Optical Transition and Momentum Transfer in Atomic Wave Packets

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It is shown that the population Rabi-floppings in a lossless two-level atom, interacting with a monochromatic electromagnetic field, in general are convergent in time. The well-known continuous floppings take place because of the restricted choosing of initial conditions, that is when the atom initially is chosen on ground or excited level before the interaction, simultaneously having a definite value of momentum there. The convergence of Rabi-floppings in atomic wave-packet-states is a direct consequence of Doppler effect on optical transition rates (Rabi-frequencies): it gradually leads to "irregular" chaotic-type distributions of momentum in ground and excited energy levels, smearing the amplitudes of Rabi-floppings. Conjointly with Rabi-floppings, the coherent accumulation of momentum on each internal energy level monotonically diminishes too.

I. INTRODUCTION

It is well known that due to interaction with a plane travelling wave the atomic momentum may be changed in limits of one photon momentum \(\hbar k\). This limitation follows from momentum conservation law, and, concerns to total atomic momentum. As to momentum per each internal energy level, as was shown recently [1], its change may be great and even much-more surpass the coupling photon's momentum \(\hbar k\). It is the case, when the atom initially is in superposition state of lower and upper energy levels with some (different in general) momentum distributions there. In other words, optical transition between atomic wave packet-states is accompanied by large-scale coherent accumulation of momentum in internal energy levels (CAMEL).

This phenomenon, which, as we hope, will have far-going consequences for atomic and molecular physics, is presented in [1] in the form as simple as possible. In particular, the operator of kinetic energy of atomic transitional motion was not included into the Hamiltonian of the atom-field system. Nevertheless this operator has not only quantitative, but also qualitative contribution into the picture of interaction. For instance, such an important phenomenon as is the Doppler-shift of frequencies, is introduced into the theory (in laboratory frame) by means of mentioned operator. Therefor, in all cases, the more logical theory of atom-field interaction, concerning the atomic wave packet-states, should contain the atomic kinetic energy operator unquestionably. Just this is done in presented paper, that is here we consider an optical transition in the two-level atom, which has been prepared in general quantum-mechanical transitional states for lower and upper internal energy levels. The behavior of level population and momentum transfer between energy levels is considered in details. It is shown, that the CAMEL-phenomenon, which exhibits regular-periodic behavior in time when the kinetic energy operator is not taken into account, really has a damping-periodic behavior and it is due to influence of the Doppler-shift of frequencies on the rate of optical transitions (Rabi-frequencies). Simultaneously a strictly important result has been obtained for atomic internal levels populations (population amplitudes), according to which the Rabi-floppings of populations for wave-packet atomic states have a damping behavior in general. It is worthwhile to remind that the well-known continuous periodic behavior takes place for "ordinary", fully unexcited or fully excited pure initial states, that is when the time evolution of populations begins from only one populated internal energy level, and, in addition, this populated state has definite value of momentum.

The quantum-mechanical behavior of a two-level atom in the near-resonant, plane-wave monochromatic radiation, taking into account the atomic kinetic energy operator, was considered earlier many times, in most close-staging to our, in papers [2], [3]. In [2] the atom initially is on one energy level and the analysis is limited by narrow momentum distributions and short times of interaction. As a consequence, a splitting of extra-narrow wave-packet into two subpackets has been created due to interaction. In paper [3] the authors restricted the analysis by the definite momentum and one energy level population case. As a consequence, only the continuous periodic behavior, taking into account the energy level splittings due to photon momentum exchange, has been obtained there for energy level’s populations.

Taking into account the results of this paper, the following may be stated about the role of initial conditions. For more general quantum-mechanical initial states, including the atomic transitional states, a) the Rabi-floppings have damping in time character and b) the optical transitions are accompanied by coherent accumulation of momentum on the internal energy levels (CAMEL). Choosing population only on the one internal energy level annihilates the
possibility of CAMEL-phenomenon. If, in addition, the populated state has only a definite value of momentum, then the damping behavior of Rabi-flopings disappears too. Only for simplest initial conditions the behavior of interaction obtains continuous periodic nature with exchange of momentum between internal energy levels restricted by one photon momentum $\hbar k$.

