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FAST TRACK COMMUNICATION

NO-γ emissions from streamer discharges: direct electron impact excitation versus resonant energy transfer

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Abstract
It has been established that production of NO-γ emission in pulsed corona discharges is dominated by the energy transfer from $N_2(A\,^3\Sigma_u^+)$ to the NO ground state $NO(X\,^2\Pi_r)$ while direct excitation by electron impact is negligible. However, recent studies suggest that the electron impact excitation plays a more important role. In this work, we report modelling results of NO-γ emission associated with streamer discharges using two cross section data sets available in the literature. The first set was originally reported by Mojarrabi *et al* (1996 *Phys. Rev.* A **54** 2977–82) and later updated by Brunger *et al* (2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 809–19); the second set was published by Hayashi (1990 *Nonequilibrium Processes in Partially Ionized Gases* (*NATO Advanced Science Institutes Series, Series B, Physics* vol 220) ed M Capitelli and J N Bardsley (New York: Plenum) pp 333–40). According to the results, the role played by the electron impact excitation in the production of NO-γ is drastically different when different cross sections are used. The results indicate that the first data set leads to better agreement with experimental measurements.

1. Introduction

The emission of NO-γ, $NO(A\,^2\Sigma^+) \rightarrow NO(X\,^2\Pi_r) + h\nu$, has been used as a diagnostic tool for the density of an important metastable state, $N_2(A\,^3\Sigma_u^+)$, produced in N_2 based plasma discharges (e.g., Simek *et al* (1998), Simek (2002, 2003a, 2003b)). In those plasma discharges, the upper excited states $NO(A\,^2\Sigma^+)$ are generated through two pathways: resonant energy transfer from $N_2(A\,^3\Sigma_u^+)$ to the NO ground state and direct excitation by electron impact. It has been generally accepted that the energy transfer process plays a dominant role (e.g., Tochikubo and Teich (2000), Simek (2002) and Ono and Oda (2005)). Measuring the intensity of NO-γ emission from the discharges therefore gives information on the concentration of $N_2(A\,^3\Sigma_u^+)$. Because the wavelength of NO-γ emission overlaps with N_2 Lyman–Birge–Hopfield (LBH) emission, special attention must be paid to distinguish between those two emissions in experiments (e.g., Tochikubo and Teich (2000)).

The NO-γ emission has also been studied in the context of upper atmosphere science as NO is an important minor species for the formation of lower ionosphere. For example, Minschwaner *et al* (2004) collected satellite data on the intensity of NO-γ band emission to derive the concentration of nitric oxide in air. The production of this emission during periods of auroral activity was modelled by Cartwright *et al* (1998). Liu and Pasko (2007) recently reported a modelling study on the NO-γ emission produced in large air discharges in the upper atmosphere known as sprites (Sentman *et al* 1995). Sprites are known to consist of filamentary discharges with a transverse size of

tens of metres, which are pressure scaled analogues of much smaller streamer discharges at atmospheric pressure (Stenbaek-Nielsen and McHarg 2008). It is found that the integral intensity of the NO-γ emission associated with sprite streamers is substantially weaker than that of the N_2 LBH emission and the production of NO($A\,^2\Sigma^+$) is predominantly by the resonant energy transfer process. The resultant NO-γ emission in sprite discharges is most likely to be observable from space only by a dedicated narrow band photometer with a passband of 240–260 nm in which N_2 LBH emissions are absent. However, a recent modelling study by Gordillo-Vázquez (2008) on sprite chemistry raised a different point of view that in sprite streamer heads the dominant production reaction of NO($A\,^2\Sigma^+$) is the direct electron impact reaction with the resonant energy transfer playing a negligible role.

In this paper, we present modelling results of the NO-γ emission produced during streamer discharges at ground pressure. Our goal is to identify the cause of the difference between our previous study and the work by Gordillo-Vázquez (2008). We show that different cross sections for electron impact excitation of NO($A\,^2\Sigma^+$) used in the two studies result in the disagreement. By comparing with experiments, it is found that the cross section reported in Mojarrabi *et al* (1996) and Brunger *et al* (2000) gives more consistent results with experimental measurements than that by Hayashi (1990). The electron impact excitation plays a minor role in the production of NO-γ emission in streamer discharges when the cross section from Mojarrabi *et al* (1996) and Brunger *et al* (2000) is used in the streamer model.

