Dynamics of excitation of multilevel systems of the band type in a laser field

V. M. Akulin and A. M. Dykhne

P. N. Lebedev Physics Institute, USSR Academy of Sciences
(Submitted 28 June 1977)
Zh. Eksp. Teor. Fiz. **73**, 2098–2106 (December 1977)

With the population of high-lying vibrational state of polyatomic molecules as an example, we consider the problem of the kinetics of population of multilevel systems of the band type. Physical arguments are advanced to show that for sufficiently complex molecules the dipole-moment matrix elements can be regarded as random and satisfying the Wigner "microcanonical" distribution. Within the framework of this hypothesis we consider the limiting cases of strong and weak interactions of the system with a laser field. For strong interactions we obtain and analyze an expression for the population distribution over the bands. For the case of weak interaction we derive a balance equation that describes the kinetics of the population of a multilevel system. It is shown that multilevel systems of the band type exhibit simultaneously both coherent and stochastic properties.

PACS numbers: 33.80.Be, 42.50.+q

The action of laser radiation on vibrational degrees of freedom of molecules was first considered in[1]. The model of the anharmonic oscillator[2, 3] is extensively used at present. This model describes the excitation of diatomic molecules and the buildup of the nondegenerate modes of polyatomic molecules. At the same time, much attention is being paid to a process recently observed in experiment, collisionless dissociation of polyatomic molecules under the influence of a high-power electromagnetic field that is at resonance with the degenerate modes of polyatomic molecules.[4–6] The anharmonic-oscillator model turned out to be inadequate for the explanation of this effect and it was found necessary to take into account a peculiarity of polyatomic molecules that manifest themselves in a high density of the vibrational states.[7–9]

A direct approach to this problem would be to calculate the spectrum of the vibrational states of the polyatomic molecule and the matrix elements of the dipole moments, followed by a solution of the Schrödinger equation. However, owing to the large number of different vibrational states that actually take part in this process, none of the stages of such an analysis can be realized in practice. To describe the effect of collisionless dissociation one uses therefore various model approaches. A distinction can be made between two groups of model approaches. In the first[7, 9] are considered almost-resonant single levels and a coherent energy acquisition. In the other one uses a stochastization hypothesis[10] and thermodynamic considerations.[11] These models have a limited applicability. Thus, in coherent models no account is taken of the large number of closely-lying levels (which are also resonant), and one neglects effects of interference of a large number of excitation channels that proceed via different levels with comparable amplitudes. Allowance for this circumstance can cause the energy acquisition by the molecule to have in a number of cases a random-walk character. Statistical methods, in turn, ignore completely the coherent effects that can cause, for example, only narrow vicinities of the resonances to be substantially populated. Yet in the problem of excitation of a multilevel system with a complex spectrum an important role may be assumed (and is in fact assumed) simultaneously by coherent and stochastic process. As a first step we wish to propose solvable models that do not ignore either of these aspects of the phenomenon.

The purpose of solving this type of model problems is to find those characteristics of the spectrum and of the Hamiltonian of the interaction of the system with the field, to which the results are most sensitive (e.g., the efficiency of energy acquisition). There are physical grounds for assuming that the number of these characteristics is small. In this case the behavior of an entire ensemble of multilevel systems with different spectra and interaction operators turn out to be the same. It is therefore natural to use the method of averaging over the ensemble. This part of the problem is general in character, does not depend on the actual structure of the molecules, and can be used to describe any system with a complex spectrum and with a "stepwise" character of the interaction. In this paper we restrict ourselves only to this part.

The subsequent program could consist of a direct calculation of the aforementioned characteristics for concrete molecules. In this approach there is hope of getting rid of the influence of a large number of degrees of freedom, so that the problem might be solved by a direct method.

1. THE MODEL

We have chosen the following model of the spectrum:

1) the level groups are localized in the vicinities of the energy values $n\hbar\omega$;

2) in each vicinity there is a large number $N_n \gg 1$ of levels, which are numbered by the index m;

3) the dipole moment differs from zero only for $\Delta n = \pm 1$ transitions between neighboring bands.

