Quasiclassical calculation of the cross sections for resonant vibrational excitation of the molecules N_2 and CO

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We consider the vibrational excitation of a diatomic molecule from which a slow electron is scattered. A quasiclassical expression is proposed for the cross section of the process, in the form of the first term of the asymptotic expansion in the parameter $(2M)^{1/2}$ (M is the reduced mass of the nuclei, and $\hbar = m = e = 1$). The calculated cross sections for scattering by molecular nitrogen and carbon monoxide are presented. The agreement between the quasiclassical calculation and the available numerical quantum calculations is satisfactory.

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The development of methods for calculating the cross sections for vibrational excitation of diatomic molecules by slow electrons is a vital problem in collision theory. Substantial progress in this direction resulted from the work by Herzenberg,[1,2] Bardsley,[3] and others.[4,5] They proposed to calculate the cross sections for resonant vibrational excitation of the molecules by the following method. The molecule goes over to the final state via an intermediate stage in which the incident electron is captured for some time by the electron shell of the molecule. The nuclear motion in the intermediate molecular ion can be considered on the basis of the notion of an adiabatic term of the system, $W(R)$, which is complex because this ion is unstable:

$$W(R)=U_I(R)-i\Gamma(R)/2, \quad \Gamma(R)\geqslant 0. \tag{1}$$

The behavior of the nuclear subsystem is described by a wave function $\xi(R)$ that satisfies, after separation of the angle variables, the following equation[1-4] (in atomic units):

$$\left[-\frac{1}{2M}\frac{d^2}{dR^2}+W(R)-E\right]\xi(R)=-\zeta_0(R)V(R). \tag{2}$$

Here M is the reduced mass of the nuclei, E the total energy of the system, R the distance between the nuclei, $\zeta_0(R)$ the vibrational wave function of the molecule initial state, and $V(R)$ the amplitude for the production of a molecular ion in the bound-nuclei approximation:

$$V(R)=[4\pi^2\Gamma(R)/k(R)]^{1/2}, \quad k(R)=[2(U_I(R)-U(R))]^{1/2},$$

and $U(R)$ is the potential energy of the molecule.

Knowing the function $\xi(R)$ we can obtain the cross sections for the vibrational transition $n_0 \to n$:

$$\sigma_{n_0\to n}=\frac{v_n}{v_{n_0}}\frac{1}{8\pi^3}\left|\int \xi(R)V(R)\zeta_n(R)dR\right|^2, \tag{3}$$

where $\zeta_n(R)$ is the vibrational wave function of the molecule in the final state; v_{n0} and v_n are respectively the velocities of the incident and outgoing electrons.

Equation (2) was solved in Refs. 2, 4, and 5 numerically, and the terms $U(R)$ and $U_I(R)$ were approximated by Morse potentials. The width $\Gamma(R)$ of the ion term was determined by the penetrability of the centrifugal barrier to the electron with the smallest orbital momentum compatible with the symmetry of the resonance. Proper choice of the model parameters led in Refs. 4 and 5 to very good agreement with the experimental data.

The analytically solvable models of the problem (2) and (3) were also considered. In Refs. 6 and 7 the terms $U(R)$ and $U_I(R)$ were taken to be potentials of oscillators having equal frequencies and shifted minima. The width $\Gamma(R)$ of the ion term was assumed to depend linearly on the distance between the nuclei. In Ref. 8 the terms $U(R)$ and $U_I(R)$ were approximated by Morse potentials, and the width $\Gamma(R)$ was assumed to be almost constant (the dependence of the width on the distance between nuclei was taken into account by perturbation theory). Comparison of the results of Refs. 6–8 with those of Ref. 4 shows that the analytically solvable models lead to good qualitative agreement with the results of the exact calculation.

In a preceding paper[9] we proposed to find the cross sections, within the framework of the considered scheme, by a quasiclassical approach that yielded satisfactory quantitative results for transitions from the ground state in the N_2 molecule. In the present paper we use the quasiclassical method to investigate transitions between excited vibrational states in the molecules N_2 and CO.

