SOVIET PHYSICS JETP VOLUME 26, NUMBER 5 MAY, 1968

RESONANCE INTERACTION BETWEEN TWO IDENTICAL DIPOLE EMITTERS

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Submitted April 28, 1967

Zh. Eksp. Teor. Fiz. 53, 1630–1640 (November, 1967)

The problem of the radiation spectrum of two isotropic oscillators located at an arbitrary distance from each other is solved. Resonance dipole interaction between two two-level fixed centers is considered and values are obtained for the energies and widths of collective states corresponding to excitation of one of the centers. The symmetry of the collective-state wave functions is discussed. The structure of the energy levels of a diatomic molecule with two identical nuclei, one of which is in the excited state and the other in the ground state, is considered. It is shown that the level width of the excited nucleus depends on the quantum numbers characterizing the nuclear motion in the molecule. In this case the relative difference in the lifetimes for various states of the molecule at γ-transition energies on the order of 10–60 keV may reach several per cent.

In an earlier paper[1] the author solved the problem of the scattering of electromagnetic waves by an aggregate of immobile dipole centers, and considered the resonance scattering of electromagnetic waves by two identical isotropic oscillators located a finite distance R from each other. The present paper is a direct continuation of[1] and is devoted to the classical and quantum calculations of the emission spectrum of two identical dipole emitters.

The question of the emission from two identical isotropic oscillators was considered earlier in part by Podgoretskiĭ and Roĭzen[2] in the short-wave limit (see also[3], Sec. 32).

1. EMISSION OF TWO IDENTICAL ISOTROPIC OSCILLATORS

From the classical point of view, the scattering of electromagnetic waves should be regarded as emission resulting from induced oscillations of the dipole moment of the system under the influence of the electric field of the incident wave. It is well known that the resonant parameters characterizing the induced oscillations coincide exactly with the frequencies and the widths of the normal oscillations of the given system. In[1], in the investigation of the problem of resonant scattering, we considered in the long-wave approximation the induced oscillations of two isotropic oscillators that are resonantly coupled with each other by the radiation field. In considering the emission of electromagnetic waves by a system of two immobile dipole centers, we can use directly the method of[1] also for arbitrary wavelengths. The results reduce to the following:

A. If the dipole moments of two identical oscillators are parallel to each other and are perpendicular to the vector R, and the amplitudes and phases of their oscillations coincide, then the emission frequency $\omega^{(1)}$ and the spectral line width $\gamma^{(1)}$ are connected with the emission frequency ω_0 and the spectral line width γ of the isolated oscillator by the relations1)

$$\omega^{(1)} = \omega_0 + \frac{3}{4}\gamma\left[\left(\frac{1}{(kR)^3} - \frac{1}{kR}\right)\cos kR + \frac{1}{(kR)^2}\sin kR\right], \qquad (1)$$

$$\gamma^{(1)} = \gamma - \frac{3}{2}\gamma\left[\left(\frac{1}{(kR)^3} - \frac{1}{kR}\right)\sin kR - \frac{1}{(kR)^2}\cos kR\right], \qquad (2)$$

where

$$k = \omega_0 / c, \quad R = |\mathbf{R}| = |\mathbf{R}_1 - \mathbf{R}_2|.$$

B. If the dipole moments of the two oscillators are parallel to each other and perpendicular to the vector **R**, and the amplitudes of their oscillations are the same while the phases are opposite, then the emission frequency $\omega^{(2)}$ and the spectral line width $\gamma^{(2)}$ are

$$\omega^{(2)} = 2\omega_0 - \omega^{(1)}, \quad \gamma^{(2)} = 2\gamma - \gamma^{(1)}. \qquad (3)$$

C. If the dipole moments of the two oscillators are parallel to the vector **R**, and the amplitudes and the phases of their oscillations are the same, then the emission frequency $\omega^{(3)}$ and the spectral line width $\gamma^{(3)}$ are

$$\omega^{(3)} = \omega_0 - \frac{3}{2}\gamma\left[\frac{1}{(kR)^3}\cos kR + \frac{1}{(kR)^2}\sin kR\right], \qquad (4)$$

$$\gamma^{(3)} = \gamma + 3\gamma\left[\frac{1}{(kR)^3}\sin kR - \frac{1}{(kR)^2}\cos kR\right]. \qquad (5)$$

D. If the dipole moments of two oscillators are parallel to the vector **R**, and the amplitudes of their oscillations are the same, while the phases are opposite, then the emission frequency $\omega^{(4)}$ and the spectral-line width $\gamma^{(4)}$ are

$$\omega^{(4)} = 2\omega_0 - \omega^{(3)}, \quad \gamma^{(4)} = 2\gamma - \gamma^{(3)}. \qquad (6)$$

