Relaxation of Velocity Distribution of Electrons Cooled (Heated) by Rotational Excitation (De-excitation) of N_2

By

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Summary: The relaxations of the velocity distribution and temperature of electrons cooled or heated by the rotational excitation or de-excitation of nitrogen molecules are studied using the Monte Carlo simulation in the system where the electrons with the initial temperature $T_e(0)$ are diluted in the heat-bath molecules with the temperature T [$100 \leqslant T$, $T_e(0) \leqslant 3000$ K] and the rotational transition cross section is given by the Gerjuoy-Stein formula. The initial Maxwell electron velocity distribution is perturbed considerably in the cooling process and slightly in the heating process. The electron temperature is higher in the cooling process and lower in the heating process than that of Mentzoni and Row for the local Maxwell distribution. The temperature equilibration time is larger than that of Mentzoni and Row to the extent of about 25%. The initial δ-function electron velocity distribution approaches the Maxwell distribution through the rotational excitation and de-excitation only. The Maxwellization is much faster than the temperature equilibration.

1. Introduction

It is well known [1] that low-energy electrons in molecules lose or gain the energy mainly through the rotational excitation or de-excitation of molecules. When the electrons are so diluted in molecules that the effect of the electron-electron collision is negligible as compared with that of the electron-molecule collision, the behavior of the electron velocity distribution in the electron cooling or heating process is not well understood. It is not obvious that the electrons cooled or heated by the molecular rotational excitation or de-excitation obey the Maxwell velocity distribution, although the Maxwell distribution is often assumed for the theoretical interpretation of the measured results in the cross-modulation experiments [2, 3] and the D region of the ionosphere [4]. In fact, it is shown [5] that the stationary electron velocity distribution, which depends on the initial velocity distribution, in molecules with two energy levels is not the Maxwell distribution and reveals the saw-tooth pattern for the initial Maxwell distribution, where the electron-electron and electron-molecule elastic collisions are ignored as compared with the electron-molecule inelastic collisions.

The purpose of this paper is to study the relaxation of the velocity distribution of low-energy electrons cooled or heated by the rotational excitation or de-excitation of nitrogen molecules with much more rotational energy levels than two [5] using the Monte Carlo simulation [6] in the system where the electrons are so diluted in the heat-bath molecules that the effect of the electron-electron collision is negligible as compared with that of the electron-molecule collision and the effect

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of the electron-molecule elastic collision is ignored. It is investigated whether the electrons maintain the initial Maxwell distribution in the cooling process, which corresponds to the cross-modulation experiment, or in the heating process, which corresponds to the shock-wave heating, and the electron temperature obeys that of Mentzoni and Row [3] for the local Maxwell distribution. It is examined whether the initial δ-function velocity distribution, being far from the Maxwell distribution, approaches the Maxwell distribution through the rotational excitation and de-excitation only.

2. Relaxation Model

The time evolution of the electron velocity distribution function $f(\boldsymbol{v})$ in the spatially homogeneous system, where the electrons with the number density $n_e=\int f(\boldsymbol{v})d\boldsymbol{v}$ are diluted in the heat-bath molecules with the number density $n(\gg n_e)$ and the electron-electron collision is ignored, is described by the Boltzmann equation [7]

$$\partial f(\boldsymbol{v})/\partial t=\sum_J \sum_{J'} \int [f(\boldsymbol{v}')f_{J'}(\boldsymbol{v}'_{J'})g_J/g_{J'}-f(\boldsymbol{v})f_J(\boldsymbol{v}_J)]gI_{JJ'}(g,\Omega)d\Omega d\boldsymbol{v}_J, \quad (1)$$

where t is the time, $I_{JJ'}(g,\Omega)$ is the differential cross section for the electron scattering into the solid angle Ω with the molecular rotational transition $J\rightarrow J'$ [$e+M(J)\rightarrow e+M(J')$, $M(J)$ being the heat-bath molecules M in the Jth rotational level] and the change in the electron velocity ($\boldsymbol{v}\rightarrow\boldsymbol{v}'$) and the molecular velocity of $M(J)$ ($\boldsymbol{v}_J\rightarrow\boldsymbol{v}'_{J'}$), and $g=|\boldsymbol{v}-\boldsymbol{v}_J|$ is the relative velocity. $f_J(\boldsymbol{v}_J)$ is the velocity distribution function of $M(J)$ taken as the Maxwell distribution at the heat-bath temperature T

