Accommodation coefficient of a diatomic molecular gas

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The interaction is considered between a diatomic symmetrical molecule in a vibrationally excited state and a surface. The sudden-perturbation method is used to determine the transition probabilities in the vibrational and rotational spectra of the molecule. These are used to obtain an expression for the energy transferred by the molecular gas to the surface, the latter being simulated by a set of harmonic oscillators. The accommodation coefficient is calculated for collisions accompanied by energy redistribution in all degrees of freedom for a non-equilibrium gas.

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In the analysis of the interaction between a gas and a surface one encounters frequently the question of the accommodation coefficient, i.e., the fraction of the total particle energy transferred to the surface in the collision. It appears that the correct approach to the calculation of this coefficient was first indicated by Landau,[1] in an analysis of the interaction of a structureless particle with a surface, accompanied by excitation of long-wave acoustic oscillations in the surface. This model yielded expressions for the accommodation coefficient in the case of small and large energy transfers compared with the thermal energy.

A structureless particle can transfer to a surface only kinetic energy. The collision of the molecule with the wall is accompanied, generally speaking, by the interaction of all its degrees of freedom with the excitations of the surface. In addition, the degrees of freedom of the molecule can interact with one another on the surface and the energy can be redistributed among them.

In the general case, the total energy of a molecule includes the energy of the translational, rotational, vibrational, and electronic motions. The latter usually corresponds to the electronic ground state at temperatures lower than the dissociation temperatures (tens of thousands of degrees). In most real situations it can therefore be assumed that the electronic degrees of freedom are not excited, nor do they become excited in the course of the interaction with the surface. Under these condition, the accommodation coefficient must be determined with account taken of the change of the energy of the translational, rotational, and vibrational motions, as well as of the surface properties.

Interaction between a molecule and a surface without a change of its vibrational state is connected with transitions in only the rotational and translational spectrum. The translational motion was accounted for in[1]. Additional inclusion of the rotations, which are practically always in equilibrium with the translational motion, cannot change the results of[1] significantly. It appears that a physically significant difference arises in the region of very low temperatures in the case of the light gases H_2 and D_2.

We consider below the contribution of the internal degrees of freedom to the accommodation coefficient. We note right away that the contribution of the internal degrees of freedom to the accommodation coefficient is substantial if the masses M of the incident molecule and m of the surface particles differ substantially. If the masses are comparable, one can expect the accommodation coefficient to be close to unity and to depend little on the temperature of the gas or the surface.

To obtain concrete results it is necessary to choose a physical model of the surface and of the interaction with it. We shall, just as in[1], describe the states of the surface by a set of harmonic oscillators corresponding to its normal oscillations, and treat the interaction of the molecule with them as impact against a hard wall, with a duration much shorter than the characteristic times of variation of the energy states of the molecule. The assumption that the interaction is short-timed makes it possible to regard the transitions between the internal states of the molecules as the result of "jarring" one of the nuclei. In fact, impact against a solid wall changes the momentum of the colliding nucleus. Since the collision is short-timed, the remaining nuclei remain practically immobile. The probability of a transition between the internal degrees of freedom can be obtained by the method of sudden perturbations[2]:

$$w_{ss'}=\left|\int \psi_{s'}^{*}\exp\left(-i\frac{\mathbf{pr}}{\hbar}\right)\psi_s d\tau\right|^2, \tag{1}$$

ψ_s and $\psi_{s'}$ are the wave function of the inital and final states of the molecule, respectively, s is the set of quantum numbers of the internal motion, $\mathbf{p}$ is the momentum acquired by the nucleus as a result of the impact, and $\mathbf{r}$ is the coordinate of the nucleus.

