phase matching (dephasing) of waves. In the case of a fixed length of an optically inhomogeneous nonlinear crystal L, the doubler efficiency first rises with the incident power, reaches a certain maximum value (for a given length of the crystal), and then falls when the pump power is increased still further. For a given pump power and given properties of the crystal there is an optimal length of the crystal $L_{\max}$ for which the doubler efficiency has its maximum (under these conditions) value $\eta_{\max}$. Under given conditions in the nonlinear crystal the maximum possible efficiency $\eta_{\max}$ rises with the pump power but this value of $\eta_{\max}$ can be obtained only by selecting the optimal length of the crystal $L_{\max}$ which varies with the pump power. The conclusion of the rise of $\eta_{\max}$ with increasing pump power ceases to be valid at powers such that the thermal self-interaction effects and diffraction-induced loss of phase matching become important.

The authors are deeply grateful to S. A. Akhmanov for valuable discussions of some of the problems.

[1] S. A. Akhmanov and R. V. Khokhlov, Kvantovaya Elektron. (Moscow) 1, 1022 (1974) [Sov. J. Quantum Electron. 4, 554 (1974)].
[2] B. V. Zhdanov, A. I. Kovrigin, and S. M. Pershin, Prib. Tekh. Eksp. No. 3, 206 (1972).
[3] A. H. Kung, J. F. Young, G. C. Bjorklund, and S. E. Harris, Phys. Rev. Lett. 29, 985 (1972).
[4] Yu. N. Karamzin and A. P. Sukhorukov, Pis'ma Zh. Eksp. Teor. Fiz. 20, 734 (1974) [JETP Lett. 20, 339 (1974)].
[5] Yu. N. Karamzin and A. P. Sukhorukov, Zh. Eksp. Teor. Fiz. 68, 834 (1975) [Sov. Phys.-JETP 41, 414 (1975)].
[6] M. N. Resenbluth, Phys. Rev. Lett. 29, 565 (1972).
[7] S. A. Akhmanov and A. S. Chirkin, Statisticheskie yavleniya v nelineĭnoĭ optike (Random Phenomena in Nonlinear Optics), Moscow State University, 1971.
[8] N. N. Filonenko, Abstracts of Papers presented at Second Intern. Conf. on Plasma Theory, Kiev, 1974, Institute of Theoretical Physics, Academy of Sciences of the Ukrainian SSR, Kiev, 1974, p. 144.
[9] N. N. Filonenko and L. P. Mel'nik, Zh. Eksp. Teor. Fiz. 69, 1183 (1975) [Sov. Phys.-JETP 42, (1975)].
[10] A. A. Galeev and R. Z. Sagdeev, Voprosy teorii plazmy (in: Problems of Plasma Theory, No. 7, Atomizdat, M., 1973, p. 3.
[11] N. Bloembergen, Nonlinear Optics, Benjamin, New York, 1965 (Russ. Transl., Mir, M., 1966).
[12] V. I. Bespalov, Izv. Vyssh. Uchebn. Zaved. Radiofiz. 9, 1117 (1966).
[13] N. N. Filenonko, Tezisy dokladov, predstavlennykh na 7-yu Vsesoyuznuyu konferentsiyu po kogerentnoĭ i nelineĭnoĭ optike (Abstracts of Papers presented at Seventh All-Union Conf. on Coherent and Nonlinear Optics), Moscow State University, 1974, p. 169.
[14] V. V. Tamoĭkin and S. M. Faĭnshteĭn, Zh. Eksp. Teor. Fiz. 62, 213 (1972) [Sov. Phys.-JETP 35, 115 (1972)].
[15] N. G. Basov, O. N. Krokhin, V. V. Pustovalov, A. A. Rupasov, V. P. Silin, G. B. Sklizkov, V. T. Tikhonchuk, and A. S. Shikanov, Zh. Eksp. Teor. Fiz. 67, 118 (1974) [Sov. Phys. -JETP 40, 61 (1975)].
[16] A. D. Piliya and V. I. Fedorov, Zh. Eksp. Teor. Fiz. 68, 987 (1975) [Sov. Phys.-JETP 41, 489 (1975)].

