

## TWO-PHOTON DISSOCIATION OF MOLECULES

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The problem is considered of the two-quantum photo-dissociation of diatomic heteronuclear molecules. A method is proposed for taking into account the complete vibration-rotational spectrum of such molecules for arbitrary two-photon transitions. The method is based on the application of the explicit expression of the Green's function for the internuclear potential. The nuclear potential curve is approximated by the Kratzer potential  $V(r) = A/r^{+2} - B/r$ . An analytic expression is derived for the two-photon dissociation cross section, which occurs without change in the ground electron term; the expression is valid for arbitrary values of the vibrational and rotational quantum numbers of the initial state.

1. A large number of theoretical and experimental papers have appeared to date which are devoted to the interaction of laser radiation with atoms. It is evident that the description of the interaction of intense electromagnetic radiation with molecules would also be of interest from other points of view. Theoretically, many-photon transitions of atoms in a strong field have been considered on the basis of the simplest atom—the hydrogen atom, which permits the most complete solution of the problem. At the same time, even very simple molecules, such as the hydrogen-molecule ion  $H_2^+$ , represent complicated quantum-mechanical systems with electronic, vibrational, and rotational degrees of freedom. Moreover, the solution of the many-photon problem for molecules is connected with the necessity of especially careful summation over the complete set of intermediate states, including the continuous spectrum. Approximate calculation of the matrix elements (summation over several “near resonance” intermediate levels) cannot be regarded as satisfactory even for atoms, let alone a system of molecular levels that are significantly more dense.

In 1959, Schwartz and Tiemann<sup>[1]</sup> first initiated the accurate calculation of the components of the matrix elements of second order for the hydrogen atom. These concerned not only the method of numerical integration of the inhomogeneous differential equations of second order,<sup>[2]</sup> but also the equivalent analytic expressions<sup>[3]</sup> based on the use of the integral representation of the Coulomb Green's function.<sup>[4]</sup> We shall show that similar methods can also be used for calculation of some two-photon processes in molecules.

In the present work, the two-quantum photodissociation of heteronuclear two-atomic molecules is considered using as an example a model that describes satisfactorily the vibrational-rotational states of the ground electron term  $^1\Sigma^+$  and results in an accurate analytic solution for arbitrary vibrational ( $v$ ) and rotational ( $K$ ) quantum numbers of the initial state. It is assumed that the two-quantum transition of the molecule to the continuous spectrum takes place without change in the electron state; the complete vibrational-rotational spectrum of the ground electron term is taken as the intermediate level. The contribution of the excited electron levels, if

necessary, can be considered similarly. We note that the possibility of many-photon dissociation of the molecules from transition to the continuous spectrum of the ground electron term was considered previously in the example of the one-dimensional Morse oscillator.<sup>[5,6]</sup> The electron state of the molecule in the vibration process is regarded only as changing adiabatically with changing distance  $r$  between the nuclei. As the effective field of the electrons and nuclei we use in the present work the potential  $V(r) = A/r^2 - B/r$ . The values of the parameters  $A$  and  $B$  are determined by the dissociation energies of the molecule  $D$  and the half-width of the potential well  $\Delta$  (or by the equilibrium distance between the nuclei  $r_0 = \Delta/2\sqrt{2}$ )  $A = D\Delta^2/8$ ,  $B = D\Delta/\sqrt{2}$ .

The potential  $V(r) = A/r^2 - B/r$  (the so-called Kratzer potential) plays a decisive role in the theory of band spectra in the old quantum mechanics and, along with the Morse potential  $V(r) = D[e^{-2\alpha(r-r_0)} - 2e^{-\alpha(r-r_0)}]$  is useful for the modern quantum-mechanical consideration of molecules.<sup>[7,8]</sup> The method proposed here for calculating the probabilities of two-photon transitions in molecules, based on the use of the explicit expression for the Green's function of the internuclear potential, is also applicable for the Morse potential<sup>[9,10]</sup> and can be employed for specific calculations. The potential  $V(r) = Ar^{-2} - Br^{-1}$  allows us to take accurate account of the simultaneous presence of vibrations and rotations in the set of eigenfunctions and in the energy spectrum, and also to clarify the effect of the rotational spectrum of the molecule on the probability of one or another multi-quantum transition. The Morse potential leads to an exactly-solved eigenvalue problem only in the case  $K = 0$ . Therefore, the practical use of the extra parameter  $r_0$  in the Morse potential, which permits us to change the distance between neighboring rotational levels  $\hbar^2/mr_0^2$  for fixed values of  $D$  and  $\alpha$  and, consequently, of the vibrational quantum  $\hbar\omega/2$ , is shown to be impossible.

