COLLISIONLESS LASER EXCITATION OF MOLECULAR VIBRATIONAL TRANSITIONS WITH A COMPLICATED ROTATIONAL STRUCTURE

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The collisionless excitation of the vibrational transitions in slightly-asymmetric-top molecules in the field of a spectrally limited laser pulse is studied analytically. Formulas for the probability of vibrational photoexcitation are derived on the basis of Schrodinger's equation for the probability amplitudes of the vibrational-rotational states and the statistical description of the complicated structure of the spectrum of molecular absorption bands. It is shown that there exist three regimes of interaction of the molecules with the field. These regimes differ by the dependence of the excitation probability on the radiation intensity. The efficiency of excitation of ozone molecules in the presence of collisions is compared with that under collisionless conditions.

When short pulses of light propagate in air the radiation can interact with the molecules under collisionless conditions. Thus already at atmospheric pressure the average time between the gas-kinetic collisions of air molecules is greater than the duration of picosecond laser pulses. There are a large number of works on the collisional interaction of IR laser radiation with molecules. These works pertain primarily to multiphoton excitation and dissociation of large molecules having a vibrational quasi continuum (see, for example, Refs. 1 and 2). At the same time IR excitation of small molecules, such as the atmospheric gases (H_2O, CO_2, O_3, SO_2, NO_2, and others), has not been adequately studied. The specific nature of their spectrum is determined by the low molecular symmetry: there is no quasicontinuum and vibrational-rotational (VR) fine splitting, which makes it impossible to represent the lower vibrational levels in the form of "bands".[2] In modeling photoinduced processes in small molecules each separate vibrational transition can be represented as a collection of two-level VR transitions. In Refs. 3 and 4 the photoexcitation of the vibrations of triatomic molecules was analyzed, taking into account the rotational structure of the molecules, for absorption in the presence of collisions.

The purpose of this work is to investigate analytically collisionless pulsed laser excitation of vibrational transitions with a complicated rotational structure of the spectrum, characteristic for asymmetric-top atmospheric molecules. The analysis is performed on the basis of Schrodinger's equation for the probability amplitudes and a statistical approach to the description of complicated VR spectra.[4]

1. We shall first study the interaction of pulsed IR radiation with separate VR transitions of the vibrational band $|0\rangle - |1\rangle$. We shall write the intensity of the electric field of the light wave as $E(t) = \varepsilon f(t) \cdot \cos\omega t$ and we shall assume that the spectral width of the envelope of the pulse $f(t)$ is much smaller than the carrying frequency ω. The equations for the probability amplitudes of a nondegenerate two-level system $|a\rangle - |b\rangle$ have the following form in the resonance approximation:[5,6]

$$\dot{a} = iV_{ab} f(t) b\, e^{-i\delta t}$$

$$\dot{b} = iV_{ab}^{ab} f(t) a\, e^{i\delta t} \qquad (1)$$

where $\delta = \omega_{ab} - \omega$ is the detuning of the frequency of the transition $|a\rangle - |b\rangle$ from the radiation frequency; $V_{ab} = \vec{a}_{ab}\vec{\varepsilon} / 2h$ is the field-induced broadening; and, $\vec{a}_{ab}$ is the matrix element of the transition dipole moment (in what follows simply the transition dipole moment).

The solution of Eqs. (1) is strongly dependent on the form of the function $f(t)$. If the field is switched on "fast" the populations (i.e., $|a|^2$ and $|b|^2$) oscillate in time, and if the field is switched on "slowly" the populations can be assumed to be constant.[6] A general solution of Eqs. (1) for the envelope $f(t) = \frac{1}{2}(1 + \text{th}\, t/2T)$, where T is the characteristic time over which the field is switched on, was obtained in Ref. 7. It was shown that the field is switched on rapidly if $|\delta|T/2 \ll 1$, and "slowly" in the opposite case. We stress that in order to calculate correctly the excitation of the system of VR transitions, forming the