2. Model formulation

A simple kinetic model to study NO chemistry associated with streamer discharges in air is used in this work and its detailed description is given by Liu and Pasko (2007). This model includes neutral species such as N_2, O_2, N(^2D), N(^4S), O(^3P), $N_2(A\,^3\Sigma_u^+)$, $N_2(B\,^3\Pi_g)$, $N_2(C\,^3\Pi_u)$, $N_2(a\,^1\Pi_g)$, NO($X\,^2\Pi_r$) and NO($A\,^2\Sigma^+$). It takes into account the electron impact excitation of the N_2 excited states and the deactivation of those states by radiative transition and collisional quenching, and the dissociation of N_2 and O_2 as well. It also includes the two excitation mechanisms of NO($A\,^2\Sigma^+$) discussed above. This model is solved simultaneously with a streamer discharge model consisting of Poisson's equation and continuity equation of charged particles (see, e.g., Liu and Pasko (2004) and Bourdon *et al* (2007)). However, different from the model used in Liu and Pasko (2004, 2007), the streamer model used in this study adopts the recently developed SP_3 method to calculate the photoionization production rate of electron–ion pairs (Bourdon *et al* 2007, Liu *et al* 2007). As a whole, the model is able to simulate not only the development of streamers but also the major emission band systems of N_2 and N_2^+ from streamers as well as the NO-γ emission (Liu and Pasko 2004, Liu *et al* 2008).

To our knowledge, two cross section data sets for electron impact excitation of NO($A\,^2\Sigma^+$) are available in the literature. The cross section originally reported by Mojarrabi *et al* (1996) was later updated by Brunger *et al* (2000). This cross section was used by Simek (2002) to study the NO-γ emission from the streamer discharges at atmospheric pressure and by Cartwright *et al* (1998) to model NO-γ production in auroras. Liu and Pasko (2007) used the excitation rate obtained by Simek (2002) to model the same emission from sprite streamers. The other cross section was published by Hayashi (1990), and Gordillo-Vázquez (2008) employed this set for their model. The two cross section sets are shown in figure 1. Hayashi's cross section is about two orders of magnitude larger than Brunger's. The corresponding excitation rate coefficients as a function of electric field are calculated using the Boltzmann equation solver BOLSIG+ (Hagelaar and Pitchford 2005). These coefficients are shown in figure 2, where the rate coefficient of N_2 LBH emission is also included for comparison. The excitation rate coefficient calculated with the cross section from Hayashi (1990) is again about two orders of magnitude higher than that from Brunger *et al* (2000). This difference has also been noted by Gordillo-Vázquez (2008), but the author incorrectly attributed it to different electron energy distributions used. The data points can be fitted by a function in a form of $A\exp(B/|EN_0/N|)$. For Brunger's set, A and B take the values of $A = 2.5127 \times 10^{-16}\,\mathrm{m^3\,s^{-1}}$ and $B = -9.3416 \times 10^6\,\mathrm{V\,m^{-1}}$, respectively, while $A = 3.1493 \times 10^{-14}\,\mathrm{m^3\,s^{-1}}$ and $B = -9.2699 \times 10^6\,\mathrm{V\,m^{-1}}$ for Hayashi's set. We use those two functions in our model.

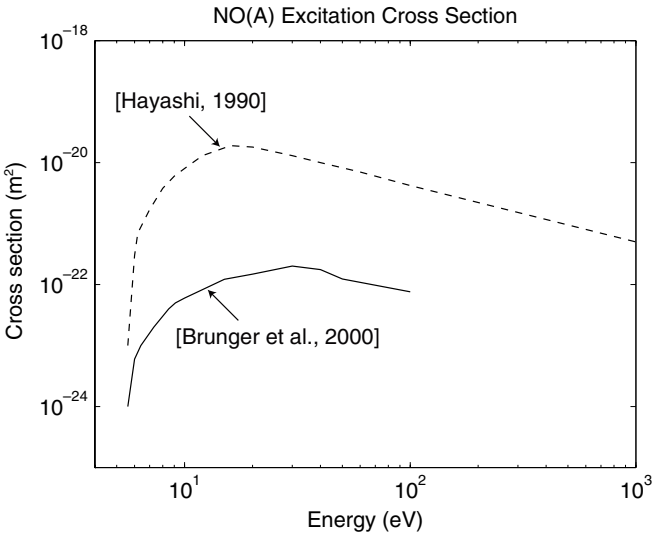

Figure 1. Solid line: the cross section reported by Brunger *et al* (2000); dashed line: the cross section reported by Hayashi (1990).

