Kinetic models of nonequilibrium nitrogen and hydrogen plasma for diagnostics of gas discharges

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Abstract. Computation-experimental methods for plasma diagnostics of nonequilibrium nitrogen and hydrogen gas discharges are presented. These methods are based on a combined use of collisional-radiative models and of experimental results on optical plasma diagnostics. Nonstationary self-consistent level-to-level collisional-radiative models include computer codes: for the processing and the simulation of the emission and the coherent anti-Stokes Raman scattering spectra; for the solution of the kinetic Boltzmann equation for plasma electrons, the thermal conductivity equation, and the balance equations for most of plasma neutral, excited, and charged particles. Banks of data were created for the processing and the simulation of the spectra, and the solution of the kinetic equations. The models were validated by the comparison of obtained results with numerous known results of experiments and with calculations for different discharges.

1. Introduction

Optical interferometry (OI), coherent anti–Stokes Raman scattering (CARS) spectroscopy, and optical emission spectroscopy (OES) have been widely used in diagnostics of nonequilibrium low temperature plasmas (NLTPs) [1]. The validation of these methods and the processing of measured data require the use of nonstationary semi-empirical self-consistent level-to-level collisional-radiative models (CRMs) of plasmas predicting population densities of the investigated diatomic molecules in the ground and electronically-excited states with resolution over vibrational and rotational levels. These models together with spectroscopic techniques allow the creation of computation-experimental methods gaining information on plasma parameters that could not be provided by any of these methods used separately [1-9]. Despite a wealth of publications [10-12] devoted to the development of the methods for describing collisional-radiative processes in NLTPs, the task remains topical up to this day. In the present work the nonstationary semi-empirical self-consistent level-to-level CRMs are developed for diagnostics of NLTPs of direct-current glow discharges, radio-frequency, and microwave discharges in N₂ and H₂. For plasmas in N₂ the discussion of the CRM is more detailed than for plasma in H₂, for which preliminary results will be given.

2. Collisional-radiative models of nonequilibrium low temperature plasma in N₂ and H₂

A comprehensive report on the semi-empirical self-consistent level-to-level CRM application to achieve plasma parameters for different types of discharges in N₂ from the spectral measurements can be found in [1, 4–6]. A detailed discussion and the development of CRMs in N₂, O₂ and H₂ are contained in [1–9, 13–15]. In brief, the CRM includes computing codes for modeling: the electron
energy distribution function (EEDF) with the corresponding database on level cross sections of elementary processes involving electrons and heavy particles (molecules, atoms and their ions); the emission and CARS spectra with the corresponding spectroscopic database; the kinetic processes involving neutral molecules and molecular ions with resolution over vibrational levels of the ground and electronically-excited states, atoms and atomic ions in the ground and electronically-excited states with the corresponding database on level rate coefficients. Important components of the CRM are the database on the level cross sections and rate coefficients of elementary processes, the models for physical and chemical processes, and the radiative models of NLTP. These components of the CRM are formed and developed on the basis of the generalization of the accessible information in the literature on the spectroscopic data, rate coefficients, and cross sections of elementary processes, as well as on results of investigations on physical and chemical processes. A special attention is accented on the reproducibility of the results from measurement using the CRM. The reliability of the developed CRM is tested by comparing the calculated results with those from numerous data found in literature on plasma parameters of dc glow, radio-frequency, and microwave discharges. Thus the CRM includes a database on experiments devoted to the determination of EEDFs, vibrational and rotational functions of molecules and their ions in the ground and electronically-excited states, rate coefficients and cross sections of elementary processes, translational and rotational temperatures of molecules, specific power absorbed by plasma, etc.

In the following discussion we consider examples illustrating the application of the CRMs in N$_2$ and H$_2$ for the modeling of plasma parameters for different types of discharges: dc glow discharge (DCGD); capacitive and inductive-capacitive radio-frequency discharges (CRFD and ICRFD); electrode microwave discharge (EMD); microwave discharge excited by the surface wave; microwave discharges generated in the rectangular waveguide and in a prismatic resonator (MDRW and MDPR).

![Figure 1](image.jpg)

**Figure 1.** Measured (symbols) [10, 18–21] and calculated (solid curve by CRM, dotted line [22], and dash-dot [23]) for nitrogen: (a), (c), (d) values of excitation rate coefficients $K_C$ and $K_A$ of electronic states C$^1\Pi_u$ and A$^1\Sigma_u^+$, respectively (see text); (b) electron characteristic energy, $D/\mu$, and temperature, $T_e$; (e) electron drift velocity, $v_{dr}$.