vibrational transition and having different frequency detunings from the radiation frequency, the general expressions for a and b, i.e., with arbitrary ratio of δ and T, must be used. Among the pulsed envelopes of the field only the solution for $f(t)$ in the form of a hyperbolic secant is known.[8] We point out, however, that in the case of spectrally limited (see Ref. 9) laser pulses it can be assumed, without introducing a significant error, that the contribution of VR lines lying outside the spectrum of the pulse to the excitation of the vibrational transition is small. For absorption lines within the spectral contour of the radiation δ may be set equal to zero. This makes it possible to solve Eqs. (1) for an arbitrary envelope $f(t)$.[6,8] If the radiation is switched on and off at $t = \mp\omega$ and the initial conditions are such that $a(-\infty) = 1$, $b(-\infty) = 0$, then the probability of excitation of the state $|b\rangle$ after the pulse terminates is given by

$$|b(\infty)|^2 = \sin^2(V_{ab}S), \quad S = \int_{-\infty}^{+\infty} f(t)dt, \tag{2}$$

i.e., the probability of excitation oscillates with a frequency proportional to the field-induced broadening and the area of the pulse. We note that the area of the pulse is inversely proportional to the width of the spectrum of the pulse.

When there is no external magnetic field the real VR levels of molecules are $(2J + 1)$ –fold degenerate with respect to the magnetic quantum number M. This means that the dipole moment vector $\vec{d}_i$ of the i-th VR transition has $2J + 1$ different projections on a distinguished direction in space. For simplicity we shall confine our attention to the case of radiation polarized linearly along the z-axis of the laboratory coordinate system and we shall study VR transitions in the parallel ($\Delta K = 0$) bands of slightly-prolate-asymmetric-top molecules (for example, the ν_3 bands of the molecules O_3 and SO_3). For the projections of $\vec{d}_i$ on the z-axis we have $\Delta M = 0$ and (see Ref. 10)

$$d_{iz} = d_i \frac{M}{J} = d_i\cos\varphi, \quad d_i \approx d_{01}\frac{K}{J};$$

$$d_{iz} = d_i\sqrt{1 - \left(\frac{M}{J}\right)^2} = d_i\sin\varphi,$$

$$d_i \approx \frac{d_{01}}{2}\sqrt{1 - \left(\frac{K}{J}\right)^2}, \tag{3}$$

$$d_i = |\vec{d}_i|, M = -J, -J + 1, \ldots, J; \; K = 0, 1, \ldots, J.$$

The index i enumerates the transitions $|0, J, K\rangle - |1, J + \Delta J, K + \Delta K\rangle$ with different J and K (J is the total angular momentum quanta number, K is the projection of J on the axis of the top). The quantity d_{01} is the dipole moment of the vibrational transition $|0\rangle - |1\rangle$. In writing Eq. (3) we assumed for simplicity that $J \geq 1$ and that the top was approximately symmetric. We note that when $J \geq 1$ it is also possible to transfer to a quasiclassical description of the rotational notion, i.e., the distribution of the orientations of J can be assumed to continuous with $0 \leq \varphi \leq \pi$. Taking into account the M degeneracy the probability of excitation of the i-th VR transition will assume the form

$$\omega_i = \frac{1}{2J + 1}\sum_{M=-J}^{J}\sin^2\left(\frac{d_{i2}\varepsilon}{2\hbar}S\right), \quad \varepsilon = |\varepsilon| \tag{4}$$