The Schrödinger equation for such a system is written in the form

 0038-5646/77/4606-1099$02.40

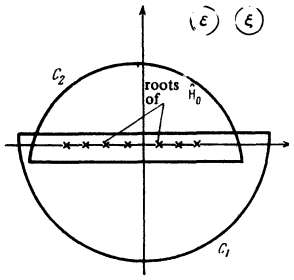


FIG. 1.

$$i\dot{\tilde{\psi}}_{n,m}=\varepsilon_{n,m}\tilde{\psi}_{n,m}+\sum_{n',m'}E\mu_{n,m}^{n',m'}\tilde{\psi}_{n',m'}\cos\omega t,\quad \hbar\equiv 1, \tag{1}$$

where $\mu_{n,m}^{n',m'}$ are the matrix elements of the transitions.

Carrying out the transformation $\tilde{\psi}_{n,m}=\psi_{n,m}\exp(-in\omega t)$ and neglecting the oscillating terms, we reduce Eq. (1) to the form

$$i\dot{\psi}_{n,m}=\Delta_{n,m}\psi_{n,m}+\sum_{n',m'}E\mu_{n,m}^{n',m'}\psi_{n',m'}, \tag{2}$$

where $\Delta_{n,m}=\varepsilon_{n,m}-n\omega$. We write down the solution of (2):

$$\psi_{n,m}(t)=\sum_{n',m'}\{\exp-i\hat{H}t\}_{n,m}^{n',m'}\psi_{n',m'}(0), \tag{3}$$

where

$$\hat{H}=\hat{H}_0+\hat{V}\equiv\delta_n{}^{n'}\delta_m{}^{m'}\Delta_{n,m}+E\mu_{n,m}^{n',m'}.$$

From (3) follows an expression for the diagonal elements of the density matrix

$$\rho_{n,m}^{n,m}=\langle n,m|e^{-i\hat{H}t}\hat{\rho}(0)e^{i\hat{H}t}|n,m\rangle \tag{4}$$

or, using the formula for the function of the operator,

$$\rho_{n,m}^{n,m}=\frac{1}{4\pi^2}\int_{C_1}d\varepsilon\int_{C_2}d\xi\,e^{-i(\varepsilon-\xi)t}\langle n,m|\frac{1}{\varepsilon-\hat{H}}\hat{\rho}(0)\frac{1}{\xi-\hat{H}}|n,m\rangle, \tag{5}$$

where the integration contours C_1 and C_2 are shown in Fig. 1. If we use the expansion

$$(x-\hat{H}_0-\hat{V})^{-1}=(x-\hat{H}_0)^{-1}+(x-\hat{H}_0)^{-1}\hat{V}(x-\hat{H}_0)^{-1}+\ldots,$$

then the integrand in (5) can be represented as a sum over the trajectories

$$\sum_{(i)(k)}(\varepsilon-\Delta_{n,m})^{-1}V_{i,k}^{n,m}\ldots V_{n_0,m_0}^{i',k'}(\varepsilon-\Delta_{n_0,m_0})^{-1}\rho_{n_0,m_0}^{n_0,m_0}\cdot$$
$$\cdot(\xi-\Delta_{n_0,m_0})^{-1}V_{i'',k''}^{n_0,m_0}\ldots V_{n,m}^{i''',k'''}(\xi-\Delta_{n,m})^{-1}. \tag{6}$$

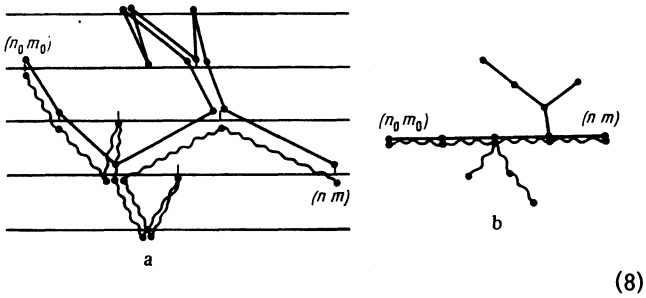
If each term of the series is represented in graphic form, then we obtain diagrams of the type shown in Fig. 2. The wavy lines in this figure correspond to the right-hand operator bracket and the straight lines to the left-hand bracket. Each straight or wavy line joining the point (a, b) with the point (c, d) corresponds to the matrix element $V_{c,d}^{a,b}=E\mu_{c,d}^{a,b}$ of the interaction operator. To each point (a, b) on a wavy line corresponds to a factor $(\xi-\Delta_{a,b})^{-1}$, and on a straight line to a factor $(\varepsilon-\Delta_{a,b})^{-1}$. The wave and straight lines start out from points corresponding to levels populated at the instant time $t=0$, and terminates at the point (n, m) of interest to us. If only a certain level (n_0, m_0) is populated at the instant of time $t=0$, then the population of the (level (n, m) is expressed as the sum of all the possible trajectories that begin at the point (n_0, m_0) and end at the point (n, m).