This paper is organized as follows. The Schrodinger equation and the stationary solutions in momentum representation are presented in Sec. 2. The time evolution of level populations and the transfer of momentum and kinetic energy between internal energy levels are examined in Sec. 3 and 4 respectively. The paper is finished by short Conclusions.

II. ROTATING-WAVE APPROXIMATION STATIONARY SOLUTIONS OF SCHRODINGER EQUATION IN MOMENTUM REPRESENTATION.

Let us consider the near-resonant interaction of a two-level atom with a plane-wave radiation field. The Hamiltonian of this system is well known and, taking into account the translational motion of atom, can be presented in the form

$$\hat{H} = \frac{\hat{p}^2}{2M} + \frac{\hbar \omega_0}{2} \hat{\sigma}_3 + \hat{V},$$

where $M$ and $\omega_0$ denote respectively the atomic mass and optical transition frequency, $\hat{\sigma}_3$ is quasispin (Pauli) operator. Second term presents the free atom with $-\frac{\hbar \omega_0}{2}$ and $\frac{\hbar \omega_0}{2}$ energies in lower and upper energy levels. Last term, $\hat{V}$, presents the interaction of atom with external travelling-wave field, and can be written in dipole approximation as

$$\hat{V} = -\hat{d}E(t, z),$$

where $\hat{d}$ is dipole moment operator for optical transition and the intensity of the plane travelling wave we’ll present in the form

$$E(t, z) = E_0 \exp(ikz - i\omega t) + c.c.$$

with $E_0$ is constant, $\omega$ and $k = \omega/c$ represent the wave frequency and the wave number. Polarization effects aren’t included into the field of investigation. Such approach is valid, as is well known, for purely linear or circular polarizations of the wave.

Denoting by $\varphi_g(\vec{\rho}, t)$ and $\varphi_e(\vec{\rho}, t)$ the wave functions of ground ($g$) and excited ($e$) energy levels ($\vec{\rho}$ is atomic internal coordinate, i.e. the radius vector of optical electron relative to atomic center-of-mass), the wave function of interacting atom may be written in the following form

$$\Psi(\vec{\rho}, z, t) = A(z, t) \varphi_g(\vec{\rho}, t) + B(z, t) \varphi_e(\vec{\rho}, t),$$

where $A(z, t)$ represents the atomic probability amplitude to be on lower level and have a space-coordinate $z$ at the time moment $t$; and the other coefficient $B(z, t)$ represents the same for upper level atom. Note, that the coordinate $z$ in (4) represents the atomic center-of-mass position in wave direction; and hence the plane wave (3) includes only this single variable $z$, the case can be considered as a question of one dimension.

In this paper our attention will be focused onto the time evolution of atomic momentum distributions on lower and upper internal energy levels and their physical consequences. So hereafter it is worthwhile to deal with atomic amplitudes in the momentum representation. Moreover, just in momentum representation the eigenvalue problem for the system under consideration has analytic solutions [2], [3].

Expanding $A(z, t)$ and $B(z, t)$ amplitudes into momentum space on the basis of definite momentum states

$$\chi(p) = \frac{1}{\sqrt{2\pi\hbar}} \exp\left(\frac{i}{\hbar}pz\right),$$

that is

$$A(z, t) = \int a(p, t)\chi(p)dp; \quad B(z, t) = \int b(p, t)\chi(p)dp,$$

we substitute all related quantities [1]–[3] into the Schrodinger equation.
After standard transformations we arrive for seeking amplitudes \(a(p, t)\) and \(b(p, t)\) to