Using the quasiclassical approximation, replacing in Eq. (4) the summation by integration, and substituting Eq. (3) we obtain:

$$\omega_i = \frac{1}{2}\left[1 - y(z_i)\right];$$

$$y(z_i) = \begin{cases} \sin z_i/z_i, & \text{Q-branch} \\ \frac{\pi}{2}\vec{E}_1(z_i), & \text{P-, R-branch} \end{cases} \tag{5}$$

where $z_i = d_i\varepsilon S/h$ and $\vec{E}_1(x) = \frac{1}{\pi}\int_0^{\pi}\sin(\theta - x\sin\theta)d\theta$ is the first-order Weber function.[11] One can see from Eq. (7) that the M degeneracy of the VR lines causes the oscillations of the probability ω_i to decay as the parameter z_i increases. In addition, as follows from the asymptotic behavior, $\vec{E}_1(x) \sim x^{-1/2}\sin\left(x - \frac{3\pi}{4}\right)$ for $x \gg 1$, the decay is weaker in the P and R branches than in the Q-branch. The effect of degeneracy on the oscillations of the populations of the VR levels of the molecules BCl_3 and SF_6 was confirmed experimentally in Ref. 12.

2. We obtain the probability of excitation of the vibrational transition $|0\rangle - |1\rangle$ by summing the probabilities ω_I for all VR transitions falling within the width Ω of the radiation spectrum:

$$\omega_1 = \sum_i q_i\omega_i, \tag{6}$$

where q_i is the rotational occupation factor of the bottom VR level. Assuming that the parameters q_i, d_i, and ω_i of the absorption lines are statistically distributed over the interval $\omega \pm \Omega/2$, we have[3]

$$\omega_1 = N\int q_i\omega_i p_\omega(q_i)p_\omega(d_i)p(\omega_i)dq_i d(d_i)d\omega_i, \tag{7}$$

where N is the number of lines in the $|0\rangle - |1\rangle$ band in the region $\omega \pm \Omega/2$; $p_\omega(q_i)$, $p_\omega(d_i)$, $p(\omega_i)$ are the distribution functions of q_i, d_i, and ω_i. The index ω

indicates that the distribution function is frequency dependent (i.e., the distribution function is different in different sections of the band). Further, for simplicity we shall assume that the molecular band is limited by the width Δ and that the distribution of the parameters of the lines within the region of overlapping of the spectrum of the band and the spectrum of the radiation Ω^* is spectrally uniform, i.e., $p(\omega_i) = (\Omega^*)^{-1}$; $p_\omega(q_i) = p(q_i)$; $p_\omega(d_i) = p(d_i)$. In this case $N = \Omega^*/L$, where L is the average separation between the lines. Substituting Eq. (5) transforms the expression (7) into the form

$$\omega_1 = U\left[1 - \sum_{\alpha=P,Q,R} \xi_\alpha \int_{d^\alpha_{min}}^{d^\alpha_{max}} y_\alpha(z_1) p_\alpha(d_i)\, d(d_i)\right] \tag{8}$$

$$U = \frac{q}{2L}\Omega^*,$$

$$\Omega^* = \begin{cases} \Omega, & |\delta_0| \le \dfrac{\Delta - \Omega}{2}, \quad \Delta \ge \Omega, \\ \Delta, & |\delta_0| \le \dfrac{\Delta - \Omega}{2}, \quad \Delta \le \Omega, \\ \dfrac{\Delta + \Omega}{2} - |\delta_0|, & \dfrac{\Omega - \Delta}{2} < |\delta_0| \le \dfrac{\Omega + \Delta}{2}, \\ 0, & |\delta_0| > \dfrac{\Omega + \Delta}{2}, \end{cases}$$

where δ_0 is the frequency detuning of the center of the band from the radiation frequency and q is the average value of the factor q_i. The quantity U is proportional to the region of overlapping of the spectrum of the pulse and the band (Fig. 1). Estimates showed that taking into account the Doppler broadening of the VR lines for $\Delta\omega_D \ll \Omega, \Delta$ ($\Delta\omega_D$ is the Doppler linewidth) does not significantly change Ω^*. The summation in Eq. (8) is performed over the index denoting the rotational branch of the band; $(d^\alpha_{min}, d^\alpha_{max})$ is the spread in the values of d_1; and, ξ_α is the relative number of lines belonging to the branch α in the region of overlapping of Ω and Δ. We note that $\sum_\alpha = 1$.

