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Ionization of excited nitrogen molecules by electron impact

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Abstract

The coincidence of the classical and quantum cross-sections of elastic scattering for charged particles allows one to use the classical approach for ionization of atomic particles by electron impact. Comparison of experimental data for electron-impact ionization of atoms, ions and molecules with identical valence electrons gives an universal function for such process, considered as a result of interaction of an incident electron with independent valence electrons of an atomic particle. The method is applied to the ionization cross-sections of vibrationally and electronically excited nitrogen molecules.
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1. Introduction

Non-equilibrium plasma chemistry requires detailed information on cross-sections of elementary processes involving vibrationally and electronically excited states [1]. Among them, the ionization process is of paramount importance [2,3] for the description of ion kinetics [4]. In particular, the ionization processes involving electrons and vibrationally excited molecules of nitrogen can be useful for the air gas-discharge plasma and for a plasma of the Earth atmosphere including aurora phenomena and glow processes in the atmosphere.

The peculiarity of ionization of an atomic particle by electron impact results in the character of interaction of a free incident electron with bound electrons of an atomic particle. Since as a result of the ionization process an initially bound electron of an atomic particle becomes free, this process corresponds to transfer the

energy to a bound electron that exceeds the electron binding energy. The analysis of this process is simplified because the cross-sections of elastic scattering of two particles interacting through the Coulomb interaction potential are identical for the classical and quantum cases [5]. This allows us to use the classical methods for evaluation of the ionization cross-sections of an atomic particle by electron impact. We apply the classical method for evaluation of the electron-impact ionization cross-section for vibrationally and rotationally excited nitrogen molecules.

2. Classical ionization cross-section of atoms by electron impact

The first and simplest version of the atom ionization cross-section in electron–atom collisions was constructed by Thomson [6]. Neglecting interaction of bound and incident free electron in the course of their scattering and assuming the bound electron to be motionless at $t = 0$, he used the Rutherford cross-section for elastic scattering under Coulomb interaction

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$$d\sigma = \frac{\pi e^4 \, d\Delta\varepsilon}{\Delta\varepsilon^2 E}. \tag{1}$$

Here, E is the energy of an incident electron, $\Delta\varepsilon$ is the energy transferred to a bound electron from the incident one. The release of a bound electron takes place when $J \ll \Delta\varepsilon \ll E$ (J is the electron binding energy, i.e. the ionization potential), so that the cross-section of ionization can be expressed as

$$\sigma_{\text{ion}} = \int_J^E d\sigma = \frac{\pi e^4}{E}\left(\frac{1}{J} - \frac{1}{E}\right). \tag{2}$$

A general Thomson expression for the ionization cross-section of an atomic particle containing several valence electrons can be written as

$$\sigma_{\text{ion}} = \sum_i \frac{\pi e^4}{J_i^2} f\left(\frac{E}{J_i}\right), \tag{3}$$

where the index i relates to the ith electron, and J_i is the binding energy of the ith electron. The universal function $f(E/J_i) = f(x)$ for the Thomson case has the form

$$f(x) = \frac{1}{x} - \frac{1}{x^2}, \quad x \geqslant 1. \tag{4}$$

The interest to the classical theory of atom ionization by electron impact decreased when Bethe [7] showed, within the framework of a strict quantum theory, that the ionization cross-section for high electron velocities contains an additional logarithmic factor. This logarithmic factor is present also in the cross-section of ionization of highly excited atom when the applicability of the classical theory seems to be valid. A detailed analysis [8,9] for the ionization cross-sections of highly excited atom by electron impact shows that classical and quantum cross-sections are close, though the analytic expression for the cross-sections has different forms. Hence, the classical approach is valid for highly excited atoms, and in other cases of ionization processes the classical approach can be considered as a model.