Following Ref. 9, we introduce the Green's function of Eq. (2), expressing it in terms of the homogeneous-equation solutions $\chi_1(R)$ and $\chi_2(R)$, which are regular at zero and infinity, respectively. We have for the $n_0 \to n$ transition cross section

$$\sigma_{n_0\to n}=\frac{v_n}{v_{n_0}}\frac{(2M)^2}{8\pi^3|Q|^2}\left| -\int_0^\infty dR\chi_2(R)V(R)\zeta_n(R)\int_0^R \chi_1(R')V(R')\zeta_{n_0}(R')dR' - \int_0^\infty dR\chi_1(R)V(R)\zeta_n(R)\int_R^\infty \chi_2(R')V(R')\zeta_{n_0}(R')dR'\right|^2, \tag{4}$$

$$Q=\chi_1'\chi_2-\chi_1\chi_2'.$$

The integrals encountered are calculated by using quasiclassical representations of the functions χ_1, χ_2, ζ_{n_0}, and ζ_n (see Ref. 9). The main contribution to the integrals are made by the vicinities of the (complex) transition points z_{n_0} and z_n, which are the saddle points for the corresponding integral, and satisfy the generalized Franck-Condon principle:

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$$E-W(z_i)=\varepsilon_i-U(z_i),\quad i=n_0,n, \tag{5}$$

ε_i is the vibrational energy of the molecule in the corresponding state.

We consider next the vibrational excitation case ($n_0 \to n$). The cross sections for the superelastic transitions ($n_0 > n$) can be obtained similarly, or determined with the aid of the detailed balancing principle. The resonant contribution to the elastic scattering ($n_0 = n$) will be considered separately.

If the minimum of $U_I(R)$ is located to the right of the equilibrium position of the molecule we find, confining ourselves to transitions in the region of classically allowed nuclear motion, that z_{n_0} lies to the left of z_n (Fig. 1). In this case only the first term of (4) is significant (cf. Ref. 9).

We obtain ultimately the following representation for the cross sections:

$$\sigma_{n_0\to n}=\frac{v_n}{v_{n_0}}\frac{(2M)^2}{8\pi^3}\frac{|I_{n_0}|^2|I_n|^2}{|Q|^2}, \tag{6}$$

$$I_{n_0}=\int dR\chi_1(R)V(R)\zeta_{n_0}(R),\qquad I_n=\int dR\chi_2(R)V(R)\zeta_n(R) \tag{7}$$

(we have extended the integration to the entire axis, cf. Ref. 9). Calculation of the integrals in (7) by the saddle-point method yields

$$\begin{gathered}
I_{n_0}=\frac{(\omega/\pi)^{1/2}V(z_{n_0})\,(2\pi/\Psi_{n_0}''(z_{n_0}))^{1/2}}{2[\varepsilon_{n_0}-U(z_{n_0})]^{1/4}[E-W(z_{n_0})]^{1/4}}\cos\left(\Psi_{n_0}(z_{n_0})+\frac{\pi}{4}\right),\\
I_n=\frac{(\omega/\pi)^{1/2}V(z_n)\,(2\pi/\Psi_n''(z_n))^{1/2}}{2[\varepsilon_n-U(z_n)]^{1/4}[E-W(z_n)]^{1/4}}\cos\left(\Psi_n(z_n)-\frac{\pi}{4}\right),\\
\Psi_{n_0}=\int_{a^-}^{z}(2M[E-W(z')])^{1/2}dz'-\int_{a}^{z}(2M[\varepsilon_{n_0}-U(z')])^{1/2}dz',\\
\Psi_n=\int_{z}^{b^-}(2M[E-W(z')])^{1/2}dz'-\int_{z}^{b}(2M[\varepsilon_n-U(z')])^{1/2}dz'.
\end{gathered} \tag{8}$$

[The difference in the signs in front of $\pi/4$ is connected with the choice of the slope of the integration contour in the vicinity of the saddle point; this slope depends on the real part of $\Psi_i''(z_i)$, $i=n_0, n$.] The Wronskian determinant Q can be calculated in the region of the classically allowed motion of the nuclei

$$|Q|^2=\left|\sin\left[\int_{a^-}^{b^-}(2M[E-W(z')])^{1/2}dz'-\frac{\pi}{2}\right]\right|^2.$$

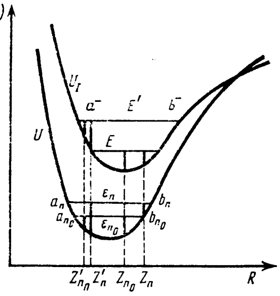

FIG. 1. Molecule potential energy U and real part of the molecular-ion energy U_I vs. the distance between the nuclei. The positions of the transition points z_{n_0} and z_n at different values of the system energy E are shown arbitrarily (at $\Gamma = 0$).

