Coherent excitation of equidistant multilevel systems in a resonant monochromatic field

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The time evolution of certain equidistant multilevel systems in a resonant monochromatic field is investigated. For a system having a constant dipole moment of the successive transitions it is shown that unlimited excitation takes place and the mean value increases in proportion to the time. The threshold characteristics of the passage of the system through a given level are investigated. For a system with a transition dipole moment that decreases like $(n+1)^{-1/2}$ it is found that the quasienergy spectrum is discrete and escape to infinity does not occur. Interesting features are also observed in the excitation of systems in which the dipole moment of the lowest transition differs from the dipole moment of the subsequent transitions. Analytic solutions are obtained for all of the cases under consideration.

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1. INTRODUCTION

The phenomenon of the collisionless dissociation of polyatomic molecules in the intense field of radiation from a pulsed CO_2 laser was discovered in 1973.[1,2] The subsequent discovery of isotopically selective dissociation[3,4] revealed the practical importance of the effect. These experiments stimulated theoretical investigations[5–12] of the interaction of polyatomic molecules with a quasiresonant field.

One of the fundamental questions in the theory of this phenomenon is, by what manner can a polyatomic molecule in principle be excited to the dissociation limit? At the present time the point of view which has already been expressed in Refs. 13 and 1 is generally accepted. It is noted that the excited vibrational states of polyatomic molecules have a high density (see, for example, Ref. 14) which is due to the superposition of levels belonging to different vibrational modes, the presence of composite vibrations, and also a splitting of degenerate levels due to the Coriolis interaction. Therefore, starting at a certain energy which is different for different molecules, resonant cascade transitions from a given vibrational-rotational level right up to the dissociation limit are possible within the limits of the width of the laser spectrum. Apparently the dipole moments of such transitions should be[5,7,9,10] much smaller than typical dipole moments for allowed transitions. But the complete forbiddenness, which exists in the harmonic approximation, may in principle be removed due to the interaction between the different vibrational modes.

It is natural to assume that the quasiresonant sequences of transitions make the largest contribution to the excitation of the quasicontinuum of high vibrational levels of polyatomic molecules. Therefore, an investigation of the interaction of various multilevel systems with a quasiresonant field is of practical interest.

In the present article exact results are obtained concerning the coherent excitation of certain equidistant multilevel systems consisting of nondegenerate levels, the field being exactly resonant for successive transitions between these levels. The systems under consideration differ with regard to the dependence of the dipole moment of the transitions on the quantum number of the transition. The form of this dependence for transitions between highly excited vibrational-rotational levels of a polyatomic molecule is not clear beforehand.

Although the approach developed here is simple, it reveals a number of interesting qualitative effects and quantitative relationships which are also of independent theoretical value.

The formulation of the problem and certain general properties of the solutions are formulated in Sec. 2. The dependence of the transition dipole moment $\mu_{n-1,n}$ on the transition quantum number n is concretely spelled out in the following sections. One of these dependences—$\mu_{n-1,n}=\mu\sqrt{n}$, that is, the harmonic oscillator—is well investigated.

The exact solution is well known for the harmonic oscillator in an arbitrary variable field.[15] The probabilities for occupation of the levels of an oscillator that is in the ground state at $t=0$, are given by a Poisson distribution:

$$W_n=\frac{\bar{n}^n}{n!}\exp(-\bar{n}). \quad (1.1)$$

In the special case of a field of the form $\mathscr{E}(t)\cos\Omega t$, where the field frequency Ω coincides with the oscillator frequency, the mean value increases without limit with time according to the law

$$\bar{n}\approx\left[\frac{\mu}{2\hbar}\int_0^t\mathscr{E}(\tau)d\tau\right]^2. \quad (1.2)$$

We note that the oscillator's escape to infinity, which has a clear physical meaning, is also understandable within the framework of the quasienergetic approach.[15,16] If the field is at exactly resonance with the oscillator, the quasienergy spectrum is continuous and an analogy exists with the escape to infinity of stationary quantum systems having a continuous spectrum.[17]

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The excitation of an infinite system having a constant dipole moment ($\mu_{n-1,n} = \mu$) for all successive transitions is considered in Sec. 3. Just as for the harmonic oscillator, in this case the quasienergy spectrum is continuous and escape of the system to infinity takes place. But in contrast to the harmonic oscillator, the mean value increases according to the law

$$\bar{n} \propto \frac{\mu}{\hbar} \int_0^t \mathscr{E}(\tau) d\tau. \tag{1.3}$$

The case when the dipole moment decreases like $\mu_{n-1,n} = \mu/(n+1)^{1/2}$ is investigated in Sec. 4. This case has a qualitative difference from the preceding cases. Here the quasienergy spectrum is discrete and there is no escape to infinity. Thus, to some extent the reduction of the dipole moment is analogous to an increasing potential for stationary quantum systems.