We shall carry out the calculations for the case of a diatomic symmetrical molecule. It is known that the dipole transitions between the vibrational and rotational states of such a molecule without a change in the electronic state are forbidden.[3] Its internal excitations are therefore long-lived and the transitions between them result only from collisions. The wave function that describes the internal states of the molecule, neglecting the interaction between them, constitutes the product

$$\psi_n(\rho')\psi_{lm}(\theta,\varphi)$$

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of the wave function of the vibrational motion of the nuclei by the wave function of their rotations. In a coordinate system fixed in the mass center of the molecule we have $\mathbf{r}=(\boldsymbol{\rho}_0+\boldsymbol{\rho}')/2$, where $\boldsymbol{\rho}'$ is the instantaneous deviation from the equilibrium distance $\boldsymbol{\rho}_0$ between the nuclei; the angles θ and φ determine the orientation of the vector $\boldsymbol{\rho}=\boldsymbol{\rho}_0+\boldsymbol{\rho}'$ relative to the z axis, which we choose in the direction of the outward normal to the surface. For not too high vibrational excitations, $\psi_n(\rho')$ are the wave functions of the harmonic oscillator and ψ_{lm} are the normalized spherical functions $Y_{lm}(\theta,\varphi)$, where l and m are the orbital and azimuthal quantum numbers.

The total change of the momentum of the colliding nucleus of a molecule of mass M is $M(v+v')/2$, where v and v' are the normal components of the velocities before and after the collisions. The matrix element of the transition from the state nlm to the state $n'lm'$, corresponding to the probability (1):

$$\int \psi_n\psi_{n'}d\rho' \int Y_{lm}{}^*Y_{l'm'}\exp(-i\mathbf{q}\boldsymbol{\rho})\sin\theta\,d\theta\,d\varphi,\qquad q=M(v+v')/4\hbar, \tag{2}$$

differs from zero at $m=m'$. Thus, only one rotational degree of freedom connected with the change of the molecule orientation relative to the z axis, interacts with the surface. We note that $q\rho\gg i$ already at room temperature, and we use the asymptotic representation[4]

$$e^{-iq\rho\cos\theta}\approx\frac{e^{iq\rho}}{2iq\rho}\sum_{l=0}^{\infty}(2l+1)P_l(\cos\theta)-\frac{e^{-iq\rho}}{2iq\rho}\sum_{l=0}^{\infty}(-1)^l(2l+1)P_l(\cos\theta), \tag{3}$$

where $P_l(\cos\theta)$ is a Legendre polynomial. The first sum in (3) reduces to $4\delta(1-\cos\theta)$, and the second to $4\delta(1+\cos\theta)$; $\delta(x)$ is the Dirac delta function.

It is now easy to integrate over the angles in (2) and obtain

$$\frac{[(2l+1)(2l'+1)]^{1/2}}{2iq\rho}[e^{iq\rho}-(-1)^{l+l'}e^{-iq\rho}]. \tag{4}$$

We have used here the known relation from the theory of associated Legendre polynomials $P_l^{|m|}(1)=\delta_{0m}$, $P_l^{|m|}(-1)=(-1)^l\delta_{0m}$, where δ_{0m} is the Kronecker symbol. Subsequent integration with respect to ρ' in (2) with allowance for (4) and (1) yields a final expression for the transition probability:

$$w_{nl}^{n'l'}=\frac{(2l+1)(2l'+1)}{q^2\rho_0^2}\frac{n'!}{n!}x^{n-n'}e^{-x}[L_n^{n-n'}(x)]^2\Delta_{nn'},\qquad x=\frac{q^2\hbar}{2\mu\omega_0},\quad \Delta_{nn'}=1-\cos 2q\rho_0\exp[i\pi(l+l'+n-n')], \tag{5}$$

ω_0 is the frequency of the vibrational transition of the molecule, $L_n^{n-n'}$ are Laguerre polynomials, and $n\geqslant n'$.[5] In the integration we have retained ρ' in the argument of the exponential but neglected it in comparison with ρ_0 in the denominator of (5).

The expression obtained for $w_{nl}^{n'l'}$ can be of independent of interest as the probability of the transition of a molecule to vibrational and rotational states when jarred.