Note that the form (3) of the ionization cross-section follows from dimensionality considerations under assumption of the classical character of electron scattering, so that the Planck constant is not included in the cross-section expression. The interest to the classical model is determined by its agreement with the experiment. The Thomson version of the classical approach may be widened by accounting for energy distributions of bound electrons and by using other real details [6,10,11]. Additional assumptions to the classical approach allow one to obtain various versions of the classical theory of atom ionization (for example [12–14]). However, within the framework of the classical theory such complications are not grounded, so that new assumptions cannot improve the accuracy of the ionization cross-sections. Moreover, because the advantage of the classical theory is its agreement with the experimental data, we will use the simplest version of the classical theory [15], when the universal function $f(E/J_i)$ in formula (3) is taken from the experiment.

In this manner, we use formula (3) in the case of ionization of an atomic particle by electron impact if the ionization process is determined by release of several identical electrons, and for each bound electron this process proceeds independently. Then, formula (3) takes the form

$$\sigma_{\text{ion}} = n \frac{\pi e^4}{J^2} f\left(\frac{E}{J}\right), \tag{5}$$

where n is the number of identical valence electrons. Using formula (3) for the classical ionization cross-section, we find the universal function $f(x)$ on the basis of the experimental data. Fig. 1 demonstrates this operation for ionization of s-electron if ionizing atoms or ions contains only s-electrons, and also if the liberation of an s-electron gives the main contribution to the ionization cross-section (as it takes place in the Li case). From this, we take the universal function $f(x)$ that describes the experimental data in the form [15],

$$f(x) = \frac{10(x-1)}{\pi x(x+8)} \tag{6}$$

and the scattering of data for different experiments characterizes the accuracy of approximation of the cross-section by formula (3).

Analyzing the data of Fig. 1, note that formula (6) follows from statistical treatment of experimental data represented on the basis of the reduced variable. Hence,
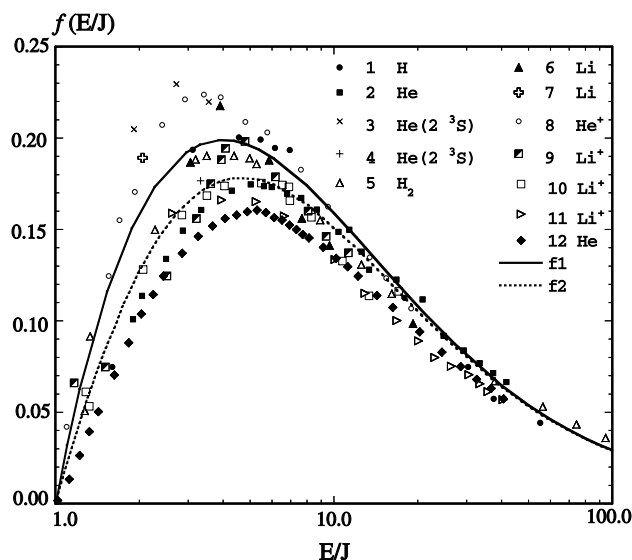

Fig. 1. The reduced cross-sections of ionization of atoms and ions with valence s-electrons by electron impact versus the reduced electron energy. *Experiment*: 1 – [27]; 2 – [18]; 3 – [28]; 4 – [29]; 5 – [30]; 6 – [31]; 7 – [31]; 8 – [32]; 9 – [33]; 10 – [34]; 11 – [35]; 12 – [19]. Solid curve corresponds to the formula: $1 - f(x) = 10(x-1)/[\pi x(x+8)]$, dashed curve to the formula: $2 - f(x) = 10(x-1)/[\pi(x+0.5)(x+8)]$.

the coincidence of this function with experimental data is determined by the accuracy in expressing the ionization cross-section through this variable only. This accuracy, which depends on the electron energy, can be estimated as ∼20% near the maximum of the function. The accuracy is better at high electron energy, even though the contribution from internal electrons to the ionization cross-section increases with electron energy. For example in the case of Li^+ (see Fig. 1), the accuracy is within ∼10%. Systems containing one and two valence electrons are characterized by the same accuracy. Nevertheless, the universal function $f(x)$ is higher and its maximum shifts to small energies for systems with less ionization potentials. Ignoring this fact, we obtain an error that is determined by the possibility to use the reduced variable for the ionization cross-section. Next, some atoms and ions included in Fig. 1 have internal electrons, but their contribution to the ionization cross-section does not exceed ∼10%.