It must be borne in mind that the region of applicability of the representation (8) is limited. First, the transition points must be located in the region of the classically allowed motion of both systems far from the turning points; second, the distance between z_n and z_{n_0} must be large enough. If the first condition is violated, the representations used for χ_1, χ_2, ζ_{n_0}, and ζ_n must be valid in the classically forbidden region or near the turning points. When the second condition is violated, both terms in (4) must be taken into account.

The investigation of the excluded cases, which follows, shows that the representation (6) is valid over a wider range than (7) and (8). From the point of view of physical clarity, an important role is played by the factorization of the cross section (6), which makes it possible to separate three stages in the vibrational excitation. Since the factor $|I_{n_0}|^2$ depends only on the total energy of the system and on the initial state, it can be interpreted as the probability of formation of an intermediate state; the factor $|I_n|^2$, which depends on n and E, has the meaning of the ion-decay probability. The denominator $|Q|^2$ describes the motion of the system in the nonstationary state from the instant of formation to the instant of decay.

It is of interest to trace the dependences of all the factors in (6) on the energy E. In the case of a small width, the factor $|Q|^{-2}$ has Breit-Wigner maxima connected with the long-lived resonant vibrational states of the molecular ion. With increasing Γ, this factor becomes a smoother function of the energy (thus, in the case of the molecules N_2 and CO the oscillatory dependence of the cross sections on E is determined mainly by the factors $|I_{n_0}|^2$ and $|I_n|^2$). In the vibrational excitation case discussed by us ($n_0 < n$) the factor $|I_{n_0}|^2$ depends on the energy more smoothly than $|I_n|^2$, so that $|I_{n_0}|^2$ is responsible for the envelope of the cross section, and its oscillator structure is determined by the factor $|I_n|^2$.

Numerical calculations in accord with Eqs. (6)–(8) (Fig. 2) show good agreement with the results of numerical integration of Eq. (2) (Ref. 4) at intermediate energies. (We have used the same terms U and U_I as in Ref. 4.) Considerable discrepancies are observed at low and high energies, i.e., at these energies the formal quasiclassical approximation is found to be untenable. If the system energy is high enough, the point z_n, remaining in the region of the classically allowed motion of the nuclei, approaches the right-hand turning points, and this leads to an increase of the quasiclassical cross section on account of the decrease of the velocity of the nuclei in the vicinity of the turning points. The rapid decrease of the cross sections at high energies is due to the proximity of the transition points z_n and z_{n_0}, so that the second term of (4) cannot be discarded. It is seen from Fig. 1 that with increasing energy the points z_n and z_{n_0} do indeed come closer together, since the terms $U(R)$ and $U_I(R)$ increase rapidly with decreasing distance between the nuclei. In addition, the transition points come closer to the stopping points and the cross section starts to grow. The corresponding section of the curve is not shown in Fig. 2.

