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LOCALIZED LONG-LIVED VIBRATIONAL STATES IN MOLECULAR CRYSTALS

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It is pointed out that localized long-lived molecular vibrational states may exist in simple molecular crystals (H_2, O_2, N_2, NO, CO). Stabilization of such excitations is connected with the anharmonicity of the intramolecular vibrations. Their mobilities are estimated. The mobility is found to depend strongly on the excitation quantum number. For example, in an N_2 crystal the vibrational excitation of a molecule with $n = 7$ can go over to a neighboring molecule (completely, without any change) within a time of the order of ~1 sec. The finite time of such states is connected with the following processes: a) radiation involving a transition of the molecule to the $(n-1)$ level which occurs in the course of $\sim 10^5-10^6$ sec for N_2, O_2, H_2, and in the course of 1–10 sec for CO and NO; b) transition of the molecule to the $(n-1)$ level involving transfer of a quantum to a neighboring molecule and absorption of several phonons from the crystal. The process depends strongly on n and on the temperature. At $T = 30°K$ and $n = 7$ the time required for the process is ~10 sec. The role of rotation of hydrogen molecules in an H_2 crystal is discussed. It is also shown that in such crystals an excitation with $n = 1$ exists up to the moment of the emission of radiation. An experiment is proposed for detecting such states.

THE purpose of this paper is to point out the possibility of the existence of localized long-lived highly excited vibrational levels of molecules in molecular crystals. The physical reason for the localization of such states consists in the following. Let us consider two weakly interacting anharmonic oscillators. At the initial instant let the first oscillator be excited and the second at rest. If the excitation amplitude of the first oscillator is small, then the frequencies of both oscillators are close to one another. As a result of this, in a time inversely proportional to their coupling force the entire excitation energy will be transferred to the vibrations of the second oscillator[1]. If, on the other hand, the initial amplitude of the first oscillator is large, then the frequencies of the two oscillators differ strongly, since the eigenfrequency of the anharmonic oscillator depends on its amplitude. This leads to the breakdown of resonance and to strong inhibition of the energy transfer from one oscillator to the other.

There is a quantum analog of this phenomenon. Let, for instance, the first oscillator be at the n-th level and the other at the ground level. It is well known that the levels of an anharmonic oscillator are not equidistant, i.e.,

$$E_n - E_{n-1} \neq E_1 - E_0.$$

Therefore, by virtue of the law of conservation of energy the first oscillator cannot transfer to the second oscillator a quantum and thus transfer part of the energy (we note that this is completely feasible for harmonic oscillators). Thus, for anharmonic oscillators only the entire energy and not part of it can be transferred from one oscillator to the other. The probability of such a process is, of course, small and shall be estimated below. The indicated difficulties in the energy transfer do not appear in the case of an oscillator in the $n = 1$ state. All this is, of course, only valid in the case when the anharmonicity energy is larger than the coupling energy of the two oscillators. This is always true for typical molecular crystals (such as H_2, N_2, O_2, CO, etc.) In fact, the energy of the vibrational interaction of two neighboring molecules (w) can be estimated from the formula $w \sim \epsilon_v(\xi/r_e)^2$ where ϵ_v is the energy of the van der Waals interaction of the two molecules (~1 kcal) and ξ/r_e is the ratio (~0.1) of the amplitude of the zero-point vibrations to the bond length of the molecule. Taking this into account, we find $w \sim 5\ \text{cm}^{-1}$. At the same time, the anharmonicity energy for a not too strongly excited molecule amounts to $\sim 30\ \text{cm}^{-1}$.

For highly excited vibrational states with a large amplitude one should apparently take ξ to be the amplitude of these vibrations. However, in this case the anharmonicity energy would also increase correspondingly. The cited criterion is of a qualitative nature, a more rigorous expression for it is given below [see the explanation of (9)].