Thus, expressing the ionization cross-section of atomic particles by electron impact through the reduced electron energy, we simplify the problem, but this gives an error in the cross-section that drops increasing the electron energy.

The universal function $f(x)$ can depend on the orbital momentum of valence electrons, and we give it in Fig. 2 for p-electron separately on the basis of the data for the ionization cross-section of the nitrogen atom [16], for the oxygen atom [17] and for the neon atom [18,19]. The degree of coincidence of this function for different elements testifies about the accuracy of this approach and its dependence on the orbital momentum of valence

electrons. One can see the difference between the ionization cross-sections involving s- and p-electrons because of a different character of electron distribution inside the atomic particles. Joining these results, we represent the approximated ionization function of p-electrons in the form,
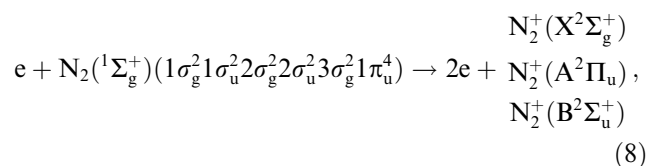
$$f(x) = \frac{10(x-1)}{\pi(x+1.5)(x+9)}. \tag{7}$$

Note that the presence of internal valence s-electrons increases the accuracy of the scaling law for the ionization cross-section. Because the ionization potential of s-electrons is greater for these atomic particles than that for p-electrons, and the number of s-electrons is less than the number of p-electrons, the contribution of s-electrons is restricted. Nevertheless, they give a contribution of 10–30% to the cross-section, and therefore the scaling law for p-electrons is worse than for s-electrons.

3. Ionization cross-section of molecules by electron impact

In considering the ionization process involving diatomic molecules, we are based on the one-electron model of the molecule when two-center orbitals of valence electrons are composed from atomic orbitals. One should note the principal peculiarity of molecule ionization in comparison with ionization of atoms. Though in molecules we locate electrons on shells, like that in the atom case, the number of electrons of one shell is less than that for atoms, and therefore several electron shells participate in the ionization process even at not high energies of an incident electron. Let us consider in detail the nitrogen case.

For moderate electron energies, we consider the following ionization processes:

$$e + N_2(^1\Sigma_g^+)(1\sigma_g^2 1\sigma_u^2 2\sigma_g^2 2\sigma_u^2 3\sigma_g^2 1\pi_u^4) \rightarrow 2e + \begin{matrix} N_2^+(X^2\Sigma_g^+) \\ N_2^+(A^2\Pi_u) \\ N_2^+(B^2\Sigma_u^+) \end{matrix}, \tag{8}$$

where the threshold electron energies for these final states of the molecular ion are 15.581, 16.718 and 18.737 eV correspondingly, and these ionization processes are accompanied by a release of an electron from the shells $3\sigma_g$, $1\pi_u$ and $2\sigma_u$, respectively. In other words, for these processes we consider the nitrogen molecule in the ground state as a bound state of the nitrogen molecular ion and an electron in the following manner:

$$N_2(^1\Sigma_g^+)(1\sigma_g^2 1\sigma_u^2 2\sigma_g^2 2\sigma_u^2 3\sigma_g^2 1\pi_u^4) = \begin{matrix} N_2^+(X^2\Sigma_g^+) - e(3\sigma_g) \\ N_2^+(A^2\Pi_u) - e(1\pi_u) \\ N_2^+(B^2\Sigma_u^+) - e(2\sigma_u) \end{matrix} \tag{9}$$
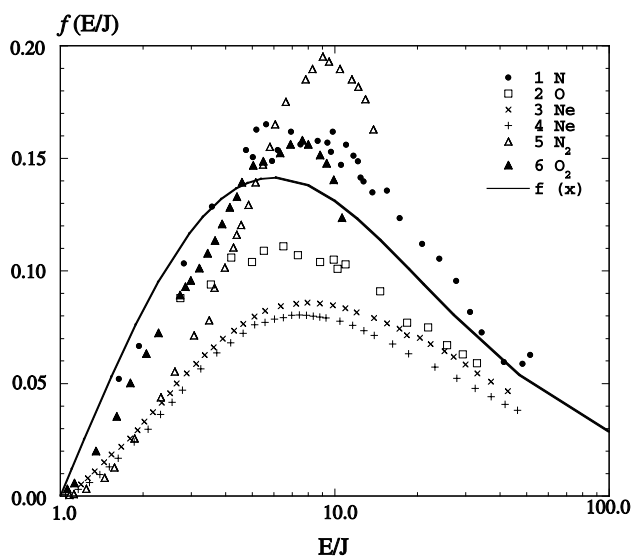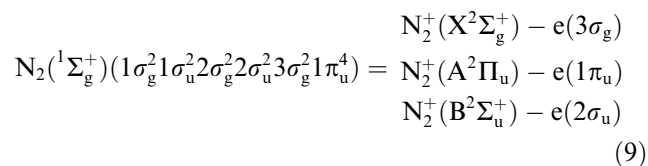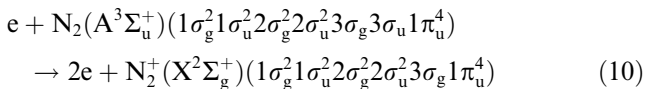

Fig. 2. The ionization reduced cross-sections for atoms and molecules with valence p-electrons by electron impact as a function of the reduced electron energy. *Experiment:* 1 – [16]; 2 – [17]; 3 – [18]; 4 – [19]; 5,6 – [25]. Solid curve corresponds to the formula: $1-f(x) = 10(x-1)/[\pi(x+1.5)(x+9)]$.

and we ignore other excited states of the forming molecular ion in the total ionization cross-section of the nitrogen molecule because other final states correspond to electron release from internal shells or require simultaneous excitation of the forming molecular ion.

Along with ionization of the nitrogen energy in the ground electronic state, we consider ionization of the metastable molecule in the electronic state $A^3\Sigma_u^+$ that proceeds according to the scheme

$$e + N_2(A^3\Sigma_u^+)(1\sigma_g^2 1\sigma_u^2 2\sigma_g^2 2\sigma_u^2 3\sigma_g 3\sigma_u 1\pi_u^4)$$
$$\rightarrow 2e + N_2^+(X^2\Sigma_g^+)(1\sigma_g^2 1\sigma_u^2 2\sigma_g^2 2\sigma_u^2 3\sigma_g 1\pi_u^4) \quad (10)$$

and a $3\sigma_u$ electron releases as a result of this process. The threshold of this ionization process is 9.357 eV that corresponds to the transition between the ground vibrational states.

From formula (3), the partial cross-section for ionization of a diatomic molecule by electron impact with transition between given vibrational states of the molecule and molecular ion has the following form:

$$\sigma_{\text{ion}} = n \frac{\pi e^4}{J_{v,v'}^2} f\left(\frac{E}{J_{v,v'}}\right) S_{v,v'}. \quad (11)$$

This cross-section corresponds to transition between given electronic states of the molecule and molecular ion, n is the number of identical valence electrons of the molecule, so that a release of these electrons can lead to transition between these electronic states, v, v' are the initial and final vibrational quantum numbers of the molecule and of the molecular ion respectively, $S_{v,v'}$ is the Franck–Condon factor, characterized by the overlapping of the vibrational wavefunctions of the molecule and molecular ion in these states. The total ionization cross-section with transition from a given electronic and vibrational v state of a molecule in all the electronic and vibrational states can be expressed as

$$\sigma_{\text{tot}} = \sum_i \sum_{v'} n_i \frac{\pi e^4}{J_{v,v'}^2} f\left(\frac{E}{J_{v,v'}}\right) S_{v,v'}, \quad (12)$$

where an index i relates to a given electronic shell, and n_i is the number of electrons in the ith shell of the molecule.

