

Emission and absorption of γ radiation by nuclei in molecules interacting with a light field

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In the present work, probabilities of emission (absorption) of nuclear γ radiation in molecules are calculated. In this process, molecular vibrations are excited and deexcited because of recoil of the nucleus. For vibrationally excited molecules, the existence of vibrational satellite shift is opposite in sign, and it allows compensation for the misalignment of the absorption and emission lines. The parameters (width, intensity, sign) of narrow frequency-tuned resonances of γ radiation with the molecules excited by laser emission in a low-pressure gas are calculated. The contributions of vibrational anharmonicity and rotation-vibration interaction are discussed.

I. INTRODUCTION

Recently^{1,2} the author has discussed the appearance of narrow γ -ray resonances for nuclei in molecules under the action of a coherent light wave. The principle of the effect is as follows: The coherent light wave excites a normal molecular vibration with the frequency ω_0 . If the gas pressure is low, then the rotational-vibrational molecular absorption line is broadened primarily by the Doppler effect rather than by collisions. In this case the light wave with its wave vector $\vec{K} = \vec{n}\omega/c$ excites molecules with a specific component of the velocity in the propagation direction:

$$\vec{K} \cdot \vec{v}_r = \omega - \omega_0 = \Omega. \quad (1.1)$$

From the classical point of view, nuclei in vibrationally excited molecules vibrate with the frequency ω_0 , which may be regarded as the average frequency of molecular transitions from one turning point of the molecular potential curve to the other. The amplitude of atomic vibrations depends on the force constants and the vibrational mode of the molecule, as well as on its orientation about the light-wave polarization vector. Its value lies in the range from 0.1 to 10^{-3} Å.³ It is significant that the average value of this displacement is of the same order of magnitude or larger than the wavelength λ_γ for nuclear γ transitions whose energy ranges from 10 keV to 1 MeV. Vibrations of a nuclear radiator (absorber) with such an amplitude might set up a considerable frequency modulation of γ radiation. In this case the modulation will extend only to the nuclei in excited molecules which have velocity satisfying condition (1.1). The frequency of nuclear transitions in molecules satisfying the condition for resonance with light wave is given by

$$\omega_{\text{res}}^{(0)} = \omega_\gamma \pm \Delta_\gamma + \vec{K}_\gamma \cdot \vec{v}_r, \quad (1.2)$$

where ω_γ is the frequency of the nuclear transition without recoil; $\hbar\Delta_\gamma$ is the recoil energy; the plus and minus signs correspond to absorption and emission of γ quanta; \vec{K}_γ is the wave vector of the γ quantum; and \vec{v}_r is the velocity of a resonantly excited molecule, determined from condition (1.1). When observing the emission (absorption) line of γ radiation collinearly with the light wave we have

$$\vec{K}_\gamma \cdot \vec{v}_r = \vec{K} \cdot \vec{v}_r \frac{\omega_\gamma}{\omega_0} = \Omega \frac{\omega_\gamma}{\omega_0}. \quad (1.3)$$

If the position of a nucleus is modulated at frequency ω_0 , and with amplitude $a \gg \lambda_\gamma$, the nuclear line $\omega_{\text{res}}^{(0)}$ splits into a set of spectral components at frequencies $\omega_{\text{res}}^{(m)} = \omega_{\text{res}}^{(0)} \pm m\omega_0$ ($m=0, 1, 2, \dots$). If the nuclear radiation is observed in a direction collinear with the propagation direction of the light wave, only a narrow portion of the Doppler width at frequency $\omega_{\text{res}}^{(0)}$ with half-width

$$\Gamma_{\text{nucl}} = \Gamma(1+G)^{1/2}\omega_\gamma/\omega_0 \quad (1.4)$$

will be modulated by the light wave. Here, Γ is the homogeneous half-width of the vibrational transition, which depends on molecular collisions, and $2\Gamma \ll \Delta\omega_D$, $\Delta\omega_D$ is the Doppler width of the optical line, and G is the saturation parameter of a vibrational transition determined by the light wave intensity. As a result, the Doppler profile of the nuclear resonance is considerably distorted (Fig. 1). At frequency $\omega_{\text{res}}^{(0)}$ a dip of half-width Γ_{nucl} is formed and at frequencies $\omega_{\text{res}}^{(m)} = \omega_{\text{res}}^{(0)} \pm m\omega_0$ peaks of the same width appear, the sum of their amplitudes being equal to the depth of the dip.

In Refs. 1 and 2 the intensities of the spectral components at $\omega_{\text{res}}^{(m)}$ were estimated using a simple classical model of frequency modulation. It is natural that any complete theory should be quantum mechanical. The aim of this paper is to present the quantum theory of emission and absorption of nu-

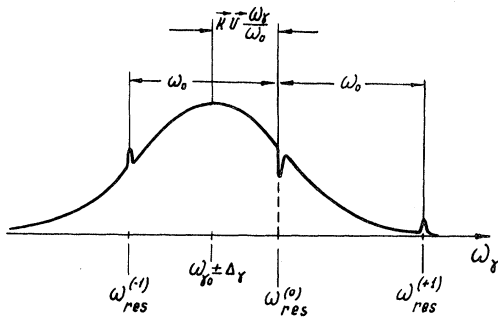


FIG. 1. Narrow resonances induced by an optical field in the absorption line of a γ transition in a nucleus located in a molecule, according to the classical model of Refs. 1 and 2.

clear γ radiation in excited and unexcited molecules. The theory predicts a so-called "vibrational shift" of nuclear lines which increases the noncoincidence between the lines of nuclear absorption and emission due to the recoil effect. Excitation of molecular vibrations reverses the sign of the vibrational shift, and thereby makes it possible to compensate for the noncoincidence between the absorption and emission lines. Finally, the results of the calculations make possible the evaluation of the parameters of narrow γ resonances for molecules excited by laser radiation in a low-pressure gas.

II. GENERAL RELATIONS

For a free nucleus the emission and absorption lines are shifted from each other by the recoil energy value R :

$$R = \hbar\Delta_\gamma = (\hbar\omega_\gamma/2Mc^2)\hbar\omega_\gamma, \quad (2.1)$$

where M is the nuclear mass, assuming that $\hbar\omega_\gamma \ll Mc^2$. Each line is broadened owing to the Doppler effect. If the nucleus is inside an atom the appearance of the spectrum is unchanged: The electron cloud does not affect the nucleus, and *vice versa*, so the electron motion is practically unaffected by changes in nuclear state.⁴ However, if the nucleus is located in a molecule then momentum and angular momentum conservation require a change of vibrational and rotational states in addition to the change in the translational motion. Changes in the internal state of the molecule must be taken into account from the very beginning, since recoil alters both the translation of the molecule and its internal state, the relative amounts depending upon the location of the nucleus inside the molecule. Therefore, the nuclear transition spectrum must

be qualitatively different in this case.⁵

The laws of conservation of momentum and energy for the system "nucleus in molecule plus γ quantum" have the following form in the nonrelativistic approximation:

$$M\vec{v}_0 \pm \hbar\vec{k}_\gamma = M\vec{v}, \quad (2.2)$$

$$\pm \hbar\omega_\gamma + \mathcal{E}_a + \frac{1}{2}Mv_0^2 = \mathcal{E}_b \pm E_0 + \frac{1}{2}Mv^2,$$

where \vec{v}_0 and \vec{v} are the initial and final translational velocities of the molecule, E_0 is the energy of the excited level in the nuclear transition under study, \mathcal{E}_a and \mathcal{E}_b are the initial and final internal energies of the molecule, and the plus and minus signs correspond to absorption and emission of γ quanta. It follows from Eq. (2.2) that the energy of a γ quantum which resonates with the nucleus in a molecule is given by

$$\hbar\omega_\gamma = E_0 + \hbar\vec{k}_\gamma \cdot \vec{v}_0 \pm \hbar\Delta_\gamma \pm (\mathcal{E}_b - \mathcal{E}_a). \quad (2.3)$$

The first term corresponds to the unshifted frequency of transition, the second one gives the frequency shift due to the Doppler effect (it is the same for both absorption and emission lines), the third term gives the line shifts caused by the recoil effect, and the last one represents those due to changes of molecular internal state.