Let us proceed to a quantitative description of the phenomenon. Let us first consider two coupled classical anharmonic oscillators. The equations of motion are in this case of the form

$$\begin{aligned} \ddot{x}_1 + \omega_0^2 x_1 + \varepsilon\lambda x_1^3 &= \varepsilon\beta x_2, \\ \ddot{x}_2 + \omega_0^2 x_2 + \varepsilon\lambda x_2^3 &= \varepsilon\beta x_1. \end{aligned} \qquad (1)$$

Here x_1 and x_2 are the coordinates of the first and second oscillator, ω_0 are their zero-point vibrational frequencies, ϵ is a small parameter, and λ and β are parameters characterizing the anharmonicity and coupling force of the two oscillators respectively. Let the oscillator mass be unity. In order to solve the system (1), we make use of the method of averaging.[1] Let us introduce the new complex amplitudes p_1 and p_2 using the equalities

$$x_1 = p_1 e^{i\omega_0 t} + \overset{*}{p}_1 e^{-i\omega_0 t},$$

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[1] This phenomenon is often demonstrated by using as an example two identical pendulums suspended on a common thread.

$$x_2 = p_2 e^{i\omega_0 t} + p_2^{*} e^{-i\omega_0 t} \tag{2}$$

and the auxiliary conditions

$$\begin{aligned} \dot{p}_1 e^{i\omega_0 t} + \dot{p}_1 e^{-i\omega_0 t} &= 0, \\ \dot{p}_2 e^{i\omega_0 t} + \overset{*}{\dot{p}_2} e^{-i\omega_0 t} &= 0. \end{aligned} \tag{3}$$

Substituting p_1 and p_2 in (1) and averaging over vibrations with a frequency ω_0, we obtain the following equations:

$$\begin{aligned} i\frac{dp_1}{d\tau} + \gamma|p_1|^2 p_1 = p_2, &\quad \gamma = \frac{3\lambda}{\beta}, \\ i\frac{dp_2}{d\tau} + \gamma|p_2|^2 p_2 = p_1, &\quad \tau = \frac{t\varepsilon\beta}{\omega_0}. \end{aligned} \tag{4}$$

The first integral of motion of the system of equations (4) is connected with the law of conservation of energy

$$\begin{gathered} E = E_1 + E_2 = 2a^2\omega_0^2, \\ E_1 = 2|p_1|^2\omega_0^2, \quad E_2 = 2|p_2|^2\omega_0^2. \end{gathered} \tag{5}$$

Introducing new variables by means of the relations

$$p_1 = a\cos\theta e^{i\psi_1}, \quad p_2 = a\sin\theta e^{i\psi_2}, \quad \varphi = \psi_1 - \psi_2, \tag{6}$$

we obtain the following system of equations:

$$\begin{gathered} \dot{\theta} = \sin\varphi, \\ \varphi = a^2\gamma\cos 2\theta + 2\cos\varphi \operatorname{ctg} 2\theta. \end{gathered} \tag{7}$$

The absolute values of the phases ψ_1 and ψ_2 are obviously unimportant. This system can be integrated analytically with arbitrary initial conditions. For example, let the first oscillator have at t = 0 the maximum amplitude and zero velocity and let the second oscillator be at rest, i.e. $\psi_1(0) = \theta(0) = 0$, $\varphi(0) = \pi/2$, $\dot{\theta}(0) = 1$. We then have

$$\cos\varphi = -\frac{a^2\gamma}{4}\sin 2\theta, \tag{8a}$$

$$\tau = \int_0^\theta \frac{d\eta}{\sqrt{1 - (a^2\gamma/4)^2\sin^2 2\eta}}. \tag{8b}$$

The first of these equations limits the region of possible values of θ. For $a^2\gamma/4 \leq 1$, i.e. when the anharmonicity is weaker than the coupling between the oscillators, arbitrary values of θ are possible, including $\theta = \pi/4$—the energies of both oscillators are equal, and $\theta = \pi/2$—the entire energy has passed to the second oscillator. The time required for this is

$$T = \frac{\omega_0}{\varepsilon\beta}\int_0^{\pi/2} \frac{d\eta}{\sqrt{1 - (a^2\gamma/4)^2\sin^2\eta}} = \frac{\omega_0}{\varepsilon\beta} K\left(\frac{a^2\gamma}{4}\right). \tag{9}$$

K(y) is a complete elliptical integral of the first kind. If the anharmonicity is stronger than the coupling between the oscillators, i.e. $a^2\gamma/4 > 1$, θ cannot attain a value of $\pi/4$ and in this case the energy of the first oscillator is always larger than that of the second oscillator[2)].