In considering the rotational state of a diatomic molecule, we note a low momentum of an incident electron in comparison with nuclear ones. This fact and the character of the ionization process as a result of transition at nearby distances between nuclei for transition states and correspondingly nearby rotation molecular momenta allows us to separate the rotational degrees of freedom from other ones. Therefore, for not low rotational momenta of the molecular particles one can ignore below the variation of their rotational state during the ionization process.

In analyzing the ionization process as a transition between two electronic terms, we are based on the potential

curves of molecular particles and on the spectroscopic data. We consider that ionization of the nitrogen molecule in the ground electronic state $X^1\Sigma_g^+$ proceeds in accordance with the process schemes (8). We are based on potential curves [20] of these electronic terms, and the spectroscopic parameters for the transition states are given in Table 1 [21]. Here, T_e is the excitation energy of a given electron state, ω_e, $\omega_e x_e$ are the vibrational frequency and the anharmonic parameter, B_e, α are the rotational constant and the correction to it, D_e is the dissociation energy. Vibrational levels of the molecule are expressed through spectroscopic parameters [22].

The ionization cross-section (11) is proportional to the Franck–Condon factor $S_{v,v'}$. The analysis on the basis of the values [23,24] for this quantity shows for the nitrogen molecule that only three–five vibrational states of the molecular ion mainly contribute to the total ionization cross-section. Hence, one can use only the average vibrational parameters for the final state of the transition. This operation allows us to simplify the analysis and extract the main information for the description of ionization kinetics of molecular particles. As a result, we use the average vibrational quantum number of the molecular ion state v', v'' and the average ionization potential $J_{v'}, J_{v''}$ for the transition from a given vibrational state v of the molecule as the parameters of the ionization process under consideration. For high vibrational levels, this method becomes precise and the optimal vibrational states of the molecular ion corresponds to coincidence of the turning points for nuclear motion in the molecule and molecular ion (see Fig. 3).

Basing on this method, one can simplify the expression (11) for the ionization cross-section, assuming that the nuclear wave function is large mostly near the turning points. Then, formula (11) for the ionization cross-section takes the form

$$\sigma_{\text{ion}} = \frac{n\pi e^4}{2} \left[\frac{1}{J_{v,v'}^2} f\left(\frac{E}{J_{v,v'}}\right) + \frac{1}{J_{v,v''}^2} f\left(\frac{E}{J_{v,v''}}\right) \right], \quad (13)$$

where v', v'' are the vibrational quantum numbers of the molecular ion for states where either the inner or the outer turning point coincide with a turning point of the molecule with a given vibrational quantum number v, n is the number of electrons involved in a given elec-

Table 1
Spectroscopic parameters (cm^{-1}) for different electronic states of the nitrogen molecule and its molecular ion [21]

Molecule (state)	T_e	ω_e	$\omega_e x_e$	B_e	α_e ($\times 10^{-2}$)	D_e
$N_2(X^1\Sigma_g^+)$	0	2359	14.3	1.998	1.73	79884
$N_2^+(X^2\Sigma_g^+)$	0	2207	16.1	1.932	1.88	71387
$N_2^+(A^2\Pi_u)$	9167	1904	15.0	1.744	1.88	62217
$N_2^+(B^2\Sigma_u^+)$	25461	2420	23.2	2.071	1.4	45919