Systems in which the dipole moments for certain transitions differ markedly from the characteristic dipole moment of the remaining transitions are also of interest. The case of an infinite system for which $\mu_{n-1,n} = \mu = \text{const}$ for $n > 1$ and $\mu_{01} \neq \mu$ is investigated in Sec. 5. The most interesting effect, arising here for $\mu_{01} \gg \mu$, is that if the system is in the ground state at the moment of time $t = 0$, it subsequently remains in the two lowest states, with a probability close to unity.

A system with $\mu_{01} \gg \mu$ is a particular case of a more general class of multilevel systems consisting of "fast" and "slow" subsystems, whose investigation is important in regard to understanding the mechanism for the excitation of polyatomic molecules.[5,9,10] Certain qualitative aspects of the kinetics of the excitation of such systems are discussed in Sec. 6.

2. FORMULATION OF THE PROBLEM. SOME GENERAL PROPERTIES OF THE SOLUTIONS

Let us assume that the wave function of a system situated in a field $\mathscr{E}(t) \cos\Omega t$ is at any instant of time a superposition of stationary wave functions of the equidistant levels, the field being exactly resonant for successive transitions between these levels. The standard procedure[17] for the substitution of a general expression for the wave function into the time-dependent Schrödinger equation and averaging over the high-frequency terms leads to the following system of equations for the amplitudes $a_n(t)$:

$$\begin{gathered} da_0/dt = i\gamma_{01}a_1, \\ da_n/dt = i(\gamma_{n-1,n}a_{n-1} + \gamma_{n,n+1}a_{n+1}), \quad n \geq 1. \end{gathered} \tag{2.1}$$

Here $\gamma_{n-1,n} = \mu_{n-1,n}\mathscr{E}/2\hbar$; $\hat{\mu}$ denotes the dipole moment operator; for the sake of definiteness it is assumed that $\gamma_{n-1,n} = \gamma_{n,n-1} > 0$. In addition, we shall assume later for simplicity that the amplitude of the field is constant. The corresponding changes in the solutions can be obtained by the following substitution:

$$\mathscr{E}t \to \int_0^t \mathscr{E}(\tau) d\tau. \tag{2.2}$$

The general solution of the system (2.1) can obviously be written in the form

$$a_n(t) = \sum u(\lambda) p_n(\lambda) \exp(i\lambda t), \tag{2.3}$$

where λ and $p_n(\lambda)$ are the eigenvalues and eigenvectors of the following system of equations:

$$\begin{gathered} \lambda p_0 = \gamma_{01}p_1, \\ \lambda p_n = \gamma_{n-1,n}p_{n-1} + \gamma_{n,n+1}p_{n+1}, \quad n \geq 1. \end{gathered} \tag{2.4}$$

The summation in formula (2.3) runs over all eigenvalues, and in the case of a continuous spectrum the summation should be replaced by an integral.

The eigenvalues of the system (2.4) are, to within a sign, none other than the eigenvalues of the quasienergy[15,16] in units of $\hbar$, and the eigenvectors characterize the structure of the quasienergy eigenstates (QES). For infinite systems the spectrum λ may be both discrete and continuous.

The eigenvectors of the system of equations (2.4) with arbitrary $\gamma_{n-1,n}$ possess the following important orthogonality property:

$$\sum p_n(\lambda) p_n(\lambda_1) = 0 \quad \text{for} \quad \lambda \neq \lambda_1, \tag{2.5}$$

which may be obtained directly from Eqs. (2.4) and is also a special case of the general orthogonality properties of QES wave functions.[15,16] Just as for the time-independent states of quantum systems, the eigenvectors of the discrete QES spectrum can be normalized to unity, and those of the continuous spectrum can be normalized to a δ-function. It will be more convenient for us to normalize the eigenvectors such that

$$p_0 = 1, \tag{2.6}$$

and to include the renormalization in the function $u(\lambda)$ in the general solution (2.3).

