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Radiative collisions between atoms in a bichromatic field

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We investigate the influence of nonmonochromaticity of radiation on transitions between atomic terms in radiative collisions between atoms. As the simplest analytically solvable example of a nonmonochromatic field we consider the bichromatic field. An analysis is carried out of Landau-Zener-type transitions in such a field at different field amplitudes and at various differences between the harmonic frequencies. New combination Landau-Zener transitions are described. The results are generalized to include the case of arbitrary nonmonochromaticity of the field.

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1. INTRODUCTION

Radiative collisions, i.e., collisions between atoms and molecules, which occur in the presence of an optical field, have recently attracted great interest. In the course of radiative collisions it is possible to have inelastic collisions that are adiabatically forbidden in the absence of an optical field by virtue of the slowness of the collisions. A review of the theory of radiative collisions is contained in a paper by Yakovlenko.[1] The theory of radiative collisions in the presence of a monochromatic perturbing field has been well-developed. At a certain instant of time, the difference between two energy terms of a quasimolecule made up of two colliding atoms becomes equal to the energy of the optical photon and crossing of the terms takes place in the "quasimolecule + field" system. As a result of this crossing, a nonadiabatic transition becomes possible. The mathematical description of the process is similar to the theory of Landau-Zener term crossing (see Ref. 2, Sec. 90).

The task undertaken in the present paper is to investigate the influence of nonmonochromaticity of the radiation on transition between terms in radiative collisions. By way of the simplest example of nonmonochromaticity we consider a bichromatic field, which is a superposition of two harmonic waves with close frequencies ω_1 and ω_2. The matrix element of the bichromatic field between the lower (a) and upper (b) levels of the quasi-molecule then takes the form

$$V_{ab}(t)=2V_{ab}^{(1)}\cos\omega_1 t+2V_{ab}^{(2)}\cos(\omega_2 t+\alpha). \quad (1)$$

The quantity α is the phase difference at the instant of time $t=0$.

We denote the time dependent energies of the terms of the quasimolecule by $E_a(t)$ and $E_b(t)$. Following the usual approach, we expand the term difference near the point of crossing in a series, and confine ourselves to the linear term

$$\omega_{ba}(t)=E_b(t)-E_a(t)=\omega_1+(F_b-F_a)t.$$

For the sake of argument, we reckoned the time from the crossing point due to the first field:

$$E_b(0)-E_a(0)=\omega_1.$$

The derivatives F_a and F_b are small quantities, since they are proportional to the velocities of the colliding atoms u, which are small compared with the atomic velocities. Namely, it is assumed that

$$F_a,F_b\sim\frac{u}{u_a}\frac{\omega_1}{\tau_a}\ll\frac{\omega_1}{\tau_a},$$

where u_a and τ_a are the characteristic atomic velocities and times.

We use henceforth the dimensionless time $\varphi=(F_a-F_b)^{1/2}t$ and the dimensionless frequency difference

$$\Delta\omega=(\omega_1-\omega_2)(F_a-F_b)^{-1/2}.$$

The dimensionless amplitude of the field

$$v_{1,2}=V_{ab}^{(1,2)}(F_a-F_b)^{-1/2}$$

then coincides with the known Landau-Zener parameter.

In Sec. 2 we consider the case of a weak field, and in Sec. 3 the case when the difference between the frequencies of the bichromatic field is large enough, and

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the field itself can be strong. Finally, in Sec. 4 we investigate the case of very strong fields at not too large differences between the frequencies of the bichromatic perturbation. The conclusion is devoted to a generalization to the case of a realistic multimode laser radiation.

2. WEAK FIELD

In this section we shall assume that the conditions $v_{1,2} \ll 1$ are satisfied. Then, just as for a monochromatic field, the probability of the transition between the terms can be obtained within the framework of perturbation theory.[1] The amplitude of the transition in first-order perturbation theory is given by

$$A_{ab}^{(1)} = -i \int_{-\infty}^{\infty} V_{ab}(t) \exp\left[i \int_{0}^{t} \omega_{ba}(t')\,dt' \right] dt. \quad (2)$$

The time dependence of the wave functions in the integrand is given in the adiabatic approximation. Therefore direct integration with respect to time yields the correct result only if there are no fast oscillations in the region that is important for the integration (see Ref. 2, Sec. 131). The perturbation-theory series constitutes in this case simply an expansion in powers of $v_{1,2}$. This is precisely the situation in the Landau-Zener transition in a weak field, where the essential time interval is of the order $(F_a - F_b)^{1/2}$.