If a molecule is initially unexcited then additional spectral components will arise at the blue edge of an absorption line and the red edge of an emission line. Their locations can be predicted from the energy-level diagram of the "nucleus-plus-molecule" system [Fig. 2(a)]. Thus an excitation of the molecular energy levels increases the detuning of the emission and absorption lines of the γ radiation. If the molecule is initially excited then transitions in which the molecular internal energy is transferred to the nucleus or the γ quantum also occur. In this case the additional emission and absorption lines are brought closer together [Fig. 2(b)]. The latter effect is of particular interest for compensating for the detuning caused by recoil. For that purpose it suffices to excite vibrational levels of the molecule with $\mathcal{E}_a \approx R$ by laser radiation. However, this method is only practical if the intensity of the additional components is rather high. The probability for a nuclear γ transition in molecules in which vibrational state changes is calculated below.

III. QUANTUM FORMULATION OF THE PROBLEM

A quantum formulation of the problem of emission of nuclear γ radiation from a nucleus in a molecule accompanied by a change in the molecular internal state is related to the problem of γ radiation from nuclei in a crystal. This problem arises in the theory of the Mössbauer effect.^{5,6} The internal

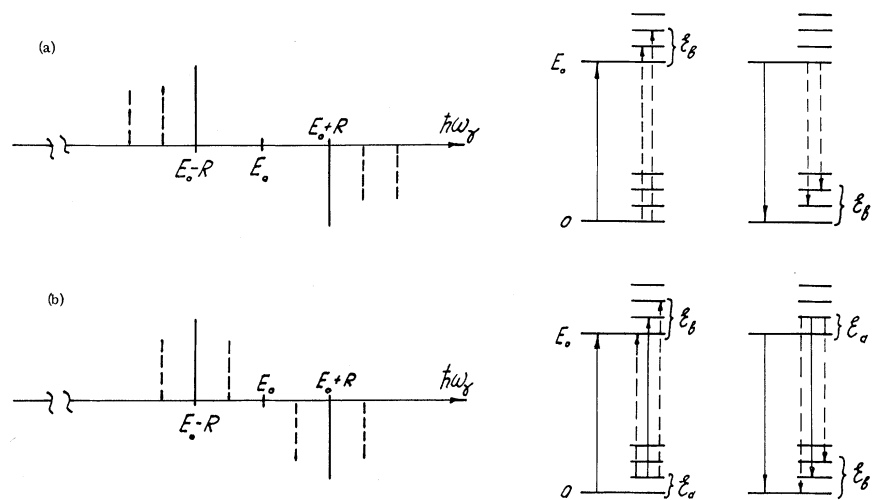


FIG. 2. Nuclear-vibrational transitions of a nucleus in (a) unexcited and (b) excited molecular systems. The patterns of absorption (downward lines) and emission (upward lines) spectra are shown on the left. Energy-level diagrams showing quantum transitions are on the right.

motion of the nucleons in a nucleus and the motion of the nuclear center of mass may be considered to be independent. The state of a nucleus in a molecule can be expressed as a product of the nuclear wave function $\psi(\vec{r}'_n)$, depending only on the internal nuclear coordinates of the nucleons \vec{r}'_n , and the wave function $\psi(\vec{R})$, which depends on the coordinates of the nuclear center of mass \vec{R} :

$$\psi(\vec{r}_n) = \psi(\vec{r}'_n)\psi(\vec{R}), \quad (3.1)$$

where $\vec{r}_n = \vec{r}'_n + \vec{R}$ is the coordinate of the n th nucleon. The interaction Hamiltonian for a system of nucleons and γ radiation, $H(\vec{r}_n)$ is related to the interaction Hamiltonian $H(\vec{r}'_n)$, which consists only of internal nuclear coordinates of the nucleons, \vec{r}'_n , by the relation⁶

$$H(\vec{r}_n) = e^{-i\vec{k}\gamma \cdot \vec{R}} H(\vec{r}'_n). \quad (3.2)$$

Separation of the internal motion in the nucleus makes it possible to express the matrix element for the transition from initial state a to final state b as a product of the two matrix elements:

$$\begin{aligned} \langle \psi_b^*(\vec{r}_n) | H(\vec{r}_n) | \psi_a(\vec{r}_n) \rangle \\ = \langle \psi_b^*(\vec{r}'_n) | H(\vec{r}'_n) | \psi_a(\vec{r}'_n) \rangle \langle \psi_b^*(\vec{R}) | e^{-i\vec{k}\gamma \cdot \vec{R}} | \psi_a(\vec{R}) \rangle. \end{aligned} \quad (3.3)$$

The initial and final states of a nucleus in a molecule are given by sets of quantum numbers $\{a, v^a, J^a\}$ and $\{b, v^b, J^b\}$, where the letters a and b describe the quantum states of the free nucleus; v^a and v^b are the vibrational quantum numbers of the molecule; and J^a and J^b are the rotational quantum numbers. The first factor depends only on the nuclear structure, and its value is constant for a particular γ transition. The second factor takes into account the change in the position of the

nuclear center of mass as the molecular state changes. The relative probability P_{ba} of a molecular transition from $|a\rangle$ to $|b\rangle$ is determined by the expression

$$P_{ba} = |\langle \psi_b^*(\vec{R}) | e^{-i\vec{k}\gamma \cdot \vec{R}} | \psi_a(\vec{R}) \rangle|^2, \quad (3.4)$$

with the total transition probability to all possible final states equal to 1.

The position of the nuclear center of mass \vec{R} in a molecule can be written conveniently as

$$\vec{R} = \vec{R}_0 + \vec{r} + \vec{u}, \quad (3.5)$$

where \vec{R}_0 is the position of the molecular center of mass, \vec{r} is the vector connecting the molecular center of mass with the nuclear equilibrium position, and \vec{u} is the vibrational shift of the nuclear center of mass about the equilibrium position. In such a coordinate system the matrix element Eq. (3.4) will have the form

$$\begin{aligned} \langle \psi_b^*(\vec{R}) | e^{-i\vec{k}\gamma \cdot \vec{R}} | \psi_a(\vec{R}) \rangle \\ = \langle T_b | e^{-i\vec{k}\gamma \cdot \vec{R}_0} | T_a \rangle \langle \psi_b^*(\vec{r}, \vec{u}) | e^{-i\vec{k}\gamma \cdot (\vec{r} + \vec{u})} | \psi_a(\vec{r}, \vec{u}) \rangle, \end{aligned} \quad (3.6)$$

where T_a and T_b are functions describing the initial and final molecular translational states, and the function $\psi(\vec{r}, \vec{u})$ describes the molecular vibrational and rotational states. The change in molecular translation is determined by the momentum and energy conservation laws (Sec. II). The problem reduces to calculating the rotational-vibrational matrix element in Eq. (3.6).