We note also that for molecules dissociating into ions, the potential  $V(r) = Ar^{-2} - Br^{-1}$  gives the necessary asymptotic behavior for large distances between the nuclei, and our analysis refers, strictly speaking, to this case. In the general case, the parameters  $A$  and  $B$  are determined by the values of the dissociation energy and the half-width  $\Delta$  of the potential well. Bound-bound

processes and processes for which the final state belongs to the electron repulsion term are considered analogously.

2. In the non-relativistic dipole approximation, the interaction of the molecule with a plane electromagnetic wave has the form  $\mathcal{E}_0(\boldsymbol{\epsilon} \cdot \mathbf{d}) \cos \omega t$ , where  $\mathcal{E}_0$  is the amplitude,  $\omega$  the frequency,  $\boldsymbol{\epsilon}$  is the unit polarization vector of the radiation, and  $\mathbf{d} = \sum e_i \mathbf{r}_i$  is the dipole moment operator of the molecules. We represent the wave function of the molecule in the Born-Oppenheimer approximation in the form of the product of the wave function  $\psi$  of the electrons for a given distance  $\mathbf{r}$  between the nuclei and the nuclear wave function  $\xi(\mathbf{r})$ . The differential cross section of two-photon dissociation is determined in second-order perturbation theory and, after integration over the coordinates of the electrons, has the form

$$\frac{1}{I} \frac{d\sigma}{d\Omega} = \frac{mp\omega}{c^2 \hbar^3} \left| \sum_i \frac{(\bar{\mathbf{d}}\boldsymbol{\epsilon})_{i1}(\bar{\mathbf{d}}\boldsymbol{\epsilon})_{i0}}{E_0 - E_i + \hbar\omega} \right|^2, \quad (1)$$

where  $mpd\Omega/(2\pi\hbar)^3$  is the density of final states of the continuous spectrum, corresponding to the flight of the dissociation products with momenta equal to  $\mathbf{p}$  relative to the center of gravity in an element of solid angle  $d\Omega$ ,  $p^2/2m = E_0 + 2\hbar\omega > 0$  is the kinetic energy of the dissociation products in the center-of-mass system,  $E_0$  is the initial energy,  $m = m_1 m_2 / (m_1 + m_2)$  is the reduced mass of the molecule,  $I = c \mathcal{E}_0^2 / 8\pi$  is the intensity of the incident radiation, and  $\bar{\mathbf{d}}(\mathbf{r}) = \int \psi^* \mathbf{d} \psi d\tau_e$  is the mean dipole moment along the axis of the molecule. The matrix elements in (1) are computed only over the nuclear wave functions; summation over the intermediate states is carried out over the complete vibrational-rotational spectrum of the ground electron term, including integration over the states of the continuous spectrum.

We limit ourselves to the case of a linear dependence of the dipole moment  $\bar{\mathbf{d}}$  of the molecule on the distance between the nuclei or on the vibrational coordinates  $(\mathbf{r} - \mathbf{r}_0)$ :  $\bar{\mathbf{d}} = \mathbf{d}_0 + \mathbf{e}_{\text{eff}} r$ , where  $\mathbf{e}_{\text{eff}}$  is the effective charge of the polar bond.<sup>1)</sup> For transitions connected with the change in the vibrational (and with it, the rotational) state of the molecules, the zero term of the expansion drops out of the matrix element in view of the orthogonality of the wave functions of the vibrational motion. For convenience, we express the calculation of the matrix elements  $(\boldsymbol{\epsilon} \cdot \mathbf{r})_{mn}$  in (1) in terms of the matrix elements of the momentum operator  $\mathbf{p} = -i\hbar\nabla_{\mathbf{r}}$ :

$$\frac{1}{I} \frac{d\sigma}{d\Omega} = \frac{\hbar p e_{\text{eff}}^4}{c^2 m^3 \omega^3} \left| \iint \nabla_{\xi_i}^* (\mathbf{r}') \nabla_{\xi_0}(\mathbf{r}) G(\mathbf{r}, \mathbf{r}' | E_0 + \hbar\omega) d^3 r d^3 r' \right|^2, \quad (2)$$

where

$$G(\mathbf{r}, \mathbf{r}' | E) = \sum_i (E_i - E)^{-1} \xi_i^*(\mathbf{r}) \xi_i(\mathbf{r}') \quad (3)$$

is the Green's function of the internuclear potential  $V(\mathbf{r})$ .