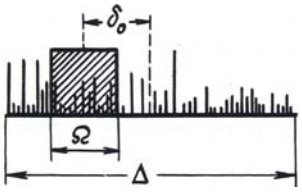

FIG. 1. Schematic diagram of the overlapping of the radiation spectrum, and the band spectrum for the calculation of the function U (see Eq. (8)).

We shall perform the integration in Eq. (8) for two limiting cases; "narrow" and "wide" distribution $p(d_i)$ (see Fig. 2). If within the distribution $p(d_i)$ of characteristic width Δd the function $y(d_j)$ does not change much, then in the interval (d_{min}, d_{max}) $p(d_i)$ can be assumed to be "narrow". With the help of Eq. (5) it can be easily shown that the conditions for the distribution $p(d_i)$ to be narrow have the form

$$|z_0 - x_n| \ll \eta, \quad \frac{\Delta d}{d_0} \ll \frac{\eta}{z_0}; \tag{9}$$

where d_0 is the most probable value of d_i and η and x_n are the half-period of the oscillations and the coordinates of the successive extrema of the function $y(z_i)$. In particular, for $y(z_i) = \sin z_i / z_i$ we have $\eta \approx \pi$, $x_n = 0$, 4.4934, and 7.7253, etc.[11] The sufficient condition for $p(d_i)$ to be "wide" on the interval (d_{min}, d_{max}) is

$$\frac{\Delta d}{d_0} \gg \frac{\eta}{z_0}. \tag{10}$$

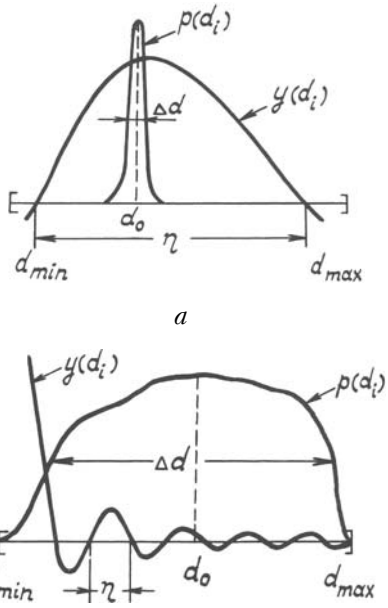

FIG. 2. The relation of the functions $y(d_i)$ and $p(d_i)$ for the cases of "narrow" (a) and "wide" (b) distributions $p(d_i)$.

It is easy to see that the conditions for a "narrow" distribution (9) are more easily satisfied for small values of z_0 while the condition for a "wide" distribution (10) is morel easily satisfied for large values of z_0.

In the case of a "narrow" distribution $p(d_i)$ ω_1 does not depend on the spread of d_i. The decay of the oscillations of ω_1 is determined solely by the parameter $z_0 = d_0\, \varepsilon\, S/h$:

$$\omega_1 = U\left[1 - \sum_\alpha \xi_\alpha y_\alpha(z_0)\right]. \tag{11}$$

Using the expansions $\sin z_0/z_0 \approx 1 - z_0^2/6$ and $\vec{E}_1(z_0) \approx \frac{2}{\pi}(1 - z_0^2/3)$ for $z_0 \ll 1$ (the weak-field regime), we obtain the following law, quadratic in ε, for the change in the probability ω_1:

$$\omega_1 = \frac{U}{6}\left[\xi_Q + 2(\xi_P + \xi_R)\right] z_0^2. \quad (12)$$

For $z_0 \gg 1$ (the strong-field regime) the quantity in Eq. (11) depending periodically on ε becomes small for all branches of the vibrational band:

$$\omega_1 = U\left[1 - \xi_Q \frac{\sin z_0}{z_0} - (\xi_P + \xi_R)\sqrt{\frac{\pi}{2z_0}}\cos\left(z_0 - \frac{\pi}{4}\right)\right] \quad (13)$$

In this case $\omega_1 \approx U$, i.e., the vibrational excitation is determined solely by the overlap function of the radiation and band spectra and does not depend on the field strength and the dipole moments of the VR transitions. The decay of the oscillations of ω_1 in the Q-branch is proportional to z_0, i.e., εS.