In the general case the molecular rotational and vibrational states change simultaneously because of recoil. However, for

$$\vec{r} = 0, \quad (3.7)$$

which corresponds to the case when the nucleus is in equilibrium at the molecular center of mass, conservation of angular momentum forbids any change in the molecular rotational state ($J^a = J^b = J$). It is assumed here that the angular momentum of the γ quantum does not cause a change in the molecular rotational state. This is true provided that the angular momentum of molecular rotation and that of nuclear internal motion are independent and, therefore, that the angular momentum of a γ quantum can only cause a change in the nuclear angular momentum. As a matter of fact, even if $\vec{R} = \vec{u}$, when $J^a = J^b$ the probabilities for the change of the molecular vibrational state during absorption or emission of a γ quantum depend on the molecular rotational state J . The proper expression with regard to J will be given below, in Sec. VII. For now we neglect this rather weak dependence on J in order not to complicate the expression. In this case under condition (3.7) the matrix element (3.6) reduces to

$$\langle \psi_b^*(\vec{R}) | e^{-i\vec{k}\gamma \cdot \vec{R}} | \psi_a(\vec{R}) \rangle = \langle \psi_b^*(\vec{u}) | e^{-i\vec{k}\gamma \cdot \vec{u}} | \psi_a(\vec{u}) \rangle, \quad (3.8)$$

where the wave function $\psi(\vec{u})$ gives the vibrational shift of the nucleus in question.

It should be noted that the rotations can be eliminated only in polyatomic molecules with symmetry, e.g., in linear molecules of the form XY_2 , in planar triangular molecules of the form XY_3 , and in tetrahedral molecules of the form XY_4 , where X is the nucleus in question. For diatomic molecules it is impossible to eliminate rotations given an arbitrary orientation of the molecular axis about \vec{K}_γ , since recoil changes vibrational and rotation simultaneously. Condition (3.7) limits the class of molecules under study and the possible locations of the nucleus in them, but still includes molecules of great practical interest (OsO_4 , WF_6 , etc.). It is important to note that condition (3.7) provides additional spectral components with the simplest structure, enabling one to obtain narrow frequency-tuned γ resonances. Therefore, the present paper will limit itself to molecules and nuclear positions satisfying condition (3.7).

In the general case the vibrational displacement of the n th nucleus can be expressed as a superposition of displacements caused by various normal molecular vibrations:

$$\vec{u}_n = \sum_l Q_l \vec{\rho}_{ln}, \quad (3.9)$$

where Q_l is the l th normal molecular coordinate and $\vec{\rho}_{ln}$ is the displacement of the n th nucleus due to the l th normal vibration. For simplicity in notation, the index l numbers here all nondegenerate

and degenerate normal vibrations. In other words, the number l takes on all values from 1 to $\sum_i d_i l$ in succession, where d_i is the degeneracy of an l th normal vibration, rather than from 1 to l . It is natural that when using the deduced expressions for the normal vibration l with the degeneracy ratio d_l , it is necessary to consider all d_l at once. In Sec. IV this is demonstrated by the doubly degenerate vibration ν_2 of the XY_2 molecule. For example, the triply degenerate vibration ν_3 of the XY_4 molecule is treated in perfect analogy to this (see the notes in the conclusion, Sec. VIII). If we adopt the usual quantum-mechanical designation for Q_l , then $\vec{\rho}_{ln}$ has dimension $M^{-1/2}$. In the following, nuclear index n is omitted to simplify the notation. In the harmonic approximation using normal coordinates, the wave function $\psi(\vec{u})$ is a product of harmonic-oscillator wave functions⁷:

$$\psi = \prod_l \psi_l, \quad \psi_l = N_{v_l} e^{-\alpha_l Q_l^2 / 2} H_{v_l}(\sqrt{\alpha_l} Q_l), \quad (3.10)$$

where $N_{v_l} = [2^{v_l} v_l! (\pi/\alpha_l)^{1/2}]^{-1/2}$ is the normalization constant and $\alpha_l = \omega_l/\hbar$, where ω_l is the characteristic frequency of the l th vibration. Then the matrix element assumes the form:

$$\begin{aligned} \langle \psi_b^*(\vec{u}) | e^{-i\vec{k}\gamma \cdot \vec{u}} | \psi_a(\vec{u}) \rangle \\ = \prod_l \langle \psi_b^*(Q_l) | e^{-i\vec{k}\gamma \cdot \vec{\rho}_l Q_l} | \psi_a(Q_l) \rangle. \end{aligned} \quad (3.11)$$

Thus the problem reduces to calculating matrix elements of the type

$$S_{v_l v_l'} = \langle \psi_b^*(Q_l) | e^{-i\vec{k}\gamma \cdot \vec{\rho}_l Q_l} | \psi_a(Q_l) \rangle, \quad (3.12)$$

with the harmonic-oscillator wave functions Eq. (3.10).

IV. PROBABILITIES OF γ TRANSITIONS IN AN UNEXCITED MOLECULE

Let us first consider nuclear γ transitions in molecules in the ground vibrational state: $v_l^a = 0$ for all values of l . For simplicity we introduce the following notation: $v_l^a = v_l^0$, $v_l^b = v_l$. In this case the matrix element (3.12) reduces to the integral

$$\begin{aligned} S_{v_l, 0} = 4 \left(\frac{\alpha_l}{\pi} \right)^{1/2} N_{v_l} \int_{-\infty}^{\infty} e^{-\alpha_l Q_l^2} H_{v_l}(\sqrt{\alpha_l} Q_l) \\ \times e^{-i\vec{k}\gamma \cdot \vec{\rho}_l Q_l} dQ_l. \end{aligned} \quad (4.1)$$

Equation (4.1) may be evaluated using the integrals given in Ref. 8. One, then, obtains

$$S_{v_l, 0} = (2^{v_l} v_l!)^{-1/2} e^{-f_l^2/4} f_l^{v_l} (-1)^{v_l/2}, \quad (4.2)$$

for even v_l , and the same expression with the additional factor $(-1)^{(v_l-1)/2} (-i)$ for odd v_l , where f_l is given by

$$f_l = \vec{K}_\gamma \cdot \vec{\rho}_l / \sqrt{\alpha_l}. \quad (4.3)$$

The probability of a γ transition accompanied by the vibrational transition $0 \rightarrow v_i$ is given by

$$P_{v_i,0} = |S_{v_i,0}|^2 = \frac{Z_i^{v_i}}{v_i!} e^{-Z_i}, \quad (4.4)$$

with the parameter Z_i given by

$$Z_i = \frac{1}{2} f_i^2 = (\vec{K}_\gamma \cdot \vec{\rho}_i)^2 / 2\alpha_i. \quad (4.5)$$

Thus, the probability of transition accompanied by excitation of the vibrational level v_i is given by a Poisson distribution. It is evident that a group of levels centered near Z_i is always excited. When $Z_i \gg 1$ the peak of the distribution occurs at $v_i \approx Z_i$, and the average value of v_i is close to the peak, i.e.,

$$v_{\max} \approx \bar{v}_i \approx Z_i \quad \text{with} \quad Z_i \gg 1. \quad (4.6)$$