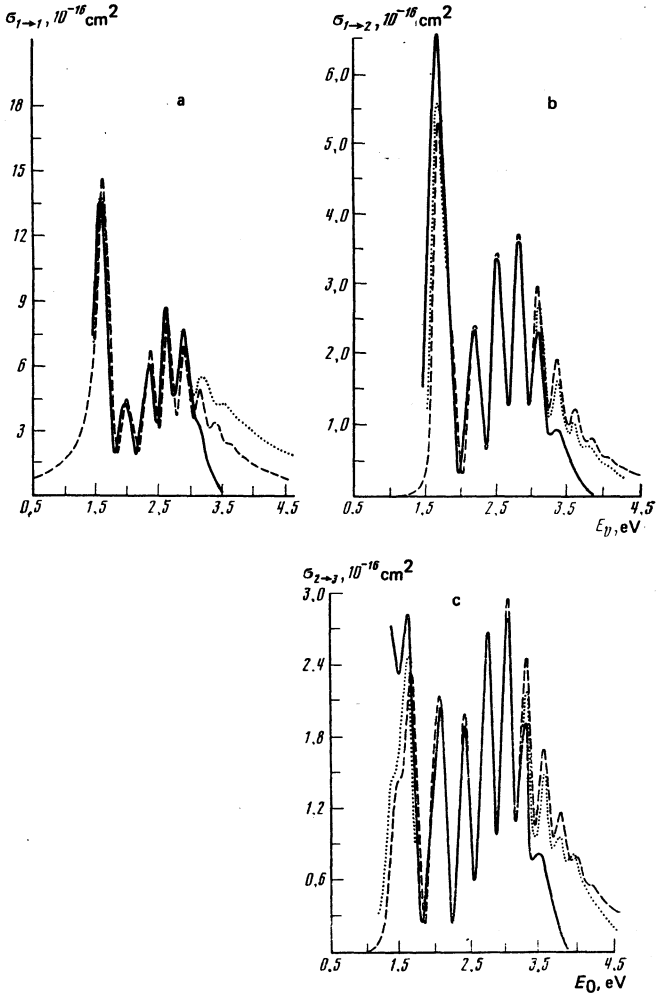

FIG. 2. Dependences of the cross section $\sigma_{v\to v'}$ for the vibrational excitation of the nitrogen molecule on the energy of the incident electron. Transitions between excited states: a) 1→1; b) 1→2; c) 2→3. Solid line—calculations by Eqs. (6)–(8) of the present paper; dotted curve—calculations using the standard-equation method with Eqs. (20) and (21); dashed line—results of Ref. 4.

We proceed now to a generalization of Eq. (6). We consider to this end a standard problem, within the framework of which it is possible to investigate the case when the transition points and turning points are close enough. The solution of the standard problem, in contrast to the true solution $\xi(R)$ of Eq. (2), will be designated $Z(R)$. Assuming that the major contribution to the cross section is made by the vicinities of these points, we can confine ourselves to a linear approximation for the potential curves, and use for the functions ζ_{n_0} and ζ_n their representations in terms of the Airy function.

We have thus the following equation for the determination of $Z(R)$

$$\left[\frac{1}{2M}\frac{d^2}{dR^2}+F^-(R-a^-)\right]Z(R)=V(a_{n_0})v(R-a_{n_0}), \tag{9}$$

where $F^-=-dW/dR$, a^- is the left-hand turning point of the ion, a_{n_0} is the left-hand turning point for the molecule in the state n_0. The function v satisfies the equation

$$\left[\frac{1}{2M}\frac{d^2}{dR^2}+F(R-a)\right]v(R-a)=0, \tag{10}$$

where $F=-dU/dR$.

We seek the solution of (9) in the form

$$Z(R)=\int C(a)v(R-a)\,da. \tag{11}$$

We then obtain for C the equation

$$\frac{1}{2M}\frac{d^2}{da^2}C(a)-\bar{F}(a-a^-)C(a)=-V\frac{F}{F^--F}\delta(a-a_{n_0}),$$

$$\bar{F}=F^-F/(F^--F). \tag{12}$$

Near the left-hand turning points (which are located in the lower half-plane, see Ref. 9), we have $F>0$, $\operatorname{Re}F^->0$, and $\operatorname{Im}F^-<0$. It is obvious that in this approach the function $v(R-a_k)$ which imitates the function $\zeta_k(R)$ must be suitably normalized. From a comparison of the asymptotic forms of these functions it follows that

$$v(R-a_k)=\left(\frac{2M\omega}{\pi}\right)^{1/2}(2MF)^{-1/6}\operatorname{Ai}[(2MF)^{1/3}(a_k-R)]$$

$$=\left(\frac{2M\omega}{\pi}\right)^{1/2}\frac{(2MF)^{-1/6}}{2\pi^{1/2}}\int_{-\infty}^{\infty}dq\exp\left[i\frac{q^3}{3}-iq(2MF)^{1/3}(R-a_k)\right], \tag{13}$$

i.e., $v(R-a)$ is proportional to the Airy function $\operatorname{Ai}(\eta)$ of argument $\eta=(2MF)^{1/3}(a-R)$ and satisfies the relation

$$\int v(R-a)v(R-a')\,dR=\frac{\omega}{F}\delta(a-a'). \tag{14}$$

This normalization allows us to establish a correspondence between the discrete spectrum of the molecule and the continuous spectrum of the standard problem, since it takes the molecule level density into account.