\[
i\hbar \frac{d a(p, t)}{dt} = \left( \frac{p^2}{2M} + \frac{\hbar \omega_0}{2} \right) a(p, t) - \frac{\hbar \Omega}{2} e^{-i\omega t} b(p + \hbar k, t),
\]

\[
i\hbar \frac{d b(p, t)}{dt} = \left( \frac{p^2}{2M} - \frac{\hbar \omega_0}{2} \right) b(p, t) - \frac{\hbar \Omega}{2} e^{i\omega t} a(p + \hbar k, t),
\]

where \(\Omega = 2dE_0/\hbar\) is the parameter of induced transitions and commonly referred to as Rabi frequency \([3]\). This system fully coincide in form with the system of equations (8), (9) in \([3]\), and its stationary form should be coincide with the system (5) in \([3]\).

\[
a(p, t) = - \left( \frac{\alpha(p) - \beta(p)}{2\beta(p)} a(p, 0) + \frac{\Omega}{2\beta(p)} b(p + \hbar k, 0) \right) e^{-i\omega_g(p)t} + \\
+ \left( \frac{\alpha(p) + \beta(p)}{2\beta(p)} a(p, 0) + \frac{\Omega}{2\beta(p)} b(p + \hbar k, 0) \right) e^{-i\omega_e(p)t},
\]

\[
b(p + \hbar k, t) = \left( - \frac{\Omega}{2\beta(p)} a(p, 0) + \frac{\alpha(p) + \beta(p)}{2\beta(p)} b(p + \hbar k, 0) \right) e^{-i\omega_g(p)t} + \\
+ \left( \frac{\Omega}{2\beta(p)} a(p, 0) - \frac{\alpha(p) - \beta(p)}{2\beta(p)} b(p + \hbar k, 0) \right) e^{-i\omega_e(p)t}.
\]

Here

\[
\alpha(p) = \frac{\hbar k^2}{2M} + \frac{pk}{M} + \Delta
\]

and may be viewed as a generalized detuning, which involves the field-atom detuning \(\Delta = \omega_0 - \omega\), Doppler- and recoil-shifts \(\frac{pk}{M}\) and \(\frac{\hbar^2 k^2}{2M}\), respectively. It really represents the usual field-atom detuning viewed from atomic center-of-mass frame of reference. The second term

\[
\beta(p) = \sqrt{\left( \frac{\hbar k^2}{2M} + \frac{pk}{M} + \Delta \right)^2 + \Omega^2}
\]

and represents merely the so called generalized Rabi frequency, including the generalized detuning \(\alpha(p)\) instead of common frequency detuning \(\Delta\). Primed and nonprimed frequencies in exponents are

\[
\omega_{g,e}'(p) = \frac{1}{2\hbar} \left( \frac{p^2}{2M} + \frac{(p + \hbar k)^2}{2m} \mp \hbar \omega \right) - \frac{\beta(p)}{2},
\]

\[
\omega_{g,e}(p) = \frac{1}{2\hbar} \left( \frac{p^2}{2M} + \frac{(p + \hbar k)^2}{2m} \mp \hbar \omega \right) + \frac{\beta(p)}{2},
\]

and represent the energies of system quasistationary states, double-splitted, as in familiar theory, in both excited and ground levels. The size of splittery is \(\omega_g - \omega_e = \omega_{e,g} - \omega_{e,g} = \beta(p)\). Note, that plugging in (10), (11) \(b(p, 0) = 0\), \(a(p, 0) = \delta(p - p_0)\), we arrive to the case, analyzed in paper \([3]\).