Translated by A. Tybulewicz

Classical heteropolar molecule in the field of circularly polarized laser radiation

V. I. Gorchakov and V. N. Sazonov

P. N. Lebedev Physics Institute, USSR Academy of Sciences, Moscow

(Submitted July 14, 1975)
Zh. Eksp. Teor. Fiz. 70, 467–476 (February 1976)

An approximate system of equations is obtained for the evolution of the state of a classical molecule. It is shown that the rotation of such a molecule has a strong influence on the buildup of radial vibrations: under certain conditions the rotation may compensate the radial vibration anharmonicity which disturbs the buildup. The dependence of the energy of this molecule on the radiation field intensity is considered; it is shown, in particular, that under certain conditions the rotation energy may be much greater than the vibration energy.

PACS numbers: 32.20.Tg

1. FORMULATION OF THE PROBLEM, DESCRIPTION OF THE MODEL, AND SOLUTION METHOD

We shall consider a diatomic heteropolar isolated molecule subjected to the field of monochromatic circularly polarized laser radiation. The question is what energy is transferred from the field to the molecule and how is this energy distributed between vibrational and rotational motion.

We shall consider this problem on the basis of the following model. We shall assume that the molecule consists of two point charged atoms. The force exerted on the atoms by the laser radiation is weak compared with the intramolecular force. Radial vibrations (vibrations of the distance between the atoms) are generally anharmonic but the amplitude of these vibrations is small compared with the equilibrium distance. The action of the laser radiation, rotation, and vibration of the molecule will all be considered ignoring the quantum effects.

The approximate nature of this model is self-evident. However, this model should not be considered just as the limiting case to which the results of a quantum investigation (not yet carried out) should reduce. The classical model can give a quantitative description of highly excited states of a molecule; moreover, it is much clearer than the quantum model and has a certain heuristic value.

We shall use a system of exact equations of motion and the averaging method (see, for example, [1]) to go over from rapidly oscillating to slowly varying unknowns. We shall obtain an approximate closed system of equations for the slow unknowns (Sec. 2) and we shall consider its solution (Sec. 3). In Sec. 4 we shall discuss the results and give some numerical estimates.

2. SYSTEM OF EQUATIONS OF MOTION

2.1. Let $r = r_1 - r_2$, $m = m_1 m_2/(m_1 + m_2)$, $e = m(e_1/m_1 - e_2/m_2)$, where $r_{1,2}$, $m_{1,2}$, and $e_{1,2}$ are, respectively, the radius vectors, masses, and charges of the first and second atoms. We shall introduce a coordinate system at rest (x, y, z) with z axis parallel to the wave vector of the radiation. Ignoring the spatial inhomogeneity, we shall describe the electric field of the laser radiation in the (x, y, z) coordinate system as follows:

$$\mathbf{E}(t) = E(\cos\psi_l, \sin\psi_l, 0),$$

where

$$\psi_l = \omega_l t + \psi_l(0)$$

is the phase of the laser field in the (x, y) plane; ω_l is the laser frequency.

We shall consider only the case when the kinetic momentum of the molecule is parallel to the radiation wave vector. We shall introduce, for the vector $\mathbf{r}$, the phase ψ_r of the rotation in the (x, y) plane; then,

$$\mathbf{r}(t) = r(\cos\psi_r, \sin\psi_r, 0).$$

In the case of free rotation, we have

$$\psi_r = \omega_r t + \psi_r(0),$$

where ω_r is the rotation frequency.

The equations of motion are as follows:

$$\left.\begin{aligned} m\ddot{r} &= -\frac{dU}{dr} + F\cos(\psi_l - \psi_r) + mr(\dot{\psi}_r)^2, \\ mr\ddot{\psi}_r &= F\sin(\psi_l - \psi_r) - 2m\dot{r}\dot{\psi}_r, \end{aligned}\right\} \qquad (1)$$

where $U = U(r)$ is the potential energy of the radial motion and $F = eE$.

2.2. The exact system (1) is difficult to investigate analytically. We have to invoke the following simplifying conditions.

The rotation frequency ω_r is much less than the characteristic vibration frequency ω_{ve}:

$$\omega_r \ll \omega_{ve}. \qquad (2)$$

The amplitude of the radial vibration a is much less than the equilibrium distance between the atoms $\mathbf{r}_e$:

$$a \ll r_e. \qquad (3)$$

We shall seek the solution of Eq. (1) in the form

$$r = r_e + a\cos(\psi_l - \psi_r + \varphi). \qquad (4)$$

Let the external force F be so weak that the vibration amplitude a, phase shift φ, and rotation frequency ω_r be relatively unaffected during the characteristic vibration period. Thus,

$$\frac{|\dot{a}|}{a}, \quad |\dot{\varphi}|, \quad \left|\frac{\dot{\omega}_r}{\omega_r}\right| \ll \omega_{ve}. \qquad (5)$$