The function $u(\lambda)$ should be chosen such that the solution (2.3) satisfies the initial conditions. We shall be interested in the initial conditions when the system is in the ground state at time $t = 0$, that is,

$$a_n(0) = \delta_{n0}. \tag{2.7}$$

If the eigenvalues and eigenvectors are known, it is no work to find the general form of the function $u(\lambda)$ by using the orthogonality property (2.5) and the normalization condition (2.6). The relationship for the determination of the function $u(\lambda)$ has the form

$$u(\lambda) \sum_{n=0}^{\infty} p_n(\lambda) p_n(\lambda_1) = \delta(\lambda - \lambda_1). \tag{2.8}$$

After these preliminary remarks, let us proceed to the investigation of specific systems.

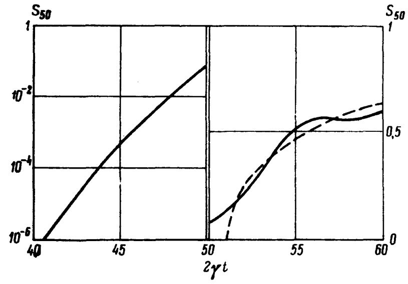

FIG. 1. Time dependence of the probability S_{50} for finding a system with a constant dipole moment in levels with quantum numbers $n \geqslant 50$. For comparison the dashed line denotes the function $g(t)$ determined by relation (3.6).

3. A SYSTEM WITH $\gamma_{n-1,n} = \gamma = \text{const}$

By comparing Eqs. (2.4) for this case with the recurrence relations for Tschebyscheff polynomials, [18] we find that $p_n(\lambda)$ can be expressed in terms of polynomials of the second kind:

$$p_n(\lambda) = U_n(z), \quad z = \lambda/2\gamma. \tag{3.1}$$

In this case the spectrum of eigenvalues is contained in the segment $-1 \leqslant z \leqslant 1$.

The expression for $a_n(t)$ satisfying the initial conditions (2.7) is determined by the following integral which reduces to a Bessel function:

$$a_n(t) = \frac{2}{\pi}\int_{-1}^{1}(1-z^2)^{1/2}U_n(z)\exp(2i\gamma tz)\,dz = i^n\frac{n+1}{\gamma t}J_{n+1}(2\gamma t). \tag{3.2}$$

As $t\to\infty$ the amplitudes $a_n(t)$ tend to zero, i.e., the system escapes to infinity.

Let us investigate the properties of the system's distribution $W_n(t) = a_n^* a_n$ over its levels. From the well known smooth asymptotic expansions of the Bessel functions, [19] one can conclude that at each instant of time for $\gamma t \gg 1$ the maximum of the distribution corresponds to levels with quantum numbers $n_{\max}$ occurring in the interval

$$2\gamma t-(2\gamma t)^{1/3} < n_{\max}+1 < 2\gamma t+(2\gamma t)^{1/3}. \tag{3.3}$$

Outside of this interval the probability W_n falls off exponentially in the region $n+1 > 2\gamma t$ and oscillates in the region $n+1 < 2\gamma t$, with its mean value smoothly decreasing toward the side of smaller n according to the law

$$W_n \sim (n+1)^2/\pi\gamma^2 t^2[4\gamma^2t^2-(n+1)^2]^{1/2}. \tag{3.4}$$

It is clear from what has been said that passage of the system through a given level has a clearly expressed threshold character. For $k \gg 1$ the time for attainment of the k-th level is $t_k \sim k/2\gamma$. We note that a similar estimate $t_{\text{char}} \sim M/2\gamma$ was made in Ref. 12 for the characteristic time associated with the establishment of a quasiequilibrium distribution in a finite M-level system with a constant dipole moment of the transitions.

It is convenient to characterize the passage of the system through a given level by the quantity

$$S_k(t) = \sum_{n=k}^{\infty} a_n^* a_n = 1 - \sum_{n=0}^{k-1} a_n^* a_n. \tag{3.5}$$

One can obtain a rough expression for $S_k(t)$ at an excess above threshold $(k+1 < 2\gamma t)$ by substituting expression (3.4) for levels below the k-th one into (3.5) and replacing the summation by an integration. As a result we find that for arbitrary k the quantity S_k is the following universal function of the variable $\eta = 2\gamma t/(k+1)$:

$$S_k(t) \sim g(\eta) = 1 - \frac{2}{\pi}\left[\arcsin\frac{1}{\eta} - \frac{1}{\eta^2}(\eta^2-1)^{1/2}\right], \quad \eta > 1. \tag{3.6}$$

The function $g(\eta)$ falls off rapidly, reaching a value ≈ 0.94 already at twofold excess above threshold.