### III. POPULATION AND MOMENTUM PER INTERNAL GROUND AND EXCITED ENERGY LEVELS

Let’s now proceed to calculation of such physical quantities, as population and mean momentum in each atomic internal energy level, and to their distributions in momentum space. Time evolution of population distributions has been determined and is presented by \([10]\) and \([11]\). Main peculiarity of these formulas is their \(p\)-dependence due to Doppler effect, which will play the key role in further presenting results. It, first of all, disturbs the population distribution in momentum space and gradually transforms the initial smooth distribution into the modulated, chaotic-like one. A behavior of time-evolution is illustrated in Fig. \([4]\). Curve \(a\) represents the initial distribution, curve \(b\)
after 4 Rabi floppings (for central range of distribution) and curve c - after 12 Rabi floppings. For simplicity the excited level was assumed to be initially empty, and the preserving symmetry about \( p = 0 \) value is conditioned by this assumption. In general, when both energy levels are populated, even symmetric with respect to some values of momentum, any symmetry in distribution is being lost rapidly.

Total population of internal energy level is

\[ n_g = \int |a(p, t)|^2 dp \]  

for ground energy level, and is

\[ n_e = \int |b(p, t)|^2 dp \]

for excited energy level. The typical form of time evolution for these populations is presented in Fig. 2. As is seen, the Rabi-floppings (oscillations) are gradually flattened due to redistributions of momentum states for interaction time. So, the momentum-dependence of probabilities for optical transitions (Rabi-floppings), arising due to Doppler-shift of frequencies, leads in general to damping in population oscillations and to establishment of definite-value populations in internal energy levels without any mechanism of relaxation.

The atomic momentum

\[ \langle p \rangle = \int \Psi^* \hat{p} \Psi d\vec{r} dz, \]  

after elementary substitution of general expression (4) and respective standard transformations can be expressed as a sum of two terms [1],

\[ \langle p \rangle = \langle p \rangle_g + \langle p \rangle_e \]  

first of which represents the amount of contribution of ground level states into the atomic momentum and is presented in general as

\[ \langle p \rangle_g = \int |a(p, t)|^2 p dp, \]

the second term has the same sense for excited level and is

\[ \langle p \rangle_e = \int |b(p, t)|^2 p dp \]

It should be mentioned that these quantities, besides being the ingredients of total atomic momentum, in accordance with first principles of quantum mechanics, have own physical meaning and are measurable quantities [1].

Typical form of time-evolutions, obtained by means of numerical calculations, is illustrated in Fig. 3 and 4, respectively for ground and excited levels. Solid curves represent the case, when only one energy level (ground), is initially populated, and dashed curves - when both levels are populated. Herewith, the same forms of momentum distributions are chosen in the last case and these distributions are shifted with each other by 

\[ p_0 \gg \hbar k. \]  

In both cases, particular A, and more general B, the oscillatory behavior is being depressed.

To carry out the behavior of CAMEL-phenomenon from the behavior of mean momentums \( \langle p \rangle_g \) and \( \langle p \rangle_e \), it is necessary to pick out from these momentums the parts, conditioned by population evolution. To this end a pair of new momentums can be introduced into the theory [1],

\[ p_g = \langle p \rangle_g / n_g \quad \text{and} \quad p_e = \langle p \rangle_e / n_e \]

time evolution of which should be solely conditioned by redistributions in wave-packet momentum states, referred to as coherent accumulation of momentum on internal energy level [1]; for familiar optical transitions with continuous periodic Rabi-floppings \( p_g \) and \( p_e \) are constant in the course of time and emerge as mean values of normalized momentum distribution per each energy level. It is readily also verified that for familiar optical transitions, beginning from fully populated energy levels, these momentums can be varied in limits of one photon momentum \( \hbar k \). In the contrary, in general case of initially populated states, there are essential redistributions in each energy level momentum states (see, for instance, Figs. 3 and 4) and \( p_g \) and \( p_e \) characteristic momentums are being changed in essentially exceeding the photon momentum limits. The graphs of normalized momentums \( p_g \) and \( p_e \), corresponding to the
IV. TRANSLATIONAL ENERGY PER INTERNAL GROUND AND EXCITED ENERGY LEVELS.