$$f_J(\boldsymbol{v}_J)=n_J(m_M/2\pi kT)^{3/2}\exp(-m_M v_J^2/2kT), \quad (2)$$

where $v_J=|\boldsymbol{v}_J|$, m_M is the molecular mass, k is the Boltzmann constant, and n_J is the number density of $M(J)$ taken as the Bolzmann distribution at the heat-bath temperature T

$$n_J/n=g_J\exp(-E_J/kT)/Q_r, \quad (3)$$

in which $n=\Sigma_J n_J$, $Q_r=\Sigma_J g_J\exp(-E_J/kT)$, $g_J=(2s+1)(s+a)(2J+1)$ is the degeneracy, s is the nuclear spin ($s=1$ and $a=0$ and 1 for odd and even J, respectively, for N_2) [8], $E_J=B_0J(J+1)$ is the rotational energy, and B_0 is the rotational constant ($B_0=2.00\,\mathrm{cm}^{-1}$ for N_2) [8].

Since the electron mass m is much smaller than m_M, it may be assumed that $g\simeq v(=|\boldsymbol{v}|\gg v_J)$, $g'=|\boldsymbol{v}'-\boldsymbol{v}'_{J'}|\simeq v'(=|\boldsymbol{v}'|\gg|\boldsymbol{v}'_{J'}|)$, and $\boldsymbol{v}_J\simeq\boldsymbol{v}'_{J'}$. On this assumption, Eq. (1) is simplified for the isotropic electron velocity distribution $P(v)=4\pi v^2 f(\boldsymbol{v})/n_e$ $\left[\int P(v)dv=1\right]$ as [9]

$$\partial P(v)/\partial t=\sum_J \sum_{J'} [n_{J'}P(v')v\sigma_{J'J}(v')-n_J P(v)v\sigma_{JJ'}(v)], \quad (4)$$

where $\sigma_{JJ'}(v)=\int I_{JJ'}(v,\Omega)d\Omega$ is the total cross section for the rotational transition

$J \to J'$ and satisfies the detailed balance $v^2 g_J \sigma_{JJ'}(v) = v'^2 g_{J'} \sigma_{J'J}(v')$, and the electron velocities v and v' are related by the energy conservation $mv^2/2 + E_J = mv'^2/2 + E_{J'}$. It is noted that the electron-molecule elastic collision ($J'=J$) is ignored in Eq. (4).

Multiplying Eq. (4) by the electron energy $\varepsilon = mv^2/2$ and integrating over v yield the usual electron energy-loss (gain) rate equation [2, 3]

$$d\bar{\varepsilon}/dt = \sum_J \sum_{J'} n_J (E_J - E_{J'}) \overline{v\sigma_{JJ'}(v)}, \tag{5}$$

where $\bar{\varepsilon} = \int \varepsilon P(v) dv$ is the average electron energy related to the electron temperature T_e by $\bar{\varepsilon} = 3kT_e/2$ and

$$\overline{v\sigma_{JJ'}(v)} = \int v\sigma_{JJ'}(v) P(v) dv. \tag{6}$$

When $P(v)$ is the Maxwell distribution at the electron temperature T_e

$$P_{MD}(v) = (m/2\pi kT_e)^{3/2} 4\pi v^2 \exp(-mv^2/2kT_e), \tag{7}$$

and $\sigma_{JJ'}(v)$ is given by the Gerjuoy-Stein formula ($J' = J \pm 2$) [2]

$$\begin{aligned} \sigma_{J,J+2}(\varepsilon) &= \{[(J+2)(J+1)]/[(2J+3)(2J+1)]\}\sigma_0 [1 - (4J+6)B_0/\varepsilon]^{1/2}, \\ \sigma_{J,J-2}(\varepsilon) &= \{[J(J-1)]/[(2J-1)(2J+1)]\}\sigma_0 [1 + (4J-2)B_0/\varepsilon]^{1/2}, \end{aligned} \tag{8}$$

where $\sigma_0 = 8\pi Q^2 a_0^2/15$, Q is the electric quadrupole moment in units of ea_0^2, and a_0 is the Bohr radius, $\overline{v\sigma_{JJ'}(v)}$ is analytically written as [3]