In the long-wave limit $kR \ll 1$ we arrive at the formulas of[1]:

$$\omega^{(1)} - \omega_0 = \omega_0 - \omega^{(2)} = {}^3/_4\gamma(kR)^{-3}, \qquad (7)$$

$$\gamma^{(1)} = 2\gamma, \quad \gamma^{(2)} = {}^1/_5\gamma(kR)^2, \qquad (8)$$

$$\omega^{(3)} - \omega_0 = \omega_0 - \omega^{(4)} = -{}^3/_2\gamma(kR)^{-3}, \qquad (9)$$

$$\gamma^{(3)} = 2\gamma, \quad \gamma^{(4)} = {}^1/_{10}\gamma(kR)^2. \qquad (10)$$

When $kR \gg 1$, expressions (1)–(3) coincide with the results of Podgoretskiĭ and Roĭzen[2]

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1) Relation (2) is given in the book of Faĭn and Khanin (see [3], Sec. 32).

$$\omega^{(1)}-\omega_0=\omega_0-\omega^{(2)}=-\frac{3}{4}\gamma\frac{1}{kR}\cos kR,$$

$$\gamma^{(1)}-\gamma=\gamma-\gamma^{(2)}=\frac{3}{2}\gamma\frac{1}{kR}\sin kR. \qquad (11)$$

In the case of oscillations of dipole moments parallel to the vector **R** (these oscillations were not considered in[2,3]) we get for $kR \gg 1$

$$\Delta\omega \sim \Delta\gamma \sim \gamma(kR)^{-2}.$$

For arbitrary oscillation-excitation conditions the emission spectrum of two identical isotropic oscillators consists, generally speaking, of four spectral lines. In the case of long waves ($\lambda = 1/k \gg R$), when $\Delta\omega \gg \gamma$, these lines are separated. When $\lambda \ll R$ they overlap and coalesce into a single line having a non-Lorentz form.

2. QUANTUM TREATMENT OF THE EMISSION OF TWO IMMOBILE DIPOLE CENTERS

The quantum analog of the isotropic oscillator is a two-level center, which has a spin 0 in the ground state and a spin 1 in the excited state, the parities of both levels being opposite. It is easy to see that in this case pure dipole electric emission takes place, on going over from the excited state to the ground state.

Assume that two such two-level centers are rigidly fixed at points $\mathbf{R}_1$ and $\mathbf{R}_2$, and that one of them is in the excited state and the other in the ground state. We denote the internal wave function of the excited state of the i-th center by $B_m(i)$, and that of the ground state by $A(i)$ (m—projection of the spin on the direction of the vector $\mathbf{R} = \mathbf{R}_1 - \mathbf{R}_2$, $i = 1, 2$). It is easy to see that in the absence of electromagnetic interaction between the centers we have six degenerate orthogonal states corresponding to the excitation of one of the centers. The degeneracy is lifted if the electromagnetic interaction is taken into account.

The wave functions of the quasistationary states, corresponding to definite energies and lifetimes, are of the form

$$\psi_m^{(\pm)}=2^{-1/2}(B_m(1)A(2)\pm A(1)B_m(2)) \qquad (m=+1,0,-1). \qquad (12)$$

Let us calculate the energy and the decay probability of these collective excited states. We assume that the energy of the resonant interaction between the centers is much smaller than the emission frequency $\omega_0 = (E_B - E_A)/\hbar$. In second order in the electromagnetic constant, the matrix element of the resonant interaction between the centers B and A, in accord with the principle of correspondence with classical theory[4], can be represented in the form

$$U_{AB;\,BA}=-\mathbf{E}_{AB}^{(1)}\mathbf{d}_{BA}^{(2)}, \qquad (13)$$

where $\mathbf{E}_{AB}(\omega_0)$ is a complex vector whose magnitude is equal to the Fourier component of the electric field that produces the dipole moment of the transition of the first center at the point where the second center is situated; $\mathbf{d}_{BA}^{(2)}$ is the dipole moment of the transition of the second center. We substitute in (13) the explicit form of $\mathbf{E}_{AB}(\omega_0)$ (see[5]), and obtain2)

$$U_{AB;\,BA}=-\left[k^2\frac{\mathbf{d}_{AB}^{(1)}\mathbf{d}_{AB}^{(2)*}}{R}e^{ikR}+(\mathbf{d}_{AB}^{(1)}\nabla)(\mathbf{d}_{AB}^{(2)*}\nabla)\frac{e^{ikR}}{R}\right], \qquad (14)$$