The ionization cross-sections are calculated below for the transitions involving these states.
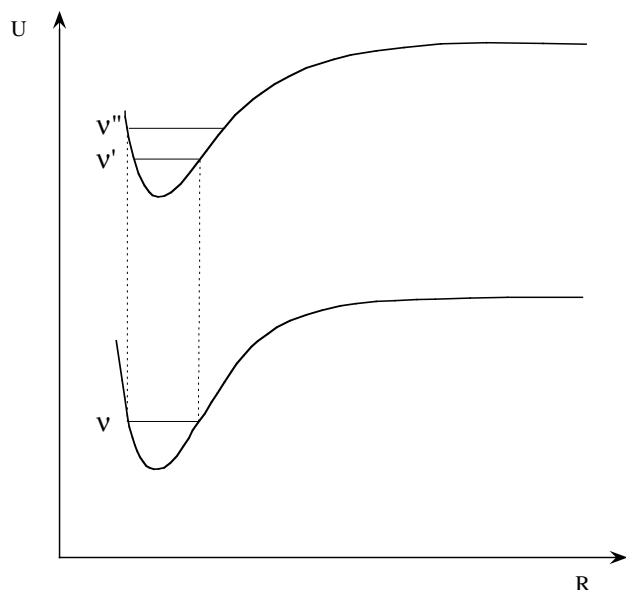
Fig. 3. Scheme of ionization transitions between given electronic states and from a given vibrational state. The lower potential curve corresponds to the electronic term of a molecule, and the upper potential curve relates to its molecular ion.

Table 3
Parameters and cross-sections (10^{-17} cm^2) of the ionization transitions e + N$_2$(X$^1\Sigma_g^+$, v) → 2e + N$_2^+$(A$^2\Pi_u$)

v	$J_{v,v'}$ (eV)	$J_{v,v''}$ (eV)	σ ($E = 20$ eV)	σ ($E = 30$ eV)	σ ($E = 50$ eV)
0	17.8	16.8	0.79	2.9	5.2
4	19.0	15.7	0.90	3.1	5.3
8	20.0	15.2	0.95	3.2	5.4
12	20.6	14.8	1.0	3.3	5.5
16	20.4	14.5	1.2	3.5	5.8
20	20.0	14.2	1.4	3.7	6.1
24	19.6	14.1	1.5	3.9	6.4
28	19.2	14.0	1.6	4.1	6.6
32	18.8	13.9	1.8	4.3	6.8
36	18.4	13.8	1.8	4.4	6.9
40	18.0	13.7	1.9	4.5	7.3

Table 4
Parameters and cross-sections (10^{-17} cm^2) of the ionization transitions e + N$_2$(X$^1\Sigma_g^+$, v) → 2e + N$_2^+$(B$^2\Sigma_u^+$)

v	$J_{v,v'}$ (eV)	$J_{v,v''}$ (eV)	σ ($E = 20$ eV)	σ ($E = 30$ eV)	σ ($E = 50$ eV)
0	18.7	19.1	0.13	1.0	2.0
4	18.5	18.8	0.16	1.1	2.1
8	18.3	18.4	0.20	1.2	2.2
12	18.1	17.9	0.26	1.3	2.3
16	17.9	17.4	0.32	1.4	2.5
20	17.7	16.9	0.39	1.5	2.6
24	17.5	16.4	0.47	1.6	2.8
28	17.1	15.9	0.56	1.7	2.9
32	16.9	15.4	0.65	1.9	3.1
36	16.7	15.0	0.75	2.0	3.3
40	16.5	14.7	0.83	2.1	3.4

tronic transition. Tables 2–4 contain the diabatic ionization potentials $J_{v,v'}$, $J_{v,v''}$ for transitions between given electronic states and from a given initial vibrational state.