$$\overline{v\sigma_{J,J\pm2}(v)} = (8kT_e/\pi m)^{1/2} s_J b_J K_1(b_J) \exp(\mp b_J), \tag{9}$$

where $s_J = \{[(J+2)(J+1)]/[(2J+3)(2J+1)]\}\sigma_0$ and $\{[J(J-1)]/[(2J-1)(2J+1)]\}\sigma_0$ and $b_J = (2J+3)B_0/kT_e$ and $(2J-1)B_0/kT_e$ for $J' = J+2$ and $J-2$, respectively, and $K_1(b_J)$ is the modified Bessel function of the second kind. Using the fact of the smallness of the rotational constant B_0 in Eqs. (5) and (9), Mentzoni and Row [3] obtained the simple rate equation $dT_e/dt = (T - T_e)/\tau$ with the electron energy relaxation time $\tau/t_0 = 3k(TT_e)^{1/2}/8B_0$, where $t_0 = [n\sigma_0(8kT/\pi m)^{1/2}]^{-1}$ is the rotational collision time.

Since the rotational transition cross section is usually given [2] by $\sigma_{JJ'}(v)$ instead of $\sigma_{JJ'}(g)$, the simplified Eq. (4) is solved by the Monte Carlo simulation, although the Monte Carlo simulation [6, 7] is applicable to the exact Eq. (1). $\sigma_{JJ'}(v)$ for nitrogen molecules is taken as the Gerjuoy-Stein formula [Eq. (8)] with [10, 11] $Q \simeq -1.04\ ea_0^2$ in correspondence to the analysis of Mentzoni and Row [3]. The initial electron velocity distribution $P(0)$ at $t=0$ is taken as the Maxwell distribution [Eq. (7)] at the initial electron temperature $T_e(0)$ or the δ-function distribution

$$P(0) = \delta(v - v_1), \tag{10}$$

where v_1 is the initial electron velocity related to $T_e(0)$ by $v_1 = [3kT_e(0)/m]^{1/2}$.

3. Monte Carlo Simulation

The isotropic electron velocity distribution $P(v)$ described by Eq. (4) is obtained by the Monte Carlo simulation [6]: $P(v)$ at the time $t+\Delta t$ is obtained from $P(v)$ at the time t by following a large number of simulated electrons during the time step Δt through the molecular rotational excitation and de-excitation collisions. Since the molecular motion is ignored in Eq. (4), the Monte Carlo procedure is somewhat simplified:

(i) At the time t, an electron velocity v is assigned to each of simulated electrons of the number of N by the probability

$$p(v)=P(v). \tag{11}$$

(ii) A time interval Δt_c between successive electron-molecule collisions is assigned by the probability

$$p(\Delta t_c)=\nu \exp(-\nu \Delta t_c), \tag{12}$$

where ν is the total collision frequency of electron-molecule collisions given by

$$\nu=N \sum_J n_J \int v S_J(v) P(v) dv, \tag{13}$$

$S_J(v)=\Sigma_{J'} \sigma_{JJ'}(v)$ being the total scattering cross section. Since ν is evaluated at the time t and taken to be constant during Δt, the time step Δt should be so small that the difference between ν at $t+\Delta t$ and ν at t, $\Delta \nu=\nu(t+\Delta t)-\nu(t)$, satisfies the condition

$$|\Delta \nu|/\nu(t) \ll 1. \tag{14}$$

(iii) A collision pair of electron with a velocity v and molecule in a rotational level J is selected by the probability

$$p_J(v)=N n_J P(v) v S_J(v)/\nu. \tag{15}$$

(iv) A rotational level J' after collision is assigned by the probability

$$p_{J'}=\sigma_{JJ'}(v)/S_J(v). \tag{16}$$

(v) The electron velocity v' after collision is obtained from the energy conservation as

$$v'=[v^2+2(E_J-E_{J'})/m]^{1/2}. \tag{17}$$

(vi) The procedures (ii)–(v) are carried out until the accumulated time $\Sigma \Delta t_c$ exceeds Δt. The electron velocity distribution $P(v)$ at the time $t+\Delta t$ is obtained from the velocity distribution of simulated electrons.