As has already been noted, in the case in which we are interested $a^2\gamma/4 \gg 1$. This means that θ differs only little from zero. In this case $\theta \lesssim 2/a^2\gamma$ and the energy of the second oscillator $E_2 \leq 8\omega_0^4/a^2\gamma^2$ at all times, i.e., there is practically no energy transfer. The smallness of θ means that in the integrand of (8b) one can expand the sine, following which the time dependence of the amplitudes becomes harmonic. Consequently, in this case one could solve the problem by replacing the anharmonic oscillators with zero-point frequencies ω_0 by two harmonic oscillators with different frequencies $\omega_1 = \omega_0 + 3\epsilon\lambda a^2/2\omega_0$ and $\omega_2 = \omega_0$ with the same coupling between the oscillators. This rule can be used in the case of many interacting anharmonic oscillators. Replacing the system of anharmonic oscillators by a system of harmonic oscillators with frequencies $\omega_i = \omega_0$ for the oscillators which are not excited at the initial moment and $\omega_1 = \omega_0 + 3\epsilon\lambda a_1^2/2\omega_0$ for a strongly excited oscillator (excitation amplitude a_1), we arrive at the problem of the local vibration. It is precisely the local vibration mode which is excited at the initial moment. In this case too, there is no energy dissipation[3)].

Let us now go over to the quantum case. Although a transfer of the energy from a strongly excited anharmonic oscillator to an unexcited oscillator is classically impossible, such a transfer occurs for quantum oscillators (of course with a very low probability). Let us consider a system of two quantum anharmonic oscillators. The Hamiltonian of the system is of the form

$$H = \sum_{i=1,2}\left(\frac{p_i^2}{2\mu} + U(x_i)\right) + \beta x_1 x_2. \tag{10}$$

Let at the initial moment the first oscillator be in the n-th state with an energy E_n and a wave function $\psi_n(x_1)$. The time during which the entire energy is transferred (as has been indicated above it cannot be divided) from the first to the second oscillator is

$$\tau_{0n} = \frac{\hbar}{\beta} f_{0n}^{-2}, \quad f_{0n} = \int \psi_0(x)\, x\, \psi_n(x)\, dx. \tag{11}$$

The matrix element f_{on} is very small for large n and depends strongly on the form of the anharmonicity. For a harmonic oscillator only the element f_{10} differs from zero. Let us estimate it for a Morse potential.

In this case we have

$$U(x) = D_e[1 - \exp\{-\beta_e(x - r_e)\}]^2, \tag{12}$$

D_e is the dissociation energy and r_e is the equilibrium separation of a diatomic molecule. The energy spectrum is of the form

$$\begin{gathered} E_n = \hbar\omega_e[n + {}^1\!/_2 - x_e(n + {}^1\!/_2)^2], \\ \hbar\omega_e = \sqrt{2D_e\beta_e^2\hbar^2/\mu}, \quad x_e = \hbar\omega_e/4D_e. \end{gathered} \tag{13}$$

The parameter x_e characterizes the anharmonicity of the molecule. For the strongly anharmonic molecule H_2 the quantity $x_e = 0.027$ and for the weakly anharmonic molecule N_2 we have $x_e = 0.0061$.[2] For heavier molecules (I_2, K_2, etc) x_e can be even smaller. The matrix elements f_{on} for the Morse potential have been calculated in[3]. We present the answer for $nx_e \ll 1$:

$$|f_{0n}|^2 = \frac{\hbar n! x_e^{n-1}}{2\mu\omega_e n^2}.$$

Thus we have for the time of the transfer of the excitation from one molecule to another

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[2)] We note that although a value $\theta = \pi/2$ does not contradict (8a), it is in a classically unattainable region.