If the distribution $p(d_i)$ is wide, then within its width Δd we can set $p(d_i) = \Delta d^{-1}$ for other values of d_i. Carrying out the integration in Eq. (8) we obtain

$$\omega_1 = U\left\{1 - \xi_Q \frac{\hbar}{\Delta d \varepsilon S}\left[Si(z_+^0) - Si(z_-^0)\right] - \right.$$

$$-\frac{1}{\Delta d}\sum_{\alpha=P,R}\xi_\alpha\left[d_+^\alpha\, {}_2F_3\left(1, \frac{1}{2}; \frac{3}{2}, \frac{3}{2}, \frac{1}{2}; -\left(\frac{z_+^\alpha}{2}\right)^2\right) - \right.$$

$$\left.\left. - d_-^\alpha\, {}_2F_3\left(1, \frac{1}{2}; \frac{3}{2}, \frac{3}{2}, \frac{1}{2}; -\left(\frac{z_-^\alpha}{2}\right)^2\right)\right]\right\}, \quad (14)$$

where $z_\pm^\alpha = (d_0^\alpha \pm \Delta d^\alpha/2)ES/\hbar$; $Si(x)$ is the sine integral;[11] and, ${}_2F_3\left(1, \frac{1}{2}, \frac{3}{2}, \frac{3}{2}, \frac{1}{2}, -x\right)$ is the generalized hypergeometric function.[13] As follows from Eq. (14) the spread in the values of d_i in the interval $\omega \pm \Omega/2$ results in additional decay of the oscillations of ω_1. We shall analyze the formula (14) for the case of the excitation of the Q-branch of the vibrational band, i.e., in what follows we shall set $\xi = 1$ and we shall drop the index Q. In the case $z_- \ll 1$ and $z_+ \ll 1$ we find with the help of the expansion $Si(x) \approx x - x^3/18$ for $x \ll 1$, like in the case of the "narrow" distribution, that the probability ω_1 increases quadratically as a function of the field strength:

$$\omega_1 = \frac{U}{18}\left(\frac{\varepsilon S}{\hbar}\right)^2\left[3d_0^2 + \left(\frac{\Delta d}{2}\right)^2\right]. \quad (15)$$

Using the expansion $Si(x) \approx \pi/2 - \cos x/x$ for $x \gg 1$ and assuming that $z_- \gg 1$, $z_+ \gg 1$, and $z_0 \gg 1$ we find that the oscillations of ω_1 have a high frequency and a small amplitude:

$$\omega_1 = U\left[1 - \left(\frac{\hbar}{\varepsilon S}\right)^2 \frac{(d_+\cos z_- - d_-\cos z_+)}{\Delta d\, d_-\, d_+}\right] \quad (16)$$

i.e., it can be assumed that $\omega_1 \approx U$ as in the case of the "narrow" distribution. The decay of the oscillations of ω_1, however, is proportional not to εS but to $(\varepsilon S)^2$.

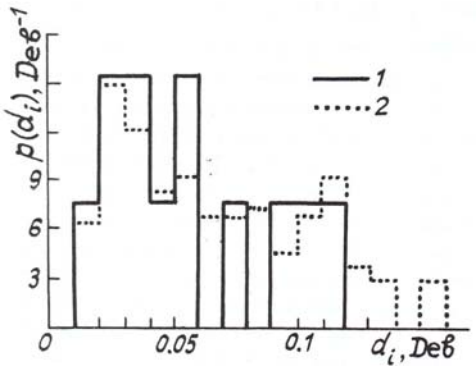

FIG. 3. The distribution functions $p(d_i)$ for the VR lines of the (000)–(0017 band of O_3: 1) lines in the range 1037, 2674–1037, and 6008 cm^{-1} (τ_p = 100 ps); 2) 109 lines in the range 1035, 7674–1039, and 1008 cm^{-1} (τ_p = 10 ps). The sampling interval is 0.01 D wide.