In order to gain some physical feeling for the parameter Z_i , let us consider the simple case of a linear symmetric molecule XY_2 in more detail. The nucleus X participates in two of the three normal molecular vibrations, namely, the doubly degenerate bending mode ν_2 (nucleus X moves in a plane perpendicular to the molecular axis) and in the nondegenerate antisymmetric vibration ν_3 (nucleus X moves along the molecular axis). For simplicity, consider two selected molecular orientations (Fig. 3) in which only one normal vibration is excited. If the molecular axis coincides with the propagation direction of the γ quantum [Fig. 3(a)] then the recoil of nucleus X may excite only the ν_3 vibration. The displacement of nucleus X along the molecular axis is related to the amplitude of normal vibration by⁹

$$u_z = \rho_3 Q_3, \quad \rho_3 = [2m_Y/m_X M]^{1/2}, \quad (4.7)$$

where $M = m_X + 2m_Y$ is the molecular mass. So, the parameter Z_3^0 can be written as

$$Z_3^0 = \left(\hbar \omega_\gamma \frac{\hbar \omega_\gamma}{2Mc^2} \right) \left(\frac{M \rho_3^2}{\hbar \omega_3} \right) = \frac{R}{\hbar \omega_3} \frac{2m_Y}{m_X}, \quad (4.8)$$

where R is the recoil energy and $\hbar \omega_3$ is the energy spacing of the ν_3 vibration. With $Z_3^0 \gg 1$, so that a well-defined peak occurs in the $P_{v_i,0}$ distribution, an average energy of

$$E_{\text{vib}} = \bar{v}_3 \hbar \omega_3 = R \times 2m_Y/m_X \quad (4.9)$$

is required to excite vibrations during emission or absorption of a γ quantum. If the γ radiation interacts with the lighter nucleus ($m_X \ll m_Y$), then vibrational excitation requires an energy much larger than the recoil energy R . Thus, the vibrational degrees of freedom may consume a larger energy than the translational degrees of freedom. The opposite occurs when the γ quantum interacts with the heavier nucleus ($m_X \gg m_Y$).

If the molecule is oriented in a plane perpendi-

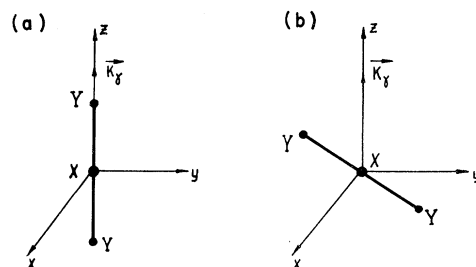


FIG. 3. Excitation of normal modes in the linear symmetric molecule XY_2 due to the recoil of the nucleus X : (a) excitation of the antisymmetric vibrational mode; (b) excitation of the bending mode.

cular to the propagation direction of the γ quantum then the recoil of nucleus X excites only normal vibration ν_2 . In this case the relation between the displacement along the OZ axis [Fig. 3(b)] and the amplitude of the normal mode is given by the coefficient $\rho_2 = \rho_3$. Hence, the parameter Z_2^0 is given by

$$Z_2^0 = (R/\hbar \omega_2) 2m_Y/m_X, \quad (4.10)$$

i.e., the average energy required to excite the ν_2 normal mode is the same as in Eq. (4.9).

It should be noted that rotation-vibration interaction removes the degeneracy of the vibration ν_2 (l doublet), and we may study, in principle, the excitation of each level of the l doublet, say, for $v_2 = 1$. The corresponding estimates of this effect are given below, in Sec. VII.

If the linear symmetric molecule XY_2 is oriented in an arbitrary direction about \vec{K}_γ then

$$Z_2 = Z_2^0 \cos^2 \phi \quad \text{and} \quad Z_3 = Z_3^0 \sin^2 \phi, \quad (4.11)$$

where ϕ is the angle between \vec{K}_γ and the molecular axis. The total energy required to excite both normal vibrations is then equal to

$$E_{\text{vib}} = \bar{v}_2 \hbar \omega_2 + \bar{v}_3 \hbar \omega_3 = R \times 2m_Y/m_X. \quad (4.12)$$

Thus the total vibrational energy transferred to a molecule is independent of molecular orientation, although the distribution between the two modes does vary with molecular orientation. The total vibrational energy transferred to the molecule depends on the ratio of the mass of the nucleus of interest to the total molecular mass. An analogous assertion can be proved for the more general case of a polyatomic molecule with the nucleus located at the center of mass.

To calculate the intensities of the vibrational satellites of the γ transition for a given normal vibration, one must average the function $P_{v_i,0}(Z_i)$ over all possible molecular orientations. Let \vec{K}_γ

be oriented along the OZ axis and define θ as the angle between the displacement vector $\vec{\rho}_i$ and OZ and ϕ as the azimuthal angle in the XY plane. In this case the value of $P_{v_i,0}$ averaged over molecular orientations is given by

$$\bar{P}_{v_i,0}(Z_i^0) = \frac{1}{4\pi} \int_{-\pi}^{\pi} d\phi \int_0^{\pi} \sin\theta P_{v_i,0}(Z_i^0 \cos^2\theta) d\theta, \quad (4.13)$$

where Z_i^0 is the maximum value of Z_i for molecules with $\theta=0$:

$$Z_i^0 = (\vec{\rho}_i \cdot \vec{K}_\gamma)^2 / 2\alpha_i. \quad (4.14)$$

The integration occurring in Eq. (4.13) cannot be performed analytically, but it can be reduced to a form convenient for numerical evaluation:

$$\bar{P}_{v_i,0} = \frac{(Z_i^0)^{v_i}}{v_i!} \int_0^1 y^{2v_i} e^{-Z_i^0 y^2} dy. \quad (4.15)$$

Figure 4 gives values of the averaged probability distribution $\bar{P}_{v_i,0}$ calculated for $v_i=0, 1, \dots, 8$ in the interval $0 \leq Z_i^0 \leq 5$. For comparison, the dashed line in Fig. 4 plots the unaveraged probability $P_{0,0}$. The averaging process increases the intensity of the lowest vibrational satellites and decreases that of the higher satellites. This effect is due to contributions from molecules whose vectors $\vec{\rho}_i$ are nearly perpendicular to \vec{K}_γ . For such molecules the parameter $Z_i = Z_i^0 \cos^2\theta \ll Z_i^0$, and the maximum of the $P_{v_i,0}$ distribution is shifted towards $v_i=0$.

Finally, let us compare the quantum-mechanical expression obtained here for the intensity of vibrational satellites of the γ lines with that derived in Refs. 1 and 2. For example, the classical and quantum-mechanical relative intensities of the first satellite are given by

$$I_{cl} = J_1^2(\vec{K}_\gamma \vec{\alpha}_{cl}), \quad I_{qu} = \frac{(\vec{K}_\gamma \cdot \vec{\rho})^2}{2\alpha} e^{-(\vec{K}_\gamma \cdot \vec{\rho})^2 / 2\alpha}, \quad (4.16)$$

where α_{cl} is the classical amplitude of vibration.

Since $\alpha_{cl} = \rho/\sqrt{\alpha}$, then for $Z \ll 1$ (or $\vec{K}_\gamma \cdot \vec{\alpha} \ll 1$) these expressions agree. However, the discrepancy becomes noticeable for $Z \geq 1$, as well as for subsequent vibrational satellites.