[3)] The solutions cited above are in formal agreement only for $t < 1/\epsilon^2\omega_0$; however, with the aid of theorems developed in [1] one can show that the exact solution on an infinite time interval will be close to the family of solutions of Eqs. (4).

$$\tau_{0n} = \frac{2\mu\omega_e n^2}{\beta n!\, x_e^{n-1}}. \tag{14}$$

Numerical estimates carried out for a series of crystals (see below) show that such excitations can move across the crystal (with a diffusion coefficient $D_n \sim l^2/\tau_{0n}$, l is a length of the order of the lattice parameter) for a considerable time without exchanging energy.

Let us enumerate certain processes which can lead to an energy redistribution, i.e., to a change in the quantum number n. First of all radiative transitions belong to such processes. For molecules with a small dipole moment (CO, NO, and others) the radiative lifetime for the n → n – 1 transition is about 1 sec and proportional to n. For molecules which have no dipole moment (such as O_2, H_2, and N_2) the dipole transition is forbidden and only the quadrupole transition is possible. The lifetime for the quadrupole transition n → n – 1 is proportional to n^2 which amounts to $\sim 10^6$–10^7 sec for the 1 → 0 transition.[4]

The most important processes facilitating a change of n are thermal transitions taking place with the absorption of several phonons corresponding to translational vibrations of the molecules. In fact, since $E_n - E_{n-1} < E_1 - E_0$ [see (13)], the process of transition of one quantum to a neighboring molecule is necessarily accompanied by absorption of phonons from the crystal. The radiationless transition n → n – 2, although energetically possible at zero temperature, is, however, most unlikely since it is connected with the production of a very large number of crystal phonons. The number of phonons produced N_{ph} can be estimated from the formula

$$N_F = \Omega / \omega_0 \sim 10 \div 20, \tag{15}$$

where Ω are the intramolecular vibration frequencies (~ 1–2×10^3 cm^{-1}) and ω_0 are the highest optical frequencies of the intermolecular vibrations of the crystal ($\sim 10^2$ cm^{-1}).

Let us estimate the probability of a thermal transition with the transfer of one quantum of energy to the neighboring molecule. We shall assume that the very high-frequency optical phonons are absorbed. It has been shown in the theory of radiationless transitions[5,6] that just such processes are the most probable. Accordingly we shall restrict ourselves to a consideration of translational vibrations of two diatomic molecules located in one unit cell of the crystal.

The Hamiltonian of such a system is of the form

$$H = \sum_{i=1}^{2} H_0(x_i) + H_q + U(x_1x_2)q. \tag{16}$$

Here $H_0(x_i)$ is the Hamiltonian of the individual molecule which has a quasiharmonic form (for example, with a Morse potential), H_q is the oscillatory Hamiltonian of intermolecular optical vibrations

$$H_q = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial q^2} + \frac{m\omega_0^2 q^2}{2}, \tag{17}$$

m is the reduced mass of the two molecules and q is some relative coordinate of these[4)].

The third term in (16) represents the Hamiltonian of the interaction of translational and intramolecular vibrations. We shall represent it in the following form:

$$U(x_1x_2)q = q(\alpha_1x_1 + \alpha_2x_2 + \beta_{11}x_1^2 + \beta_{22}x_2^2 + \beta_{12}x_1x_2), \tag{18}$$

α_1 and α_2 are linear coupling coefficient and β_{ij} are the anharmonic coupling coefficients of the intermolecular and intramolecular vibrations. The frequencies of the intramolecular vibrations are considerably higher than the frequencies of the intermolecular vibrations. As a result of this one can consider the motion of the molecular oscillators to be adiabatic. Their wave functions $\psi_s(x_1, x_2)$ and eigenvalues $E_s(q)$ will depend on q as a parameter. In first-order perturbation theory in $U(x_1x_2)q$ we obtain for them

$$E_s(q) = E_s^0 + qU_{ss},$$
$$\psi_s(x_1x_2; q) = \psi_s^0(x_1x_2) + q\sum_{s'} \frac{U_{ss'}}{E_s^0 - E_{s'}^0}\psi_{s'}^0(x_1x_2). \tag{19}$$