The subsequent analysis of the problem depends essentially on the statistics of the dipole moments. We confine ourselves in this paper to an analysis of a system with random dipole moments. The physical arguments favoring such a model are the following: The presence of anharmonicity of the vibrations of polyatomic molecules causes the levels of various degenerate and multiple modes to experience Fermi resonances, as a result of which the dipole moment of the oscillatory transition becomes smeared out between them. This smearing has a complicated and irregular character. The only requirement that the matrix elements of the transition dipole moments must satisfy is the sum rule (the sum of the squares must be constant). A natural model that describes this situation is the total ignorance model, which leads to the "microcanonical" Wigner distribution function[1)]

$$g(\{\mu_{n,m'}^{n+1,m}\})=\delta\Big[\sum_{m,m'}(\mu_{n,m'}^{n+1,m})^2-(\mu_n^{n+1})^2\Big]\prod_{m,m'}d\mu_{n,m'}^{n+1,m}. \tag{7}$$

According to the distribution (7), the matrix elements of the dipole moments do not correlate with one another.

As a result of the distribution (7) the essential contribution to the sum (6) is made only by those trajectories in which each pair of points is joined by two lines (see (8), where diagram b is the schematic representation of diagram a).



(8)

In addition, the condition $N_n \gg 1$ over times shorter than the Poincare return point allows us to sum only the

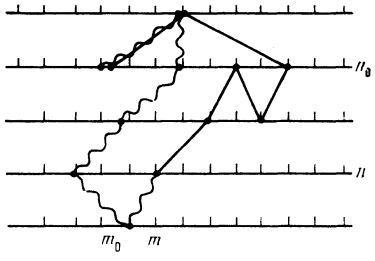
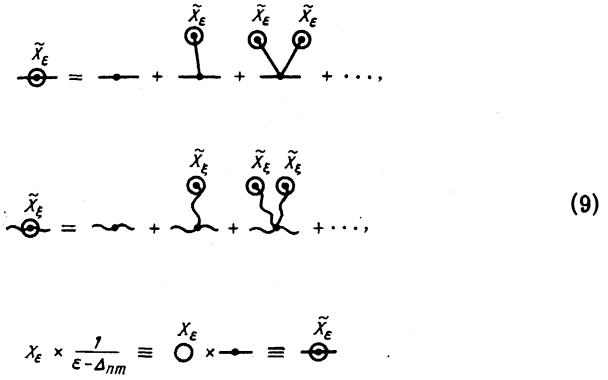


FIG. 2.

principal sequence of the series (6) and neglect the self-intersection of the trajectories. This form of the diagrams allows us to renormalize the operators corresponding to points on the straight and wavy lines. The operators are obtained from the recurrence relations (9):



(9)

The concrete form of these operators depends on the relation between the interaction Hamiltonian and the dimension of the band of levels γ_n. If $V \gg \gamma_n$, the interaction is "strong" and we can neglect the size of the band and regard it as degenerate. In the opposite limiting case we can let the band dimension tend to infinity, after fixing the average level density.

The quantity $\rho_{n_0 m}^{n m}$ is self-averaging. In fact the small contribution $O(1/N)$ of the self-intersecting trajectories means that the variance is small:

$$\langle \rho_{n,m}^{n,m} \rangle^2 - \langle (\rho_{n,m}^{n,m})^2 \rangle = O(1/N).$$

This means that for the overwhelming majority of systems with noncorrelating matrix elements of the dipole-moment operator the distribution of the populations over the bands is the same. This justifies the use of the technique of averaging over the ensemble for the description of a concrete system with random dipole moments.