where

$$R=|\mathbf{R}-\mathbf{R}_2|, \quad k=\omega_0/c=E_B-E_A)/\hbar c.$$

We now write down the matrix elements (14) in the representation of the wave functions (12). In this representation, the matrix U is diagonal. We take into account the fact that the width of the excited level of the isolated center is $\gamma = 4k^3|d_{AB}|^2/3\hbar$ and does not depend on the magnetic quantum number m. Here $\mathbf{d}_{AB_{+1}}^{(1)} = \mathbf{d}_{AB_{+1}}^{(2)}$ and $\mathbf{d}_{AB_{-1}}^{(1)} = \mathbf{d}_{AB_{-1}}^{(2)}$ are perpendicular to each other and to the vector **R**, and the dipole moment of the transition $\mathbf{d}_{AB_0}^{(1)} = \mathbf{d}_{AB_0}^{(2)}$ is directed along the vector **R**. As a result we obtain

$$U_{+1}^{(\pm)}=U_{-1}^{(\pm)}=\pm U^{(\perp)}(R), \qquad U_0^{(\pm)}=\pm U^{(\parallel)}(R), \qquad (15)$$

where

$$U^{(\perp)}(R)=-\frac{3}{4}\gamma\left(\frac{1}{kR}+i\frac{1}{(kR)^2}-\frac{1}{(kR)^3}\right)e^{ikR}, \qquad (16)$$

$$U^{(\parallel)}(R)=-\frac{3}{2}\gamma\left(\frac{1}{(kR)^3}-i\frac{1}{(kR)^2}\right)e^{ikR}. \qquad (17)$$

The (+) sign in formula (15) pertains to the symmetrical state of (12), and the (–) sign to the antisymmetrical state.

The real and imaginary parts of the quantity

$$U_m^{(\pm)}=\pm\left(\Delta E_m-i\frac{\Delta\Gamma_m}{2}\right)=\pm\hbar\left(\Delta\omega_m-i\frac{\Delta\gamma_m}{2}\right) \qquad (18)$$

have the usual physical meaning: ΔE_m characterizes a shift of the energy level, similar to the Lamb shift[3,6], and $\Delta\gamma_m$ characterizes the change in the level width. It is easy to verify that for frequencies $\omega_m^{(\pm)}$ and the widths $\gamma_m^{(\pm)}$ of the dipole γ radiation, which takes place on going over from the symmetrical and antisymmetrical states of (12) into the ground state $\psi_0 = A(1)A(2)$, we obtain the relations (1)–(6) of the classical theory (here $\omega_{+1}^{(+)} = \omega_{-1}^{(+)} = \omega^{(1)}$, $\omega_{+1}^{(-)} = \omega_{-1}^{(-)} = \omega^{(2)}$, etc.).

We emphasize that this difference between $\gamma_m^{(\pm)} - \gamma$ and zero is due exclusively to the interference of the radiation of the two centers in the γ decay of the collective excited states (12). Indeed, direct calculation of the total emission probability per unit time in the transition from the state (12) to the ground state ψ_0 yields

$$\gamma_m^{(\pm)}=\frac{k^3}{4\pi\hbar}\int|\mathbf{d}_{AB_m}e^{i\mathbf{kR}/2}\pm\mathbf{d}_{AB_m}e^{-i\mathbf{kR}/2}|^2\,d\Omega_{\mathbf{k}}=$$

$$=\gamma\pm\frac{2}{\hbar}\operatorname{Im}\left(k^2\frac{|\mathbf{d}_{AB_m}|^2}{R}e^{ikR}+(\mathbf{d}_{AB_m}\nabla)(\mathbf{d}_{ABm}^{*}\nabla)\frac{e^{ikR}}{R}\right)$$

$$=\gamma-\frac{2}{\hbar}\operatorname{Im}U^{(\pm)}. \qquad (19)$$

Let us consider now two identical anisotropic centers

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2)Formula (14) for rigidly fixed dipole centers follows directly from the general expression for the retarded electromagnetic interaction [6]:

$$U_{AB;BA}=-\int\frac{j_{AB}(\mathbf{r}_1)j_{AB}^{*}(\mathbf{r}_2)}{|\mathbf{r}_1-\mathbf{r}_2|}\exp\left(i\frac{E_B-E_A}{\hbar c}|\mathbf{r}_1-\mathbf{r}_2|\right)d^3\mathbf{r}_1\,d^3\mathbf{r}_2,$$

where $j_{AB} = \{\mathbf{j}_{AB}, ic\rho_{AB}\}$ is the four-dimensional density of the transition current."