The probability $W_{nlv}^{n'l'v',\nu\pm1}$ of the transition in the states of the molecule and of the surface is obtained by multiplying (5) by the probability of the transition in the continuous spectrum of the translational energy of the molecule and of the surface oscillators. The latter was calculated in[1] as the probability of the molecule velocity changing from v to v' with simultaneous change $\nu\rightarrow\nu\pm1$ of the state of the surface oscillator in first order of quantum-mechanical perturbation theory:

$$\frac{8}{3}\frac{vv'}{V\hbar\omega\gamma}(\nu\delta_{\nu,\nu-1}+(\nu+1)\delta_{\nu,\nu+1}), \tag{6}$$

ν is the vibrational number of the surface oscillator, ω is its frequency, V is the volume of the wall, γ is its density, and v and v' are the velocities of the molecule mass center before and after the collision, respectively. The number of surface oscillator in the state ν is given by

$$\frac{V}{2\pi^2c^3}\frac{\omega^2d\omega}{\exp(\hbar\omega/T_{\rm sur})-1},$$

c is the sound velocity and $T_{\rm sur}$ is the surface temperature. The total differential probability of the transition in molecule states accompanied by absorption of a surface quantum $\hbar\omega$ is given by the relation

$$dW_{nlv}^{n'l'v'}=\frac{4}{3}\frac{vv'}{\pi^2c^3\gamma\hbar}\frac{\omega\,d\omega}{\exp(\hbar\omega/T_{\rm sur})-1}w_{nl}^{n'l'}. \tag{6a}$$

If the surface gives up a quantum $\hbar\omega$ to the molecule, then

$$dW_{nlv}^{n'l'v'}=\frac{4}{3}\frac{vv'}{\pi^2c^3\gamma\hbar}w_{nl}^{n'l'}\frac{\omega\,d\omega}{1-\exp(-\hbar\omega/T_{\rm sur})}. \tag{6b}$$

In these expressions, it is assumed that the acoustic phonons have the simplest dispersion law $\omega=kc$ and that the oscillators have an equilibrium distribution in energy.

To obtain the total transition probability averaged over the surface states it is necessary to integrate (6) with respect to $d\omega$ with allowance for the conservation law for the energies of the interacting molecules and the surface:

$$\hbar\omega=\hbar\omega_0+\frac{M}{2}(v^2-v'^2)+\frac{\hbar^2}{2I}[l(l+1)-l'(l'+1)]=\hbar\omega_0+\varepsilon-\varepsilon', \tag{7}$$

I is the moment of inertia of the molecule.

The calculation of the accommodation coefficient can be simplified by taking into account the following circumstance: At not too high a temperature the populations of the vibrational states of the molecules (thousands of degrees), a noticeable number of molecules are in the first-excited state. The fraction of particles in higher states is exponentially small. Therefore an important role in the energy balance of the interacting molecule + surface systems is played by single-quantum transitions between the ground and first excited states. In addition, since the energy conservation law calls for

$$q^2\hbar/2\mu\omega_0=M(v+v')^2/8\hbar\omega_0\ll 1,$$

$Mv'^2/2\lesssim\hbar\omega_0$. All this enables us to simplify (5) and obtain for single-quantum transitions the following expressions for the probabilities:

$$w_{nl}^{n-1,l'}\approx(2l+1)(2l'+1)\frac{(n+1)^2}{n}\Delta_{n,n-1}\frac{\hbar}{2\mu\omega_0\rho_0^2}\tag{8a}$$

$$w_{nl}^{n+1,l'}\approx(2l+1)(2l'+1)\frac{(n+2)^2}{n+1}\Delta_{n,n+1}\frac{\hbar}{2\mu\omega_0\rho_0^2},\tag{8b}$$

$$w_{nl}^{nl'}=\frac{(2l+1)(2l'+1)}{q^2\rho_0^2}\Delta_{nn}.\tag{8c}$$

It follows from the foregoing relations that the probability of collision without a change of the vibrational state does not depend on n and exceeds the probability of the single-quantum transition, since

$$\mu\omega_0/q^2\hbar\gg 1.$$

Expressions (6) and (8) allows us to calculate the total energy transferred from the gas to the surface. We obtain first the energy transferred in collisions accompanied by transitions in the vibrational, rotational, and translational motions of the molecule. It is equal to the difference $dE=dE_1-dE_2$ between the energy given up by the molecule to the surface and the energy drawn from the surface. The energy dE_1 corresponds to the transition $n,l,v,\nu\to(n-1),l',v',(\nu+1)$ with excitation of the surface-oscillator quantum (7).