Substituting Eq. (4) into Eq. (1) and averaging over the rapidly oscillating quantities (see[1]), we obtained the following convenient closed system of equations for the slow unknowns:

$$\dot{a} = -F\sin\varphi/2m\omega_l, \qquad (6)$$

$$\dot{\varphi} = \omega_v(a) - \omega_l + \omega_r - \frac{F\cos\varphi}{2m\omega_l a}, \qquad (7)$$

$$\dot{\omega}_r = -\frac{aF\sin\varphi}{2mr_e^2}, \qquad (8)$$

where $\omega_v(a)$ is the frequency of the radial vibrations which generally depends on the amplitude. If $a = 0$, then $\omega_v(a) = \omega_{ve}$. It follows from Eq. (3) that

$$|\omega_{ve} - \omega_v(a)| \ll \omega_{ve}.$$

In the derivation of Eqs. (6)–(8) we have confined ourselves to the case when the laser frequency differs only a little from the characteristic vibration frequency:

$$|\omega_{ve} - \omega_l| \ll \omega_{ve},$$

and whenever possible we have replaced $\omega_v(a)$ and ω_{ve} with ω_l. Moreover, we shall not distinguish between ω_{ve} and ω_l in all those cases when this is allowed. The anharmonicity becomes important when

$$|\omega_{ve} - \omega_v(a)| \gtrsim |\omega_{ve} - \omega_l|.$$

Thus, the condition (3) does not free us of the need to allow for the dependence of ω_v on a.

The vibrational-rotational state of a classical diatomic molecule is described completely by the following three quantities: $a(t)$, $\varphi(t)$, and $\omega_r(t)$. It follows from the condition (2) that the value of the rotation phase ψ_r is unimportant. Equations (6) and (8) have the integral:

$$\omega_r(t) - \omega_l \frac{a^2(t)}{2r_e^2} = c, \qquad (9)$$

which makes it possible to reduce the order of the system of equations (6)–(8).

3. MOTION OF A MOLECULE UNDER BEAT AND STEADY-STATE CONDITIONS

3.1. We shall substitute the value of $\omega_r(t)$ from Eq. (9) into Eq. (7) and we shall introduce

$$\omega_v^*(a)=\omega_v(a)+\frac{\omega_l}{2}\frac{a^2}{r_e^2}+c. \tag{10}$$

We can see that the whole influence of the rotation can be described by replacing the dependence $\omega_v(a)$ with some effective dependence $\omega_v^*(a)$.

If follows from the condition (3) that the amplitude is $a \ll r_e$ so that we shall assume

$$\omega_v(a)\approx\omega_{ve}\left(1-\rho\frac{a^2}{r_e^2}\right), \tag{11}$$

where ρ is a numerical coefficient representing the radial vibration anharmonicity. According to Eqs. (10) and (11), the role of the rotation reduces the replacement of ρ by $\rho^*=\rho-\frac{1}{2}$ and to a change in ω_{ve}.

In the quantum case, when the energy of a term is expressed in the form

$$E\approx\hbar\omega_{ve}(v+{}^1/_2)-x_e\hbar\omega_{ve}(v+{}^1/_2)^2+B_eK(K+1),$$

where $B_e=\hbar^2/2mr_e^2$, this numerical coefficient is $\rho=x_e\hbar\omega_{ve}/2B_e$.

Let us assume that initially the molecule is at rest: $a=0$ and $\omega_r=0$ [then $c=0$ in Eq. (9)]. The radial vibrations begin to build up in the laser radiation field. The radial vibration amplitude a exhibits beats from zero to $a_{\max}$. The value of $a_{\max}$ can be found as follows.

We shall introduce an action variable $I=m\omega_l a^2/2$ and rewrite the system (6)–(7) in the Hamiltonian form

$$\left.\begin{aligned}\dot{I}&=-\frac{\partial H}{\partial\varphi}=-\frac{F\sin\varphi}{(2m\omega_l)^{1/2}}\sqrt{I},\\ \dot{\varphi}&=\frac{\partial H}{\partial I}=\omega_v^*(I)-\omega_l-\frac{F\cos\varphi}{2(2m\omega_lI)^{1/2}};\end{aligned}\right\} \tag{12}$$

where

$$H=H(I,\varphi)=\int^{I}dI_1(\omega_v^*(I_1)-\omega_l)-\frac{F\cos\varphi}{(2m\omega_l)^{1/2}}\sqrt{I}.$$