The simulation domain is identical to the one employed by Liu and Pasko (2006), which is a rectangular box corresponding to a cross section of the discharge region. An external uniform electric field is established in the simulation domain by two remote electrodes and a small conducting sphere is then placed below the bottom boundary to enhance the field in a small region around it for the initiation of the streamer. The air pressure is fixed at 760 Torr. The external homogeneous field E_0 is set at $1.5 \times 10^6\,\mathrm{V\,m^{-1}}$ and pointing

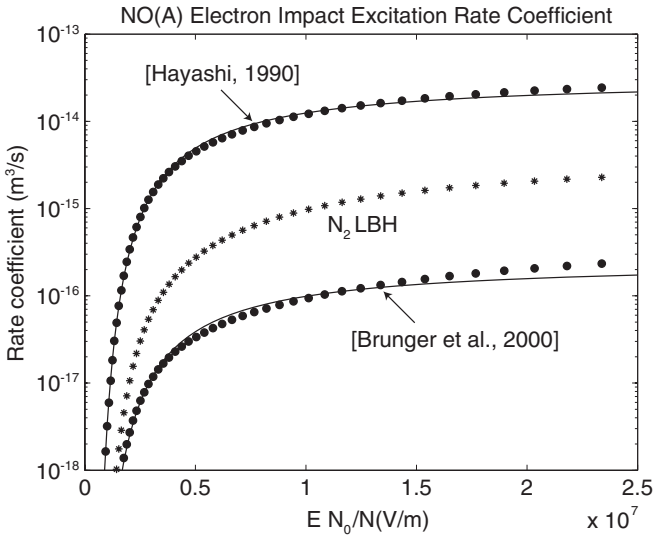

Figure 2. Dots and stars are data points calculated using BOLSIG+. Solid lines are fitting function in a form of $A\exp(B/|E|)$ (see text for the values of A and B for each case). N_0 and N are densities of gas at atmospheric pressure and pressure under consideration, respectively.

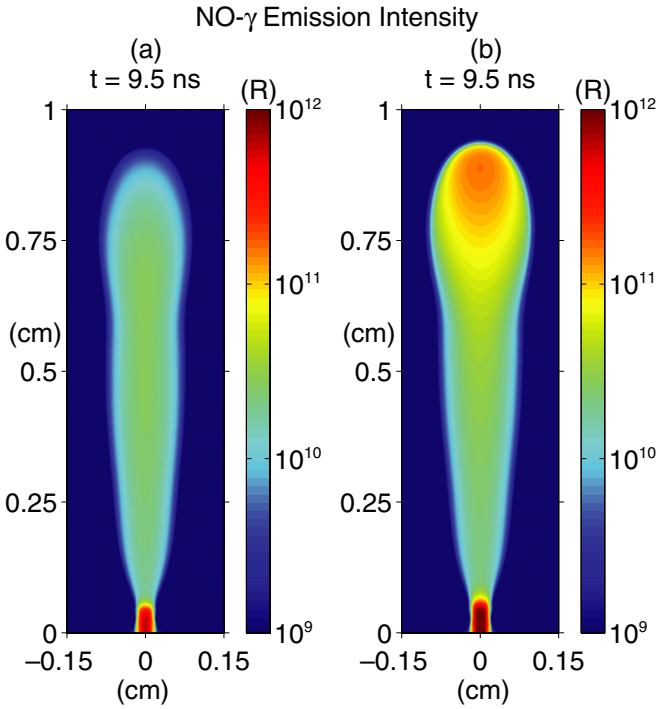

Figure 3. NO-γ emission intensity distribution in Rayleigh units of the model streamer calculated using the cross section from (*a*) Brunger *et al* (2000); (*b*) Hayashi (1990).