Here $\psi_s^0(x_1x_2)$ and E_s^0 are wave functions and eigenvalues of the Hamiltonian $\sum_i H_0(x_i)$ and

$$U_{ss'} = \iint dx_1\, dx_2\, \psi_s^{0*}(x_1x_2)U(x_1x_2)\psi_s^0(x_1x_2). \tag{20}$$

The complete wave function in the adiabatic approximation is the product of the wave functions of intra and intermolecular motion

$$\Phi_{sv}(x_1x_2; q) = \psi_s(x_1x_2; q)\varphi_v^s(q),$$
$$\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial q^2} + \frac{m\omega_0^2 q^2}{2} + E_s(q)\right]\varphi_v^s(q) = \left[\hbar\omega_0\left(v + \frac{1}{2}\right) + E_s^0\right]\varphi_v^s(q). \tag{21}$$

The nonadiabaticity operator giving rise to transitions in the system has the following matrix elements:

$$M_{vs}^{v's'} = -\frac{\hbar^2}{m}\frac{U_{ss'}}{E_s^0 - E_{s'}^0}\left(\int \varphi_v^s(q)\frac{\partial}{\partial q}\varphi_{v'}^{s'}(q)\,dq\right), \tag{22}$$

and the transition probability depends on them in the following manner[5,6]:

$$W_s^{s'} = \frac{2\pi}{\hbar Z}\sum_{v,\,v'} |M_{vs}^{v's'}|^2 e^{-E_{vs}/kT}\delta(E_{sv} - E_{s'v'}), \quad Z = \sum_v e^{-E_{vs}/kT}. \tag{23}$$

Evaluation of this sum yields the following result:

$$W_s^{s'} = \frac{\pi p[n(T)]^p\omega_0}{(p-1)!}\gamma\delta^{p-1},$$
$$p = \frac{E_s^0 - E_{s'}^0}{\hbar\omega_0}, \quad \gamma = \frac{\hbar|U_{ss'}|^2}{m\omega_0(E_s^0 - E_{s'}^0)^2}, \tag{24}$$
$$\delta = \frac{(U_{ss} - U_{s's'})^2}{2m\hbar\omega_0^3}, \quad n(T) = [e^{\hbar\omega_0/kT} - 1]^{-1}.$$

Here p represents the number of phonons which must be absorbed from the crystal in order that the transition from s to s′ be possible energetically. In general p is not an integer; therefore in order to satisfy the conservation law an acoustic phonon is usually also absorbed. Account of this circumstance would complicate the estimate considerably. Consequently, following[8], we shall assume that p can take on arbitrary values in accordance with (24).

In the initial state let the first oscillator be on the n-th level and the second one on the ground level; in the final state the first oscillator will be on the (n – 1) level and the second one on the first level. Then, using (13), we find

$$p = p_n = \frac{2x_e\Omega(n-1)}{\omega_0}, \quad \delta = \frac{\xi^4(\beta_{11} - \beta_{22})^2}{2\hbar m\omega_0^3},$$
$$\gamma = \frac{\xi^4\beta_{12}^2 n}{2\hbar m\omega_0^3 p_n^2}, \quad \xi^2 = \frac{\hbar}{\mu\Omega}. \tag{25}$$

[4)] q can also be the relative quasirotational coordinate.