with oriented axes. We assume that the excited state of the isolated center is nondegenerate. The resonant electromagnetic interaction lifts the degeneracy between the symmetrical and antisymmetrical states of the two centers. For the energy shifts and for the change of the level widths we obtain, in accordance with the general result (14),

$$\Delta E^{(\pm)} - {}^1\!/_2 i \Delta\Gamma^{(\pm)} = \pm[U^{(\perp)}(R)\cos\theta + (U^{(\parallel)}(R) - U^{(\perp)}(R))\cos\psi_1\cos\psi_2]. \tag{20}$$

Here $U^{(\perp)}(R)$ and $U^{(\parallel)}(R)$ are determined respectively from formulas (16) and (17), θ is the angle between the dipole moments of the transitions $\mathbf{d}^{(1)}$ and $\mathbf{d}^{(2)}$, ψ_1 and ψ_2 are the angles between the vectors **R** and $\mathbf{d}^{(1)}$ and **R** and $\mathbf{d}^{(2)}$ respectively ($|\mathbf{d}^{(1)}| = |\mathbf{d}^{(2)}|$).

In the long-wave limits ($kR \ll 1$) we have

$$\Delta\omega^{(\pm)} = \pm\left[{}^3\!/_4\gamma\frac{1}{(kR)^3}(\cos\theta - 3\cos\psi_1\cos\psi_2)\right], \tag{21}$$

$$\gamma^{(\pm)} = \gamma(1 \pm \cos\theta) \mp {}^1\!/_{10}\gamma(kR)^2(2\cos\theta - \cos\psi_1\cos\psi_2). \tag{22}$$

If the directions of the axes of the two centers are parallel to each other, then, according to (22), $\gamma^{(-)} \ll \gamma^{(+)} = 2\gamma$, and in the case of pulsed irradiation the antisymmetrical state of the two centers is not excited in practice. The same takes place also in the case of isotropic centers (narrow resonances in the long-wave limit, see Sec. 1 and[1]).

If $\cos\theta \neq 1$, then $\gamma^{(+)} \sim \gamma^{(-)}$. In this case, for the same radiation direction, it can be readily seen that the polarizations of photons with frequencies $\gamma^{(+)}$ and $\gamma^{(-)}$ are mutually perpendicular. We note that in fact we have considered the model of the so-called "Davydov" splitting of the collective energy levels in an anisotropic crystal[7].

Assume now that at the initial instant of time the center 1 is in an excited state and center 2 in the ground state. Expanding the wave function B(1)A(2) in terms of the quasistationary states (12), we obtain the following expression for the probability of the excitation transfer from the first center to the second:

$$P(t) = {}^1\!/_4|\exp(-i\omega^{(+)}t - {}^1\!/_2\gamma^{(+)}t) - \exp(-i\omega^{(-)}t - {}^1\!/_2\gamma^{(-)}t)|^2. \tag{23}$$

In formula (23) t denotes the time elapsed from the instant when the delayed interaction (14) is switched on[3].

It is easy to see that in the long-wave limit multiple migration of excitation from one center to the other will occur during the lifetime of the excited state $t \sim 1/\gamma$, with a frequency $\sim\gamma(kR)^{-3}$.

3. SYMMETRY OF QUASISTATIONARY STATES OF THE EXCITED AND UNEXCITED CENTERS

The matrix (14) for the spins $s_B = 1$ and $s_A = 0$ can be represented in the form

$$U(R) = [C(R) + D(R)(\mathbf{Sn})^2]\mathcal{P}_{A\rightleftarrows B}, \tag{24}$$

where $\hat{\mathbf{S}}$ is the spin operator of the excited nucleus,

$$C(R) = U^{(\parallel)}(R) = -\frac{3}{2}\gamma\hbar\left[\frac{1}{(kR)^3} - i\frac{1}{(kR)^2}\right]e^{ikR}, \tag{25}$$

$$D(R) = U^{(\perp)}(R) - U^{(\parallel)}(R) = -\frac{3}{4}\gamma\hbar\left[\frac{1}{kR} + \frac{3}{(kR)^2}i - \frac{3}{(kR)^3}\right]e^{ikR} \tag{26}$$

(see (16) and (17)), $\mathcal{P}_{A\rightleftarrows B}$ is the exchange operator whose eigenfunctions are the states (12), and the corresponding eigenvalues are +1 and −1, and $\mathbf{n} = \mathbf{R}/R$.