It is noted that the velocity distribution $P(v)$ obtained by the Monte Carlo simulation is the average distribution in the velocity region $(v-\Delta/2,\ v+\Delta v/2)$ with the width of Δv,

$$\bar{P}(v)=\int_{v-\Delta v/2}^{v+\Delta v/2} P(v)dv/\Delta v. \tag{18}$$

Owing to the fact that there exist practical limits in the values of the velocity width Δv ($\rightarrow 0$) and the number N ($\rightarrow\infty$) of simulated electrons and the heat-bath molecules have much more rotational energy levels than two, the fine structure of $P(v)$ such as the saw-tooth pattern indicated by Peyraud [5] for the two-energy-level molecules may not be obtained for $\bar{P}(v)$. The electron temperature T_e is obtained from the average $\overline{v^2}=\int v^2P(v)dv\simeq\Sigma_N v^2/N$ as $T_e=m\overline{v^2}/3k$.

4. Results and Discussion

Since the electron energy should not exceed the molecular vibrational excitation threshold [1–3] in order that the molecular rotational excitation or de-excitation is the dominating electron energy loss or gain mechanism, the results are obtained for the temperature range $100\leqq T_e(0)$, $T\leqq 3000$K for the electron cooling $[T_e(0)>T]$ and heating $[T_e(0)<T]$ in nitrogen molecules. The time step Δt is taken as $0.5\lesssim\Delta t/t_0\lesssim 10$ so that the variation of the collision frequency $|\Delta\nu|/\nu(t)$ is within a few percent; the convergence of result is checked by decreasing $\Delta t/t_0$. The upper limit of the rotational level $J_{\max}$ is taken so that $(n_{J\max}/n)/(n_J/n)_{\max}\lesssim 0.001$, where $(n_J/n)_{\max}$ is the maximum value of the rotational distribution [Eq. (3)]; the convergence of result is checked by increasing $J_{\max}$. The velocity width and the number of simulated electrons are taken as $\Delta v/v_0=0.2$ and $10^3\lesssim N\lesssim 10^4$, where $v_0=(2kT_e/m)^{1/2}$ is the most probable speed.

A. Cooling Process

The time evolutions of the electron velocity distribution $\bar{P}\equiv\bar{P}(v/v_0)[=\bar{P}(v)v_0]$ for the initial Maxwell distribution $[P(0)=MD]$ in the cooling process are presented in Figs. 1–3. $\bar{P}$ deviates considerably from the Maxwell distribution (MD): $\bar{P}>MD$ at $v/v_0\lesssim 1$ or $v/v_0\gtrsim 2$ and $\bar{P}<MD$ at $1\lesssim v/v_0\lesssim 2$. The degree of deviation in-

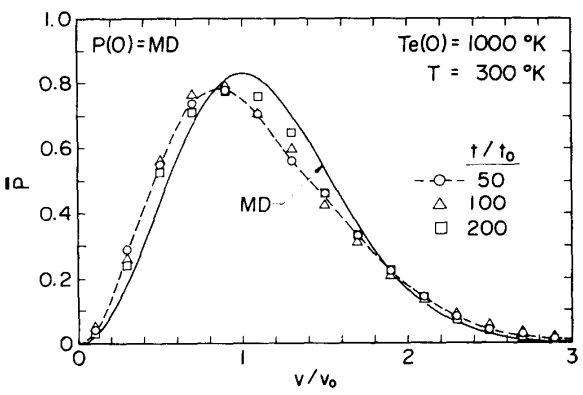

Fig. 1. Time evolution of electron velocity distribution for the initial Maxwell distribution (MD) in the cooling process.

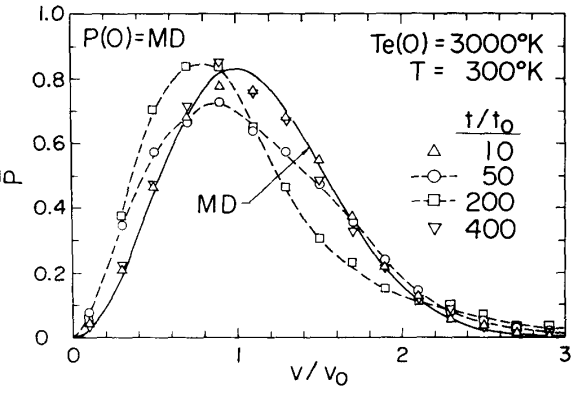

Fig. 2. Time evolution of electron velocity distribution for the initial Maxwell distribution (MD) in the cooling process.

creases with increasing $T_e(0)$ (Figs. 1 and 2) or decreasing T (Figs. 1 and 3) and is remarkable at $T_e(0)/T \sim 10$. [Even for $T_e(0)=500$K and $T=300$K, a little deviation is observed.] $\bar{P}$ deviates from *MD* to the largest extent at the middle stage of cooling (see Fig. 5) and approaches *MD* within the statistical uncertainty at the nearly same time when the electron temperature T_e reaches T (Fig. 5).