3. **Application of CRMs for modeling of nonequilibrium low temperature plasma parameters in N$_2$ and H$_2$**

The modeling and processing of the measured data are based on the self-consistent modeling of the kinetic processes in NLTP via the joint solution of the kinetic Boltzmann equation for the electron energy distribution function, the thermal conductivity equation, and the balance equations for heavy-
particles in the ground and excited states considering heterogeneous processes on the surface of the reactor wall, as well as in the simulation of CARS and of the emission spectra.

Figure 2. Measured (symbols) [10,17,18] and calculated (solid curve by CRM, and dotted line [22]) vs. $T_v$ and the reduced electric field $E/N$: (a), (b), (c) EEDF in DCGD in $N_2$; (d) first Townsend ionization coefficient $\alpha/N$ in $N_2$.

Figure 3. CRM calculated (solid curve) and measured (symbols) EEDF: a) in a nitrogen microwave discharge excited in a prismatic resonator [5]; b) in a nitrogen plasma excited by a microwave surface-wave [24].

Figure 4. Measured (symbols) [18, 19] and calculated by the CRM (solid curve) in $H_2$: (a) electron drift velocity $v_{dr}$; (b) electron characteristic energy $D/\mu$; (c) EEDF vs. $E/N$ at $T_g=T_v=325$ K.

3.1. Electron energy distribution function

In an overwhelming number of the papers, which are reviewed in [10–13], on the solution of the kinetic Boltzmann equation for the EEDF are taken into account summary cross sections of excitation and deactivation for the total population densities of the electronically-excited states of diatomic
molecules without resolution on vibrational levels. Preference is given to experimental data in the formation of the database. For the refinement of the absolute values of the cross sections the method of formation of the self-consistent set of cross sections is used \[16\]. Absolute values of the cross sections are determined via a comparison of the EEDF-based calculated data with the experimental one for the electron average energy, \(<e>\), the first Townsend ionization coefficient, \(\alpha/N\), the electron temperature, \(T_e\), the drift velocity, \(v_{dr}\), and the characteristic energy, \(D/\mu\) (\(D\) is the electron diffusion coefficient, and \(\mu\) is the electron mobility), as well as for the summary values of the electron-impact excitation rate coefficients, \(K_Y\), of electronic states \(Y\) of molecules. However this method is ambiguous \[17\]. A more sequential approach for the database formation is presented in \[3,5,8,13\]. It implies that along with the data for \(<e>\), \(\alpha/N\), \(T_e\), \(v_{dr}\), \(D/\mu\), and \(K_Y\) additional experimental data on NLTP of different discharges is used: EEDF; vibrational distribution functions (VDFs) of molecules in the ground and electronically-excited states; translational, \(T_g\), and vibrational, \(T_v\), temperatures; specific power consumption, etc. In contrast to most of papers \[10–13\] (see also references therein) the level cross sections of excitation of the electronic excited states of the molecules are directly taken into account while solving the kinetic Boltzmann equation for the EEDF. This improves the quantitative description of experiments. The results of the database formation for the level cross sections and the comparison of their absolute values with the experimental data are given in \[13\].

![Figure 5](image-url.com)

**Figure 5.** Measured (symbols) \[1,7,25–27\] and calculated by CRM (solid curve): (a) EEDF vs. \(T_v\) in DCGD in hydrogen at \(E/N = 20\) Td; (b) VDF in inductive-capacitive RF discharge in hydrogen at a pressure 1.5 Torr.

Figures 1–5 show results from measurements \[5,10,18-21,24\] and calculations (in frame of CRM and from \[22,23\]) versus \(T_v\) and the reduced electric field \(E/N\): EEDF in DCGD, radio-frequency and microwave discharges in \(H_2\) and \(N_2\); \(\alpha/N\) and summary values of the excitation rate coefficients \(K_Y\) of the electronic state \((Y=A'^3\Sigma^+_u, C'^3\Pi_x)\) of nitrogen in DCGD and for the conditions of electron swarm experiments in \(N_2\); the characteristic energy \(D/\mu\), the temperature \(T_e\), and drift velocity \(v_{dr}\) of electrons for the conditions of electron swarm experiments in \(N_2\). As is seen from figures 1–5, there is a good agreement between theory and experiment for the formed self-consistent sets of level cross sections of elementary processes. The calculated values of \(\alpha/N\), \(T_e\), and \(D/\mu\) coincide also well with their calculated results by authors \[22,23\]. It is important to emphasize that the agreement between the measured and calculated EEDFs takes place at values of \(T_v\) in the range 4000–6000 K. The same values of \(T_v\) are obtained in calculations by the CRM and from experiments using Raman scattering technique \[28\], CARS spectroscopy \[1–3, 29–37\], and absorption spectroscopy \[38\] in DCGD and microwave discharges and their afterglows in \(N_2\).
Table 1. Computational and experimental results for $T_v$ and $T_g$.