(This figure is in colour only in the electronic version)

in the positive z direction. The radius and the potential applied to the conducting sphere are 0.1 cm and 5000 V, respectively. To initiate the streamer, as a common practice, we place a cloud of plasma with spherically symmetric Gaussian spatial distribution on the axis of symmetry in the vicinity of the sphere, i.e. $n_e = n_p = n_0 \exp[-(r/\sigma_r)^2 - ((z - z_0)/\sigma_z)^2]$, where n_e and n_p are the densities of electrons and positive ions, respectively, $n_0 = 10^{18}\,\mathrm{m}^{-3}$, $\sigma_r = \sigma_z = 0.01\,\mathrm{cm}$ and $z_0 = 0.02\,\mathrm{cm}$. The size of the computational domain is $1.0 \times 0.15\,\mathrm{cm}^2$. The computational grid is uniform in both radial and axial directions with 1401 and 211 grid points, respectively. The air is a mixture of 78.11% N_2, 20.91% O_2 and 0.98% Ar gases with a density of $2.688 \times 10^{25}\,\mathrm{m}^{-3}$ at the ground pressure. The concentration of NO is set at 400 ppm, the same value as in the experiments of Tochikubo and Teich (2000). The coefficients for ionization, attachment and excitation used in the simulation are taken from the work by Moss *et al* (2006) unless otherwise specified.

3. Results and discussion

To study the difference in the NO-γ intensity when the two cross sections are used, two streamer simulations were conducted using the respective excitation rate coefficients. As the NO chemistry in our model does not affect the dynamics of the streamer, the modelling results for electric field and electron density are exactly the same for the two simulations. The streamer initiates in the strong field region near the bottom boundary and propagates along the direction of the external homogeneous field. Although from direct comparison of cross sections (figure 1) and rate coefficients (figure 2) significant differences are expected in resultant densities of NO($A\,^2\Sigma^+$) molecules, we conduct modelling of the spatially and temporally resolved chemical dynamics of NO($A\,^2\Sigma^+$) produced by a model streamer in order to accurately quantify the relative importance of the direct and the resonant energy transfer production channels of NO-γ emissions for the two cross sectional data sets shown in figure 1. The results for NO-γ emission intensity distribution of the model streamers are shown in figure 3. The intensity for Brunger's case is relatively uniform along the streamer channel. However, Hayashi's case shows enhancement in the region corresponding to the streamer head where the intensity is about two orders of magnitude stronger than Brunger's case. This enhancement is similar to those present in the emissions from N_2 and N_2^+ excited states shown in Liu *et al* (2008). It indicates effective production of the excited states NO($A\,^2\Sigma^+$) by electron impact in the streamer head, which are then quickly deactivated due to collisional quenching and radiative transition. The natural lifetime of NO($A\,^2\Sigma^+$) is $\sim$0.2 μs, and these species are mainly quenched by O_2 with a coefficient of $1.62 \times 10^{-16}\,\mathrm{m}^3\,\mathrm{s}^{-1}$ (Simek 2003a). If the quenching effect is taken into account, the total lifetime of NO($A\,^2\Sigma^+$) is about 1.1 ns at ground level. The presence of the enhancement for Hayashi's case therefore is consistent with such a short lifetime. In the channel behind the streamer head, the NO($A\,^2\Sigma^+$) states are mainly produced by the resonant energy transfer from $N_2(A\,^3\Sigma_u^+)$ metastable states. The metastables $N_2(A\,^3\Sigma_u^+)$ are mainly produced by the passage of the streamer head. Their total lifetime at 760 Torr accounting for both radiative transition and quenching (see, e.g., Guerra *et al* (2001) and Liu and Pasko (2007)) for quenching rates) is approximately 70 ns, which is much longer than the simulation time, so that the density of $N_2(A\,^3\Sigma_u^+)$ stays at a constant level after the passage of the streamer head and

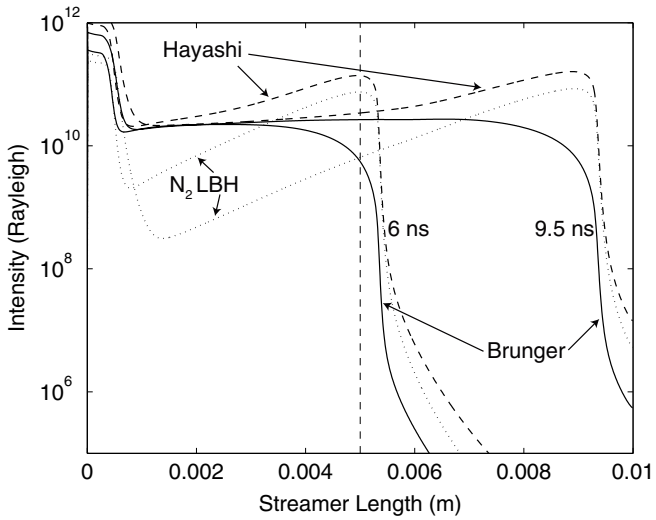

Figure 4. NO-γ emission intensity profiles at 6 ns and 9.5 ns calculated using two separate electron impact cross sections from Brunger *et al* (2000) and Hayashi (1990) along the symmetry axis of the streamer. The profile for N_2 LBH emission is also given for comparison.