It is easy to understand that in the general case (arbitrary spins s_A and s_B, and arbitrary multipolarity of the L radiation), the resonant interaction can always be represented in the form $U = \widetilde{M}\mathcal{P}_{A\rightleftarrows B}$, where $\widetilde{M}$ is an operator in the spin state of the two particles[4]. It follows therefore that the quasistationary states of a system of two immobile centers, one of which is excited, has a definite symmetry with respect to permutation of the first and second centers (or a definite value of the total energy spin, equal in this case to 1 or 0; see[3]). We emphasize that the last statement is valid in all cases when there exists a spontaneous transition B → A (not necessarily of electromagnetic character).

If the centers A and B are not immobile and take part in the oscillatory motion in the vicinity of the points $\mathbf{R}_1^{(0)}$ and $\mathbf{R}_2^{(0)}$, and the value of the resonant interaction (14) is much smaller than the distance between the neighboring vibrational levels s and s′ (t and t′), while the frequency of the γ radiation is much larger than this distance, the operator (24) for the nonstationary centers must simply be averaged over the wave functions of the quasistationary states of the system AB (see Sec. 4). The wave functions of the collective excited states have in this case the form

$$\psi_m^{(\pm)}(1,2) = 2^{-1/2}[B_m(1)A(2) \pm A(1)B_m(2)]\psi_s(\mathbf{R}_1 - \mathbf{R}_1^{(0)})\psi_t(\mathbf{R}_2 - \mathbf{R}_2^{(0)}) \tag{27}$$

(m—projection of the spin on the vector $\mathbf{R}_2^{(0)} - \mathbf{R}_1^{(0)}$). Formulas (24)—(26), when averaged over the functions of the type (27), describe, for example, the splitting of the energy levels of molecules with two identical nuclei, "frozen-in" as impurities in a solid.

It is easy to see that owing to the presence of the imaginary exponential (see (24)), the mean values C(R) and D(R) are exponentially small in the case when the amplitude of the oscillations is essentially larger than the wavelength of the γ radiation $\lambda\!\!\!^{-} = 1/k$. This denotes in fact that the change of the energy and of the lifetime of the excited nucleus can take place in principle in the case when the probability of the Mössbauer effect (γ transition without excitation of the bond) deviates noticeably from zero.

Let us consider now the question of the symmetry of the total wave function of non-identical particles, which depends on the spatial coordinates of both particles and

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[3] A simple analysis shows that, in accordance with the causality principle, the instant of excitation of one of the centers does not coincide with the instant of switching on the retarded interaction (14). The time difference between them is R/c. When $\gamma R/c \ll 1$, this difference is insignificant. We shall henceforth assume throughout that the inequality $\gamma \ll c/R$ is satisfied.

[4] In electric or magnetic radiation of arbitrary multipolarity L, the explicit form of the matrix $\widetilde{M}$ can be obtained with the aid of a method similar to that used by us in Sec. 2 for E1 transitions (see footnote[2]). In particular, in the case of magnetic dipole radiation the interaction $U = \widetilde{M}\mathcal{P}_{A\rightleftarrows B}$ has the same form (24) - (26) as before. In view of the complexity of the formulas for $L > 1$ (the corresponding expressions for pure electric and magnetic transitions of the same multipolarity coincide) we shall not present them here. We emphasize that, regardless of the multipolarity of the radiation, the resonance interaction between the centers has in the shortwave limit the order of magnitude γ/kR.

on the discrete internal coordinates5). If we permute the coordinates of the two particles, and then make the substitution A $\rightleftarrows$ B, then the quantum state of the system of two particles does not change. This operation is obviously equivalent to a permutation of the spatial and discrete internal coordinates. Repeating the reasoning which is customarily employed in such cases, we arrive at the conclusion that the total wave function of the particles A and B can be either symmetrical or antisymmetrical with respect to such a permutation.

It is easy to show that the spontaneous transition BA → AA + γ can take place only if the total wave function of the system BA has the same symmetry as the wave function of the two identical particles A. Since the system AB is unstable in the presence of the decay B → A + γ, we should take into consideration the fact that

a) the total wave function of the particles A and B is symmetrical, if A and B are bosons;

b) the total wave function of the particles A and B is antisymmetrical, if A and B are fermions.

Theorems a) and b) follow, under sufficiently general assumptions, from the formalism of quantum field theory6).