Then, considering H as the integral of motion, we eliminate φ from Eq. (12). In this way we obtain the equation of motion for I in the potential form:

$$\ddot{I}=-\frac{dV(I;I(0),\varphi(0))}{dI}, \tag{13}$$

where

$$V(I;I(0),\varphi(0))=-\frac{IF^2}{4m\omega_l}$$

$$+\int_0^I dI_1(\omega_v^*(I_1)-\omega_l)\left[\int_0^{I_1}dI_2(\omega_v^*(I_2)-\omega_l)+\frac{F\cos\varphi(0)\sqrt{I(0)}}{(2m\omega_l)^{1/2}}\right]. \tag{14}$$

The oscillations of the quantity I, described by Eq. (13), correspond to beats of the amplitude a.

It is interesting to note that the potential V depends on the initial values of $I(0)$ and $\varphi(0)$ [the condition $a(t=0)=0$ will not be used yet]; moreover, V depends on the external force F and on the frequency detuning

$$\Delta\omega=\omega_{ve}-\omega_l.$$

If initially we have $a(0)=0$, we then find that $I(0)=0$ and $\dot{I}(0)=0$. We shall substitute in Eq. (14) the value

$$\omega_v^*(I)=\omega_{ve}-\frac{2\rho^*}{mr_e^2}I.$$

The dependence of V on I is shown schematically in Fig. 1. If $\rho^*\neq 0$ then, for sufficiently large values of $\nabla\omega$, the potential curve $V(I)$ has a local maximum which is located above the abscissa if $\Delta\omega>\Delta\omega_c$. We can easily show that the value of $\Delta\omega_c$ at which the curve $V(I)$ touches the abscissa from below is

$$\Delta\omega_c=\omega_l\left(\frac{27}{8}\frac{\rho^*F^2}{m^2r_e^2\omega_l^4}\right)^{1/3}.$$

When the condition $\Delta\omega=\Delta\omega_c$ is satisfied, the molecule at a beat maximum has a vibrational energy E_v equal to

$$E_v\approx\frac{ma_{\max}^2\omega_l^2}{2}=\frac{mr_e^2\omega_l^2}{2}\left(\frac{64F^2}{\rho^{*2}m^2r_e^2\omega_l^4}\right)^{1/3}. \tag{15}$$

Since

$$\omega_r(t)=\frac{\omega_l}{2}\frac{a^2(t)}{r^2}.$$

It follows that the buildup of the radial vibrations is accompanied by untwisting of the molecule. However, the rotational energy is

$$E_r=\frac{mr_e^2\omega_r^2}{2}=E_v\frac{a^2}{4r_e^2},$$

i.e., it is much lower than the vibrational energy.

3.2. The system (6)–(8) is derived ignoring the relaxation processes and it cannot describe motion for a fairly long time, particularly the attainment of steady-state conditions when $\dot{a}=\dot{\varphi}=\dot{\omega}_r=0$. We shall now introduce into our system some terms which describe the damping of the motion.

In a consistent development of the classical model we must allow for the force of radiation friction. Then, instead of Eq. (8), we obtain

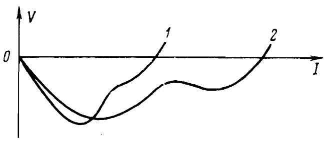

FIG. 1. Potential $V(I)$ describing beats of the amplitude a of radial vibrations; the action is $I=m\omega_l a^2/2$ [compare with Eqs. (13) and (14)]. Two curves are shown and for these curves the frequency detuning is $\Delta\omega_1<\Delta\omega_2<\Delta\omega_c$.

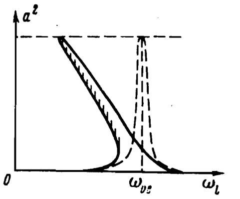

FIG. 2. Dependence of the amplitude a of radial vibrations on the laser frequency ω_l under steady-state conditions. The continuous curve is the dependence $a^2(\omega_l)$ obtained ignoring rotation for $\rho > 0$; the states shown shaded are not obtained because of their instability. The dashed curve is the dependence $a^2(\omega_l)$ obtained allowing for rotation in the case of exact compensation of the anharmonicity by rotation, e.e., in the case when $\rho^* = \rho - \gamma_v/{}^2\gamma_r = 0$.

$$\dot{\omega}_r = -\tau\left(\omega_r^2 + \frac{3}{2}\omega_l^2\frac{a^2}{r_e^2}\right)\omega_r - \frac{Fa\sin\varphi}{2mr_e^2},$$

where $\tau = 2e^2/3mc^3$. The equation for $\dot{a}$ becomes

$$\dot{a} = -\frac{1}{2}\tau\omega_l^2 a - \frac{F\sin\varphi}{2m\omega_l};$$

the equation for $\dot{\varphi}$ remains unchanged.