To calculate the cross sections of the vibrational transition in accord with (3) it is necessary to integrate $Z(R)$ with the final-state function $\zeta_n(R)$. Taking the normalization (14) into account, we obtain

$$\sigma_{n_0\to n}=\frac{v_n}{v_{n_0}}\frac{1}{8\pi^3}|V(a_n)|^2\frac{\omega^2}{F^2}|C(a_n)|^2. \tag{15}$$

$C(a)$ is thus directly connected with the cross section.

The solution (13) can be expressed in terms of Airy functions that satisfy definite boundary conditions as $a\to\pm\infty$:

$$C(a)=\frac{C_1(a_<)C_2(a_>)}{C_1'C_2-C_1C_2'}\cdot 2M\frac{V(a_{n_0})F}{F^--F},$$

$$a_<=\min(a_{n_0},a),\qquad a_>=\max(a_{n_0},a). \tag{16}$$

Since $\{\operatorname{Re}[F^-F/(F^--F)]\}^{1/2}>0$, the expansion (11) is meaningful only under the condition that $C_2(a)\to 0$ as $a\to\infty$; this defines uniquely the function C_2:

$$C_2(a)=\operatorname{Ai}[(2M\bar{F})^{1/3}(a-a^-)]. \tag{17}$$

The boundary condition as $a\to-\infty$ is less trivial. Let us calculate the asymptotic value of $Z(R)$ as $R\to\infty$. Using (13) and (16), we write down the asymptotic representations of the functions $v(R-a)$ and $C(a)$:

$$v(R-a)=\frac{1}{2}\frac{(2M\omega/\pi)^{1/2}}{(2MF)^{1/4}(R-a)^{1/4}}\left\{\exp\left(i\left[\frac{2}{3}(2MF)^{1/2}(R-a)^{3/2}-\frac{\pi}{4}\right]\right)\right.$$
$$\left.+\exp\left(-i\left[\frac{2}{3}(2MF)^{1/2}(R-a)^{3/2}-\frac{\pi}{4}\right]\right)\right\},$$
$$C(a)=\frac{S(a_{n_0})}{2(2M\tilde{F})^{1/12}(a^--a)^{1/4}}\left\{A_+\exp\left(i\left[\frac{2}{3}(2M\tilde{F})^{1/2}(a^--a)^{3/2}+\frac{\pi}{4}\right]\right)\right.$$
$$\left.+A_-\exp\left(-i\left[\frac{2}{3}(2M\tilde{F})^{1/2}(a^--a)^{3/2}+\frac{\pi}{4}\right]\right)\right\}. \tag{18}$$

By S are meant all the factors in (16), with the exception of $C_1(a)$.

We obtain the integral (11) by the saddle-point method. Omitting an inessential factor, we get

$$Z(R)\sim(R-a^-)^{-1/4}\left\{A_+\exp\left(-i\left[\frac{2}{3}(2MF^-)^{1/2}(R-a^-)^{3/2}+\frac{\pi}{4}\right]\right)\right.$$
$$\left.+A_-\exp\left(i\left[\frac{2}{3}(2MF^-)^{1/2}(R-a^-)^{3/2}+\frac{\pi}{4}\right]\right)\right\}.$$

In the spirit of the standard problem, this asymptotic relation should be compared with the quasiclassical representation of the true wave function $\xi(R)$, which is valid within the well at a sufficiently large distance from both the left- and right-hand turning points a^- and b^-. When setting up such a representation we must impose the condition that $\xi(R)$ decrease as $R\to+\infty$ i.e., take into account the reflection of the particle at the point b^-. As a result of this comparison, we find that

$$A_+=\exp\left\{i\int_{a^-}^{b^-}[2M(E-W)]^{1/2}dz\right\}\equiv\exp(i\lambda),$$
$$A_-=\exp\left\{-i\int_{a^-}^{b^-}[2M(E-W)]^{1/2}dz\right\}\equiv\exp(-i\lambda). \tag{19}$$

By the same token, the solution $C_1(a)$ in (16) is uniquely defined.