V. PROBABILITIES OF γ TRANSITIONS IN AN EXCITED MOLECULE

Let us consider a molecule which is initially in the first vibrational level of the l th normal vibration: $v_l^0=1$; $v_l^0=0$, and $l' \neq l$. The matrix element for a molecular transition to another level of the l th vibration can be reduced to the form

$$S_{v_l,1} = (2\pi 2^{v_l} v_l!)^{-1/2} \int_{-\infty}^{\infty} e^{-x^2} H_1(x) H_{v_l}(x) e^{if_l x} dx. \quad (5.1)$$

A calculation using the integrals given in Ref. 8 results in the following expression¹⁰ for the probability of transition $P_{v_l,1}$:

$$P_{v_l,1} = \frac{1}{v_l!} Z_l^{v_l} e^{-Z_l} [L_1^{(v_l-1)}(Z_l)]^2, \quad (5.2)$$

where $L_m^{(n)}$ is the generalized Laguerre polynomial. In the present case $L_1^{(n)}(Z) = n+1-Z$,¹¹ and expression (5.2) has a simple form:

$$P_{v_l,1} = \frac{1}{v_l!} Z_l^{v_l} (Z_l - v_l)^2 e^{-Z_l}. \quad (5.3)$$

The most essential is the γ transition in which the vibrational state changes from $v_i=1$ to $v_i=0$. Here, the vibrational energy of the molecule is transferred to the nucleus or the γ quantum. This process gives rise to spectral components at $E_0 \pm (R - \hbar\omega)$, thus bringing absorption and emission lines closer together. When $R = \hbar\omega$, the vibrational satellite lines of absorption and emission overlap. As can be seen from Eq. (4.4), the transition probability $P_{0,1}$ is the same as that of the reverse process $P_{1,0}$. The maximum intensity of the vibrational satellites at $E_0 \pm (R - \hbar\omega)$, which

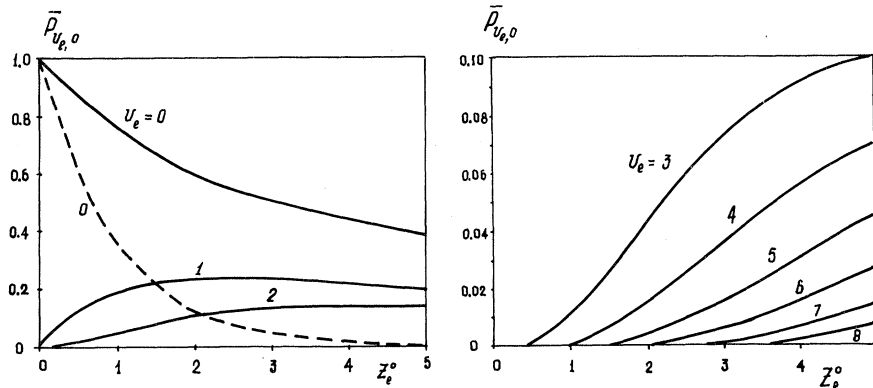


FIG. 4. Molecular orientation-averaged probabilities of nuclear-vibrational transitions giving the intensity distribution of vibrational satellites of nuclear γ transitions in unexcited molecules in a gas.

occurs when $Z_i = 1$, is found to be 37%. In this case the intensity of the primary lines at $E_0 \pm R$ is equal to 0. For larger values of Z the maximum intensity occurs for satellites with $v \approx Z$. Just as in the case of unexcited molecules it is necessary here to average over molecular orientations. Averaged probability values can be obtained from the following expression:

$$\bar{P}_{v_i,1} = \frac{1}{v_i!} (Z_i^0)^{v_i-1} \int_0^1 y^{2(v_i-1)} e^{-Z_i^0 y^2} (Z_i^0 y^2 - v_i)^2 dy. \quad (5.4)$$

Figure 5 shows the results of numerical calculations for $\bar{P}_{v_i,1}(Z_i^0)$. Averaging over orientations eliminates the sharp minima and increases the intensity of lower-order satellites.

In an analogous manner we are able to deduce expressions for probabilities of γ transitions in a molecule which is initially in a higher vibrational level. In this case vibrational satellites arise at frequencies $\omega_{\gamma_0} \pm [\Delta_{\gamma} + (v_i - v_i^0)\omega_i]$, where v_i^0 is the initial vibrational state of the molecule. The expressions for the relative intensities are cumbersome, however.

The large relative intensities of the vibrational satellites indicates that vibrational excitation of molecules may become a new method of compensating for the recoil-induced shift of emission and absorption lines of γ radiation. This method should be more efficient than the existing methods of atomic heating¹² and rapid relative motion of source and target.¹³ In practice, molecular vibrational levels can be excited easily using infrared laser radiation. If the molecules are excited in a gas at pressures above a few Torr, where the collisional broadening of molecular vibrational-rotational transitions is comparable to or greater than Doppler broadening, then the molecules become excited regardless of their velocity. In this case the broadening of vibrational satellites of γ transi-

tions will be determined by the Doppler effect. However, at low pressures the widths of the vibrational satellites of the γ transitions may be considerably narrower than the Doppler widths.

VI. STRUCTURE OF NARROW γ RESONANCES INDUCED BY A LIGHT WAVE

In a low-pressure gas the coherent light wave excites molecules of velocity \vec{v} satisfying condition (1.1). As a result, the molecular velocity distributions of both levels of the vibrational-rotational transition under consideration can depart significantly from equilibrium. The initial velocity distribution of the lower level of the transition is given by

$$n_0(\vec{v}) = qN_0W(\vec{v}), \quad (6.1)$$

where N_0 is the total number of particles in the ground vibrational state in the absence of the field, $W(\vec{v})$ is the Maxwell velocity distribution, and q is the fraction of molecules in the vibrational level. Under the action of a resonant field the velocity distributions of the lower (1) and upper (2) levels of the transition become

$$n_1(\vec{v}) = n_0(\vec{v}) \frac{2 + p(\vec{v})}{2[1 + p(\vec{v})]}, \quad (6.2)$$

$$n_2(\vec{v}) = n_0(\vec{v}) \frac{p(\vec{v})}{2[1 + p(\vec{v})]},$$

where $p(\vec{v})$, the line-shape factor of a moving molecule, is a function of the detuning of its transition frequency ω_0 from the field frequency ω and the saturation parameter G :

$$p(\vec{v}) = \frac{G\Gamma^2}{(\omega - \omega_0 - \vec{k} \cdot \vec{v})^2 + \Gamma^2}. \quad (6.3)$$

Note that at exact resonance [Eq. (1.1)] $p(\vec{v}) = G$. The width of the resonance is determined by Eq.

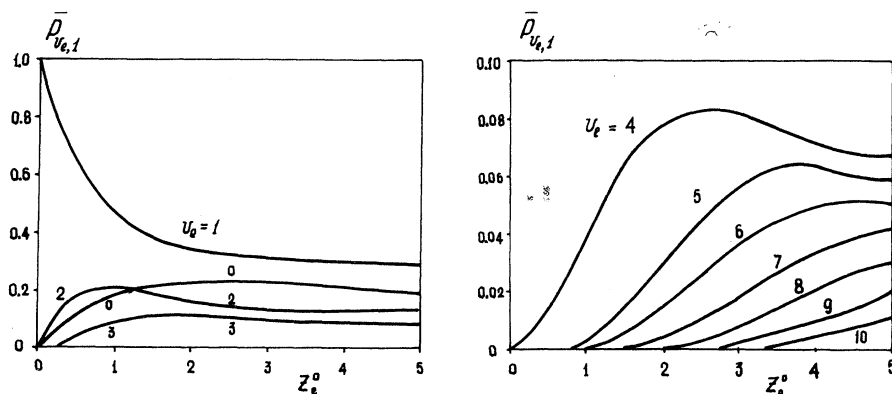


FIG. 5. Molecular orientation-averaged probabilities of nuclear-vibrational transitions giving the intensity distribution of vibrational satellites of nuclear γ transitions in vibrationally excited molecules in a gas.