Under steady-state conditions the relationship between ω_r and a is

$$\omega_r \approx \omega_l (a^2/2r_e^2)^{1/3}.$$

The effective frequency dependence becomes

$$\omega_v^*(a) = \omega_{ve} - \omega_{ve}\rho\frac{a^2}{r_e^2} + \omega_l\left(\frac{a^2}{2r_e^2}\right)^{1/3} \approx \omega_{ve} + \omega_{vc}\left(\frac{a^2}{2r_e^2}\right)^{1/3}.$$

We shall not consider this case in detail but simply note that under steady-state conditions a large fraction of the energy of a molecule is carried by the rotational motion:

$$\frac{E_r}{E_v} = \frac{1}{2}\left(\frac{2r_e^2}{a^2}\right)^{1/3} \gg 1.$$

3.3. In a sufficiently dense medium the damping (relaxation) of motion can occur because of collisions. We shall allow for this possibility phenomenologically by introducing friction coefficients γ_v and γ_r. We shall write

$$\left.\begin{aligned} \dot{a} &= -\gamma_v a - \frac{F\sin\varphi}{2m\omega_l}, \\ \dot{\omega}_r &= -2\gamma_r\omega_r - \frac{Fa\sin\varphi}{2mr_e^2}. \end{aligned}\right\} \tag{16}$$

Under steady-state conditions we have

$$\omega_r = \omega_l\frac{\gamma_v}{2\gamma_r}\frac{a^2}{r_e^2},$$

where

$$\omega_v^*(a) = \omega_{ve}\left(1 - \rho^*\frac{a^2}{r_e^2}\right), \quad \rho^* = \rho - \frac{\gamma_v}{2\gamma_r}. \tag{17}$$

The dependence of the steady-state vibration amplitude on the frequency of the external force is given by the resonance curve $a^2(\omega_l)$. In the case of molecules we usually have $\rho > 0$, i.e., the resonance curves plotted ignoring rotation are deflected to the left in the direction of lower frequencies. An example of such a curve is given in Fig. 2 (for details of the properties of the resonance curves the reader is directed to[1,2]).

As shown above, allowance for the rotation under steady-state conditions reduces to the substitution $\rho \to \rho^*$ [see Eq. (17)]. It should be noted that if $\gamma_v/2\gamma_r = \rho$, then $\rho^* = 0$. This means that if $\gamma_v/2\gamma_r = \rho$, the rotation of a molecule compensates exactly its anharmonicity.

3.4. We shall now consider the energy of a molecule under steady-state conditions.

If $\rho^* \sim 1$, the vibrational energy E_v is much higher than the rotational energy E_r. The dependence of E_v on the frequency ω_l and the intensity $I = c|E|^2/4\pi$ of the laser radiation field can be found by eliminating φ from Eqs. (7) and (16). This gives the equation

$$\frac{E_v}{E_m}\left[\left(\frac{\Delta\omega}{\omega_l} - \rho^*\frac{E_v}{E_m}\right)^2 + \frac{\gamma_v^2}{\omega_l^2}\right] = \frac{I}{I_m}, \tag{18}$$

where

$$E_m = \frac{mr_e^2\omega_l^2}{2}, \qquad I_m = \frac{m^2r_e^2\omega_l^4 c}{\pi e^2}.$$

E_m is the characteristic molecular energy and I_m is the characteristic intensity at which the electric field in the laser wave becomes of the same order of magnitude as the internal molecular field.