The relaxation of $\bar{P}$ for the initial δ-function distribution [$P(0)=\delta$] in the cooling

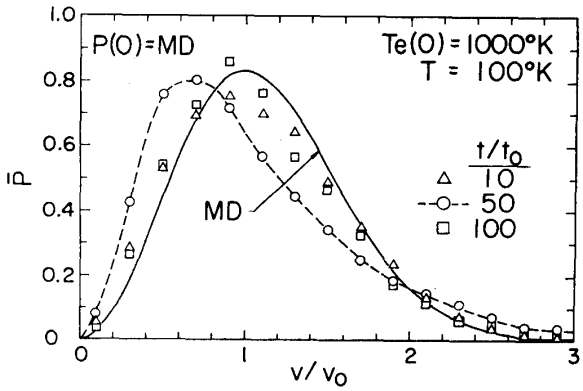

Fig. 3. Time evolution of electron velocity distribution for the initial Maxwell distribution (*MD*) in the cooling process.

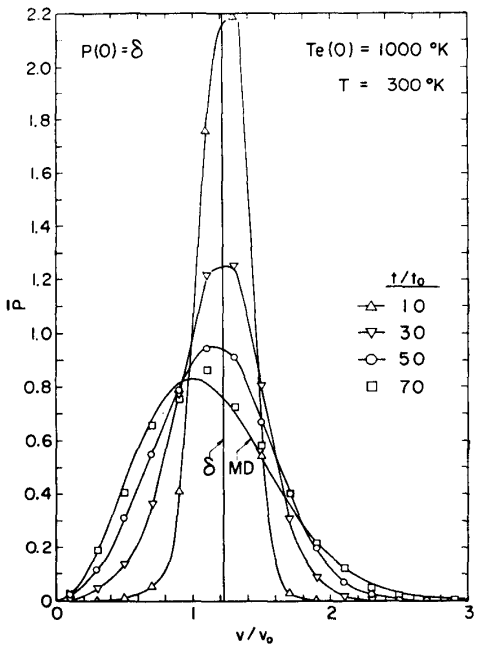

Fig. 4. Relaxation of electron velocity distribution for the initial δ-function distribution in the cooling process.

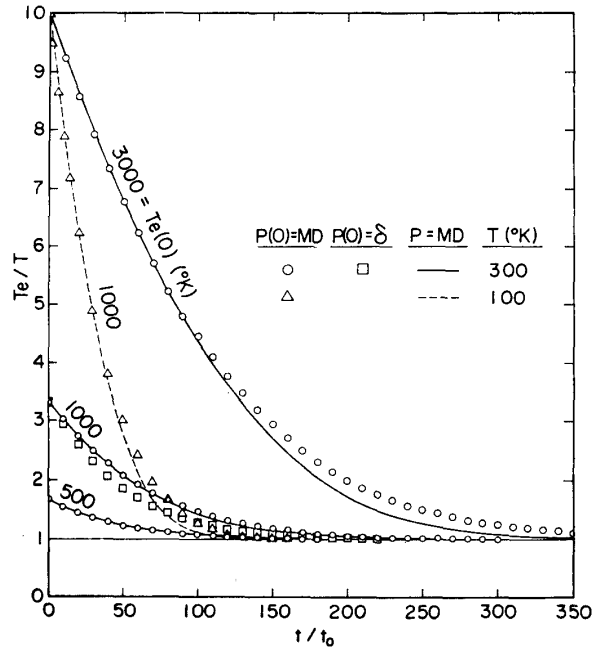

Fig. 5. Relaxation of electron temperature in the cooling process in comparison with that of Mentzoni and Row (*P=MD*).

process is presented in Fig. 4 for $T_e(0)=1000\text{K}$ and $T=300\text{K}$. The δ-function distribution, being far from the Maxwell distribution, approaches MD within the statistical uncertainty; the Maxwellization time τ_{MD} defined to be the time when $\bar{P}$ approaches MD within 5% is $\tau_{MD}/t_0 \simeq 100$. It is noted that the Maxwellization ($\delta \to MD$) is much faster than the temperature equilibration ($T_e \to T$) (Fig. 5).

