $\Phi_{\lambda,n} = \Phi_{\lambda,\mp m}\delta_{n,\mp m}$, so that according to Eq. (63) the flux in the polar directions vanishes when $m \pm n$ is nonzero, in agreement with what follows from general symmetry considerations.

[43] To next order in the iteration of the Gavrila-Kaminski scheme (in increasing powers of ω^{-1}), the Floquet-components of the decaying state of Eq. (5) are given by

$$\Psi_{\lambda,n} = \Phi_\lambda\delta_{n,0} + (1 - \delta_{n,0})(W_\lambda + n\omega - H + i\epsilon)^{-1}V_n\Phi_\lambda,$$

with $H = \mathbf{P}^2/2 + V_0$ [see Eq. (14)]. By carrying out the Floquet synthesis defined by Eq. (5) to obtain the decaying state $\Psi_\lambda(\mathbf{r}, t)$, we may write:

$$\begin{aligned} \Psi_\lambda(\mathbf{r}, t) &= \Phi_\lambda(\mathbf{r})\exp(-iW_\lambda t) \\ &\quad + \int d\mathbf{r}' \int dt' \mathcal{G}^{(+)}(\mathbf{r}, \mathbf{r}', t - t') \\ &\quad \times [V(\mathbf{r}' + \alpha(t')) - V_0(\mathbf{r}')] \\ &\quad \times \exp(-iW_\lambda t')\Phi_\lambda(\mathbf{r}'). \end{aligned}$$

Here $\mathcal{G}^{(+)}(\mathbf{r}, \mathbf{r}', t)$ denotes the time-dependent Green's function associated with the Hamiltonian $H = \mathbf{P}^2/2 + V_0$. For atomic hydrogen the atomic binding potential $V(\mathbf{r})$ equals $-1/|\mathbf{r}|$. If one is interested only in the *open-channel* part of Ψ_λ, we may omit the first term in the above equation and we may omit the subtraction of V_0 in the integral. Furthermore, if the frequency is sufficiently high, the (time-dependent) Green's function for the open-channel part of the decaying state (corresponding to electrons ejected out of the atomic system with high speeds) can be replaced by the free-particle Green's function. If we assume that (in the Kramers frame of reference) only the amplitude to find the electron at the momentary position of the harmonically oscillating proton $-\alpha(t)$ is relevant for the ionization process, we may replace $\Phi_\lambda(\mathbf{r})$ in the above equation by $\Phi_\lambda(-\alpha(t))$. If these replacements are carried out, we obtain Eqs. (64) and (65).