We should note the peculiarities of the above evaluations. Separating the electronic and vibrational degrees of freedom for molecular particles, we compose ionization of molecular particles as a result of only electron interaction at given vibrational states. This fact is taken into account in formula (11) where parameters of the vibrational states of transitions are the ionization potentials $J_{v,v'}$ and the Franck–Condon factors $S_{v,v'}$. In addition, we use the classical ionization cross-sections due to interaction of an incident electron and bound electrons of the molecule. But one can refuse from the last

assumption for relative ionization cross-sections or if we use the experimental data for the ionization transition between the ground vibration states. In this case, we use separation of electronic and vibrational states according to formula (11) and replace in this manner the classical approach for the ionization cross-section by a scaling with the experimental cross-section for the ground electronic state.

The last approach allows us improving the accuracy of the partial cross-sections using experimental information. We demonstrate this for the considered nitrogen case for incident-electron energies of 20, 30 and 50 eV when the total ionization cross-sections, σ_{tot}, are, respectively, 0.174, 0.61, and 1.09 Å^2 [25]. The total ionization cross-section is

$$\sigma_{tot} = \sigma(X^2\Sigma_g^+) + \sigma(A^2\Pi_u) + \sigma(B^2\Sigma_u^+), \qquad (14)$$

where the argument indicates the final electronic state of the molecular ion. Therefore, using the experimental values of σ_{tot}, we practically restore the function $f(x)$ in the expression (13) for molecule ionization from a given molecule state. Along with the diabatic ionization potentials, Tables 2–4 contain the ionization cross-sec-
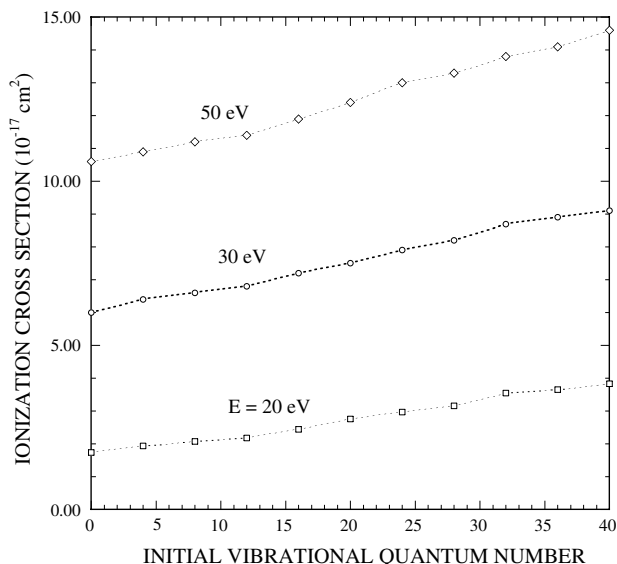
Table 2
Parameters and cross-sections (10^{-17} cm^2) of the ionization transitions e + N$_2$(X$^1\Sigma_g^+$, v) → 2e + N$_2^+$(X$^2\Sigma_g^+$)

v	$J_{v,v'}$ (eV)	$J_{v,v''}$ (eV)	σ ($E = 20$ eV)	σ ($E = 30$ eV)	σ ($E = 50$ eV)
0	15.8	15.4	0.82	2.1	3.4
4	15.8	15.1	0.87	2.2	3.5
8	15.9	14.9	0.92	2.2	3.6
12	15.9	14.7	0.92	2.2	3.6
16	16.0	14.6	0.92	2.3	3.6
20	16.1	14.4	0.97	2.3	3.7
24	16.1	14.2	1.0	2.4	3.8
28	16.2	14.1	1.0	2.4	3.8
32	16.2	13.9	1.1	2.5	3.9
36	16.2	13.8	1.1	2.5	3.9
40	16.3	13.7	1.1	2.5	3.9

Fig. 4. Total ionization cross-section for the process $N_2(X^1\Sigma_g^+, v_i)$+ e $\rightarrow N_2^+(X^2\Sigma_g^+, A^2\Pi_u, B^2\Sigma_u^+)$ + 2e, as function of the initial vibrational quantum number for different collision energies.

tions $\sigma(E)$ which are calculated by formula (13) and are expressed in 10^{-17} cm^2. In Fig. 4, the total ionization cross-section, resulting from different ionization channels, is plotted as function of the initial vibrational quantum number for different collision energies, showing the small vibrational dependence.