The relaxation of the electron temperature T_e for $P(0)=MD$ in the cooling process is presented in Fig. 5 in comparison with the electron temperature T_{MR} of Mentzoni and Row [3] for the local Maxwell distribution ($P=MD$), which is obtained by solving Eqs. (5) and (9). The effect of $T_e(0)$ on the deviation of T_e from T_{MR} is shown for $T=300\text{K}$. For $T_e(0) \lesssim 500\text{K}$, T_e is in good agreement with T_{MR}, although $\bar{P}$ indicates a little deviation from MD. For $T_e(0) \gtrsim 1000\text{K}$, T_e is in agreement with T_{MR} in the first half of the cooling process and becomes higher than T_{MR} in the latter half owing to the considerable deviation of $\bar{P}$ from MD, which is also observed for $T=100\text{K}$ [$T_e(0)=1000\text{K}$]. The degree of deviation of T_e from T_{MR} increases with increasing $T_e(0)$ in correspondence to the degree of deviation of $\bar{P}$ from MD, which is shown for $T_e(0)=1000$ and 3000K. The effect of the heat-bath temperature T on the deviation of T_e from T_{MR} is shown for $T=300$ and 100K [$T_e(0)=1000\text{K}$]. The degree of deviation increases with decreasing T in correspondence to the degree of deviation of $\bar{P}$ from MD. The temperature equilibration time τ_T defined to be the time when T_e approaches T within 10% is larger than that of Mentzoni and Row τ_{MR} to the extent of about 25%: (τ_T/t_0, τ_{MR}/t_0) at $T=300\text{K}$ are (90, 90) for $T_e(0)=500\text{K}$, (180, 170) for 1000K, and (360, 290) for 3000K; (120, 100) for $T_e(0)=1000\text{K}$ and $T=100\text{K}$.

The relaxation of T_e for $P(0)=\delta$ in the cooling process is also presented in Fig. 5 for $T_e(0)=1000\text{K}$ and $T=300\text{K}$. T_e is lower than T_{MR}; $\tau_T/t_0 \simeq 150$ is about 10% less than τ_{MR}/t_0.

B. Heating Process

The time evolution of $\bar{P}$ for $P(0)=MD$ in the heating process is presented in Fig. 6 for $T_e(0)=300\text{K}$ and $T=1000\text{K}$. $\bar{P}$ deviates slightly from MD ($\bar{P}<MD$ at $v/v_0 \lesssim 1$ or $v/v_0 \gtrsim 2$ and $\bar{P}>MD$ at $1 \lesssim v/v_0 \lesssim 2$), to the largest extent at the middle stage of cooling, and approaches MD within the statistical uncertainty at about τ_T/t_0 (see Fig. 8). The degree of deviation for the lower $T_e(0)=100\text{K}$ or the higher $T=3000\text{K}$ is as small as that for $T_e(0)=300\text{K}$ and $T=1000\text{K}$.

The relaxation of $\bar{P}$ for $P(0)=\delta$ in the heating process is presented in Fig. 7 for $T_e(0)=300\text{K}$ and $T=1000\text{K}$. The δ-function distribution approaches MD within the statistical uncertainty; $\tau_{MD}/t_0 \simeq 50$ is much less than τ_T/t_0 (Fig. 8).

The relaxation of T_e for $P(0)=MD$ in the heating process is presented in Fig. 8 in comparison with T_{MR} ($P=MD$). T_e is lawer than T_{MR}. The degree of deviation of T_e from T_{MR} remains nearly the same for the decrease in $T_e(0)$ (300→100K) or the increase in T (1000→3000K). τ_T is lorger than τ_{MR} to the extent of about 25%: (τ_T/t_0, τ_{MR}/t_0) at $T=1000\text{K}$ are (260, 220) for $T_e(0)=300\text{K}$ and (300, 240) for 100K; (800, 690) for $T_e(0)=300\text{K}$ and $T=3000\text{K}$.