(1.4). The population fraction q of the level of interest depends on the degeneracy of the level and on the rotational partition function Z_{rot} :

$$q = gZ_{\text{rot}}^{-1} e^{-hF(J)/kT}, \quad (6.4)$$

where $F(J)$ is the energy of the rotational level.

The absorption line shape of one vibrational satellite of the γ transition at frequency $\omega_{\gamma}^{(v_i)}$, caused by nuclei in molecules in the i th level ($i=1,2$), is given by

$$\Phi_i(\omega_{\gamma}) = \int \sigma(\omega_{\gamma} - \omega_{\gamma}^{(v_i)} - \vec{K}_{\gamma} \cdot \vec{v}) n_i(\vec{v}) d(\vec{v} \cdot \hat{n}_{\gamma}). \quad (6.5)$$

Here \hat{n}_{γ} is a unit vector in the propagation direction of the γ quantum, the velocity distributions $n_i(\vec{v})$ are given by Eqs. (6.2), and $\sigma(x)$ is the cross section for resonant absorption of a γ quantum:

$$\sigma(x) = \sigma_0 \frac{\Gamma_{\gamma}^2}{x^2 + \Gamma_{\gamma}^2}, \quad (6.6)$$

with σ_0 the peak value of the absorption cross section and Γ_{γ} the natural linewidth of the nuclear γ transition.

If the direction of observation of γ radiation coincides with that of the light wave ($\hat{n}_{\gamma} = \hat{n}$), then a narrow structure arises in the spectrum $\Phi_i(\omega_{\gamma})$ caused by a hole in the velocity distribution of the lower level, $n_1(v_z)$, or by a peak in the velocity distribution of the upper level, $n_2(v_z)$. Here $v_z = \vec{v} \cdot \hat{n}$ is the projection of the molecular velocity onto the light-wave direction (\hat{n} and \hat{n}_{γ} are directed along the OZ axis). When the relative natural width of the γ transition, $\Gamma_{\gamma}/\omega_{\gamma}$, is much smaller than the relative homogeneous width of the vibrational transition, that is, when

$$\frac{\Gamma_{\gamma}}{\omega_{\gamma}} \ll \frac{\Gamma}{\omega} (1+G)^{1/2}, \quad (6.7)$$

then the width $2\Gamma_{\text{nuc}}$ of the resulting narrow γ resonance is governed by a molecular "channel" and determined by Eq. (1.4). In this case the convolution integral Eq. (6.5) can be reduced in the usual manner to the simple form

$$\Phi_i(\omega_{\gamma}) = \sigma_0 n_i(\xi), \quad \xi = \frac{\omega_{\gamma} - \omega_{\gamma}^{(v_i)}}{K_{\gamma}}, \quad (6.8)$$

where for each molecular energy level i , $n_i(\xi) = n_i(v_z)$ [Eq. (6.2)] is the distribution of velocity components along the light-wave direction.

Let us first consider nuclei in excited molecules which have vibrational satellites at frequencies

$$\omega_{\gamma}^{(v_i-1)} = \omega_{\gamma_0} \pm [\Delta_{\gamma} + (v_i - 1)\omega_i]. \quad (6.9)$$

The total line shape of the γ radiation from these nuclei has the form

$$\Phi_2(\omega_{\gamma}) = \sigma_0 \sum_{v_i} P_{v_i,1} n_2 \left(\frac{\omega_{\gamma} - \omega_{\gamma_0} \pm [\Delta_{\gamma} + (v_i - 1)\omega_i]}{K_{\gamma}} \right). \quad (6.10)$$

It follows from Eqs. (6.10) and (6.3) that narrow peaks occur at resonant frequencies

$$\begin{aligned} \omega_{\text{res}}^{(v_i-1)} &= \omega_{\gamma_0} \pm [\Delta_{\gamma} + (v_i - 1)\omega_i] + \frac{\omega_{\gamma_0}}{\omega_0} (\omega - \omega_0) \\ &= \omega_{\gamma}^{(v_i-1)} + \frac{\omega_{\gamma_0}}{\omega_0} \Omega. \end{aligned} \quad (6.11)$$

Their half-width Γ_{nuc} is determined by Eq. (1.4) and by the relative amplitude given by Eq. (5.4). The typical form of the spectrum $\Phi_2(\omega_{\gamma})$ is illustrated in Fig. 6(b), where the distribution of molecular velocities $n_2(v_z)$ in excited levels is also shown.

Nuclei in unexcited molecules have vibrational satellites at frequencies

$$\omega_{\gamma}^{(v_i)} = \omega_{\gamma_0} \pm (\Delta_{\gamma} + v_i \omega_i), \quad (6.12)$$

and their total γ radiation line shape has the form

$$\Phi_1(\omega_{\gamma}) = \sigma_0 \sum_{v_i} P_{v_i,0} n_1 \left(\frac{\omega_{\gamma} - \omega_{\gamma_0} \pm (\Delta_{\gamma} + v_i \omega_i)}{K_{\gamma}} \right). \quad (6.13)$$

In this case narrow dips occur at the resonance frequencies

$$\omega_{\text{res}}^{(v_i)} = \omega_{\gamma}^{(v_i)} + \frac{\omega_{\gamma_0}}{\omega_0} \Omega, \quad (6.14)$$

with half-width Γ_{nuc} and relative amplitudes given by Eq. (4.15). The typical form of the spectrum $\Phi_1(\omega_{\gamma})$ is shown in Fig. 6(a), together with the distribution of molecular velocities $n_1(v_z)$ in the ground vibrational state.

Experimentally, one observes the total spectrum

$$\Phi(\omega_{\gamma}) = \Phi_1(\omega_{\gamma}) + \Phi_2(\omega_{\gamma}), \quad (6.15)$$

which is due to nuclei in both unexcited and excited molecules. The frequencies of all holes and peaks in the spectra $\Phi_1(\omega_{\gamma})$ and $\Phi_2(\omega_{\gamma})$ coincide, with the exception of the peak in $\Phi_2(\omega_{\gamma})$ at frequency $\omega_{\gamma}^{(-1)} = \omega_{\gamma} \pm (\Delta_{\gamma} - \omega_i)$. Because of this the relative amplitude of the vibrational satellite at frequency $\omega_{\gamma}^{(-1)}$ is determined by the probability $P_{0,1}$. The amplitudes of the remaining resonances depend on the relative amplitudes of the peaks and holes at coinciding frequencies. The hole depth in $n_1(v_z)$ is exactly equal to the peak height in $n_2(v_z)$ since, according to Eq. (6.2),

$$n_1(v_z) = n_0(v_z) - n_2(v_z). \quad (6.16)$$

Using this relation one can reduce the expression for the total spectrum $\Phi(\omega_{\gamma})$ to the form