We shall make specific estimates for the conditions of existence of the regimes of interaction of the molecules with the field studied above for the example of the excitation of the Q-branch of the (000)–(001) band of O_3 by 9P(30) radiation from a CO_2 laser ($\omega/\pi c$ = 1037.4341 cm^{-1}). Existing picosecond laser systems for the 9-μm and 10-μm range give a radiation pulse width $\tau_p \gtrsim 10$ ps.[14] If τ_p = 100 ps, then the spectral width $\Omega/2\pi c \approx 0.33$ cm^{-1} covers 13 absorption lines of O_3, of which 11 belong to the Q-branch.[15] For τ_p = 10 ps the radiation spectrum now contains 109 lines, of which 95 belong to the Q-branch. The distribution functions $p(d_i)$, calculated for these cases based on the data of Ref. 15, are presented in Fig. 3. The effective parameters d_0 and Δd for distributions in the form of a histogram can be calculated using the formulas

$$d_0 = \sum_{k=1}^{M} d_k p(d_k) D_k, \quad \Delta d = p^{-1}(d_0) = \frac{1}{M}\sum_{k=1}^{M} p(d_k), \quad (17)$$

where D_k is the width of the k-th interval of the sample and M is the total number of such intervals. Using Eq. (7), we obtain $d_0 \approx 0.062\ D$ and $\Delta d \approx 0.12\ D$ for distribution 1 and $d_0 \approx 0.067\ D$ and $\Delta d \approx 0.15\ D$ for distribution 2. For the envelope of the spectrally limited

pulse we shall use $f(t) = \mathrm{sech}(t/T)$, for which $S = \pi T = 1.19\ \tau_p$. If $\tau_p = 100$ ps and $I \lesssim 1$ MW/cm^2 (i.e., $\varepsilon \lesssim 2.7 \cdot 10^4$ V/cm for linearly polarized light), then $z_0 \lesssim 0.03 \ll 1$ and the conditions (9) hold, i.e., the regime of weak field and "narrow" distribution $p(d_i)$ is realized. The regime of strong field and "wide" distribution $p(d_i)$ obtains for $I \gtrsim 100$ GW/cm^2 ($z_0 \gtrsim 10$) with other conditions remaining the same. The theoretically possible situations $z_1 \ll 1$ (10) and $z_0 \gg 1$ (9) are not realized in this specific case.