To determine dE_1 we multiply (7) by the transition probability (6b) and by the distribution of the incident particles among the translational and rotational energies:

$$\frac{M\hbar^2}{2IT^2}e^{-\varepsilon/T}v\,dv.$$

Taking (8a) into account, we get

$$dE_1=\frac{4}{3\pi^2}\frac{M^2(2l+1)(2l'+1)}{T^2\rho_0^4\gamma c^3\omega_0}v^2v'^2(\hbar\omega_0+\varepsilon-\varepsilon')^2\Delta_{n,n-1}$$
$$\times\left\{\exp\left(\frac{\hbar\omega_0}{2T_{\text{sur}}}+\frac{\varepsilon}{T_{\text{sur}}}\right)-\exp\left(-\frac{\hbar\omega_0}{2T_{\text{sur}}}+\frac{\varepsilon'}{T_{\text{sur}}}\right)\right\}^{-1}\frac{(n+1)^2}{n}\tag{9}$$
$$\times\exp\left[\varepsilon\left(\frac{1}{T_{\text{sur}}}-\frac{1}{T}\right)+\frac{\hbar\omega_0}{2T_{\text{sur}}}\right]dv\,dv'.$$

The energy dE_2 transferred from the surface to the gave corresponds to the transition $n,l,v,\nu\to(n+1),l',v',(\nu-1)$ and is calculated similarly. The total energy transferred to the surface is

$$dE=\frac{4}{3\pi^2}\frac{M^2(2l+1)(2l'+1)v^2v'^2}{T^2\rho_0^4\gamma c^3\omega_0}(\hbar\omega_0+\varepsilon-\varepsilon')^2\cdot$$
$$\cdot\left\{\exp\left(\frac{\hbar\omega_0}{2T_{\text{sur}}}+\frac{\varepsilon}{T_{\text{sur}}}\right)-\exp\left(-\frac{\hbar\omega_0}{2T_{\text{sur}}}+\frac{\varepsilon'}{T_{\text{sur}}}\right)\right\}^{-1}\cdot$$
$$\cdot\left\{\frac{(n+1)^2}{n}\exp\left[\varepsilon\left(\frac{1}{T_{\text{sur}}}-\frac{1}{T}\right)+\frac{\hbar\omega_0}{2T_{\text{sur}}}\right]\Delta_{n,n-1}-\frac{(n+2)^2}{n+1}\cdot\right.\tag{10}$$
$$\left.\cdot\exp\left[\varepsilon'\left(\frac{1}{T_{\text{sur}}}-\frac{1}{T}\right)-\frac{\hbar\omega_0}{2T_{\text{sur}}}\right]\Delta_{n,n+1}\right\}.$$

If the vibrational states of the molecule have an equilibrium distribution with a temperature T_{vib}, then

$$\bar n=\frac{1}{\exp(\hbar\omega_0/T_{\text{vib}})-1},\quad\left\langle\frac{(n+1)^2}{n}\right\rangle\approx\bar n,\quad\left\langle\frac{(n+2)^2}{n+1}\right\rangle\approx\bar n+1.$$

In the case of complete thermodynamic equilibrium, when $T=T_{\text{sur}}=T_{\text{vib}}$, it follows from (10) that $dE=0$, i.e., there is no energy between the gas and the surface.

We emphasize that formula (10) takes into account only collisions accompanied by transitions of all the molecules degrees of freedom in the energy spectrum. Yet there exist collisions without changes of the vibrational state of the molecule. The probability of such an interaction, as follows from (8c), differs from zero. Within the limits of applicability of perturbation theory, allowance for these interactions is justified at large energy transfers from the molecule to the surface compared with the thermal energy. Such large energy transfers, connected with a change in only the vibrational and rotational states, take place at very low temperatures. At high temperatures the process of collision without a change of the vibrational state should be treated classically, as is done in[1] for a structureless particle. Allowance for the change of the vibrational energy of the molecule allows us to expand the region of temperatures within which the quantum analysis is valid, inasmuch as usually $\hbar\omega_0/T\gg 1$. Inclusion of the rotational motions of the molecules in the analysis alongside the translational ones cannot lead to qualitatively new results compared with[1]; these degrees of freedom are classical in a wide temperature range and are in equilibrium with one another. We shall dwell here only on processes that are given by relation (10).