2. TWO DEGENERATE LEVEL BANDS

We consider the problem of two degenerate levels such that dipole transitions are allowed between all the sublevel. In this case each line corresponds to a factor $V = E\mu_m^{m'}$, and each point on a wavy (straight) line corresponds to a factor $\xi^{-1}(\varepsilon^{-1})$. For this case, relation (9) takes the form

$$X_\varepsilon = \sum_{n=0}^{\infty} \left(\frac{u}{\varepsilon^2} X_\varepsilon \right)^n = \frac{\varepsilon^2}{\varepsilon^2 - u X_\varepsilon}, \quad X_\varepsilon = \frac{\varepsilon^2}{2u} - \left[\left(\frac{\varepsilon^2}{2u} \right)^2 - \left(\frac{\varepsilon^2}{u} \right) \right]^{1/2} \tag{10}$$

where $u = N\langle E\mu_m^{m'} \rangle$, and $\langle \cdots \rangle$ means averaging over the Wigner ensemble. The summary population of the upper level is given by

$$\rho_2^2 = -\frac{1}{4\pi^2} \int_{c_1} d\varepsilon \int_{c_2} d\xi \frac{X_\varepsilon X_\xi}{\varepsilon \xi} \frac{\varepsilon \xi u X_\varepsilon X_\xi}{(\varepsilon \xi)^2 - (u X_\varepsilon X_\xi)^2} e^{-i(\varepsilon - \xi)t}, \tag{11}$$

and the population of the lower level is

$$\rho_1^1 = -\frac{1}{4\pi^2} \int_{c_1} d\varepsilon \int_{c_2} d\xi \frac{X_\varepsilon X_\xi}{\varepsilon \xi} \frac{(\varepsilon \xi)^2}{(\varepsilon \xi)^2 - (u X_\varepsilon X_\xi)^2} e^{-i(\varepsilon - \xi)t}. \tag{12}$$

We define the quantities $I_1 = \rho_2^2 + \rho_1^1$ and $I_2 = \rho_1^1 - \rho_2^2$. Taking (10) into account and making the change of variables $\xi = (2u)^{1/2} \operatorname{ch}\varphi$, $\varepsilon = (2u)^{1/2} \operatorname{ch}\psi$, $\tau = (2u)^{1/2} t$, we get

$$I_{1,2} = \int_{c_1'} \int_{c_2'} (e^{\varphi + \psi} \mp 1)^{-1} \operatorname{sh}\psi \operatorname{sh}\varphi \exp[i\tau(\operatorname{ch}\psi - \operatorname{ch}\varphi)] d\varphi\, d\psi, \tag{13}$$

where the integration contour C_1' is the segment $\operatorname{Re}\varphi > 0$, $\operatorname{Im}\varphi \in (-\pi/2; 3\pi/2)$ and the contour C_2' is the segment $\operatorname{Re}\psi > 0$, $\operatorname{Im}\psi \in (3\pi/2; -\pi/2)$. The integrals (13) can be evaluated:

$$I_1 = 1, \quad I_2 = J_0(\tau) + J_2(\tau) = \frac{2}{\tau} J_1(\tau). \tag{14}$$

We ultimately get

$$\rho_1^1 = \frac{1}{2}\left[1 + \frac{2}{E(N\langle (\mu_m^{m'})^2 \rangle)^{1/2} t} J_1(E(N\langle (\mu_m^{m'})^2 \rangle)^{1/2} t) \right], \tag{15}$$
$$\rho_2^2 = \frac{1}{2}\left[1 - \frac{2}{E(N\langle (\mu_m^{m'})^2 \rangle)^{1/2} t} J_1(E(N\langle (\mu_m^{m'})^2 \rangle)^{1/2} t) \right].$$

As $t \to \infty$ the populations become equalized at a rate

$$\langle |\rho_{11} - \rho_{22}| \rangle_t \sim t^{-1/2}. \tag{16}$$

It must be noted that this result can be obtained also in another manner. For a two-band degenerate submatrix the dipole moment submatrices can be diagonalized. As a result, the band-population problem reduces to a determination of the average level population of an aggregate of two-level systems whose transition dipole-moment matrix elements are described by the known "semicircular" density distribution[2] [13]: $g(\mu) = (2\pi u)^{-1} \times (4\pi u - \mu^2)^{1/2}$. The population of the upper levels is given in this case by the expression

$$\rho_2^2(t) = \int (4\pi u - \mu^2)^{1/2} \sin^2(E\mu t)\, d\mu.$$

We emphasize that the expression for the eigenvalue density of a random matrix was obtained in[13] for a "canonical" Wigner distribution. In our problem we used a "microcanonical" Wigner distribution, which led to the same result. This result is natural, inasmuch as over times much shorter than the Poincaré return time only a small fraction of the dipole moments (subsystem) has become involved, and the microcanonical distribution reduces to a canonical one.