More accurate estimates are required near threshold. Starting from Eqs. (2.1), with (3.2) taken into account, we find the following equation for $S_k(t)$:

$$\frac{dS_k}{dt} = i\gamma(a_k^* a_{k-1} - a_k a_{k-1}^*) = \frac{2k(k+1)}{\gamma t^2}J_k(2\gamma t)J_{k+1}(2\gamma t). \tag{3.7}$$

By integrating (3.7) (see Ref. 20) and making a numerical calculation, we obtain (Fig. 1) the typical $S_k(t)$ dependence. Above the threshold, relatively rapid oscillations are superimposed on the monotonic growth of the function $S_k(t)$, but on the average the behavior of $S_k(t)$ is well described by formula (3.6).

The important characteristics of the system's distribution with respect to its levels are the mean value, the root-mean-square value, and the variance. We obtain the following equation for the mean value directly from Eqs. (2.3):

$$\frac{d^2\bar{n}}{dt^2} = \frac{d^2}{dt^2}\sum_{n=1}^{\infty} n a_n^* a_n = 2\gamma^2 a_0^* a_0, \quad \bar{n}(0) = 0, \quad \frac{d\bar{n}}{dt}(0) = 0. \tag{3.8}$$

After integration[20] we obtain the following exact expression for $\bar{n}$:

$$\bar{n} = {}^{16}/_3\gamma^2t^2[J_0^2(2\gamma t)+J_1^2(2\gamma t)] - {}^{8}/_3\gamma t J_0(2\gamma t)J_1(2\gamma t) + J_0^2(2\gamma t) + {}^{1}/_3 J_1^2(2\gamma t) - 1. \tag{3.9}$$

The asymptotic form of the Bessel functions for $\gamma t \gg 1$ reduces this expression to the form

$$\bar{n} \sim \frac{16}{3\pi}\gamma t - 1 + \frac{1}{2\pi\gamma t} + O\left(\frac{1}{\gamma^3 t^3}\right), \tag{3.10}$$

that is, $\bar{n} \sim 0.85 n_{\max}$.

There are also no difficulties in deriving a formula for the root-mean-square value. After simple transformations we obtain

$$\overline{n^2} = \sum_{n=1}^{\infty} n^2 a_n^* a_n = \frac{1}{\gamma^2 t^2}\sum_{n=2}^{\infty} n^2(n^2-1)J_n^2(2\gamma t) - 2\bar{n} = 3\gamma^2t^2 - 2\bar{n}, \tag{3.11}$$

because the sum of the series in Eq. (3.11) is well known. [20]

TABLE I.

n	V_n	n	V_n
0	0.08	3	0.07
1	0.15	4	0.05
2	0.10	5	0.04

The following formula for the variance of the distribution for $\gamma t \gg 1$ follows from formulas (3.10) and (3.11):

$$D=\overline{n^2}-\bar{n}^2\sim\left(3-\frac{256}{9\pi^2}\right)\gamma^2t^2+1-\frac{16}{3\pi^2}+O\left(\frac{1}{\gamma^2t^2}\right). \quad (3.12)$$

From here we find that the quantity $D^{1/2}$ characterizing the halfwidth of the distribution is related to $\bar{n}$ by the approximate relationship $D^{1/2}\sim 0.2\bar{n}$. Thus, for $\gamma t\gg 1$ the relative width of the distribution is constant, in contrast to the harmonic oscillator where it decreases with time (formulas (1.1) and (1.2)).

However, in summarizing the results of the present section we note that the excitation of an equidistant system having a constant dipole moment is subject to the same qualitative regularities as the excitation of a harmonic oscillator. Foremost among these regularities is the possibility, in principle, of unlimited excitation.