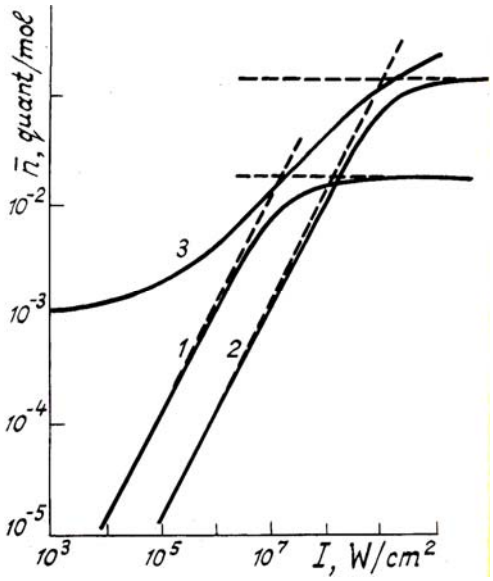

FIG. 4. The average number of quanta absorbed by an O_3 molecule versus the radiation intensity for excitation of the (000)–(001) transition. $\omega/2\pi c = 1037.4341$ cm^{-1}. 1, 2) collisionless regime: $\tau_p = 100$ ps (1), $\tau_p = 10$ ps (2), collisional regime (3) ($\tau_p \approx 75$ ns, $p = 3$ torr). The solid curves show the calculation by the method of line-by-line summation with the help of the real spectrum,[15] the dashed lines show the calculation using the formulas (12) and (16) with $q = 2.71 \cdot 10^{-3}$; $L = 2.27 \cdot 10^{-2}$cm^{-1}; $d_0 = 0.062$ D ($\tau_p = 100$ ps) and $q = 2.65 \cdot 10^{-3}$, $L = 2.95 \cdot 10^{-2}$cm^{-1} $d_0 = 0.067$ D ($\tau_p = 10$ ps).

It is of interest to compare the analytical results obtained for the probability of a vibrational transition with the results of line-by-line summation (6) using real spectroscopic information. Figure 4 shows the average number $\bar{n}$ of quanta absorbed by the O_3 molecule as a function of the intensity I of the CO_2 laser radiation. The curves 1 and 2 were calculated for a pulse $f(f) = \mathrm{sech}(t/T)$ by line-by-line summation with the help of the data of Ref. 14; In addition, the general solution[8] was used for the excitation probability of each VR transition and the M degeneracy was taken into account. The calculations showed that taking into account the absorption lines of O_3 lying outside the spectral contour of the radiation does not increase $\bar{n}$ appreciably. At the same time the formulas for the quasiclassical approximation (5) give too large a result (for $\tau_p = 100$ ps approximately by a factor of 2), without changing substantially the character of its intensity dependence. One can see from Fig. 4 that the results of the line-by-line summation are in good agreement with the calculation based on the formulas (12) and (16). We note that decreasing the pulse width by an order of magnitude proportionately decreases $\bar{n}$ in the weak-field regime and increases it in the strong-field regime.

From the viewpoint of the efficiency of the vibrational excitation it is interesting to compare the dependence $\bar{n}(I)$ for absorption of radiation under collisionless and collisional conditions. In Fig. 4 curve 3, which was taken from Ref. 3, was calculated for the collisional regime of excitation of the transition (000)–(001) in O_3. It is obvious that excitation is more efficient under collisional conditions, especially for low radiation intensities. Rotational relaxation and (with linear absorption) detuning of the frequency of the radiation from the nearest VR transitions plays the determining role here.[4]

Thus in the case of collisionless excitation of vibrational transitions in molecules by short laser pulses three characteristic regimes differing by the value of the parameter $z_0 = d_0 \varepsilon S/h$, which is proportional to the ratio of the characteristic field-induced broadening and the width of the spectrum of the pulse, can be distinguished. The weak-field regime, when the probability of vibrational excitation $\omega_1 \sim I \cdot S^2$, where I and S are the peak intensity and the area of the pulse, obtains when $z_0 \ll 1$. If $z_0 \simeq 1$, then an oscillatory regime, which is characterized by oscillations of the probability ω that decay as z_0 increases, is realized. In the strong-field regime $z_0 \gg 1$ and $\omega_1 \approx U$, i.e., vibrational excitation does not depend on the field strength and is determined by the overlap function of the radiation spectrum and the band spectrum. We stress that the decay of the oscillations of ω_1 as z_0 increases is caused both by the M degeneracy of the VR transitions and the random spread of the transition dipole moments d_i. It is significant that when the spread in the values of d_i is large ("wide" function $p(d_i)$; see the condition (10)) the oscillations of ω_1 decay more rapidly than in the case when the spread is small ("narrow" function $p(d_i)$, see the condition (9)). Thus, for example, in the Q branch the decay for the indicated cases is proportional to $(\varepsilon S)^2$ and εS, respectively. We stress that for the spectrally limited pulses studied in this work the irregularity of the position of the centers of the VR lines of the band does not affect the period and the decay of the oscillations of the probability ω_1. As regards the efficiency of vibrational photoexcitation, for laser pulses whose

width is greater than 10 ps collisional absorption conditions are apparently preferable.

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