3. INFINITE SYSTEM OF DEGENERATE LEVELS

Such a system simulates the acquisition of energy by a polyatomic molecule in the case of a strong interaction with a laser field ($\gamma_n \ll E(N_n \langle (\mu_{n-1,m}^{n,m'})^2 \rangle)^{1/2}$). In this problem, the operators X_ε and X_ξ retain the same form as in Sec. 2. To determine the population of the n-th level it is necessary to find the number of trajectories that go from the point n_0 into the point n and consist of wavy and straight line jointly. For an infinite system of levels the number of trajectories of length M, joining n and n_0, is determined by the number of combinations

$C_M^{(M-n+n_0)/2}$. This means that if at the initial instant $t=0$ only one level n_0 was populated, then the population of the n-th level at the instant of time t is given by

$$\rho_n^n = \sum_M \int_{c_1} d\varepsilon \int_{c_2} d\xi \frac{X_\varepsilon X_\xi}{\varepsilon\xi} C_M^{1/2(M-n+n_0)} \left(\frac{2uX_\varepsilon X_\xi}{\varepsilon\xi}\right)^M e^{-i(\varepsilon-\xi)t}. \quad (17)$$

If we use the relation

$$\sum_{k=0}^{\infty} x^{k+1} C_k^{1/2(k-s)} = \left(\frac{1}{4x^2}-1\right)^{-1/2} \cos\left(s \arccos\frac{1}{2x}\right), \quad (18)$$

which is obtained with the aid of the integral representation for C_b^a, then we obtain from (17) and (18) for the case $\rho_n^n(0)=\delta_n^{n_0}$:

$$\rho_n^n = \int_{c_1} d\varepsilon \int_{c_2} d\xi\, e^{-i(\varepsilon-\xi)t} \left[\left(\frac{\varepsilon\xi}{X_\varepsilon X_\xi u}\right)^2 - 1\right]^{-1/2} T_{n-n_0}\left(\frac{\varepsilon\xi}{X_\varepsilon X_\xi u}\right). \quad (19)$$

At large n this is a saddle-point integral. It can be represented in the form

$$\rho_n^n \sim t^{-1/2} G(n^2/t), \quad (20)$$

where $G(n^2/t)$ is a certain universal function that attenuates exponentially at infinity and contains oscillations in the vicinity of $n^2 \leq Et(N\langle\mu^2\rangle)^{1/2}$. The oscillations of the function G are the traces of the coherence, and the dependence on the argument n^2/t is a consequence of the stochastic behavior. In the essential regions $n^2 \sim Et(N\langle\mu^2\rangle)^{1/2}$, where $G\sim 1$, the contributions of the coherence effects and stochasticity are of the same order. We shall not present the explicit form of the function G, which can be expressed in terms of parabolic-cylinder functions. We note only that (20) enables us to show that the acquisition of energy by the molecule in the case of a strong interaction is given by

$$\mathscr{E} \sim [Et(N\langle\mu^2\rangle)^{1/2}]^{2/3}. \quad (21)$$

4. CASE OF WEAK INTERACTION

We consider the case of a relatively weak interaction, such that

$$\gamma_n/N_n \ll E[\langle(\mu_{n-l,m}^{n,m'})^2\rangle]^{1/2} = V \ll \gamma_n. \quad (22)$$

This model describes most probably the acquisition of energy by a polyatomic molecule. For simplicity we consider the case of an equidistant spectrum[3] $\Delta_{n,m} = \alpha m$ and set α equal to π. This means that the energy-dependent quantities are measured in units of α/π and the time in π/α.

Equation (9) leads in this case to the relation

$$\frac{X_\varepsilon(n,m)}{\varepsilon-\Delta_{n,m}} = \left[\varepsilon-\Delta_{n,m}-V^2\sum_{n',m'} X_\varepsilon(n',m')(\varepsilon-\Delta_{n',m'})^{-1}\right]^{-1}. \quad (23)$$

If we put

$$Q_\varepsilon = \sum_{n',m'} X_\varepsilon(n',m')(\varepsilon-\Delta_{n',m'})^{-1},$$

then each point will correspond to the operator

$$\frac{X_\varepsilon(n,m)}{\varepsilon-\Delta_{n,m}} = \frac{1}{\varepsilon-\Delta_{n,m}-V^2Q_\varepsilon} \quad (24)$$

and for Q_ε we have the equation

$$Q_\varepsilon = \sum_m \frac{1}{\varepsilon-V^2Q_\varepsilon-m\pi} = \operatorname{ctg}(\varepsilon-V^2Q). \quad (25)$$