For a given value of I, the dependence of E_v on ω_l is represented by curves such as those plotted in Fig. 2 because $E_v \propto a^2$. We shall consider the dependence of E_v on I for a given laser radiation frequency. Using Eq. (18), we can show that the dependence $E_v(I)$ is continuous if $\Delta\omega < \sqrt{3}\gamma_v$. If $\Delta\omega > \sqrt{3}\gamma_v$, the dependence $E_v(I)$ may become discontinuous for $\rho^* \neq 0$. Let us assume that the intensity I varies slowly with time. Then, at $I = I_{c1}$ or $I = I_{c2}$ the vibrations break down and the vibrational energy changes abruptly (Fig. 3). The critical field I_{c1} (or I_{c2}) at which there is a downward (or upward) sudden change is

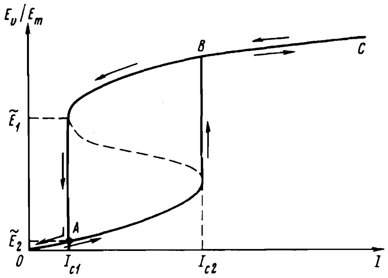

FIG. 3. Dependence of the vibrational energy E_v on the laser intensity I under steady-state conditions. When the intensity I is increased from 0, the vibrational energy varies along the curve OABC with an abrupt upward jump at $I = I_{c2}$. When I is reduced, the energy varies along CBAO with an abrupt downward jump at $I = I_{c1} < I_{c2}$.

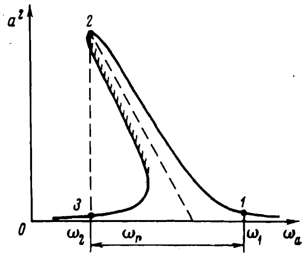

FIG. 4. Amplitude of radial vibrations $a(t)$ and effective frequency $\omega_a(t) = \omega_l - \omega_r(t)$. A molecule undergoes a transition from an initial state 1 with $\omega_r = 0$ and $\omega_a = \omega_l$ to a state 2 in which $\omega_r > 0$ and $\omega_a = \omega_l - \omega_r = \omega_2$. State 3 is assumed by a molecule which cannot rotate and is excited by a frequency $\omega_l = \omega_a = \omega_2$.

$$I_{c1,c2} = I_m \tilde{E}_{1,2}\left(\frac{\gamma_v^2}{\omega_l^2} + \left(\frac{\Delta\omega}{\omega_l} - \rho^* \tilde{E}_{1,2}\right)^2\right),$$

where

$$\tilde{E}_{1,2} = \frac{2\Delta\omega \pm ((\Delta\omega)^2 - 3\gamma_v^2)^{1/2}}{3\rho^*\omega_l}.$$

If, for example, $(\Delta\omega)^2 \approx 3\gamma_v^2$ then $\tilde{E}_1 \approx \tilde{E}_2 = 2\Delta\omega/3\rho^*\omega_l$ and

$$I_{c1,c2} \approx I_m \frac{8\sqrt{3}}{9\rho^*}\left(\frac{\gamma_v}{\omega_{ve}}\right)^3. \qquad (19)$$

4. DISCUSSION

Why is the buildup of radial vibrations of a molecule in the field of laser radiation necessarily accompanied by untwisting? Why, in spite of the condition $\omega_r \ll \omega_{ve}$ and lack of allowance for the centripetal force, does the rotation of a molecule have a strong influence on the buildup of radial vibrations?

The answer to the first question is easiest to obtain on the basis of quantum considerations. Radiation-induced untwisting appears because the absorption of circularly polarized photons results in an acquisition of an energy $\hbar\omega_l$ and also of a torque $\hbar$ per each (dipole-) absorbed photon. Consequently, a molecule begins to rotate in the same direction as the vector of the radiation electric (and magnetic) field.

The answer to the second question is as follows. So far, we have described the influence of the rotation on the amplitude of the steady-state radial vibrations by the the substitution $\rho \to \rho^*$. However, in some cases it is more convenient to adopt a different description. It is clear from Eq. (1) that radial vibrations are excited by a force whose phase is $\psi_l - \psi_r$. The corresponding frequency

$$\omega_a = \frac{d}{dt}(\psi_l - \psi_r) = \omega_l - \omega_r$$

will be called the effective frequency. This effective frequency is the frequency of an external laser field "experienced" by an observer rotating together with the molecule. Let $\omega_l > \omega_{ve}$ and let the frequency ω_r rise so slowly that the amplitude of radial vibrations can follow the effective frequency $\omega_a(t) = \omega_l - \omega_r(t)$ which "creeps" slowly to the left. As ω_a decreases, the amplitude a can grow considerably. In other words, in the process of untwisting of a molecule the point in Fig. 4 representing the state of the molecule may travel from position 1 to position 2. It should be noted that it is not possible to build up the molecular vibrations to the amplitude represented by point 2 if the frequency $\omega_a = \omega_l - \omega_r$ is independent of time, i.e., if the molecule is rigidly fixed and does not rotate. In the case we find that even if the laser frequency coincides with ω_2, the buildup from the state of rest gives a steady-state position 3 (Fig. 4).