4. A SYSTEM WITH $\gamma_{n-1,n}=\gamma/(n+1)^{1/2}$

For this case let us make a replacement of the unknowns in Eqs. (2.4) according to the formula

$$p_n=(-1)^n[(n+1)!]^{1/2}(\lambda/\gamma)^n s_n. \quad (4.1)$$

The corresponding system of equations for s_n is given by

$$-s_0=s_1,$$
$$-(n+1)s_n=xs_{n-1}+(n+1)s_{n+1},\ n\geq 1, \quad (4.2)$$

where $x=(\gamma/\lambda)^2$.

Equations (4.2) coincide with the special case of the recurrence relations[18] for Laguerre polynomials of the form $L_n^{x-n-1}(x)$, which tend to zero as $n\to\infty$ if x is equal to a positive integer, but increase without limit in the opposite case. Therefore, the spectrum of the eigenvalues λ is discrete and is given by the formula

$$\lambda_{(\pm m)}=\pm\gamma/m^{1/2},\ m=1,\ 2,\ldots. \quad (4.3)$$

The components of the eigenvectors which are expressed by the formula

$$p_n^{\pm m}=(\mp 1)^n[(n+1)!]^{1/2}m^{-n/2}L_n^{m-n-1}(m), \quad (4.4)$$

represent, apart from a factor, a special case of the Poisson-Charlier polynomials,[21] where the normalization factors may be determined from the known properties of these polynomials:

$$\sum_{n=0}^{\infty}|p_n^{(\pm m)}|^2=2e^m m^{2-m}(m-1)! \quad (4.5)$$

Combining Eqs. (4.3)–(4.5) and (2.8), after simple transformations we find that the solution of the initial system of Eqs. (2.1) satisfying the initial conditions (2.7) has the following form for the case under consideration:

$$a_n(t)=i^n[(n+1)!]^{1/2}\sum_{m=1}^{\infty}\frac{m^{m-2-n/2}}{(m-1)!}e^{-m}L_n^{m-n-1}(m)\cos\left(\frac{\gamma t}{m^{1/2}}-\frac{n\pi}{2}\right). \quad (4.6)$$

Since the quasienergy spectrum of the system under consideration is discrete, the amplitudes do not attenuate to zero with time and are so-called almost periodic functions,[22] that is, the system does not escape to infinity. One can nevertheless estimate how strongly in principle sufficiently high levels are occupied, having averaged the distribution over an infinite time interval. Such averaging leads to the following formula:

$$V_n=\lim_{T\to\infty}\frac{1}{T}\int_0^T a_n^*(t)a_n(t)\,dt=\frac{1}{2}(n+1)!\sum_{m=1}^{\infty}\frac{m^{2m-4-n}}{[(m-1)!]^2}e^{-2m}[L_n^{m-n-1}(m)]^2. \quad (4.7)$$

The results of a numerical calculation of the levels' average populations according to formula (4.7) are partially reflected in Tables I and II, where the average level population distribution is given for excitation, by a resonant field, of an equidistant multilevel system whose transition dipole moment decreases according to the law $\mu_{n-1,n}=\mu/(n+1)^{1/2}$. In the lowest transition, inversion takes place on the average. This is followed by a monotonic decrease of the populations. This decrease, however, is slow and the probability of the system being at very high levels is appreciable. The asymptotic behavior of V_n has the form

$$V_n\propto n^{-3/2}. \quad (4.8)$$

The calculation also shows that the basic contribution to the sum in formulas (4.7) and (4.6) is given by terms with m contained within the interval

$$n/3<m<n. \quad (4.9)$$

It follows from the asymptotic behavior (4.8) of V_n that the energy of the system increases without limit. The asymptotic behavior of the energy can be obtained from the following considerations. The averaging leading to formula (4.7) is carried out over an infinite time interval. Averaging over a finite interval must also take into consideration averaging of the oscillating terms containing sum and difference frequencies of the form

$$\lambda_{m_1}\pm\lambda_{m_2}=\gamma/m_1^{1/2}\pm\gamma/m_2^{1/2}. \quad (4.10)$$

TABLE II.