In the resonance approximation, leaving one exponential each from the cosines in the perturbation (1) and taking the expansion (2) into account, we obtain the transition probability

$$w_{ab} = |A_{ab}^{(1)}|^2 = 2\pi [v_1^2 + v_2^2 + 2 v_1 v_2 \cos(\alpha - {}^1/_2 \Delta\omega^2)]. \quad (3)$$

We see therefore that the magnitude of the interference term depends both on the initial phase difference α, and on the change of the phase as a result of the difference between the frequencies of the harmonics.

If the frequency difference is large, i.e., $\Delta\omega \gg 1$, then we must average the probability over small fluctuations of this quantity. The reason is that in a realistic case the two laser waves are not strictly monochromatic, but have a finite spectral width $\delta\omega$. The statement made above concerning the averaging is valid under the condition

$$\frac{F_a - F_b}{|\omega_1 - \omega_2|} \ll \delta\omega \ll |\omega_1 - \omega_2|$$

of these two inequalities, the right-hand one ensures the existence of two separated monochromatic wave, and the left-hand one ensures a large increment, compared with unity, to the argument of the cosine in expression (2), owing to the contribution $\delta\omega$, and it is this which brings about the need for averaging. As a result, the interference term vanishes, and the probability of the transition turns out to be equal to the sum of the probabilities of the Landau-Zener type from each of the harmonics:

$$\langle w_{ab} \rangle = 2\pi (v_1^2 + v_2^2).$$

We note that because of the indicated effective averaging there is no interference between two such transition points in the pre-dissociation phenomenon (Ref. 2, Sec. 90).

3. FIELD WITH HARMONICS THAT DIFFER GREATLY IN FREQUENCY

In this section we consider the case when the crossing points of the terms can be regarded as isolated. To this end it is necessary that the time interval between the crossing points $\Delta\varphi \approx \Delta\omega$ be substantially longer than the time of transition in the vicinity of the crossing points:

$$\Delta\omega \gg \max(1, v_{1,2}).$$

The amplitudes can in this case be of arbitrary magnitude: $v_{1,2} \gtrsim 1$.

Inasmuch as at $\Delta\omega \gg 1$ there is effective averaging over the phase difference of the two fields and there is no interference term, we can multiply the probabilities calculated by the Landau-Zener theory for each of the transition points. This statement was justified in Sec. 2 within the framework of perturbation theory. We shall show that it is valid also outside this framework. Assume that far to the left of the first term-crossing point t_1 the particle is on the lower level a. Then its wave function is

$$\Psi(t \ll t_1) = \psi_a \exp(-iE_a t).$$

After passing through the point t_1, since the time of buildup of the probability of population of the upper level b is small compared with the time $t_2 - t_1$ to the next crossing point, we have for the wave function in the interval between the two crossing points the expression

$$\Psi(t_1 \ll t \ll t_2) = \alpha_1^{1/2} \psi_a \exp(-iE_a t + i\varphi_a) + (1-\alpha_1)^{1/2} \psi_b \exp(-iE_b t + i\varphi_b).$$

Here $\alpha_1 = \exp(-2\pi v_1^2)$ is the probability of staying on the lower level a in accord with the Landau-Zener theory, while φ_a and φ_b are the phase shifts accumulated after the passage through the first crossing point t_1.