The calculation of the total energy E reduces to integration of (10) with respect to dv and dv' and to summation over l and l' with allowance for the energy conservation law (7). Since the rotational degrees of freedom for all molecular gases are classical, it is possible to change from summation over l to integration. The expressions for $\Delta_{nn'}$ contain a rapidly oscillating term which vanishes upon integration, so that $\langle\Delta_{n,n-1}\rangle=\langle\Delta_{n,n+1}\rangle\approx 1$. After introducing the new variables

$$x=\frac{Mv^2}{2T},\quad x'=\frac{Mv'^2}{2T},\quad y=\frac{\hbar^2l(l+1)}{2IT},\quad y'=\frac{\hbar^2l'(l'+1)}{2IT}$$

and making simple transformations, we obtain an expression for the energy transferred from the gas to the surface in collisions accompanied by transitions in the vibrational, rotational, and translational spectra of the molecule:

$$E=\frac{16}{3\pi^2}\frac{MT^5}{\hbar^4\gamma c^2\omega_0}\frac{\exp\{-\hbar\omega_0/2T_{\text{vib}}\}}{\operatorname{sh}(\hbar\omega_0/2T_{\text{vib}})}\int_0^\infty dx\int_0^\infty dy\int_0^X dx'\int_0^Y dy'(xx')^{1/2}\left(\frac{\hbar\omega}{T}\right)^2$$
$$\times\exp[-(x+y)]\frac{1-\exp[(\alpha-\beta)\hbar\omega_0/2T-\alpha\hbar\omega/T]}{1-\exp[-\hbar\omega/T_{\text{sur}}]},\tag{11}$$
$$\alpha=\frac{T}{T_{\text{sur}}}-1,\quad\beta=1-\frac{T}{T_{\text{vib}}},\quad\frac{\hbar\omega}{T}=\frac{\hbar\omega_0}{T}+x+y-x'-y',$$
$$X=\hbar\omega_0/T+x+y,\quad Y=\hbar\omega_0/T+x+y-x'.$$

At arbitrary values of the parameters α and β, the integral in (11) can be calculated by numerical methods. We confine ourselves here to approximate integration. The integration region includes all the possible values of $\hbar\omega$ from zero to ∞. We separate those parts of the region where $\hbar\omega/T \ll 1$ and $\hbar\omega/T \gg 1$. In these regions the integration can be carried out analytically. It turns out that the region of large energy transfers $\hbar\omega/T \gg 1$ makes the main contribution to (11). In fact, in the limit $\hbar\omega/T \ll 1$ we can expand the denominator of (11) in powers of this parameter. Retaining the first nonvanishing term of this expansion and omitting in the numerator the exponential $\sim\exp[-\hbar\omega_0/T]$, which is small compared with unity, we get after integrating with respect to x' and y'

$$E' \approx A\frac{T_{\text{sur}}}{T}\frac{\exp[-\hbar\omega_0/2T_{\text{vib}}]}{\operatorname{sh}(\hbar\omega_0/2T_{\text{vib}})}\int_0^\infty dx\int_0^\infty dy\, x^{1/2}\left(\frac{\hbar\omega_0}{T}+x+y\right)^{7/2}\exp[-(x+y)],$$
$$A=\frac{0.4}{\pi^2}\frac{MT^5}{\hbar^4\gamma c^3\omega_0}. \qquad (12)$$

For the subsequent integration we note that the main contribution to the integral is made by values $x \lesssim 1$ and $y \lesssim 1$, whereas $\hbar\omega_0/T \gg 1$. We can therefore use in the integrand the approximate equality

$$\left(\frac{\hbar\omega_0}{T}+x+y\right)^{7/2} \approx \left(\frac{\hbar\omega_0}{T}\right)^{7/2}\left[1+\frac{7}{2}\frac{T}{\hbar\omega_0}(x+y)\right].$$

Completing the integration, we get

$$E'=A\frac{T_{\text{sur}}}{2T}\left(\frac{\hbar\omega_0}{T}\right)^{7/2}\frac{\exp(-\hbar\omega_0/2T_{\text{vib}})}{\operatorname{sh}(\hbar\omega_0/2T_{\text{vib}})}\left[1+\frac{35}{4}\frac{T}{\hbar\omega_0}\right]. \qquad (13)$$