An analogous set of calculations we have made also for the kinetic energy of atom. It can be readily verified by standard calculations, that the (19)-type splitting is true also for kinetic energy:

\[
\langle E_{\text{kin}} \rangle = \langle E_{\text{kin}} \rangle_g + \langle E_{\text{kin}} \rangle_e,
\]

(23)

with

\[
\langle E_{\text{kin}} \rangle_g = \int |a(p, t)|^2 \frac{p^2}{2M} dp,
\]

(24)

\[
\langle E_{\text{kin}} \rangle_e = \int |b(p, t)|^2 \frac{p^2}{2M} dp,
\]

(25)

presenting the part contribution of ground and excited internal energy levels into the atomic kinetic energy. Time-behavior of (24) and (23) for one-level initial population \((a(p, 0) \neq 0, b(p, 0) = 0)\) case exhibits, of course, continuously flopping behavior for familiar one-state population case (Fig. 8a) and oscillatory saturating behavior for one-level wave-packet case (Fig. 9a). The general, two-level population case with different wave-packet distributions, is presented in Fig. 10a. For comparison we present there also the total kinetic energy of atom (solid lines in Figs. 9-10). As seen from Figs. 9 and 10, total energy has some fluctuations after instant switching on of the interaction, which gradually disappears. That is because of Heisenberg principle of indeterminacy.

The behavior of normalized quantities,

\[
E_{\text{kin,g}} = \frac{\langle E_{\text{kin}} \rangle_g}{n_g},
\]

(26)

\[
E_{\text{kin,e}} = \frac{\langle E_{\text{kin}} \rangle_e}{n_e},
\]

(27)

are presented in Figs. 8b-10b. The role of initial state preparation is obvious for kinetic energy too and is totally similar to the case of momentum behavior.

Nevertheless, there is a very important difference between the behaviors of momentum and energy, if we discriminate internal and external degrees of atom for the interaction. The coupling photonic momentum can only transfer (or is obtained from) external translational degrees of atom, while the photonic energy deals with both internal (and it is the main part) and external degrees of freedom. As a consequence, a logically correct scale, as is the photon momentum for atomic translational motion momentum, absent for atomic translational motion energy. That is why the statement of the question about CAMEL-type phenomenon in the translational energy space quantitatively looks to be problematic, at least hitherto.

V. CONCLUSIONS

The detailed analysis of a two-level atom in the field of near-resonant monochromatic radiation shows, that in more general quantum-mechanical states with some distributions in momentum space (wave-packet states) the Rabi-floppings between internal energy levels monotonically decrease for long times of interaction. The reason of decreasing is momentum dependence of optical transition probabilities, or, in more usual in laser physics terminology, due to dependence of Rabi-floppings on the Doppler-shift of frequencies.
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FIG. 1. Population distribution in momentum space for ground state.

FIG. 2. Internal levels populations for one-level wave-packet initial case. Hereafter X-axes is measured in $\tau = \frac{\hbar^2}{M}$ units.

FIG. 3. Mean momentas of internal ground energy level. $A$ - for familiar one-state wave-packet case, and $B$ - for general superpositional case.

FIG. 4. Mean momentas of internal excited energy level. $A$ - for familiar one-state wave-packet case, and $B$ - for general superpositional case.

FIG. 5. Normalized momentums $p_g$ and $p_e$ for familiar one-state case with definite momentum.

FIG. 6. Normalized momentums $p_g$ and $p_e$ for one-state wave-packet case.

FIG. 7. Normalized momentums $p_g$ and $p_e$ for general superpositional case.

FIG. 8. a) Internal levels energies for familiar one-level state with definite momentum. b) Internal levels normalized energies for one-level state with definite momentum.

FIG. 9. a) Total energy, and internal levels energies for one-state wave-packet initial case. b) Total energy, and internal levels normalized energies for one-state wave-packet initial case.

FIG. 10. a) Total energy and internal levels energies for general superpositional case. b) Total energy and internal levels normalized energies for general superpositional case.
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