The matrix elements $U_{ss'}$ are calculated in the harmonic approximation. The temperature dependence of the transition probability is given by the factor $[n(T)]^{p_n}$ for $\hbar\omega_0 \gg kT$ (this is true for all the crystals under consideration; on increasing the temperature they melt); it has an activation form

$$W_s^{s'} \sim \exp\left\{-\frac{E_s{}^0 - E_{s'}{}^0}{kT}\right\}. \tag{26}$$

We shall present numerical estimates of the lifetime of the n-th vibrational level relative to the thermal transition of one quantum to a neighboring molecule for the N_2 crystal. In the N_2 crystal there exist two optic intermolecular vibrations: a translational one with $\hbar\omega_0 = 69\ \text{cm}^{-1}$ and a quasivibrational one with $\hbar\omega_0 = 49\ \text{cm}^{-1}$.[9] For estimates we shall take the first frequency. The anharmonicity coefficients are estimated with the aid of derivatives of the Lennard-Jones potential of the N_2 molecule.[10] It was finally assumed that

$$\beta_{ij}\frac{\hbar}{\Omega\mu}\sqrt{\frac{\hbar}{m\omega_0}} \cong 1\ \text{cm}^{-1}.$$

Then $\delta \approx 10^{-4}$,

$$\gamma = \frac{\delta n}{2p_n{}^2}, \qquad W_n \cong \frac{\pi\omega_0 n\,[n(T)]^{p_n}}{2p_n!}\,\delta^{p_n}.$$

For example, for n = 7 and T = 30°K we have $W_7 \sim 10^{-1}$ sec. The transition time of the excitation to the neighboring molecule is $\tau_{07} = 1$ sec. This means that the excitation will complete ten jumps before it imparts one quantum to another molecule. At lower temperatures (for example, at helium temperatures) the temperature factor becomes very small so that the excitation energy is practically not dissipated before emission ($\sim 10^5 - 10^6$ sec). The estimates cited provide the limits of the existence of such excitations and the method of their stabilization. An analogous situation occurs also for O_2, CO, and NO crystals. The emission of the two latter molecules is rather rapid (~ 10 sec).

A very favorable situation for the existence of strongly excited vibrational states occurs in the H_2 crystal. This is connected, on the one hand, with the strong anharmonicity of the hydrogen molecule and, on the other hand, with the rotation of the hydrogen molecule in the crystal5). In this case the transfer of an energy quantum to a neighboring molecule is only possible if it has a large rotational energy. The energy which must be taken from the crystal is in this instance (for the n-th excitation) $\Delta E_n = 240\ \text{cm}^{-1}\ (n - 1)$. The transitions occur at the expense of the quadrupole-quadrupole interaction of H_2. Only changes of the rotational quantum number J by two units are possible in the first order in this interaction. The most probable transition is that with the following change of the quantum numbers of the oscillators: $(0, n|J, 0) \rightarrow (1, n - 1|J - 2, 1)$; the first two numbers in the parenthesis show the rotational and vibrational state of the first oscillator, and the following two numbers show the analogous states for the second oscillator. The transition from the state J = 0 to the state with J = 0 is impossible for the first oscillator, since it is forbidden for the quadrupole interaction. The probability of such a transition can be estimated in accordance with the formula

$$W_n{}^Q = \frac{2\pi V_Q{}^2}{\hbar B_e}\exp\left\{-B_e\frac{J(J+1)+2}{kT}\right\},$$

where $B_e \sim 60\ \text{cm}^{-1}$ is the rotational quantum of a hydrogen molecule, V_Q is the energy of the quadrupole-quadrupole interaction,[4] and J is determined from the law of conservation of energy

$$B_e[J(J+1) - (J-2)(J-1)] = 2\hbar x_e\omega_e(n-1) + 2B_e.$$

This equation can be approximately rewritten in the form $J \sim n$. Even for n = 2 this probability is smaller than the probability of a radiative transition (for $T < T_m = 14$°K). On the other hand, because of the large anharmonicity the excitations of the hydrogen molecule have a high mobility.

Let us note further that according to (24) the probability of a change of one intramolecular quantum into intermolecular vibrations is negligibly small, since in this case $p_n \sim 10-20$. [The temperature factor in (24) must be omitted.] Consequently, the excited state with n = 1 exists up to the emission of this quantum (even for polar crystals of the HC*l* type). The presence of such localized long-lived states can lead to the appearance of very narrow lines in the inelastic neutron scattering by such crystals.

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31

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5)Ya. B. Zel'dovich drew our attention to the important role of molecular rotation in the hydrogen crystal.