the production of NO($A\,^2\Sigma^+$) is maintained at a constant level in the streamer channel.

Gordillo-Vázquez (2008) conducted a modelling study on sprite chemistry using a zero dimensional but a more complete chemistry model than this study. The streamer was simulated by an electric field pulse with preset width and magnitude. The obtained results show that the NO-γ emission is enhanced in the streamer head and the production of NO($A\,^2\Sigma^+$) is dominated by the electron impact excitation, which disagrees with the work in Liu and Pasko (2007). The author argued that the difference resulted from the fact that Liu and Pasko (2007) used an excitation rate that was calculated by assuming a Maxwellian electron distribution function (Simek 2002). According to the results obtained in this work, it is clear that the disagreement is due to the use of different cross sections for electron impact excitation. When the cross section from Brunger *et al* (2000) is used, electron impact excitation plays a negligible role in the production of NO($A\,^2\Sigma^+$) in streamer discharges.

Many experiments have shown that the decay of NO-γ intensity can be fitted by a single exponential decay in the post-discharge period (e.g., Tochikubo and Teich (2000)). The decay constant matches the quenching rate of $N_2(A\,^3\Sigma_u^+)$ by O_2, which establishes that the dominant production mechanism for NO($A\,^2\Sigma^+$) is the resonant energy transfer in pulsed corona discharges. To analyse the decay of NO-γ present in the streamer head for Hayashi's case, we further investigate the emission intensity along the symmetry axis of the streamer as shown in figure 4. The intensity from Hayashi's cross section shows a peak in the streamer head, decays toward the channel and reaches a constant level in the end. The constant level agrees with the intensity from Brunger's cross section indicating the production of NO($A\,^2\Sigma^+$) is dominated by the resonant energy transfer after the passage of the streamer head even for the Hayashi case. The intensity of N_2 LBH is also peaked in the streamer head and decays towards the channel. However, as N_2 LBH is produced mainly by electron impact excitation, there is no constant intensity level in the streamer channel. At $z = 0.5$ cm (the vertical dashed line), the intensity of N_2 LBH drops from 7.5×10^{10} to 6.47×10^9 R in 3.5 ns. The factor of decrease $\frac{7.5\times10^{10}}{6.47\times10^9} = 11.6$ is very close to the exponential decay of $N_2(a\,^1\Pi_g)$ defined by its total lifetime 1.29 ns, $e^{3.5/1.29} = 15.1$. However, the decay of intensity of NO-γ after the passage of the streamer head does not follow a similar exponential decay because of the contribution from the resonant energy transfer. The intensity drops by a factor of $\frac{1.383\times10^{11}}{3.41\times10^{10}} = 4.0$ but $e^{3.5/1.1} = 24.1$. Nevertheless, if the electron impact excitation dominated over the resonant energy transfer in the production of NO($A\,^2\Sigma^+$) in the streamer head, it would be expected that the emission intensity quickly drops after the passage of the streamer head.

Spectroscopic measurements of pulsed corona discharges have shown that there are no strong peaks in NO-γ emission intensity at the active discharge period (see, e.g., Tochikubo and Teich (2000), figures 4 and 7), which are an observed feature of the first and second emission band systems of N_2 and the first negative band system of N_2^+ that are driven by electron impact excitation during this period. After the active discharge period, the NO-γ intensity slowly decays on a time scale determined by the quenching of $N_2(A\,^3\Sigma_u^+)$ as discussed above, which is inconsistent with the fast drop in intensity behind the streamer head as would be expected if the electron impact excitation is the dominant production mechanism for NO($A\,^2\Sigma^+$) in the streamer head. It can therefore be concluded that the cross section reported by Brunger *et al* (2000) is more consistent with experimental measurements.

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