The investigation of the standard problem leads to the conclusion, already stated by us, that Eq. (6) remains valid in this case, too, but with different expressions for I_{n_0} and I_n. Using (15)–(19) we have

$$I_{n_0}=\left(\frac{\omega}{F^--F}\right)^{1/2}\frac{V(z_{n_0})}{(2M\tilde{F})^{1/6}}\mathrm{Ai}(\eta_{n_0}),$$
$$I_n=\left(\frac{\omega}{F^--F}\right)^{1/2}\frac{V(z_n)}{(2M\tilde{F})^{1/6}}\cdot\frac{1}{2}[w^-(\eta_n)e^{-i\lambda}+w^+(\eta_n)e^{i\lambda}],$$
$$Q=(e^{i\lambda}+e^{-i\lambda})/2=(C_1'C_2-C_1C_2')/(2MF)^{1/3}; \tag{20}$$
$$\eta_i=(a_i'-a^-)(2MF)^{1/3},\quad i=n_0,n,$$

with a_i' standing for the turning point closest to a_i and corresponding to a positive slope of F; $w^+(\eta)$ and $w^-(\eta)$ are the Airy-equation solutions that increase and decrease at $\mathrm{Re}\,\eta<0$ and have asymptotic values

$$(-\eta)^{-1/4}\exp\left[\pm i\left(\frac{2}{3}(-\eta)^{3/2}+\frac{\pi}{4}\right)\right].$$

Obviously, at large values of the argument η_i, the factors I_{n_0} and I_n go over asymptotically to the corresponding quasiclassical expressions (8). We note in this condition the possibility of a transition from the quasiclassical representation to a representation in terms of an Airy function, independently for the factors I_{n_0} and I_n. This enables us to investigate, for example, the case when the intermediate system is produced close to the turning point, and decays in the quasiclassical region, or vice versa.

The vicinities of the right-hand turning points (the case of small E) can be considered quite similarly. The standard problem can be formulated in exactly the same manner. The difference between the signs of F and $\mathrm{Re}F^-$ in this and in the following cases is inessential, since

$$\mathrm{Re}\,\tilde{F}=\mathrm{Re}\,(F^-F/(F^--F))>0\quad\text{at}\quad\mathrm{Re}\,F^-<F<0.$$

The final result, which is valid at small E, is

$$I_{n_0}=\left(\frac{\omega}{F^--F}\right)^{1/2}\frac{V(z_{n_0})}{(2M\tilde{F})^{1/6}}\cdot\frac{1}{2}[w^-(\eta_{n_0})e^{-i\lambda}+w^+(\eta_{n_0})e^{i\lambda}],$$
$$I_n=\left(\frac{\omega}{F^--F}\right)^{1/2}\frac{V(z_n)}{(2M\tilde{F})^{1/6}}\mathrm{Ai}(\eta_n), \tag{21}$$
$$\eta_i=(b_i'-b^-)(2MF)^{1/3},\quad i=n_0,n;$$

b_i' is the turning point closest to b_i and corresponding to a linear slope in the well of the molecule.

We consider finally the resonant contribution to elastic scattering. It must be borne in mind that in addition to the resonance mechanism, when a long-lived intermediate state is produced in the course of the process, elastic scattering can take place also potentially at relatively large impact parameters of the electron. The elastic cross section will contain therefore, besides the oscillating term, also a slowly varying one. From the point of view of the method used in this paper, a feature of elastic resonant scattering is the coincidence of the transition points z_{n_0} and z_n. All the results obtained on the basis of the standard-problem method remain valid in this case, whereas the quasiclassical equations must be reviewed. In fact, in this case the two terms in (4) are equally important. To calculate the integrals it suffices nonetheless to use the quadratic representation of the phases of the wave functions ζ_m, χ_1, and χ_2 in the vicinity of the transition point z_m.