It should be noted that in fact the time for a change in ω_r and the time for a to attain its steady-state value are of the same order of magnitude. Therefore, Fig. 4 is only illustrative. The buildup of vibrations should be described by the system of equations (6)–(8) or the system (7), (16). In particular, it follows from Eq. (16) that if $\rho = \gamma_v/2\gamma_r$, and the laser frequency is $\omega_l = \omega_{ve}$, the steady-state rotation frequency ω_r is such that the effective frequency $\omega_a = \omega_l - \omega_r$ corresponds exactly to the resonance curve maximum.

We shall now obtain some numerical estimates which are applicable to a typical heteropolar molecule HCl. The parameters of this molecule are[1)]:

$$e \approx 4.8\cdot 10^{-10}\ \text{cgs esu} \quad m = 1.63\cdot 10^{-24}\ \text{g}; \quad r_e = 1.29\ \text{Å};$$
$$\hbar\omega_{ve} = 2886\ \text{cm}^{-1}; \quad B_e = 10.3\ \text{cm}^{-1}; \quad x_e = 0.0172.$$

Then

$$\rho = x_e \frac{\hbar\omega_{ve}}{2B_e} = 2.41; \quad E_m = 24.6\ \text{eV}; \quad I_m = 1.6\cdot 10^{17}\ \text{W/cm}^2.$$

According to Eq. (15), the maximum energy of a molecule under beat conditions is

$$E_{v\,\max} = E_m\left(\frac{256}{\rho^{*2}}\frac{I}{I_m}\right)^{1/3} = 84.4\left(\frac{I\ \text{W/cm}^2}{10^{17}}\right)^{1/3}\ \text{eV}.$$

The classical model can be used to describe vibrations only if $E_{v\,\max} \gg \hbar\omega_{ve} = 0.36$ eV, which is true if $I \geqslant 10^{11}$ wt/cm^2. In principle, such intensity is attainable if only for a focused laser beam.

In the case of molecules with smaller values of $\hbar\omega_{ve}$ the classical theory becomes valid at much lower intensities.

In the model with friction we usually have $\gamma_v \approx 10^{-5}\omega_{ve}$; however, even if we assume that $\gamma_v \approx 10^{-4}\omega_{ve}$, it follows from Eq. (19) that $I_{c1,c2} \lesssim 10^5$ W/cm^2. Thus, the vibrations may collapse at the intensities at which the classical theory is applicable.

Let us consider whether the conditions (2), (3), and (5) are satisfied in real situations. The frequency ω_{ve} corresponds, in the quantum language, to the characteristic frequency of transitions between vibrational levels; the frequency ω_r corresponds to purely rotational transitions. If the temperature is not too high, we have $\omega_{ve} = (30-100)\,\omega_r$ and the condition (2) is satisfied well. The condition (3) is also satisfied well as long as the vibrational energy does not approach the dissociation energy. It follows from the system (6)–(8) that the condition (5) can be reduced to

$$I \ll I_m \frac{a^2}{r^2} = 1.6 \cdot 10^{17} \frac{a^2}{r^2}\ \mathrm{W/cm^2}.$$

If $I = 10^{12}$ W/cm^2, then the condition (5) is satisifed even for $a/r_e \approx 10^{-2}$.

Clearly, the classical description of rotation is fully permissible under normal conditions since the rotational quantum numbers of the molecules are $j = 10-30$. The only serious objection against the classical model is related to the requirement $v \gg 1$, where v is the vibrational number (under normal conditions $v = 0$, 1, or 2). Therefore, it would be interesting to determine how the above relationships are affected by allowance for the quantum effect. In this connection it is interesting to consider the results obtained in[3–15]; in particular, it is pointed out in[15] that the probability of excitation of a quantum oscillator is frequently governed by the work of the external field done on corresponding classical oscillator.

The thermal rotation of a molecule governs the initial value of $\omega_r(0)$ in our model. If, in the process of buildup of radial vibrations and untwisting, the angular velocity changes only slightly compared with $\omega_r(0)$, the radiation-induced untwisting is negligible. However, under real conditions, the molecule may absorb 30–50 quanta before dissociation so that its rotational quantum number has a correction Δj, which is not less than[2)] 30–50, whereas the thermal value is $j = 10-30$. Thus, radiation-induced untwisting of a molecule cannot be ignored compared with its thermal rotation.