The relaxation of T_e for $P(0)=\delta$ in the heating process is also presented in

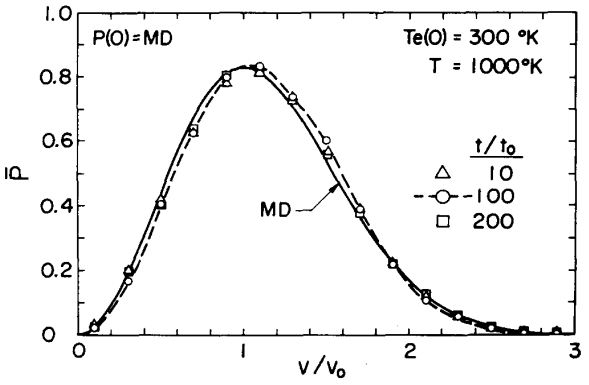

Fig. 6. Time evolution of electron velocity distribution for the initial Maxwell distribution (*MD*) in the heating process.

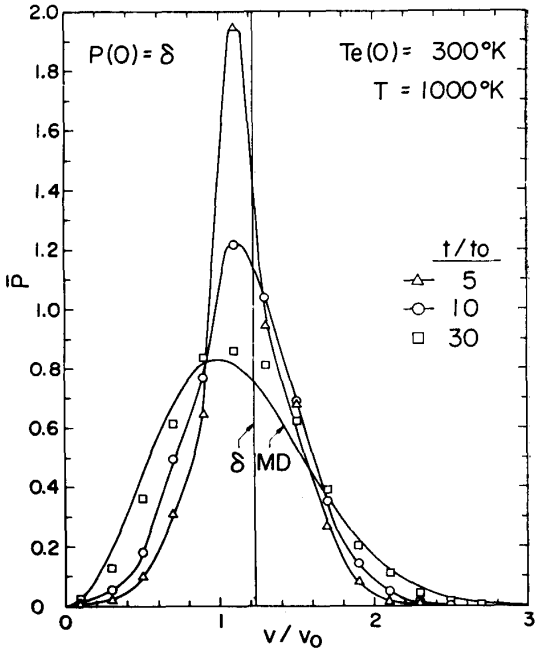

Fig. 7. Relaxation of electron velocity distribution for the initial δ-function distribution in the heating process.

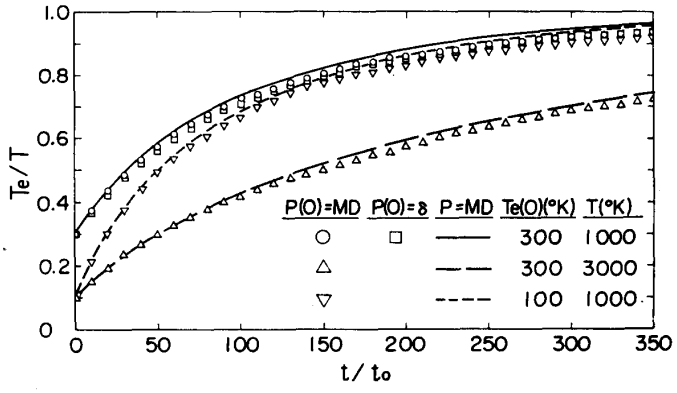

Fig. 8. Relaxation of electron temperature in the heating process in comparison with that of Mentzoni and Row (*P=MD*).

Fig. 8 for $T_e(0)=300\text{K}$ and $T=1000\text{K}$. T_e is lower than that for $P(0)=MD$; $\tau_T/t_0 \simeq 270$ is about 20% larger than τ_{MR}/t_0.

5. Concluding Remarks

The relaxations of the velocity distribution and temperature of electrons cooled or heated by the rotational excitation or de-excitation of nitrogen molecules are studied using the Monte Carlo simulation. The initial Maxwell electron velocity distribution is perturbed considerably in the cooling process and slightly in the heating process. The electron temperature is higher in the cooling process and lower in the

heating process than that of Mentzoni and Row for the local Maxwell distribution. The temperature equilibration time is larger than that of Mentzoni and Row to the extent of about 25%. The initial δ-function electron velocity distribution approaches the Maxwell distribution through the rotational excitation and de-excitation only. The Maxwellization is much faster than the temperature equilibration.

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