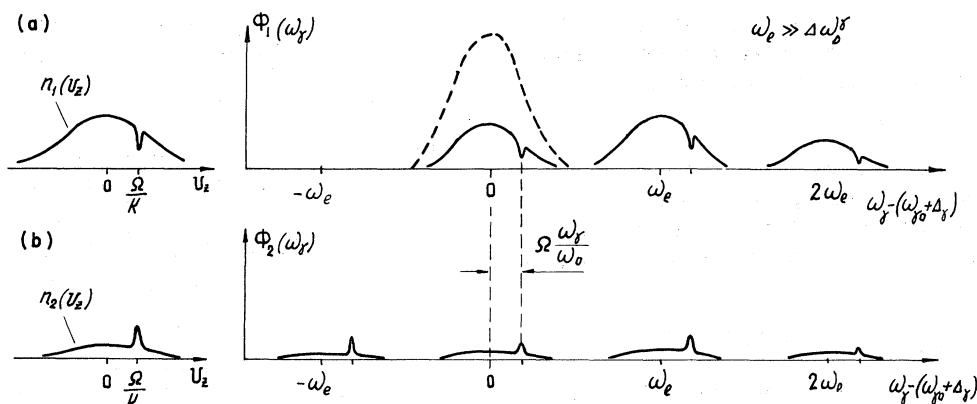


FIG. 6. Spectrum of nuclear γ transitions in (a) vibrationally unexcited and (b) excited molecules. The distribution of v_z , the component of molecular velocity in the propagation direction of the light wave (which coincides with the direction of γ radiation observation), is shown at the left.

$$\Phi(\omega_\gamma) = \sigma_0 \sum_{v_i \geq 0} [\bar{P}_{v_i,0} n_0(\omega_\gamma^{(v_i)}) + (\bar{P}_{v_i+1,1} - \bar{P}_{v_i,0}) n_2(\omega_\gamma^{(v_i)})] \quad (6.17)$$

The first term in square brackets represents the Doppler-broadened vibrational satellites of the γ transition, independent of the optical field. The second term gives the contribution due to the optical field. Against the background of the Doppler-broadened lines narrow peaks and dips arise, their amplitudes and signs determined by the difference in probabilities

$$\Delta P_{v_i} = \bar{P}_{v_i+1,1} - \bar{P}_{v_i,0}, \quad v_i \geq 0. \quad (6.18)$$

Figure 7 illustrates the dependence of ΔP_{v_i} on Z_i^0 for small values of v_i .

The spectrum of nuclear γ transitions in molecules, calculated quantum mechanically, differs significantly from the results of the classical model.^{1,2} In the classical model a dip always occurs at frequency $\omega_\gamma^{(0)} = \omega_\gamma \pm \Delta_\gamma$, whereas peaks occur at the remaining frequencies $\omega_\gamma^{(v_i)}$. In the quantum model, however, a peak occurs at frequency $\omega_\gamma^{(-1)}$, a dip occurs at $\omega_\gamma^{(0)}$, and at the remaining frequencies peaks or dips occur, depend on the value of the parameter Z_i^0 . In addition, in the quantum model the spectrum of narrow resonances is asymmetric about frequency $\omega_\gamma^{(0)}$, as expected.

The frequency of narrow resonances $\omega_{\text{res}}^{(v_i)}$, determined by the general relations (6.11) and (6.14), may be tuned through the Doppler profile of each vibrational satellite by tuning the field frequency ω through the Doppler profile of the corresponding rotational-vibrational transition centered at ω_0 .

The tuning range at the γ transition is larger than that at the optical "channel" by a factor ω_γ/ω_0 .

VII. EFFECT OF VIBRATIONAL ANHARMONICITY AND OF VIBRATION-ROTATION INTERACTION

The structure of the vibrational satellites of nuclear γ transitions in molecules was considered above in the harmonic approximation, also neglecting rotational motion. The effect of molecular rotations on the frequencies of the vibrational satellites was excluded by condition (3.7). This con-

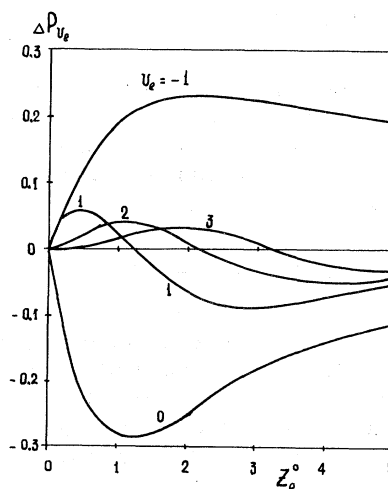


FIG. 7. The factor ΔP_{v_i} , which determines the amplitude of the γ resonance of the v_i th vibrational satellite. Positive values of ΔP correspond to narrow peaks, negative values to dips. $\Delta P_{-1} = \bar{P}_{01}$.

siderably simplifies the structure of narrow γ resonances by eliminating transitions which change rotational state. However, one must take into account the dependence of rotational energy on vibrational state. In addition, it is necessary to consider the role of vibrational anharmonicity. This may be done with the help of the scheme considered above. Let us restrict ourselves here to some remarks on changes in the spectrum caused by vibrational anharmonicity and rotation-vibration interactions, without giving detailed calculations.

In the general case the center frequency of the vibrational satellite resulting from the transition $(v_i^0, J) \rightarrow (v_i, J)$ is given by the expression

$$\omega_\gamma(v_i, J) = \omega_{\gamma_0} \pm (1/\hbar) [\hbar \Delta_\gamma + \mathcal{E}(v_i, J) - \mathcal{E}(v_i^0, J)], \quad (7.1)$$

where J is the rotational quantum number, v_i^0 is the initial vibrational state of a molecule ($v_i^0 = 0$ for unexcited molecules, and $v_i^0 = 1$ for molecules excited to the first vibrational level), and $\mathcal{E}(v_i, J)$ is the energy of the vibrational-rotational molecular state. In the simplest case \mathcal{E} is given by

$$\mathcal{E}(v_i, J) = \hbar[G_{v_i} + F_{v_i}(J)], \quad (7.2)$$

$$G_{v_i} = \omega_i(v_i + \frac{1}{2}) - \omega_i \chi_i (v_i + \frac{1}{2})^2 + \dots, \quad (7.3)$$

$$F_{v_i}(J) = B_{v_i} J(J+1), \quad B_{v_i} = B_i - \alpha_i(v_i + \frac{1}{2}), \quad (7.4)$$

where $\hbar G_{v_i}$ is the vibrational energy and $\hbar F_{v_i}(J)$ is the rotational energy.

Let us consider changes in the γ -ray spectrum due to anharmonicity and rotations in two limiting cases, depending on the pressure of the molecular gas. Consider first the high-pressure case, where narrow resonances do not occur. The only effect of anharmonicity is to cause small changes in the positions of the Doppler-broadened vibrational satellites. Furthermore, because of rotations, unexcited and excited molecules are distributed over many rotational states. A superposition of the frequencies $\omega_\gamma(v_i, J)$ of various J values causes a vibrational satellite to be broadened by an amount

$$\begin{aligned} \delta\omega_\gamma &= (B_{v_i} - B_{v_i^0}) \langle J(J+1) \rangle_{av} \\ &\simeq \alpha_i \frac{kT}{\hbar B_i} (v_i - v_i^0). \end{aligned} \quad (7.5)$$

This broadening is considerably smaller than the Doppler width of the γ line and therefore may be neglected.