After the passage through the second term crossing point t_2, we have for the wave function the following expression:

$$\begin{aligned}\Psi(t \gg t_2) = {} & (\alpha_1\alpha_2)^{1/2} \psi_a \exp(-iE_a t + i\varphi_a') \\ & + (\alpha_1(1-\alpha_2))^{1/2} \psi_b \exp(-iE_b t + i\varphi_b') \\ & + ((1-\alpha_1)\alpha_2)^{1/2} \psi_b \exp(-iE_b t + i\varphi_b'') \\ & + ((1-\alpha_1)(1-\alpha_2))^{1/2} \psi_a \exp(-iE_a t + i\varphi_a'').\end{aligned}$$

Here $\alpha_2 = \exp(-2\pi v_2^2$, and $\varphi'_{a,b}$ and $\varphi''_{a,b}$ are the phase shifts accumulated after the passage through the crossing point t_2. The transition probabilities are given by

$$w_{ab} = |(\alpha_1(1-\alpha_2))^{1/2} + (\alpha_2(1-\alpha_1))^{1/2} \exp[i(\varphi_b'' - \varphi_b')]|^2.$$

The interference term vanishes after averaging over the phase difference, in analogy with the situation in Sec. 2 for the perturbation-theory case. We thus obtain

$$\langle w_{ab} \rangle = \alpha_1(1-\alpha_2) + \alpha_2(1-\alpha_1). \quad (4)$$

For a weak field we obtain from this the result obtained at the end of Sec. 2, as was to be expected.

A specifically new effect for the bichromatic field is the appearance of combination frequencies of the type $n\omega_1-(n-1)\omega_2$. If the distance between the quasimolecular terms is close to any one of these frequencies, then transitions of the Landau-Zener type also occur in the vicinity of these points. The corresponding transition points are shown in Fig. 1. The distance between neighboring transition points is $\Delta\omega$, as is also the distance between the basic points, where the term difference is equal to ω_1 and ω_2. Therefore the very idea that these points are isolated is valid under the condition $\Delta\omega \gg \max(1, v_{1,2})$ written out above.

To consider the produced transitions, which we can call combination transitions, the standard system of two equations that describe the dynamics of a two-level system in a monochromatic external field in the vicinity of the Landau-Zener point[1] must first be subjected to the following modification: it is necessary to iterate each of the equations up to terms of third order perturbation theory with respect to $v_{1,2}$ and retain only those terms which contain the combination frequencies $2\omega_1-\omega_2$ or $2\omega_2-\omega_1$. The system of equation then assumes the traditional form in the Landau-Zener approximation, except that the matrix element $v_{1,2}$ is replaced by a three-photon combination matrix element. A similar procedure is carried out for the investigation of multiphoton bound-bound transitions in atoms in the case of term energies that are independent of the time (Ref. 3, Sec. 3.2; Ref. 4).

The matrix element for the transition at the frequency $2\omega_2-\omega_1$ is of the form

$$v_1^{(3)}=v_1v_2^2/\Delta\omega^2.$$

The corresponding Feynman diagram (Ref. 3, Sec. 2.3) is shown in Fig. 2. Similarly, in the case of the transition of the frequency $2\omega_1-\omega_2$ we have

$$v_2^{(3)}=v_1^2v_2/\Delta\omega^2.$$

Under the condition $\Delta\omega\gg v_{1,2}$ indicated above, these multiphoton matrix elements are small compared with the single-proton matrix elements $v_{1,2}$. This statement agrees with the assumption that the Landau-Zener transition points are isolated. If $v_{1,2}\lesssim 1$, then the combination transitions have a weak effect on the resultant transition probability inasmuch as we obtain in this case $v_{1,2}^{(3)}\ll 1$. They become important, however, in the case of strong fields $v_{1,2}\gg 1$. The passage through the points ω_1 and ω_2 (see Fig. 1) returns the system in this case to the initial state, so that the resultant transition probability, after passing through the four Landau-Zener transition points $2\omega_1-\omega_2$, ω_1,

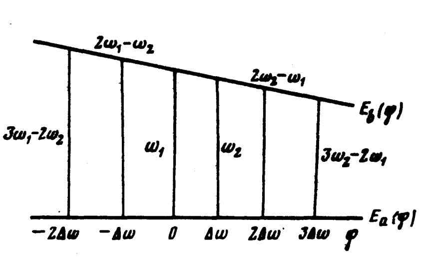

FIG. 1. Term crossing in bichromatic field and combination points of Landau-Zener transition.