3. Let us first consider the dependence of the cross section on the angular variables (the direction of flight of the dissociation products) and also the selection rules for rotational ( $K$ ) and magnetic ( $M$ ) quantum numbers of the molecule. The wave function describing the

motion of the nuclei in the initial state has the form  $\xi_0(\mathbf{r}) = R_{\nu K}(\mathbf{r}) Y_{KM}(\theta, \varphi)$ , where  $Y_{KM}$  are normalized spherical harmonics, and the radial part  $R_{\nu K}(\mathbf{r})$  of the nuclear wave function satisfies the equation

$$\frac{d^2 R}{dr^2} + \frac{2}{r} \frac{dR}{dr} + \frac{2m}{\hbar^2} \left[ E_0 - \frac{\hbar^2}{2m} \frac{K(K+1)}{r^2} - \frac{A}{r^2} + \frac{B}{r} \right] R = 0 \quad (4)$$

and the normalization condition

$$\int_0^\infty R_{\nu K}^2(r) r^2 dr = 1, \quad E_0 = E_{\nu K}.$$

The wave function  $\xi_f(\mathbf{r})$  of the final state is determined by expansion over the partial waves and has the asymptotic form of a plane wave of unit amplitude plus a diverging spherical wave<sup>[11]</sup>

$$\xi_f(\mathbf{r}') = \frac{(2\pi)^{3/2}}{Vq} \sum_{K'M'} i^{k'} \exp\{-i\eta_{K'}\} R_{qK'}(r') Y_{K'M'}(\theta', \varphi') Y_{K'M'}^*(\Theta, \Phi). \quad (5)$$

Here  $R_{qK}$  is the regular solution of Eq. (4) for  $E = \hbar^2 q^2 / 2m > 0$ , normalized in the energy scale,  $\eta_{K'}$  is the phase,  $\Theta$  and  $\Phi$  are the polar and azimuthal angles defining the direction of flight of the particles in a coordinate frame whose polar axis is directed along the polarization vector of the incident radiation. We now use the expansion of the Green's function (3) in partial waves

$$G(\mathbf{r}, \mathbf{r}' | E) = \sum_{K_i M_i} Y_{K_i M_i}^* \left( \frac{\mathbf{r}}{r} \right) Y_{K_i M_i} \left( \frac{\mathbf{r}'}{r'} \right) g_{K_i}(\mathbf{r}, \mathbf{r}' | E), \quad (6)$$

where  $g_{K_i}(\mathbf{r}, \mathbf{r}' | E)$  is a solution of the inhomogeneous equation

$$\left\{ \frac{\hbar^2}{2m} \left[ \frac{d^2}{dr^2} + \frac{2}{r} \frac{d}{dr} - \frac{2mA}{\hbar^2 r^2} - \frac{K_i(K_i+1)}{r^2} \right] + \frac{B}{r} + E \right\} g_{K_i}(\mathbf{r}, \mathbf{r}' | E) = \frac{\delta(\mathbf{r} - \mathbf{r}')}{rr'}. \quad (7)$$

Then, after averaging over the magnetic quantum number  $M$  of the initial state ( $\nu, K$ ), we can represent (2) in the form

$$\frac{1}{I} \frac{d\sigma}{d\Omega} = \frac{(2\pi)^3 \hbar^2 e_{\text{eff}}^4}{c^2 \omega^3 m^3 (2K+1)^2} \left| \sum_M \left[ M_{K+1, K} e^{i\eta_K} a^2(K) Y_{K, M}(\Theta, \Phi) - M_{K+1, K+2} e^{i\eta_{K+2}} a(K) b(K+2) Y_{K+2, M}(\Theta, \Phi) + M_{K-1, K} e^{i\eta_K} b^2(K) Y_{K, M}(\Theta, \Phi) - M_{K-1, K-2} e^{i\eta_{K-2}} a(K-2) b(K) Y_{K-2, M}(\Theta, \Phi) \right] \right|^2, \quad (8)$$

where

$$a(K) = \left[ \frac{(K-M+1)(K+M+1)}{(2K+1)(2K+3)} \right]^{1/2}, \quad b(K) = \left[ \frac{K^2 - M^2}{4K^2 - 1} \right]^{1/2}, \quad (9)$$