The normalization condition (14) is used for a more accurate evaluations of the cross-sections which accuracy is estimated as ~30%.

The results of identical calculations for the diabatic ionization potentials and ionization cross-sections of the process (10) are given in Table 5.

Note that we assume the function $f(x)$ to be identical for different electron transitions of a given molecule. In the considered nitrogen case, basing on formula (13) for the ionization cross-section involving given initial and final electronic and vibrational states, the normalization condition (14) and experimental data [18,25] for the total ionization cross-sections from the ground electronic and vibrational state, we obtain this function in the form

$$f(x) = \frac{6(x-1)}{\pi(x+10)(x+2)} \tag{15}$$

and estimate its accuracy as ~20–30% far from the threshold energy. In the framework of this theoretical approach, the ionization cross-section values depend on the set of experimental data used to refine the universal function, affecting also the accuracy. As a further check of the present calculations, we can note that the brancing ratios of the calculated cross-sections, i.e. $\sigma(X)/\sigma_{tot}$, $\sigma(B)/\sigma_{tot}$, $\sigma(A)/\sigma_{tot}$, are within the recent experimental values reported by Zyl and Pendleton [26]. Moreover the small dependence of the ionization cross-section on the vibrational quantum number confirms the previous results [4] based on Gryzinski cross-sections.

4. Conclusion

In analyzing kinetics of excited atomic and molecular particles in a plasma, we need the rate constants of the processes involving these particles. We have in particular considered ionization of molecules by electron impact taking into account vibrational states of the molecular particles. We start from the classical version for interaction of an incident electron with bound electrons of the molecule, and the classical approach is based on coincidence of the cross-sections of elastic scattering for charged particles within the framework of the classical and quantum mechanics. The approach uses experimental cross-sections of ionization for various atoms and molecules. The considered version of the classical approach gives an universal cross-section whose parameters are the ionization potential for the diabatic transition from a given electronic and vibrational state of the molecule. Accuracy is improved by using the experimental ionization cross-sections which refer to the ground vibrational state of a given molecule.

As for vibrational states of the ionization transition, the rule is valid that the turning points of initial and final vibrational states coincide, so that the ionization transition has the diabatic character for the vibrational degree of freedom. This is fulfilled strictly for excited vibrational state and is valid more or less for the ground vibrational state of the molecule. Rotational states are not significant for these processes because of low electron momenta and the nature of this electron transition when the distance between nuclei do not vary during the transition. As a result, we have a simple scheme for evaluation of the ionization cross-sections in electron–molecule collisions if colliding molecule and molecular ion are found in given initial and final electronic states. The above calculations for the nitrogen case demonstrate the possibilities of this scheme.

Table 5
Parameters and cross-sections (10^{-17} cm^2) of the ionization transitions e + $N_2(A^3\Sigma_u^+, v) \rightarrow 2e + N_2^+(X^2\Sigma_g^+)$

v	$J_{v,v'}$ (eV)	$J_{v,v''}$ (eV)	σ ($E = 15$ eV)	σ ($E = 25$ eV)	σ ($E = 40$ eV)
0	9.6	11.6	1.4	3.1	4.2
4	8.8	12.0	1.7	3.5	4.6
8	8.2	12.4	2.1	4.0	5.1
12	7.6	12.8	2.6	4.5	5.6
16	7.1	13.2	3.0	5.1	6.2
20	6.8	13.6	3.4	5.5	6.6
24	6.6	14.0	3.7	5.8	6.8

Acknowledgments

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