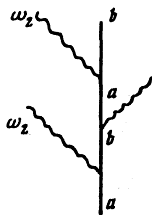

FIG. 2. The Feynman diagram for three-photon combination transition at the frequency $2\omega_2-\omega_1$.

ω_2 and $2\omega_2-\omega_1$ takes the following form

$$\langle w_{ab}\rangle=\alpha_1^{(3)}(1-\alpha_2^{(3)})+\alpha_2^{(3)}(1-\alpha_1^{(3)}). \tag{5}$$

They have introduced here the notation

$$\alpha_{1,2}^{(3)}=\exp[-2\pi|v_{1,2}^{(3)}|^2]$$

and used the same averaging over the phase difference as in the derivation of Eq. (4).

At $v_{1,2}\gg 1$ and $v_{1,2}^{(3)}\ll 1$, as seen from (4) and (5), the transition probability is low. If we remain within the framework of Eq. (4), then the probability obtained is exponentially small. The probability of combination transitions calculated by formula (5) may turn out to be much higher. In this case the probability is

$$\langle w_{ab}\rangle=2\pi(v_1v_2/\Delta\omega^2)^2(v_1^2+v_2^2)$$

and has only a power-law smallness. However, if we take $\Delta\omega$ large enough such that $\langle w_{ab}\rangle$ turns out to be very small, then we can make the combination probability small compared with the exponentially small probability obtained from formula (4) on the fundamental harmonics.

All the foregoing is valid when then following five-photon combination transitions generated by the matrix elements

$$v_{1,2}^{(5)}\sim v_1^2v_2^2v_{1,2}/\Delta\omega^4$$

at the frequencies $3\omega_1-2\omega_2$ and $3\omega_2-2\omega_1$ are small. To describe them we can repeat everything stated above concerning three-photon combination transitions. This description is necessary if they become comparable with unity.

4. FIELD WITH HARMONICS THAT DIFFER LITTLE IN FREQUENCY

We assume in this section that the condition $\Delta\omega \ll v_{1,2}$ is satisfied. The time of the Landau-Zener transition then greatly exceeds the distance $\Delta\omega$ between the transition points, and these points cannot be regarded as isolated. For the same reason, we cannot speak of combination transitions, since the respective points come close to the point corresponding to the fundamental tones. At first glance it might seem that in the case considered here we can speak of one field with amplitude

$$v_1+v_2e^{i\alpha}.$$

Actually, the inequality $\Delta\omega\ll v_{1,2}$ indicated above turns out to be insufficient. The amplitude of the bichromatic field

$$v_1+v_2\exp[i(\alpha+\varphi\Delta\omega)]$$

remains constant also during the transition under the condition $\Delta\omega\Delta\varphi_{tr} \ll 1$. In a weak field $\Delta\varphi_{tr} \sim 1$ and the transition to the limit of one field is justified, since $\Delta\omega \ll 1$. On the contrary, in a strong field we have $\Delta\varphi_{tr} \sim v_{1\ 2}$, and the two fields can be replaced by a single one when the more stringent condition $\Delta\omega \ll v_{1,2}^{-1}$ is satisfied.

When a transition to the case of a single field is possible, the probability is described by the Landau-Zener formula

$$\langle w_{ab}\rangle = 1-\exp[-2\pi(v_1^2+v_2^2+2v_1v_2\cos\alpha)]. \qquad (6)$$

This formula is valid under the condition

$$\Delta\omega \ll \min(v_{1,2}, v_{1,2}^{-1}).$$

At $v_{1,2} \ll 1$ this yields in natural fashion the perturbation-theory result (3) with $\Delta\omega \ll 1$.