At $V^2 \gg 1$ there exist roots of (25) that do not depend on the microstructure of the spectrum: $Q_{\varepsilon,\xi}=\pm i$. In this case the following recurrence relation holds for the total populations of the bands:

$$\rho_n^n = \frac{1}{2}(\rho_{n+1}^{n+1}+\rho_{n-1}^{n-1}) \sum_m \frac{V}{\varepsilon - iV^2 - m\pi}\frac{V}{\xi + iV^2 - m\pi}. \quad (26)$$

After summing (at $V^2 \gg 1$) and taking the inverse Fourier transform we arrive at the equation

$$\dot{\rho}_n^n = V^2(\rho_{n+1}^{n+1}+\rho_{n-1}^{n-1}-2\rho_n^n). \quad (27)$$

This is the usual kinetic equation. From its solution we find that at V independent of n the total acquired energy is

$$\mathscr{E}(t) = \sum_n \omega n \rho_n^n \sim (V^2t)^{1/2} = \left(\frac{\pi E^2\langle\mu^2\rangle}{\alpha}t\right)^{1/2}. \quad (28)$$

If we sum all the terms of the type (7) of the series (6), taking into account the distribution of the populations over the levels in the band, i.e., without summation over the initial and final points, then we can show that the levels that are substantially populated are those deviating by not more than $2V^2$, i.e., by $2\pi E^2\langle\mu^2\rangle/\alpha$, for the harmonic position of the energy. The population distribution over an individual band takes the form of a Lorentz curve:

$$\rho_{n,m}^{n,m} = \rho_n^n \frac{2V^2}{\Delta_{n,m}^2 + 4V^4}. \quad (29)$$

It is easy to modify the problem of Sec. 4 to accommodate the case of two bands. Then

$$\rho_{1,m}^{1,m} = \int_{c_1} d\varepsilon \int_{c_2} d\xi \frac{V}{\varepsilon-iV^2}\frac{V}{\xi+iV^2}\frac{1}{1-q^2}\frac{V}{\varepsilon-\Delta_{1,m}-iV^2}\frac{V}{\xi-\Delta_{1,m}+iV^2}, \quad (30)$$

$$\rho_{2,m}^{2,m} = \int_{c_1} d\varepsilon \int_{c_2} d\xi \frac{V}{\varepsilon-iV^2}\frac{V}{\xi+iV^2}\frac{q}{1-q^2}\frac{V}{\varepsilon-\Delta_{2,m}-iV^2}\frac{V}{\xi-\Delta_{2,m}+iV^2},$$

where

$$q = \sum_m V^2(\varepsilon-iV^2-\pi m)^{-1}(\xi+iV^2-\pi m)^{-1} = (1-i(\varepsilon-\xi)(2V^2)^{-1})^{-1}.$$

The integrals (30) yield directly an exponential equalization of the populations, with a characteristic time $(2V^2)^{-1}$, with the stationary populations localized in the $2V^2$ vicinity: $\Delta_{1,m}\sim\Delta_{2,m}\sim 0$.

The obtained kinetic equation shows that in a multi-

level system of the band type, under conditions of weak interaction, the phase shifts of the off-diagonal elements of the density become effectively randomized. At first glance this condition contradicts the localization of the populations in small vicinities of levels with $\Delta = 0$. Actually, however, these conditions do not contradict one another. The population of a band is given according to (29) by a Lorentz curve and behaves like Δ^{-2} as $\Delta \to \infty$, while the probability amplitude tends to zero like Δ^{-1}. While the integral of Δ^{-2} converges at infinity, the integral of Δ^{-1} diverges at infinity. Thus, the phase randomization, which is determined by the interference of the probability amplitudes, receives contributions from states far from resonance, while the population receives contributions only from states close to resonances.

We emphasize that the foregoing analysis was based essentially on the hypothesis that the dipole-moment matrix elements are random and have a zero mean value. If the dipole moments correlate, then the results can change significantly. Thus, in the case of complete correlation it turns out that the acquired energy relaxes exponentially to its stationary value. This question will be considered in detail separately.

One of us (V. M. Akulin) is deeply grateful to N. V. Karlov for interest in the work.

[1)] Dyson[12] advances arguments that the Wigner distribution is incorrect for Hamiltonians and has therefore no physical realization. In our problem the submatrix of the dipole moments is not a Hamiltonian and because of the sum rule it satisfies the Wigner distribution.