It should be noted that if the particles A and B form a bound system (for example, a molecule), then, in the presence of resonant electromagnetic interaction, their energy levels correspond to a relative-motion wave function of definite symmetry (parity):

$$\varphi(\mathbf{R}_1-\mathbf{R}_2)=\pm\varphi(\mathbf{R}_2-\mathbf{R}_1).$$

This is connected with the fact that, as shown above, the internal function of the quasistationary states of the particles A and B is symmetrical or antisymmetrical with respect to the substitution B $\rightleftarrows$ A in the presence of a spontaneous transition B → A. The total wave function can in this case be represented in the form of a product of three functions:

$$\Phi(1,2)=\psi_{\text{int}}^{(\pm)}\varphi(\mathbf{R}_1-\mathbf{R}_2)\chi(\mathbf{R}_{\text{c,m}}), \tag{28}$$

where $\chi(\mathbf{R}_{\text{c.m.}})$ is the wave function describing the motion of the center of mass $\mathbf{R}_{\text{c.m.}}$. The function χ, as can be readily seen, is symmetrical with respect to the complete permutation of the coordinates of the particles A and B: $\mathbf{R}_1 \rightleftarrows \mathbf{R}_2$, $M_A \rightleftarrows M_B$ (M—mass of the particle). It follows from (28) that the signs of the symmetry of the "internal" function ψ_{int} and of the function $\varphi(\mathbf{R})$ coincide in the case of integer spins, and are opposite in the case of half-integer spins.

4. CASE OF DIATOMIC MOLECULE

Let us consider now the structure of the energy levels of a free diatomic molecule, one of the nuclei of which (B) is in an excited state and the second (A) in the ground state. We assume that a pure E1 transition B → A + γ takes place, and results in the formation of a molecule with two identical nuclei.

We stipulate the satisfaction of the following inequalities:

$$\omega_0 \gg \Delta\omega_{\text{vib}} \gg \Delta\omega_{\text{rot}} \gg \gamma, \tag{29}$$

$$c/R_0 \gg \Delta\omega_{\text{vib}} \gg \Delta\omega_{\text{rot}} \gg \gamma, \tag{30}$$

$$(\hbar\omega_0/Mc^2)R_0/c \ll a_{\text{vib}} \sim \hbar/\sqrt{2\hbar\omega_{\text{vib}}M}. \tag{31}$$

Here ω_0 and γ are the frequency and the natural width of the γ radiation in the B → A + γ decay, $\Delta\omega_{\text{vib}}$ and $\Delta\omega_{\text{rot}}$ are the distances between the neighboring vibrational and rotational levels of the molecule (in $\sec^{-1}$), R_0 is the average distance between the nuclei A and B, M is the mass of the nucleus ($M_A \approx M_B = M$), and a_{vib} is the amplitude of the vibrations of the nuclei in the molecule.

The process whereby excitation is transferred from the nucleus B to the nucleus A can be visualized as being an aggregate of successive jolts: the nucleus B with coordinate $\mathbf{R}_1$ emits a photon and goes over into the nucleus A, while the nucleus A with coordinate $\mathbf{R}_2$ absorbs a photon and goes over into the nucleus B. When the conditions (29) and (3) are satisfied, the effective time of excitation transfer, the order of magnitude of which is $\tau = 2/\omega_0 + R_0/c$ ($1/\omega_0$ is the time required for a packet of γ quanta with dimensions $\sim\lambda\!\!\!^{-} = c/\omega_0$ to escape from the nucleus[10]), is negligibly small compared with the time characterizing the motion of the nuclei in the molecule. On the other hand, the inequality (31) denotes that we can neglect the change in the mutual placement of the nuclei in the molecule due to the fact that the nuclei are displaced during the course of the photon exchange by a distance $(\hbar\omega_0/Mc^2)R_0/c$.

Thus, in the approximation (29)–(31), the coordinates of the nuclei have practically no time to change during the effective time. This means that the interaction (24) for immobile centers should be regarded as an operator acting in the space of the wave functions of the relative motion of the nuclei $\varphi_{tK}(\mathbf{R})$ (t and K are the quantum numbers of the vibrational and rotational levels of the molecule). We can assume here that the interaction energy is $U \ll \hbar\Delta\omega_{\text{rot}} \ll \hbar\Delta\omega_{\text{vib}}$, and we can use perturbation theory.

The wave function of the quasistationary states of two nuclei in a diatomic molecule can be written in the form (28). Let us represent the function of the relative motion of the nuclei in the form of the product of the wave function of the vibrational motion $\varphi_t(R)$ ($R = |\mathbf{R}_1 - \mathbf{R}_2|$) and the angular function characterizing the rotation of the motion. We now average the interaction (24) over the vibrational motion. We obtain

$$\overline{U}_t = \pm\int C(R)+D(R)(\hat{\mathbf{S}}\mathbf{n})^2]|\varphi_t(R)|^2R^2dR = \pm[\overline{C}_t+\overline{D}_t(\hat{\mathbf{S}}\mathbf{n})^2], \tag{32}$$

where C(R) and D(R) are determined from formulas (25) and (26). In particular, assuming that $a_{\text{vib}} \ll R_0$, where R_0 is the distance between the equilibrium positions of the nuclei, we can write the following simple formulas for the vibrational ground state of the molecule

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5) These include the spin, charge, mass, etc. (cf. with the isotopic-spin formalism).