Let us next consider the low-pressure limit, where the γ resonances are narrow. Suppose the light wave interacts with the vibrational-rotational transition $(0, J_0) \rightarrow (1, J)$. In this case the rotational

quantum number of excited molecules, J , is fixed by the rotational-vibrational transition selected by the light wave and differs from J_0 by unity. For R -branch transitions $J = J_0 + 1$; for P -branch transitions $J = J_0 - 1$. The infrared transition $J = J_0$ (Q branch) is forbidden in the case of molecules with center-of-inversion symmetry.¹⁴ Both anharmonicity and change in rotational states of molecules interacting with the light wave lead to noncoincidence of resonant dips and peaks, since the frequencies of the transitions $(0, J_0) \rightarrow (v_i, J_0)$ for unexcited molecules differ somewhat from those of $(1, J) \rightarrow (v_i + 1, J)$ for excited molecules. For example, from Eqs. (6.19) and (7.1) expressions for the peak and dip frequencies of the first vibrational satellite can be obtained:

$$\omega_{\text{peak}}^{(1)} = \omega_{\gamma_0} \pm [\Delta_\gamma + \omega_i - 4\omega_i \chi_i - \alpha_i J(J+1)] + \frac{\omega_\gamma}{\omega_0} \Omega \quad (7.6)$$

and

$$\omega_{\text{dip}}^{(1)} = \omega_{\gamma_0} \pm [\Delta_\gamma + \omega_i - 2\omega_i \chi_i - \alpha_i J_0(J_0+1)] + \frac{\omega_\gamma}{\omega_0} \Omega. \quad (7.7)$$

The difference between these frequencies is

$$\Delta\omega_\gamma = 2\omega_i \chi_i + 2\alpha_i \begin{Bmatrix} J_0 + 1 \\ -J_0 \end{Bmatrix}, \quad (7.8)$$

where the upper term in brackets refers to the case of an R -branch infrared transition and the lower one to a P -branch transition. Figure 8 shows the "fine" structure of γ resonances caused by anharmonicity ($\omega_i \chi_i \gg \alpha_i J_0$). This splitting is observable only if $2\Gamma_{\text{nucl}} < \Delta\omega_\gamma$.

Anharmonicity and changes in rotation do not affect the structure of the resonance of the fundamental line produced by the $0 \rightarrow 0$ and $1 \rightarrow 1$ vibrational transitions, nor do they affect the resonant peak associated with the $1 \rightarrow 0$ transition.

The vibrational satellite intensities are also practically unchanged by anharmonicity and rotational-state changes, at least those due to transitions to vibrational states far from the molecular dissociation limit. Weak vibrational satellites also arise at frequencies associated with molecular transitions between the vibrational states of various normal modes.

During excitation and deexcitation of degenerate molecular vibrations a fine structure of γ resonances may appear owing to the degeneracy removal caused by rotation-vibration interaction. By way of illustration, the case of an XY_2 molecule in Sec. IV shows that the vibrational level $v_2 = 1$ is split by the value

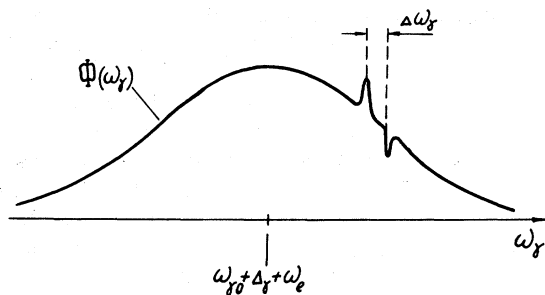


FIG. 8. Splitting of narrow γ resonances caused by vibrational anharmonicity and vibration-rotation interaction.

$$\Delta\nu_{\text{spl}} = q_2 J(J+1), \quad (7.9)$$

where q_2 is the constant of the l doublet. Such a splitting may show up only in high-resolution experiments with low-pressure gases when $2\Gamma_{\text{nucl}} < \Delta\nu_{\text{spl}}$.

Centrosymmetrical molecules with the simplest structure of γ resonances are considered in this paper. For other molecules, say, for diatomic ones, one should take into account from the very beginning the change in the molecular rotational state during emission and absorption of γ quanta. For diatomic molecules such calculations have been done in Ref. 15.

VIII. CONCLUSIONS

This work presents the theory of emission and absorption of nuclear γ radiation in molecules. The principal conclusions are as follows:

- (i) The emission and absorption lines of nuclear γ transitions in molecules have vibrational satellites arising from γ transitions accompanied by excitation of molecular vibrations by recoil. A sizable fraction of the γ transitions may be channeled into these vibrational satellites.
- (ii) The spectrum of nuclear γ transitions in a vibrationally excited molecule contains a vibrational satellite resulting from a nuclear γ transition with the vibrational energy transmitted to the nucleus or to the γ quantum. In this satellite, which has an intensity of up to 37% of that of the γ line of the free nucleus, the absorption and emission lines are brought closer together.
- (iii) As first discussed in Refs. 1 and 2, the spectrum of nuclear γ transitions in a low-pressure molecular gas subjected to an intense coherent field contains narrow γ resonances, the frequencies of which can be easily tuned over their Doppler profiles.

(iv) If the nucleus is located at the center of mass of a centrosymmetric molecule, then the narrow γ resonances are simple in structure. The structure remains rather simple even when the effects of vibrational anharmonicity and vibrational-rotational interactions are included.

Effect (i) considerably decreases the overlap of the Doppler profiles of the emission and absorption lines of nuclear γ radiation in molecules. It is essential to allow for the vibrational structure of γ transitions for a nucleus located in a molecule. Even to a first approximation this effect cannot be neglected.

Effect (ii) should be rather easy to observe in experiments in which molecular vibrations are excited by infrared laser radiation. For example, one can use γ transitions of the nuclei ^{186}Os ($\hbar\omega_\gamma = 137$ keV) or ^{188}Os ($\hbar\omega_\gamma = 155$ keV) in the OsO_4 molecule, which can be excited by CO_2 -laser radiation.

Effect (iii) is observable when a molecular gas of a very low pressure is subjected to infrared laser radiation. The classical estimates of Refs. 1 and 2 agree in order of magnitude with the quantum-mechanical formulas obtained here [Eq. (6.17)].

In much the same way one can show that excitation and deexcitation of electronic states of a molecule give rise to electronic-vibrational satellites of a γ transition. This increases the tuning region of γ resonances for a nucleus in a molecule with properly excited electronic states. Unlike the case of excitation and deexcitation of electrons in an atom,⁴ the strong electronic-vibrational coupling in molecules gives rise to electronic-vibrational satellites of large amplitude.

In this paper no consideration has been given to questions of experimental detection and application of the above mentioned effects. Some discussions are given in Refs. 1, 2, and 16. The possibility for detection of the modulation of γ radiation of Os nuclei, as the vibration ν_3 of the OsO_4 molecule is excited in the CO_2 -laser field, is considered in detail in Ref. 17. The probabilities for a vibrational-nuclear γ transition of an Os nucleus in an OsO_4 molecule are estimated here with allowance made for threefold degeneracy of the vibration ν_3 . On the basis of these estimates a conclusion has been drawn on the possibility of experimental observation of γ -radiation modulation both by detecting scattered γ quanta and by measuring the internal-conversion electron current. Here we wish only to point out that the proposed method of obtaining narrow frequency-tunable absorption and emission γ resonances opens the way for nuclear spectroscopy in a range of nuclear transitions that has not been investigated so far, namely, in the vicinity of nuclear

resonances:

$$10^{-9} < |\omega_\gamma/\omega_{\gamma_0} - 1| < 10^{-5}. \quad (8.1)$$

The Mössbauer effect¹⁸ is the most efficient method of studying the structure of resonances very close to a γ transition. For far-removed regions the universal techniques of γ spectroscopy¹⁹ are used. Between these boundaries, in the energy range from 10^{-3} to 10 eV, lies a region of great interest. If it ever becomes possible to detect the fine structure of nuclear levels in this region,

for example, by the techniques described in the present paper, it will be possible to interact with nuclear transitions of very low energies by optical laser radiation, with all ensuing effects.

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