n_1	n_2	$\sum_{n=n_1}^{n_2} V_n$
6	10	0.12
11	20	0.10
21	∞	0.29

Since terms with $m \sim n$ (see Eq. (4.9)) make the main contribution to expression (4.6) for the amplitude $a_n(t)$, the minimal difference frequencies essential for averaging are of the order of $\gamma/n^{3/2}$. Hence the characteristic time t_n for excitation of the n-th level may be estimated at

$$t_n \sim n^{3/2}/\gamma. \tag{4.11}$$

Now, using the asymptotic expression (4.8) for V_n one can easily derive

$$\bar{n} \propto (\gamma t)^{2/3}. \tag{4.12}$$

In concluding the present section we note that the time for excitation of the nth level for the systems under consideration, including the harmonic oscillator, may be expressed by the following universal formula:

$$t_n \sim n/\gamma_{n-1,\,n}. \tag{4.13}$$

5. A SYSTEM HAVING FOR THE 0–1 TRANSITION A DIPOLE MOMENT DIFFERENT FROM THE DIPOLE MOMENT FOR THE SUBSEQUENT TRANSITIONS

Let us assume $\gamma_{n-1,n} = \gamma = \text{const}$ for $n \geq 2$, $\gamma_{01}/\gamma = \beta$. For this case the system (2.4) takes the following form:

$$\lambda p_0 = \beta\gamma p_1,\ \lambda p_1 = \beta\gamma p_0 + \gamma p_2,\ \lambda p_n = \gamma(p_{n-1} + p_{n+1}),\quad n \geqslant 2. \tag{5.1}$$

In order to find the solutions of the system of Eqs. (5.1) it is convenient to investigate a truncated system from which the first two equations are eliminated. Such a truncated system obviously has two linearly independent solutions, and its general solution can be represented in the form

$$p_n = C_1 T_n(z) + C_2 U_n(z),\ n \geqslant 1, \tag{5.2}$$

where, just as in Sec. 3, $z = \lambda/2\gamma$ and $T_n(z)$ and $U_n(z)$ are Tschebyscheff polynomials of the first and second kind, respectively. Using the explicit form of the Tschebyscheff polynomials for $n = 1$, 2 and taking Eq. (2.6) into consideration, we obtain from the first two equations of the system (5.1) the following expressions for the constants C_1 and C_2:

$$C_1 = 2(\beta^2 - 1)/\beta,\ C_2 = (2 - \beta^2)/\beta. \tag{5.3}$$

We note that for $\beta = 1$ (the case investigated in Sec. 3) we have $C_1 = 0$ and $C_2 = 1$, that is, the solution (5.2) agrees with (3.1).

Just as in Sec. 3, the continuous spectrum of eigenvalues is contained in the segment $-1 \leq z \leq 1$. However, in contrast to Sec. 3, for certain values of β there are also discrete eigenvalues z which can easily be determined from the following condition:

$$\lim_{n\to\infty} p_n(z) = 0. \tag{5.4}$$

Bearing in mind that

$$T_n(\cos\theta) = \cos n\theta,\ U_n(\cos\theta) = \sin(n+1)\theta/\sin\theta,$$

and regarding $\theta = x + iy$ as a complex variable, we find that condition (5.4) reduces to the following system of equations for the determination of x and y:

$$(\beta^2 - 1 - e^{2|y|})\cos x = 0,\ (\beta^2 - 1 + e^{2|y|})\sin x = 0. \tag{5.5}$$

The system (5.5) has solutions only under the condition

$$\beta \geqslant \sqrt{2}. \tag{5.6}$$

These solutions are:

$$x = N\pi\ (N = 0, \pm1, \pm2, \ldots),\ y = \pm{}^1/{}_2 \ln(\beta^2 - 1), \tag{5.7}$$

and in the plane of the variable $z = \cos(x + iy)$ their aggregate reduces to the following two eigenvalues:

$$z_{(\pm)} = \pm\beta^2/2(\beta^2 - 1)^{1/2}. \tag{5.8}$$

The corresponding eigenvectors may be written in the form

$$p_0 = 1,\quad p_n^{(\pm)} = (\pm1)^n \beta(\beta^2 - 1)^{-n/2},\quad n \geqslant 1 \tag{5.9}$$

Once the eigenvalue spectrum and the eigenvectors of the system under consideration are found, we obtain for the amplitudes a solution satisfying the initial conditions (2.7).