6) On the basis of the general theorems of local field theory (see [8,9]) it can be shown that in a number of cases, particularly if the process B → A + γ takes place, the commutation relations between the fields A and B must have a normal character: at integer spin the operators A and B commute in spatially separated points, and for half-integer spin they anticommute. From this follow directly the theorems a) and b). The author is grateful to A. B. Govorkov for calling his attention to [8,9].

$$\overline{C}_0 = C(R_0)\exp\left(-\frac{1}{4}\frac{\hbar\omega_0^2}{Mc^2\omega_{\text{vib}}}\right),$$

$$\overline{D}_0 = D(R_0)\exp\left(-\frac{1}{4}\frac{\hbar\omega_0^2}{Mc^2\omega_{\text{vib}}}\right), \qquad (33)$$

where $C(R_0)$ and $D(R_0)$ have respectively the form (25) and (26). According to Sec. 3, the plus sign in formula (32) corresponds to an even function $\varphi_{tK}(\mathbf{R})$, and the minus sign to an odd function. In the case of Σ terms (zero projection of the electron angular momentum on the molecule axis[11]) the sign of expression (32) is determined by the factor $(-1)^K$, where K is the rotational moment of the molecule.

As a result of averaging the operator (32) over the rotation of the molecule we obtain after simple transformations

$$\overline{U}_{tK} = \pm[\overline{C}_t + \overline{D}_t\{2b_K + 2a_K(\hat{\mathbf{S}}\hat{\mathbf{K}})^2 + a_K(\hat{\mathbf{S}}\hat{\mathbf{K}})\}], \qquad (34)$$

where

$$a_K = -\frac{1}{4K(K+1)-3}(1-d_K), \quad b_K = \frac{1}{3}(1-2a_KK(K+1)), \qquad (35)$$
$$d_K = 3\Lambda^2/K(K+1).$$

Here $\hat{K}$ is the operator of the conserving angular momentum of the molecule, equal to the sum of the angular momenta of the electrons and of the orbital angular momentum of the nuclei, and Λ is the projection of the electron angular momentum on the molecule axis.

The eigenvalues of this matrix depend on the total angular momentum of the molecule $\hat{\mathbf{J}} = \hat{\mathbf{K}} + \hat{\mathbf{S}}$. We obtain finally for the energy shift and for the change in the width of the nuclear levels in a diatomic molecule the formula

$$\Delta E_{tKJ} - {}^1\!/_2 i\hbar\Delta\gamma_{tKJ} = \pm[\overline{C}_t + \overline{D}_tL_{JK}]. \qquad (36)$$

When $K \gg 1$, as can be readily seen, $L_{K+1,K} = L_{K-1,K} = 1/2$ and $L_{KK} = 1$. Consequently, at high temperatures (in practice, room temperatures), we can separate four groups of closely-lying levels of the molecule AB, each of which corresponds to a definite lifetime of the excited nucleus.

In the case of Σ terms ($\Lambda = 0$) of the square bracket in (36) is preceded by the factor $(-1)^K$. With this,

$$L_{K+1,K} = \frac{1}{2} + \frac{K - {}^1\!/_2}{4K(K+1)-3}, \quad L_{K-1,K} = \frac{1}{2} - \frac{K + {}^1\!/_2}{4K(K+1)-3},$$
$$L_{KK} = 1. \qquad (37)$$

We emphasize that the change $\Delta\gamma_{tKJ}$ of the level width includes interference effects (see Sec. 2) due to the γ transitions of the molecule BA in the state with quantum numbers t, K, and J, in all possible states of the molecule with two identical nuclei A.

According to (36), the resonance interaction between the nuclei A and B produces, besides the ordinary quadrupole interaction between the nucleus and the electrons of the molecule, also a hyperfine splitting of the molecular levels. Each component of the hyperfine structure has a different lifetime. Moreover, the nuclear interaction considered by us obviously makes an appreciable contribution to the Λ-doubling, i.e., to the splitting of the "positive" and "negative" levels of the molecule at $\Lambda \neq 0$ (see[11]).