[2)] We derive this distribution in a manner simpler than in[13]: $g(\mu) = \mathrm{Im}\,\mathrm{Sp}(\varepsilon - V - i\delta)^{-1}|_{\varepsilon=\mu} = \mathrm{Im}\,X_{\varepsilon}\varepsilon^{-1}|_{\varepsilon=\mu} = (2\pi u)^{-1}(4\pi u - \mu^2)^{1/2}$ at $|\mu| < (4\pi u)^{1/2}$. This is in fact the Rabi frequency distribution.

[3)] If the inequality (22) is satisfied, the answer does not depend on the concrete structure of the spectrum of an individual band.

[1]G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **46**, 403 (1964); **48**, 666 (1965) [Sov. Phys. JETP **19**, 273 (1964); **21**, 439 (1965)]; F. V. Bunkin, R. V. Karapetyan, and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. **47**, 216 (1964) [Sov. Phys. JETP **20**, 145 (1965)].

[2]A. N. Oaraevskiĭ and V. A. Savva, Kratk. Soobshch. Fiz. **7**, 50 (1970).

[3]V. I. Gorchakov and V. N. Sazonov, Zh. Eksp. Teor. Fiz. **70**, 467 (1976) [Sov. Phys. JETP **43**, 241 (1976)]; V. N. Sazonov and V. Yu. Finkel'shteĭn, Dokl. Akad. Nauk SSSR **231**, 78 (1976) [Sov. Phys. Dokl. **21**, 645 (1976)].

[4]N. R. Isenor, V. Merchant, R. S. Hallsworth, and M. C. Richardson, Can. J. Phys. **51**, 1281 (1973).

[5]R. V. Ambartsumyan, Yu. A. Gorokhov, V. S. Letokhov, G. H. Makarov, and A. A. Puretskiĭ, Zh. Eksp. Teor. Fiz. **69**, 72 (1975) [Sov. Phys. JETP **42**, 36 (1975)]; **71**, 440 (1976) [**44**, 231 (1976)]; Pis'ma Zh. Eksp. Teor. Fiz. **22**, 374 (1975) [JETP Lett. **22**, 177 (1975); **23**, 26 (1976) [**23**, 22 (1976)].

[6]V. M. Akulin, S. S. Alimpiev, N. V. Karlov, B. G. Sartakov, and E. M. Khokhlov, Pis'ma Zh. Eksp. Teor. Fiz. **25**, 428 (1977) [JETP Lett. **25**, 400 (1977)].

[7]V. M. Akulin, S. S. Alimpiev, N. V. Karlov, B. G. Sartakov, and L. A. Shelepin, Zh. Eksp. Teor. Fiz. **69**, 836 (1975) [Sov. Phys. JETP **42**, 427 (1975); **71**, 454 (1976) [**44**, 239 (1976)]; **72**, 88 (1977) [**45**, 47 (1977)].

[8]N. Blombergen, Opt. Commun. **15**, 416 (1975).

[9]V. S. Letokhov and A. A. Makarov, Kogerentnoe vozbuzhdenie mnogourovnevykh molekulyarnykh sistem v sil'nom kvazirezonansnom lazernom pole (Coherent Excitation of Multilevel Molecular Systems in a Strong Quasiresonant Field), Preprint ISAN SSSR, 1976.

[10]E. V. Shurak, Zh. Eksp. Teor. Fiz. **71**, 2039 (1976) [Sov. Phys. JETP **44**, 1070 (1976); J. Stone, M. F. Goodman, and D. A. Dows, J. Chem. Phys. **65**, 5063 (1976); S. Mukamel and J. Jorthner, Chem. Phys. Lett. **40**, 150 (1976); J. Chem. Phys. **65**, 5205 (1976); V. T. Platonenko, Pis'ma Zh. Eksp. Teor. Fiz. **25**, 52 (1977) [JETP Lett. **25**, 46 (1977)].

[11]V. M. Agronovich and V. I. Rupasov, Preprint No. 11/192, Akademgorodok, Podolskii Rayon, Moscow, 1976.

[12]F. E. Dyson, Statistical Theory of Energy Levels of Complex Systems (Russ. transl.), IIL, 1963.

[13]E. P. Wigner, Ann. Math. **65**, 203 (1957).

Translated by J. G. Adashko