1. First let us consider the case of a purely continuous spectrum, that is, when the condition

$$\beta \leqslant \sqrt{2}, \tag{5.6'}$$

which is the reverse of condition (5.6), is satisifed. In agreement with Eq. (2.8), the function $u(z)$, which effects the expansion of the solution in terms of eigenvectors, is sought from the relationship

$$u(z)\left\{1 + \sum_{n=1}^{\infty} [C_1 T_n(z) + C_2 U_n(z)][C_1 T_n(z_1) + C_2 U_n(z_1)]\right\} = \delta(z - z_1). \tag{5.10}$$

The simplest way of obtaining $u(z)$ in explicit form is to multiply both sides of (5.10) by $(1 - z_1^2)^{1/2}$ and integrate with respect to z_1 in the interval $-1 \leq z_1 \leq 1$. Here only the integral containing $T_2(z_1)$ gives a nonvanishing term in the sum. As a result we obtain the following expressions for the amplitudes $a_n(t)$:

$$a_0(t) = \frac{2\beta^2}{\pi}\int_{-1}^{1} \frac{(1 - z^2)^{1/2}\exp(2i\gamma tz)}{\beta^4 + 4(1 - \beta^2)z^2}\,dz, \tag{5.11}$$

$$a_n(t) = \frac{2\beta^2}{\pi}\int_{-1}^{1} \frac{(1 - z^2)^{1/2}[C_1 T_n(z) + C_2 U_n(z)]}{\beta^4 + 4(1 - \beta^2)z^2}\exp(2i\gamma tz)\,dz,\quad n \geqslant 1. \tag{5.12}$$

It is obvious that for $\beta \sim 1$ the solution does not differ essentially from that derived in Sec. 3. Let $\beta \ll 1$, i.e., the dipole moment of the 0–1 transition is considerably smaller than the dipole moment of the following transitions. In this case the rate of the system excitation is determined by the rate of attenuation of the ground-state amplitude. The neighborhood of the point $z = 0$ plays a major role in expression (5.11) for $a_0(t)$. Therefore we can retain only the oscillating term in the numerator of the integrand and extend the integration to the entire infinite axis. As a result we obtain

$$a_0(t)\approx\exp(-\beta^2\gamma t)=\exp(-\gamma_{01}^2 t/\gamma). \qquad (5.13)$$

Thus, the amplitude is damped exponentially without oscillations, as is typical of a two-level system having an upper-level width exceeding the rate of the induced transitions.[23,24] In the case under consideration the role of the width of the first excited level is played by the quantity γ which characterizes its rate of decay due to transitions to higher levels.

At $\gamma_{01}^2 t/\gamma \ll 1$ the total probability S_1 for excitation of the system increases linearly:

$$S_1=1-a_0^* a_0\sim 2\gamma_{01}^2 t/\gamma. \qquad (5.14)$$

It was shown in Sec. 3 that excitation of the n-th level has a threshold character with a time $t_n \sim n/2\gamma$. Hence the average population of the levels with $n<2\gamma t$ is given by

$$W_n\sim S_1/n_{max}\sim(\gamma_{01}/\gamma)^2. \qquad (5.15)$$

At $\gamma_{01}^2 t/\gamma > 1$, the amplitudes of the states with $n \ll 2\gamma t$ are also attenuated together with the ground state-amplitude.

2. Let us go on to an examination of the case when there are two discrete eigenvalues together with the continuous spectrum, i.e., when condition (5.6) is satisfied. In this case the amplitudes $a_n(t)$ are determined by the following expression:

$$a_n(t)=\tilde{a}_n(t)+u_{(+)}p_n^{(+)}\exp(2i\gamma z_{(+)}t)+u_{(-)}p_n^{(-)}\exp(2i\gamma z_{(-)}t). \qquad (5.16)$$

Here $\tilde{a}_n(t)$ denotes the already obtained part of the solution which is associated with the continuous spectrum (formulas (5.11) and (5.12)); $z_{(\pm)}$ and $p_n^{(\pm)}$ are determined by formulas (5.8) and (5.9). In accordance with Eq. (2.8) we obtain the following result for the coefficients $u_{(\pm)}$ of the expansion in the discrete eigenvalues:

$$u_{(\pm)}=\Big[\sum_{n=0}^{\infty}|p_n^{(\pm)}|^2\Big]^{-1}=\frac{\beta^2-2}{2(\beta^2-1)}. \qquad (5.17)$$

As $t\to\infty$ the $\tilde{a}_n(t)$ part of the solution (5.16), associated with the continuous spectrum, is attenuated. There is left the oscillating part of the solution which predominates at $\beta\gg 1$, i.e., when the dipole moment of the 0–1 transition is much larger than the dipole moment of the subsequent transitions. In fact, the probability W_d of finding the system in the two discrete QES is given by

$$W_d=u_{(+)}^2\sum_{n=0}^{\infty}|p_n^{(+)}|^2+u_{(-)}^2\sum_{n=0}^{\infty}|p_n^{(-)}|^2=\frac{\beta^2-2}{\beta^2-1}. \qquad (5.18)$$

For $\beta\gg 1$ we have $W_d\approx 1-\beta^{-2}$, i.e., the probability W_d is close to unity.