Let us estimate the magnitude of the effects indicated by us. From relations (39)—(41) it follows that when $K \sim 10$ and $M/m_e \sim 10^5$ (m_e—electron mass), formula (36) is valid under the conditions

$$10^{22}\,\text{sec}^{-1} \gg \omega_0 \gg 3\cdot 10^{13}\,\text{sec}^{-1}(\sim 0{,}03\,\text{eV}),$$
$$\gamma \ll 10^{12}\,\text{sec}^{-1}(\sim 10^{-3}\,\text{eV}).$$

It follows from (33) that the quantities $\overline{C_0}$ and $\overline{D_0}$ become exponentially small if $\omega_0 a_{\text{vib}}/c \gg 1$. It is easy to understand that this conclusion has a general character. Thus, if the probability of the excitation of the vibrational levels of the molecule in γ decay (including dissociation) is close to unity, all the effects considered above, as expected, vanish. Consequently, we should confine ourselves to the following γ-radiation frequencies

$$\omega_0 \lesssim c/a_{\text{vib}} \sim 10^{20}\,\text{sec}^{-1}(\sim 60\,\text{keV}). \qquad (38)$$

From relations (24)—(26), (32), and (36) it follows that when condition (38) is satisfied and $\omega_0R_0/c > 1$ we have

$$\Delta E_{tKJ} \sim \hbar\Delta\gamma_{tKJ} \sim \hbar\gamma c/\omega_0R_0. \qquad (39)$$

We note that γ is equal to the radiative width of the excited nucleus. Taking into account internal conversion of the electrons we get

$$\Delta E_{tKJ} \sim \hbar\Delta\gamma_{tKJ} \sim \hbar\frac{\gamma_{\text{tot}}}{1+\alpha_{\text{conv}}}\frac{c}{\omega_0R_0}, \qquad (40)$$

where α_{conv} is the internal-conversion coefficient, $\gamma_{\text{tot}} = T_{1/2}^{-1}\ln 2$, and $T_{1/2}$ is the half-life of the excited nucleus in the isolated atom.

According to (40), when $\omega_0R_0/c > 1$ (this requirement is satisfied by all the known γ transitions), the corrections to the energy levels of the molecule are very small[7]. It must be noted, however, that when $\gamma > 10^6\,\text{sec}^{-1}$ the resonant interaction between the nuclei makes the main contribution to the Λ doubling of the Π terms of the molecule. Of greatest interest is the dependence of the lifetime of the excited nucleus on the quantum numbers of the molecule. For example, in the case of the $\text{Br}^{*80}\text{Br}^{80}$ molecule (E1 transition with energy 37 keV, $T_{1/2} \sim 4\times 10^{-9}$ sec[12], $\alpha_{\text{conv}} \approx 1.5$) the relative lifetime difference for the Σ states with rotational angular momenta K and K + 1 is approximately 5%. Effects of this kind can in principle be observed experimentally (for example, by the delayed-coincidence method).

We note that although we have considered in detail only the case of resonant interaction between nuclei with spins $s_A = 0$ and $s_B = 1$, the main conclusions of the present paper remain in force for all values of the spins s_A and s_B. In particular, for the case $\lambda\!\!\!^{-} \ll R_0$ the estimates (39) and (40) are valid for any radiation multipolarity (see the footnote[4]). We can speak here not only of diatomic molecules (free or with oriented axes), but of all chemical compounds containing two identical atoms (generally speaking, there may be several such atoms), whose nuclei have low-lying excited levels. There are several dozen known "Mössbauer" γ transitions with energy smaller than 60—70 keV[12]. In particular, we can expect that in the compounds $\text{Fe}^{*57}\text{Fe}^{57}\text{O}_3$

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[7] When $\omega_0R_0/c < 1$, the following estimates are valid:

$$\Delta E_{tKJ} \sim \gamma(c/\omega_0R_0)^3, \quad \text{Im}\,\overline{C}_t \sim \gamma, \quad \text{Im}\,\overline{D}_t \sim \gamma(\omega_0R_0/c)^2.$$

In the limit $\omega_0R_0/c < 1$, the radiative width $\gamma_{tKJ} \approx 2\gamma$ (regardless of the quantum numbers of the molecular level).

(M1—E2 transition with energy 14 keV, $T_{1/2} = 1.15 \times 10^{-8}$ sec, $\alpha_{\text{conv}} = 15$), $Al^{*28}Al^{28}(SO_4)_3$ (M1—E2 transition with energy 31 keV, $T_{1/2} = 2.3 \times 10^{-9}$ sec, $\alpha_{\text{conv}} = 2$), and $As^{*72}As^{72}$ (E1 transition with energy 46 keV, $T_{1/2} = 1.5 \times 10^{-8}$ sec, $\alpha_{\text{conv}} \approx 1$), the difference between the lifetimes of the excited nucleus for different molecular levels will amount to 1—5% (we are using the data from the tables of[12,13]).

The author is deeply grateful to M. I. Podgoretskiĭ for interest in the work and for valuable discussions.

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Translated by J. G. Adashko
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