If the phase difference α of the two fields has a random character, then expression (6) can be averaged over α and we get

$$\langle w_{ab}\rangle = 1-\exp[-2\pi(v_1^2+v_2^2)]I_0(4\pi v_1 v_2). \qquad (7)$$

Here I_0 is a modified Bessel function. In particular, in strong fields, when $v_{1,2} \gg 1$, we get from (7)

$$\langle w_{ab}\rangle = 1-(8\pi^2 v_1 v_2)^{-1}\exp[-2\pi(v_1-v_2)^2].$$

We see that in the case of a very strong field at $v_1 = v_2$, for a bichromatic field with close frequencies but with randomly distributed phases, the probability that the particle will remain on the initial level can be small not exponentially in $v_{1,2}$ as for the case of the monochromatic field, but in power-law fashion. However, at $v_1 \neq v_2$ the exponential smallness is preserved.

We proceed now to the case

$$v_{1,2}^{-1} \leq \Delta\omega \ll v_{1,2},$$

when the problem cannot be reduced to the case of a single field. In this case, obviously, it is necessary to regard the field as strong, i.e., $v_{1,2} \gg 1$. To consider this much more complicated situation we confine ourselves to the particular case of identical perturbation amplitudes in both harmonics:

$$v_1 = v_2 = v.$$

This system of equations that describe the amplitudes of the populations of the levels $b(\varphi)$ and $b(\varphi)$ takes the following dimensionless form

$$i\frac{da}{d\varphi} = 2vb\cos\frac{\varphi\Delta\omega-\alpha}{2}, \quad i\frac{db}{d\varphi} = 2va\cos\frac{\varphi\Delta\omega-\alpha}{2} + \varphi b. \qquad (8)$$

In the case when

$$\cos\left(\frac{\varphi\Delta\omega-\alpha}{2}\right) \to 1,$$

we have the usual Landau-Zener system of equations[1] for a monochromatic field.

In the general case the system (8) has no analytic solution. We resort to a quasiclassical solution of this system. It is valid under the condition $d\lambda/d\varphi \ll 1$, where

$$\lambda = \left[v\cos\frac{\varphi\Delta\omega-\alpha}{2}\right]^{-1}$$

is the characteristic wavelength of the system. This means that the conditions $v \gg \Delta\omega$ and $v \gg 1$, which were already postulated before, must be satisfied.

When these conditions are satisfied, the probability that the particle will remain on the level a takes, according to adiabatic perturbation theory, the form (Ref. 2, Sec. 53)

$$w_{aa} = \exp\left\{-32v^2\int_0^{x^*}\left[x^2+\cos^2\left(x\xi-\frac{\alpha}{2}\right)\right]^{1/2}dx\right\}. \qquad (9)$$

Here $\xi = 2v\Delta\omega$, and x^* is a point in the complex plane (complex turning point), where

$$x^2+\cos^2(x\xi-\alpha/2) = 0.$$

In the case $\xi \ll 1$, i.e., $\Delta\omega v^{-1}$, we obtain from (9) the possibility of the substitution

$$\cos(x\xi-\alpha/2) \to \cos(\alpha/2),$$

so that $x^* = i\cos(\alpha/2)$. We then obtain

$$w_{aa} = \exp(-8\pi v^2\cos^2(\alpha/2)),$$

as already seen in (6) above at $v_1 = v_2$.

In the case of arbitrary $v\Delta\omega \gtrless 1$, we estimate the integral in (9) by starting from the condition $v \gg 1$. It is easy to see that under this condition w_{aa} is maximal when the phase difference α is close to π. Outside this region, the probability w_{aa} decreases rapidly, so that we confine ourselves only to the vicinity of the values of α near π. Calculating the integral in (9), we obtain

$$w_{aa} = \exp\{-2\pi v^2(\pi-\alpha)^2/(1+\xi^2)^{1/2}\}. \qquad (10)$$

At $\xi \ll 1$ we obtain from (10) the already known result,

$$w_{aa} = \exp[-2\pi v^2(\pi-\alpha)^2].$$

If $\xi \gg 1$, then we get from (10)

$$w_{aa} = \exp[-\pi(\pi-\alpha)^2/4v(\Delta\omega)^2].$$

The last formula is valid if $v^{-1} \ll \Delta\omega \ll v$.