The probability W_i of the system's escape to infinity, which is equal to the probability of finding the system in the continuous QES, is obviously given by

$$W_i=1-W_d=1/(\beta^2-1), \qquad (5.19)$$

that is, for $\beta\gg 1$ we have $W_i\simeq\beta^{-2}\ll 1$.

The physical cause of the effect becomes clear if we consider the structure of the eigenvectors corresponding to the discrete QES. For $\beta\gg 1$

$$|p_0^{(\pm)}|\approx|p_1^{(\pm)}|\approx 1,\qquad |p_n^{(\pm)}|\approx 1/\beta^{n-1}\ll 1,\quad n\geq 2. \qquad (5.20)$$

Thus, the system only oscillates between the two lowest levels. This is associated with that well known fact that a resonant field in a two-level system splits each level into two quasienergy sublevels which differ from the position of the steady-state level by the amounts $\pm\gamma_{01}$. Therefore, the transition from the QES corresponding to the first excited level to the next level automatically becomes nonresonant, and this detuning cannot be cancelled out because $\gamma\ll\gamma_{01}$.

Thus, the examples examined in the present section have shown that in multilevel systems an estimate of the rate of coherent excitation in a resonant field based on the minimal dipole moment of the subsequent transitions is not always adquate. In those cases when the dipole moment of one or several transitions differs markedly from the characteristic dipole moment of the sequence, important factors limiting the effectiveness of the excitation are:

a) the broadening of a relatively weak transition by stronger neighboring transitions;

b) splitting of a relatively strong transition, which causes the weaker neighboring transitions to deviate from resonance.

6. CONCLUSION

The results of the present work as applied to the kinetics for excitation of the quasicontinuum of a polyatomic molecule should be regarded as a first step. Further elaboration of the model should take the following factors into consideration:

1) the actual linewidth of the laser radiation;

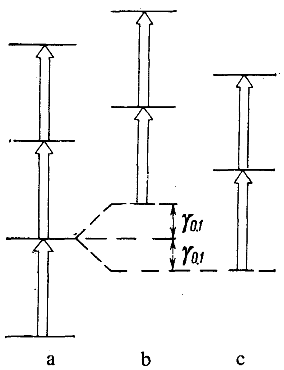

FIG. 2. Comparison of the probability W_i of escape to infinity for three multilevel systems having a dipole moment for the 0–1 transition much larger than the dipole moment of the remaining transitions: a) $W_i\ll 1$ (Sec. 5); b) and c) $W_i\sim 1/2$ (Refs. 9 and 10).

2) the contribution made to the process by all resonant transitions located within the limits of the laser line;

3) the role of not-strictly-resonant transitions, which may turn out to be substantial.

Another feasible approach is that of Ref. 8, where the influence of the quasicontinuum reduces to intramolecular decay by an excited anharmonic vibrational mode. It is not clear, however, how to allow such an approach for the evolution of finite states of decay for transitions between which the field is also quasiresonant.

The excitation of multilevel systems that simulate the molecule as a whole, and not just its quasicontinuum, is of special interest. Such an investigation[5,9,10] leads to systems consisting of two subsystems. The upper subsystem simulates the quasicontinuum. The lower subsystem consists of several levels of an excitable mode, and the dipole moment for its transitions is much larger than for the transitions to the quasicontinuum.

It was shown in Sec. 5 that in this case a situation is possible when no excitation is present even under conditions of resonance between all successive transitions. Excitation becomes effective[9,10] if the radiation is at resonance with the transition between the lower level of the upper "slow" subsystem and the upper quasilevel of the lower "fast" subsystem. This situation is illustrated in Fig. 2 as applied to the case considered in Sec. 5. In this connection for the decay of the ground state as $t\to\infty$ with probability ~ 1, two upper subsystems are required.

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