In conclusion, we shall point out that in the model with relaxation there may be conditions under which the ratio γ_v/γ_r depends strongly on pressure. For example, this may happen when the rotational relaxation is due to collisions, whereas the relaxation of radial vibrations is not only due to collisions but also due to an additional friction mechanism. Then, the reduction in pressure causes molecules to untwist to high angular velocities; the amplitude of the steady-state vibrations rises along the resonance curve (Fig. 4). Variation of the pressure alters $\rho^* = \rho - \gamma_v/2\gamma_r$ so that we can obtain $\rho^* = 0$, i.e., we can compensate the radial vibration anharmonicity.

In this connection it is interesting to point out that, according to[16], the yield of the dissociation reaction of the SF$_6$ molecules (and of some other molecules) in the field of CO$_2$ laser radiation increases when the pressure is reduced. It is possible that also in the case of polyatomic molecules the rotation compensates (to a larger or smaller degree) the anharmonicity, as shown here for diatomic molecules. If this is correct, the probability of dissociation should have a maximum at some specific pressure.

The authors are grateful to V. S. Letokhov for suggesting the subject, G. A. Askar'yan for valuable critical comments, and to participants of a seminar headed by V. L. Ginzburg for discussions.

[1)] The energy of a vibrational quantum $\hbar\omega_{ve}$ and the rotational constants are measured in units of cm^{-1}. The dimensions of energy can be restored if we multiply by $2\pi\hbar c = 1.24 \times 10^{-4}$ eV·cm.

[2)] Under certain conditions the correction Δj is much higher than the correction to the vibrational number v because in case of spontaneous emission of a quantum of frequency $\omega \approx \omega_{ve}$ the number v necessarily decreases by unity, whereas j is equally likely to decrease or increase.

[1] N. N. Bogolyubov and Yu. A. Mitropol'skiĭ, Asimptoticheskie metody v teorii nelineĭnykh Kolebaniĭ, Fizmatgiz, M, m 1963 (Asymptotic Methods in the Theory of Nonlinear Oscillations, Gordon and Breach, New York, 1964).

[2] L. D. Landau and E. M. Lifshitz, Mekanika, Nauka, M., 1973 (Mechanics, 2nd ed., Pergamon Press, Oxford, 1969).

[3] G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **46**, 403 (1964); **48**, 666 (1965) [Sov. Phys.-JETP **19**, 273 (1964); **21**, 439 (1965)].

[4] F. V. Bunkin, R. V. Karapetyan, and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. **47**, 216 (1964) [Sov. Phys.-JETP **20**, 145 (1965)].

[5] A. M. Perelomov and V. S. Popov, Teor. Mat. Fiz. **1**, 360 (1969).

[6] N. D. Artamonova, V. T. Platonenko, and R. V. Khokhlov, Zh. Eksp. Teor. Fiz. **58**, 2195 (1970) [Sov. Phys.-JETP **31**, 1185 (1970)].

[7] Yu. V. Afanas'ev, E. M. Belenov, E. P. Mrkin, and I. A. Poluěktov, Pis'ma Zh. Eksp. Teor. Fiz. **13**, 462 (1971) [JETP Lett. **13**, 331 (1971)].

[8] G. J. Pert, IEEE J. Quantum Electron. **QE-9**, 435 (1973).

[9] Ya. B. Zel'dovich, Usp. Fiz. Nauk **110**, 138 (1973) [Sov. Phys.-Usp. **16**, 427 (1973)].

[10] P. A. Braun and A. N. Petelin, Zh. Eksp. Teor. Fiz. **66**, 1581 (1974) [Sov. Phys.-JETP **39**, 775 (1974)].

[11] W. R. Salzman, Phys. Rev. A **10**, 461 (1974).

[12] R. Guccione-Gush and H. P. Gush, Phys. Ref. A **10**, 1474 (1974).

[13] I. G. Svin'in, Teor. Mat. Fiz. **22**, 97 (1975).

[14] V. N. Lugovoĭ and V. N. Strel'tsov, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 442 (1975) [JETP Lett. **21**, 202 (1975)].

[15] G. A. Askar'yan and V. A. Namiot, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 646 (1975) [JETP Lett. **21**, 305 (1975)].

[16] R. V. Ambartsumyan, Yu. A. Gorokhov, V. S. Letpkhov, and G. N. Makarov, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 375 (1975) [JETP Lett. **21**, 171 (1975)].

Translated by A. Tybulewicz