| p (Torr) | R (cm) | $N_e$ (cm$^{-3}$) | t (ms) | $E/N$ (Td) | Experiment $T_v$ (K) | Theory $T_v$ (K) | Ref. | Type of discharge |
|---------|--------|-------------------|--------|------------|----------------------|------------------|------|-----------------|
| 3.5     | 1.8    | $3.5 \times 10^9$ | 20     | 80         | 4000±400$^a$         | 3790±400$^a$     | [1, 3] DCGD in N$_2$ |
|         |        |                   |        |            | 480±30$^b$           | 460±25$^b$       |      | DCGD in N$_2$    |
| 7.0     | 1.8    | $1.2 \times 10^{10}$ | 15    | 45         | 4100±450$^a$         | 4320±400$^a$     | [1, 3] DCGD in N$_2$ |
|         |        |                   |        |            | 530±40$^a$           | 540±30$^a$       |      | DCGD in N$_2$    |
| 9.5     | 1.8    | $6 \times 10^9$    | 15    | 60         | 4150±450$^a$         | 4270±400$^a$     | [1, 3] DCGD in N$_2$ |
|         |        |                   |        |            | 600±40$^a$           | 580±30$^a$       |      | DCGD in N$_2$    |
| 15      | 1.8    | $2 \times 10^{10}$ | 30    | 70         | 1000±100$^a$         | 1140±110$^b$     | [1, 3, 33] DCGD in N$_2$ |
| 30      | 1.8    | $5 \times 10^{10}$ | 30    | 67         | 1350±130$^b$         | 1300±130$^b$     | [1, 3, 33] DCGD in N$_2$ |
| 15      | 1.8    | $6.8 \times 10^9$  | 30    | 70         | 775±60$^a$           | 760±50$^a$       | [1, 33] DCGD in N$_2$ |
| 20      | 1.8    | $4 \times 10^{10}$ | 30    | 68         | 1200±110$^a$         | 1230±120$^a$     | [3, 33] DCGD in N$_2$ |
| 2       | 1      | $2 \times 10^{10}$ | 11    | 80         | 5300±400$^a$         | 540±30$^a$       | [34] DCGD in N$_2$ |
| 12      | 0.7    | $\approx 1 \times 10^9$ | 30   | >100       | 2850±100$^a$         | 395±12$^a$       | [35] DCGD in N$_2$ |
| 20      | 1      | $3.9 \times 10^{10}$ | 30    | 60         | 1150–1200$^b$        | 1170±1200$^b$    | [36] DCGD in N$_2$ |
| 20      | 1.6    | $1 \times 10^9$    | 30    |           | 2400±400$^b$         | 330±20$^b$       | [1, 37] CIRFD in H$_2$ |
| 2       | 2$^d$  | $1 \times 10^9$    | 30    | <2000      | 540±40$^b$           | <2000±40$^b$     | [25] CIRFD in H$_2$ |
| 1.5     | 1.6    | $1 \times 10^9$    | 30    | 3400±540$^b$ | 540±120$^b$        | 3100±120$^b$     | [26, 27] CIRFD in H$_2$ |
| 8       | 1.6    | $1 \times 10^9$    | 30    | 2700±420$^b$ | 750±50$^b$         | 2700±50$^b$     | [26, 27] CIRFD in H$_2$ |
| 1       | 1      | $9 \times 10^9$    | 33    | 3500±400$^b$ | 460±50$^b$         | 460±50$^b$      | [5] MWRD in N$_2$ |
| 1.7     | 1.5    | $1.5 \times 10^{11}$ | 33   | 4500±500$^c$ | 1300±150$^c$       | 1300±150$^c$     | [5] MWWD in N$_2$ |
| 1       | 1.1    | $1.1 \times 10^{11}$ | 33   | 4500±500$^c$ | 550±50$^c$         | 550±50$^c$      | [9] EMD in N$_2$ |

$^a$Scanning CARS spectroscopy with the collinear interaction of the laser beams.
$^b$Broadband CARS spectroscopy with the collinear interaction of the laser beams.
$^c$Optical interferometry.
$^d$Optical emission spectroscopy.
$^e$Two-wavelength CARS spectroscopy with the collinear interaction of the laser beams.
$^f$Scanning CARS spectroscopy with sharp laser beam focusing (Planar BOXCARS).
$^g$RF discharge was created between two plane electrodes spaced by 2 cm.