$$M_{K_i K'}(\omega) = \iint_0^\infty dr dr' (rr')^2 g_{K_i}(\mathbf{r}, \mathbf{r}' | E_{\nu K} + \hbar\omega) \cdot$$

$$\times \left[ \frac{dR_{\nu K}(r)}{dr} + \lambda(K_i) \frac{R_{\nu K}(r)}{r} \right] \left[ \frac{dR_{qK'}(r')}{dr'} + \lambda'(K') \frac{R_{qK'}(r')}{r'} \right], \quad (10)$$

and the parameters are  $\lambda(K+1) = -K$ ,  $\lambda(K-1) = K+1$ ,  $\lambda'(K' = K_i + 1) = K' + 1$ ,  $\lambda'(K' = K_i - 1) = -K'$ . Equation (8) expresses the selection rule  $\Delta M = 0$ ,  $\Delta K = \pm 1$  and the distribution of the dissociation products over the angles of flight. If the kinetic energy of the flight of the dissociation products is much greater than the initial rotational energy of the molecule<sup>2)</sup> ( $qr \gg K + 1/2$ ), the

<sup>1)</sup> The possibility of the expansion of  $\bar{\mathbf{d}}$  in powers of the vibrational coordinates  $(\mathbf{r} - \mathbf{r}_0)$  is connected with the assumption of the smallness of the vibrations. For molecules which dissociate into ions, a linear dependence of  $\bar{\mathbf{d}}$  on  $\mathbf{r}$  evidently occurs even for large distances. The general case of a nonlinear dependence of  $\bar{\mathbf{d}}$  on  $\mathbf{r}$  is considered in [9, 10].

<sup>2)</sup> In this case, flight takes place principally in the direction of the molecular axis. Such an approximation, which is valid far from the threshold of the bound-bound transition, is called the axial recoil approximation.

factor  $iK' \exp(-i\eta_{K'})$  with accuracy to terms  $(K' + 1/2)^2 / (qr)^2$  can be regarded as independent of  $K'$ .<sup>[12]</sup> Since the radial wave functions depend only very weakly on  $K$ , then, for example, for  $K = 0$ , by assuming  $M_{10} = M_{12}$ , we obtain the following angular dependence:

$$d\sigma \sim \cos^4 \Theta d\Omega.$$

4. The radial part of the nuclear wave function of the initial bound state of the molecule has the form<sup>[11,8]</sup>

$$R_{vK}(r) = \frac{2\sqrt{\Gamma(2s+2+v)}}{a^{1/2}n^2\sqrt{\Gamma(2s+2)}} \left(\frac{2r}{an}\right)^s e^{-r/an} \Phi\left(-v, 2s+2, \frac{2r}{an}\right), \quad (11)$$

where  $s$  is the positive root of the equation

$$s(s+1) = 2mA/\hbar^2 + K(K+1), \quad (12)$$

$\Phi$  is the confluent hypergeometric function,  $n = s + 1 + v$ ,  $v = 0, 1, 2, \dots$  is the vibrational quantum number,  $a = \hbar^2/Bm$  is the Coulomb unit of length. The energy levels of the discrete spectrum are determined by the formula

$$E_{vK} = -\frac{B^2m/\hbar^2}{2n^2} = -D + \hbar\omega_0\left(v + \frac{1}{2}\right) - \frac{3}{2\epsilon}\hbar\omega_0\left(v + \frac{1}{2}\right)^2 + \frac{\hbar^2}{2J}\left(K + \frac{1}{2}\right)^2 + \dots \quad (13)$$

The latter equation is obtained by an expansion of  $E_{vK}$  in a series in the small parameter  $\epsilon^{-1} = \hbar/\sqrt{2m\bar{A}}$ . The parameter  $\epsilon$  is equal to twice the ratio of the vibrational quantum for  $v = 0$  ( $\hbar\omega_0/2$ ) to the rotational part for  $K = 1$  ( $\hbar^2/J$ ), where  $\omega_0 = (4/\Delta)(D/m)^{1/2}$ ,  $J = mr_0^2$  is the moment of inertia of the molecule. For most molecules,  $\epsilon \sim 10^2$ . From states with a given rotational quantum number  $K$  and consequently with given  $s$ , the ground state is the state with  $v = 0$ . The radial part of the partial wave of the final state is equal to

$$R_{qK'}(r) = \frac{c'(2qr)^{s'}}{\Gamma(2s'+2)} e^{-r/qa} \Phi\left(-n' + s' + 1, 2s' + 2, \frac{2r}{an'}\right) \quad (14)$$

and normalized in the energy scale

$$c' = \sqrt{2q/\pi} e^{\pi/2aq} \left| \Gamma\left(s' + 1 - \frac{i}{aq}\right) \right|, \quad (15)$$