In the region of large values of $\hbar\omega/T \gg 1$ we neglect the exponentials in the denominator and numerator and (11) and arrive in similar fashion to the relation

$$E'' \approx \frac{A}{4}\left(\frac{\hbar\omega_0}{T}\right)^{9/2}\frac{\exp(-\hbar\omega_0/2T_{\text{vib}})}{\operatorname{sh}(\hbar\omega_0/2T_{\text{vib}})}\left[1+\frac{45}{4}\frac{T}{\hbar\omega_0}\right]. \qquad (14)$$

Comparison of (13) and (14) shows that $E''/E' \approx \hbar\omega_0/T \gg 1$ in all real cases. Thus, the energy transferred in collisions accompanied by simultaneous change of the energies of all the degrees of freedom of the molecule accurate to $T_{\text{sur}}/\hbar\omega_0 \ll 1$ is determined by expression (14). It must be borne in mind here that $T_{\text{sur}} < T$. If the surface temperature exceeds the gas translational temperature T, the energy transfer can be calculated on the basis of (11).

The general expression (11) for the energy makes it also possible to consider the accommodation coefficient, which by definition[6] characterizes the energy transfer from the gas to the surface and is equal to the ratio

$$\xi=\varkappa/\varkappa_0,$$

where $\varkappa = E/(T - T_{\text{sur}})$ and $\varkappa_0$ is the energy-transfer coefficient under the condition that the gas molecules leave the surface upon attaining temperature equilibrium with it.

The maximum energy that can be transferred by the gas molecule is

$$E_{max}=3(T-T_{\text{sur}})+\hbar\omega_0. \qquad (15)$$

The first term consists of the average translational and rotational energies of the molecule moving towards the surface. It is assumed that after the collision to molecule acquires the temperature of the surface. Since the vibrational spectrum is discrete and $\hbar\omega_0/T \gg 1$, it takes many collisions to establish equilibrium between the molecule vibrations and the surface. Therefore a molecule departing from the surface cannot have time to reach thermal equilibrium with it. From this point of view, the foregoing definition of the accommodation coefficient must be revised. We shall define the accommodation coefficient as the ratio of the total energy transfer (14) to the maximum possible one (15). Then the part of the accommodation coefficient connected with the change of the vibrational state of the molecule as a result of the collision with the surface is

$$\xi \approx 0.04\frac{MT^{1/2}}{\gamma(\hbar c)^3}(\hbar\omega_0)^{5/2}\left(1+\frac{45}{4}\frac{T}{\hbar\omega_0}\right)\exp\left\{-\frac{\hbar\omega_0}{T_{\text{vib}}}\right\}. \qquad (16)$$

The limits of applicability of (16) are connected with the conditions $\hbar\omega_0/T \gg 1$ and $\hbar\omega_0/T_{\text{vib}} > 1$, and in addition $\xi \lesssim 1$. The last inequality is equivalent to the condition

$$\frac{\hbar\omega_0}{T_{\text{vib}}} \gtrsim \ln\left[0.04\frac{MT^{1/2}}{\gamma(\hbar c)^3}(\hbar\omega_0)^{5/2}\right]. \qquad (17)$$

We present the numerical value of ξ for the following parameters: $T = 600°$, $\hbar\omega_0 = 0.3$ eV, $\hbar\omega_0/T \approx 3$, $c = 5 \cdot 10^5$ cm/sec, $M = 10^{-23}$ g, and $\gamma = 5$ g/cm^3. We obtain $\xi = 0.6$, which does not contradict the available experimental data for diatomic molecules of the type N_2 and 0_2.[6]

As already indicated, (16) determines only one part of the accommodation coefficient, while its other part, which is due to collisions without change of the vibrational state of the molecule, can also be calculated in the assumed model. However, such calculations can be used only at low temperatures. The result, on the other hand, should be of the same order as obtained in[1]. At high temperatures T, for which the analysis of (17) is carried out, the fraction of the transferred energy at such collisions is of the order of $4Mm/(M+m)^2$[6] is the mass of the surface atom. Therefore the total accommodation coefficient is, with high accuracy, the sum of the last expression in (16). This is the situation when T_{vib} is not too high, when the number of vibrationally excited molecules is relatively small. This is always the case if the condition $\hbar\omega_0/T_{\text{vib}} > 1$, under which (16) was obtained, is satisfied.

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