3.2. Vibrational-translational nonequilibrium in nitrogen and hydrogen plasmas

The results of the database formation for the level rate coefficients, describing vibrational kinetics of N$_2$ and H$_2$ in the ground electronic state, and a comparison of their absolute values with the experimental data are given in [1,3]. The mechanisms causing gas heating and the formation of VDFs of these molecules in the ground electronic state as function of residence time in an active region of discharges are discussed in detail in [1,3].
Figure 6. Calculated (solid curves by CRM, dash-dot lines by Boltzmann’s, and dotted line by Treanor’s formulas) and measured (symbols) (a) [29–32], (b) [35], (c) [34], and (d) [38] VDFs in the ground state $X^1Σ^+$ of nitrogen molecules.

Figure 7. Measured (symbols) and calculated (solid line) emission spectra of $N_2(C^3Π_u,v_C → B^3Π_g,v_B)$ and $N_2(B^2Σ_u^+,v_{Bi} → X^2Σ_g^+,v_{XG})$ in nitrogen plasma of EMD at $p = 1$ Torr, electron density $N_e=10^{11}$ cm$^{-3}$ and electric field strength 139 V/cm [9] in the spectral range: (a) 340–360 nm; (b) 360–380 nm; (c) 380–435 nm.

Table 1 and figures 5(b) and 6 compare the computational results by CRMs for the relative population densities over vibrational levels, $v$, of nitrogen $N_2(X^1Σ^+_g,v)$ and hydrogen $H_2(X^1Σ^+_g,v)$ molecules, and the values of $T_g$ and $T_v$ with measurements performed by different techniques in plasmas of DCGD and contracted discharge, and radio-frequency and microwave discharges. The experimental data were obtained in our laboratory [1,3,5,9,29–33] and in a series of experiments published in [25–27,34,35,37]. The measured values of $T_g$ and $T_v$ listed in table 1 were obtained at different values of the pressure, $p$, the electron number density, $N_e$, the internal radius of discharge tube, $R$, and residence time, $t$, of the molecules in active region of discharges. The relative population densities in the $N_2(X^1Σ^+_g,v)$ and $H_2(X^1Σ^+_g,v)$ states and the values of $T_g$ and $T_v$ were determined using two approaches. In the first of them, the sought parameters are determined from the comparison
of the amplitudes and line contours in the experimental and calculated broadband CARS spectra. In the second approach, the sought temperatures and distributions are determined from the processing of scanning CARS spectra by the amplitude technique. It is seen that measured and calculated parameters in frame of the CRM are in a good mutual agreement. The measured and calculated values of $T_g$ and $T_v$ in nitrogen plasmas of DCGD, contracted discharge and radio-frequency discharges lie in the ranges $395-1350$ K and $2400-5300$ K, respectively. In nitrogen plasmas of EMD the values of $T_v$ and $T_g$, obtained from spectroscopic measurements [9], are equal to $4500\pm450$ K and $550\pm50$ K, respectively. For radio-frequency discharges in H$_2$ [25–27] the values of $T_v$ and $T_g$, obtained from broadband CARS measurements, are not higher than $3400$ K and $750$ K, respectively.

3.3. Vibrational kinetics of electronically-excited states of nitrogen molecules

The results of the database formation for the level rate coefficients, describing vibrational kinetics of nitrogen molecules in the electronically-excited states, and a comparison of their absolute values with the experimental data are given in [8,13]. The mechanisms responsible for the formation of VDFs of molecules in the electronically-excited states as function residence time in an active region of the discharges are also discussed in detail in [8,13].