$n' = -i[(B^2m/\hbar^2)/2E']^{1/2}$  is the principal quantum number of the final state of the continuous spectrum with energy  $E' = E_{vK} + 2\hbar\omega$ ,  $q = \sqrt{2mE'}/\hbar = -i/an'$  is the relative wave number of the dissociation products,  $s'$  the positive root of Eq. (12), in which  $K$  is replaced by  $K'$ .

We use the following integral representation for the radial Green's function  $g_{K'}(r, r'|E)$  of the potential  $V(r) = Ar^{-2} - Br^{-1}$ :<sup>3)</sup>

$$g_{K'}(r, r'|E_{vK} + \hbar\omega) = -\frac{2m}{\hbar^2\sqrt{rr'}} (1 - e^{-2\pi iv})^{-1} \times \int_1^{(9+)} t^{-v-1/2} (1-t)^{-1} \exp\left(-\frac{r+r'}{av} \frac{1+t}{1-t}\right) I_{2s+1}\left(\frac{4\sqrt{rr'}t}{av(1-t)}\right) dt. \quad (16)$$

<sup>3)</sup>We note that the equation for the radial Green's function  $g(r, r'|E)$  of the Morse potential in the new variables  $y = 2\beta \exp[-\alpha(r-r_0)]$  and  $\bar{g} = \alpha r' g/\sqrt{yy'}$  is identical with Eq. (7) for the potential  $V(y) = Ay^2 - By^{-1}$  with  $B = \beta\hbar^2/2m$ ,  $K_i(K_i+1) + 2mA/\hbar^2 = \zeta^2 - \frac{1}{2}$ ,  $E = \hbar^2/8m$ , where  $\zeta = [-2m(E_0 + \hbar\omega)]^{1/2}/\alpha\hbar$ . Consequently,  $\bar{g}(y, y')$  is determined by Eq. (16), in which we set  $r = y$ ,  $\nu = \beta$ ,  $s + \frac{1}{2} = \zeta$ ,  $\alpha\nu = 2$ .<sup>[9,10]</sup>

The contour of integration runs from unity on the real axis, around the point  $t = 0$  and returns to unity on the  $t$  axis. Here  $\nu = [(B^2m/\hbar^2)/\{-2(E_{vK} + \hbar\omega)\}]^{1/2}$  is the "principal quantum number" in the virtual state;  $s$  the real root of Eq. (12), in which  $K$  is replaced by  $K_i$ . Equation (16) is easily obtained by using the analog of Eq. (7) with the equation for the radial Green's function of the Klein-Gordon equation.<sup>[4]</sup> Since radial functions which describe the nuclear vibrations depend weakly on  $K$ , then one can assume  $R_{qK'} = R_{qK}$ ,  $g_{K\pm 1} = g_K$ . (We note that the functions  $R$  and  $g$  depend on  $K$  through the parameter  $s(K)$ . For  $s \sim 10^2$ , we have  $s(K) = s(K \pm 1)$  with accuracy to within  $K \cdot 0.01\%$ . In this connection, we shall omit the prime on  $s$  in the following.)

Using the standard relations for the confluent hypergeometric functions and the properties of the Laplace transform, we represent the matrix elements of (10) in the form

$$M_{K_i K'}(\omega) = 2c'(4q/an)^s \sqrt{\Gamma(2s+2+v)} \times \frac{1}{a^{1/2}n^2\sqrt{v!}} P_{v+1}\left(-\frac{\partial}{\partial x}\right) \left[ \frac{s+1+n'}{2} Q_0 + (\nu'-1)Q_1 + \frac{s+1-n'}{2} Q_2 \right]_{x=1/(s+1+v)}, \quad (17)$$

where  $P_{v+1}$  is a differential polynomial of  $(v+1)$  order:

$$P_{v+1} = \left(s+v+\lambda + \frac{1}{n} \frac{\partial}{\partial x}\right) \Phi\left(-v, 2s+2, -\frac{2}{n} \frac{\partial}{\partial x}\right) - \nu \Phi\left(-v+1, 2s+2, -\frac{2}{n} \frac{\partial}{\partial x}\right), \quad (18)$$