5. CONCLUSION

We examine in conclusion how the results of the present paper are modified if we go over from the simplest nonmonochromaticity in the form of two harmonics of a field to a realistic nonmonochromatic field containing a large number of close harmonics. The situation is simplest if perturbation theory is applicable (see Sec. 2). By generalizing formula (3) we find that the transition probability is proportional to the sum of the squares of the individual harmonics of the amplitude of the nonmonochromatic perturbation, expanded in a Fourier series, to which quadratic interference terms are added. The latter vanish in the case of a sufficiently large so-called correlation interval of radiation,[5] which constitutes the difference between the frequencies of neighboring field harmonics (the analog of the condition $\Delta\omega \gg 1$), or when the mode phases are not synchronized.

The results of Sec. 3 remain on the whole in force also for an arbitrary nonmonochromatic field, inasmuch as in a real laser the distances between the frequencies of the neighboring modes are the same. Consequently, no new combination transitions, besides those indicated in Fig. 1, arise. Of course, changes take place in the values of the probabilities of the transitions because each value of the combination frequency, including also that of the fundamental tones, is realized by a large number of pairs of radiation harmonics. We can state as a result that the time of the Landau-Zener transition increases to the effective width of the nonmonochromatic radiation

$$t_{tr}^{nonmon} \sim \left|\frac{\omega_{min}-\omega_{max}}{F_a-F_b}\right|,$$

if this width exceeds the time of the Landau-Zener transition for one harmonic.

The laser-radiation spectrum, however, contains besides the superposition of the modes also a nonmonochromaticity due to the finite duration of the pulse and the stochastic phase randomization, whose influence on the character of the Landau-Zener transition calls for a separate analysis and may alter the foregoing conclusion concerning the transition time.

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Permutation symmetry of wave functions of a system of identical particles

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It is shown that if the concept of identity of particles of a system is correctly defined, the known limitation on the permutation symmetry of the wave functions of physically possible states of the system turns out to be an automatic consequence of this definition.

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1. We consider a system of N particles and assume that the quantity q_i is the complete set of dynamic variables that describe its i-th particle. Then the Hamiltonian of this system and the wave functions $|\Psi\rangle$ of its physically possible states will depend on the quantities q_i. If the particles of the system are indistinguishable, then its Hamiltonian turns out to be a symmetrical function of the quantities q_i. Analytically, this property of H is expressed by the relation

$$\hat{P}^{-1}H\hat{P}=H\rightarrow H\hat{P}=\hat{P}H, \tag{1}$$

where $\hat{P}$ is any operator of the permutation group of the N indices of the quantities q_i. We must immediately emphasize that the explicit form of H, and in particular its property (1) is assumed to be known beforehand, since the equations of quantum mechanics become meaningful only under this condition.

Each level of the Schrödinger equation

$$H|n\rangle=E_n|n\rangle \tag{2}$$

of a system of identical particles (SIP) corresponds as a rule to a function $|n\rangle$ which has one fully defined type of permutation symmetry (PS), i.e., to each value of E_n there corresponds only one Young pattern. However, Young patterns of different levels may not coincide: Eq. (2) obviously admits of solutions $|n\rangle$ with different types of PS. On the other hand, it is known that in nature there have been encountered so far only those $|n\rangle$ for SIP, which have a maximal PS—either fully symmetrical, or fully antisymmetrical. It can be assumed that this strong limitation on the possible type of the PS of the solutions $|n\rangle$ of the Schrödinger equation for SIP is connected with the nature of its particles—an SIP of definite nature admits also solutions only of the corresponding type PS, consequently, so far only two types of particles have been observed—bosons and fermions. This raises the question of the possibility in principle of existence in nature of particles (of course, with integer or half-integer spin in units of h), aggregates of which would be described by functions with an intermediate type of PS—parabosons and parafermions.[1,2]

If the wave functions $|\Psi\rangle$ of the physically possible states of SIP admit of more than two types of PS, then there is apparently no unambiguous connection between the spin of the particles and the PS of these functions. It will be shown below, however, that the PS of the functions $|\Psi\rangle$ is an intrinsic property of the SIP as a

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