![Figure 8](image.jpg)

*Figure 8.* Calculated (solid and dotted line by CRM) and measured (symbols) VDF in the state $C\Pi_u$ of N$_2$ vs. $t$ in the active discharge region: (a), (b), and (c) [39]; (d) [40]; (e), (f) and (g) [1,13]; (h) [1,13].
Figure 9. Calculated (solid line by CRM) and measured (symbols) VDF in the state B^3Π_g of N_2 vs. t in the active discharge region: (a), (b) [41]; (c), (d) [42]; (e), (f) [43].

Figure 10. Calculated (solid line by CRM) and measured (symbols): (a) concentrations of atomic nitrogen in 4S state and nitrogen molecules in A^3Σ_u^+,v_A = 0 [45]; (c) and (d) VDF in A^3Σ_u^+ state [44].

During the modeling of the experiments, a special attention was given to the determination of populations on vibrational levels v_C = 0–4, v_B = 0–17, and v_A = 0–13 of the states C^3Π_u, B^3Π_g, and A^3Σ_u^+, of nitrogen molecules, respectively. These states are the upper ones for transitions in nitrogen: second positive system N_2(C^3Π_u,v_C → B^3Π_g,v_B), first positive system N_2(B^3Π_g,v_B → A^3Σ_u^+,v_A), and Vegard-Kaplan system N_2(A^3Σ_u^+,v_A → X^3Σ_g^+,v). The VDFs in electronically-excited states were determined from a comparison of the amplitude and contour of the vibronic bands of different sequences of measured and calculated emission spectra [6,8,9,13]. The comparison of spectra was performed at a constant value of the preliminary found rotational and translational temperatures. These temperatures were determined by an unresolved rotational structure technique. Figure 7, for example, illustrates a comparison of the calculated and experimentally recorded emission spectra of N_2(C^3Π_u,v_C → B^3Π_g,v_B), and first negative system N_2(B^3Σ_u^+,v_B → X^3Σ_g^+,v) of the molecular ion of nitrogen in an electrode microwave discharge. Values of T_g determined from bands, corresponding
to the different sequences of \( N_2 \left( C^3 \Pi_u, v_C \rightarrow B^1 \Pi_g, v_B \right) \) and \( N_2^+ \left( B^3 \Sigma^+ u_v, v_B \rightarrow X^1 \Sigma^+_u, v_X \right) \), coincided. In nitrogen DCGD, the values of \( T_g \) determined from emission spectra are in good agreement with results of measurements performed by OI and CARS spectroscopy \([4,8]\) (see table 1). Figure 8 shows the results of calculations and measurements \([1,13,39,40]\) of populations on vibrational levels \( v_C = 0 \sim 4 \) of the \( C^3 \Pi_u \) state of \( N_2 \) as a dependence on the residence time of the molecules in the discharge region. For the states \( B^1 \Pi_g \) and \( A^3 \Sigma^+_u \) of \( N_2 \) a similar comparison of the calculation results and the experimental values \([41-44]\) is shown in figure 9 and figure 10, respectively. The semi-empirical self-consistent level-to-level CRM in \( N_2 \) adequately reflects alternations of the relative populations on vibrational levels \( v_C = 0 \sim 4, v_B = 0 \sim 17, \) and \( v_A = 0 \sim 13 \) of the states \( C^3 \Pi_u, B^1 \Pi_u, \) and \( A^3 \Sigma^+_u \) measured in \([1,13,40-44]\). Figure 10(a) presents the results of calculations and measurements \([45]\) of absolute concentrations of atomic nitrogen in the ground electronic state \( N(\ ^4S) \) and of nitrogen molecules in the metastable state \( A^3 \Sigma^+_u, v_A = 0 \). It was established that the calculated structure of the vibrational distributions and the concentrations of molecules and atoms in the ground and excited states depend on the kinetic scheme, experimental conditions, the database for level rate coefficients, and (which is especially important) on their dependence on the translational temperature, as well as on set of the level cross sections for elementary processes used in the semi-empirical self-consistent level-to-level CRM.

4. Conclusion

Computation-experimental methods of investigation of the nitrogen and hydrogen plasmas of different types of discharges are developed. These methods are based on processing of the emission and CARS spectra of the plasma by kinetic level-to-level semi-empirical collisional-radiative models. The reliability of the developed models is confirmed by a comparison of the results of simulation with a great body of the published data for different types of discharges. These models have been already effectively used in diagnostics of nonequilibrium plasma of DC and microwave discharges \([1-9]\).

Acknowledgements

This study was partly supported by the Program \# 9 for Fundamental Researches of the Presidium of the Russian Academy of Sciences and RFBR Grant 07-08-00020.

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