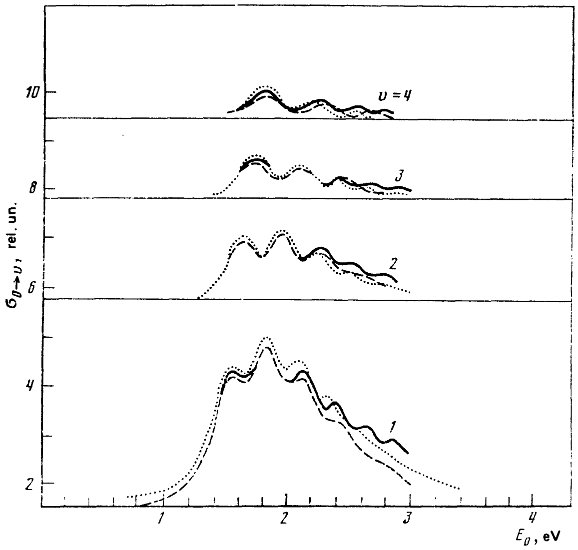

FIG. 3. Dependence of the cross sections for vibrational excitation of the ground state of carbon monoxide on the incident-electron energy. Dashed line—results of Ref. 5; solid line—calculations by Eqs. (6)—(8) of the present paper (an expression obtained in Ref. 9 was used for the factor I_0 in the regions of deviation from the dashed line); dotted line—experiment.[10]

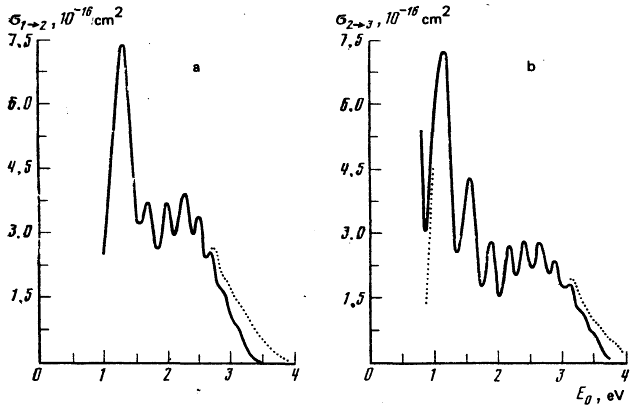

FIG. 4. Cross sections $\sigma_{v\to v'}$ of vibrational excitations of carbon monoxide vs. the incident-electron energy. Transitions between excited states: a) 1 → 2; b) 2 → 3. Solid curve—calculated from Eqs. (6)—of the present paper; dotted curve—calculations by the standard equation method using Eqs. (20) and (21).

The resultant integrals can be easily calculated, so that the final results are completely described by Eqs. (6)–(8).

The results of the numerical calculation within the framework of the present paper and the comparison with the exact calculations of Ref. 4 for the N_2 molecule are shown in Fig. 2. The agreement can be regarded as good both qualitatively and quantitatively. Attention must be called to the role of the regions in which the standard-problem method must be used. A shortcoming of this method is the somewhat arbitrary choice of the point at which the average slope of the term of the molecule is calculated. In addition, the cross section is influenced by the choice of the point at which the transition amplitude V is calculated (the transition point z_n was chosen in the present paper).

Figures 3 and 4 show the results of our calculations of the vibrational excitation of the CO molecule with the terms proposed in Ref. 5, as compared with the results of that paper. We note that in Ref. 5 they calculated only the cross sections for vibrational excitation for the ground state of the molecule, and the numerical values of the cross sections were determined by normalization to the principal maximum of the transition cross section $\sigma_{0\to 1}$ obtained from experiment.[10] The numerical values of the cross sections $\sigma_{0\to n}$ in our case turned out to be twice as large as in Refs. 5 and 10; the cross sections were therefore compared in Fig. 3 in dimensionless units. (The normalization of the experimental cross section is discussed, e.g., in Ref. 11.)

Within the framework of the model considered, the absolute values of the cross sections are closely connected with the term width and hence with the modulation of the cross sections. An attempt to reconcile the absolute values of our calculated cross sections with the results of Refs. 5 and 10, by decreasing the width Γ, increase the depth of the cross-section modulation. In our opinion, calculation for the CO molecule by the procedure of Ref. 4 might clarify the situation. We point out that the mentioned connection between the modulation depth of the cross sections and their absolute values may be useful in investigations of vibrational excitation of molecules under conditions when the experimental data are limited.

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