$$Q_l(x) = \iint_0^\infty dr dr' (rr')^{s+1} g_K(r, r'|E_{vK} + \hbar\omega) \times \exp\left(-\frac{rx}{a} - \frac{r'}{an'}\right) \Phi\left(-n'+s+l, 2s+2, \frac{2r'}{an'}\right), \quad l=0, 1, 2, \quad (19)$$

and the function  $g$  is defined in (16). Integrating over  $dr$  and  $dr'$ ,<sup>[9,13]</sup> and considering the integral  $dt$  as the integral representation for Appell functions of first order  $F_1(a, b, b', c, X, X^*)$ , we get

$$Q_l(x) = \frac{mav\Gamma(2s+2)}{\hbar^2(v-s-1)} \left(\frac{2avn'}{1+\nu x}\right)^{2s+2} \frac{\exp[(2 \arctg avq - \pi)/aq]}{(n^2 - \nu^2)^s (n'+\nu)^2} \delta^l F(l, x), \quad (20)$$

$$F(l, x) = F_1(s-\nu+1, -n'+s+l, n'+s+2-l, s+2-\nu, -\delta\beta(x), -\delta^*\beta(x)), \quad (21)$$

where

$$\beta(x) = (\nu x - 1) / (\nu x + 1), \quad \delta = -\exp(-2i \arctg avq).$$

For  $|X| < 1$ , the Appell function (21) can be computed by expansion in the rapidly converging double hypergeometric series:<sup>[14]</sup>

$$F_1(a, b, b', c, X, X^*) = \sum_{m,n=0}^{\infty} \frac{(a)_{m+n} (b)_m (b')_n}{(c)_{m+n} m! n!} X^m (X^*)^n. \quad (22)$$

Actually,  $|\delta| = 1$ ,  $0 < \beta < 1$ . We note immediately that the poles of the function (21), which appear at  $s - \nu + 2 = 0, -1, -2$ , describe resonances at the intermediate levels:

$$\nu = \sqrt{\frac{B^2m/\hbar^2}{-2(E_{vK} + \hbar\omega)}} = s + 1 + \nu + p, \quad p = 1, 2, \dots, \quad (23)$$

i.e., for  $\hbar\omega = -E_{vK} - (B^2m/\hbar^2)/2(s+1+\nu+p)^2$ ,  $|1/2 E_{vK}| < \hbar\omega < |E_{vK}|$ . It follows from (21) and (22) that  $F(0, x)$

=  $F^*(1, x)$ . Substituting (20) in (17), we get

$$M_{K, K'}(\omega) = NP_{v+1} \left( -\frac{\partial}{\partial x} \right) \left\{ (1 + vx)^{-2s-2} \left[ \frac{(1 - \lambda')F(1, x)}{\sqrt{v^2 - n'^2}} + \operatorname{Re} \frac{n' + s + 1}{(n' + v)^2} F(0, x) \right] \right\} \Big|_{x=(s+1+v)^{-1}}, \quad (24)$$

$$N = -\frac{am}{\hbar^2} \sqrt{\frac{aq}{2\pi n^2 (anq)^{s+2}}} \frac{\sqrt{\Gamma(2s + 2 + v)} |\Gamma(s + 1 - i/aq)|}{(v^2 - n'^2)^s (v - s - 1) \sqrt{v!} \Gamma(2s + 2)}. \quad (25)$$

where the functions  $F(0, x)$  and  $F(1, x)$  are defined in (21), and the differential polynomial  $P_{v+1}$  in (18). The final expression for the cross section for an arbitrary vibrational and rotational quantum numbers of the initial state of the molecule is determined by Eqs. (8), (24), and (25).

As an example of the use of the formulas obtained above in a specific calculation, we have considered the two-photon dissociation of the ground state ( $v = 0, K = 0$ ) of a molecule with the following parameters: dissociation energy  $D = 31$  eV, half-width of the potential well  $\Delta = 6.6$  Å, reduced mass of the dissociation products  $m = m_H$ . In this case, the total dissociation cross section in a field with quantum energy 2 eV amounts to  $\sigma = 10^{-18}$  I cm<sup>2</sup>/W. We note that the parameters are so chosen that the frequency of the incident radiation approaches the mean between the neighboring resonance frequencies, for which the cross section of the process goes to infinity upon neglect of damping of the levels.

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