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SOURCES OF ELECTRON ENERGY IN WEAKLY IONIZED EXPANSIONS OF NITROGEN

By: John A. Lordi and Michael G. Dunn

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FOREWORD

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1. INTRODUCTION

The sources of electron energy in thermal plasmas comprise a subject of much current interest. The electron temperature plays a fundamental role in the ionization kinetics and in the propagation of electromagnetic energy through the plasma. In particular, the influence of the electron temperature on the electron number density and collision frequency is important in expansions flows of shock-heated air through hypersonic nozzles and around blunt-nosed entry vehicles.

Several experimental[1-7] and theoretical[8-12] studies of nonequilibrium electron temperatures in expansion flows have been reported. Measurements in both nozzle[1-4] and free-jet[5-7] expansions have indicated electron temperatures significantly higher than the heavy-particle translational temperatures. The analyses[8-10] indicate that if the expansion is from an equilibrium reservoir, then the two temperatures will remain equal in the absence of electron heating sources. The purpose of this report is to determine the source terms in the electron energy equation which are responsible for the high electron temperatures observed in shock-tunnel expansions of N_2. In addition, the measured electron temperatures are compared with the corresponding vibrational temperatures. It is suggested in Refs. 1 and 12 that the electrons are closely coupled to the molecular vibrational degree of freedom.

In recent experiments[13], thin-wire Langmuir probes were used to measure the electron temperature and number density in nozzle expansions of weakly ionized nitrogen. An independent measurement of the number density was obtained using microwave interferometry. The purpose of the experiments was to infer the rate coefficient for the dissociative recombination of N_2^+ by fitting calculated and measured nozzle-flow number densities. Since both the electron number density and temperature were measured at several axial locations, these data can be used to make a detailed evaluation of the terms in the electron energy equation.

A general discussion of the electron energy equation is given in Section 2 before presenting the evaluation of the individual terms in Section 3. The electron-temperature data of Ref. 13 are then compared with calculations of the N_2 vibrational temperature in Section 4.

2. ELECTRON ENERGY EQUATION IN A WEAKLY IONIZED PLASMA

In a weakly ionized two-temperature Maxwellian plasma, the equation describing the conservation of electron energy can be decoupled from the global conservation equations. For a one-dimensional flow in the absence of applied fields, the electron energy equation may be written in the form[8-11]

$$u_e n_e \frac{d}{dx}\left(\frac{5}{2} k T_e\right) = u_e \frac{dp_e}{dx} + j^2 \eta + Q_{th} + Q_{in} + Q_c + Q_{rad} \tag{1}$$

where u_e, n_e, T_e, and p_e are the velocity, number density, temperature, and pressure of the electrons, k is the Boltzmann constant, x is the distance along the nozzle axis and $j^2\eta$, Q_{th}, Q_{in}, Q_c, and Q_{rad} are heat source terms due to ohmic heating, thermalizing collisions, inelastic collisions, thermal conduction, and radiation. Also, j and η are the conduction current and resistivity

$$j = -e n_e (u_e - u) \tag{2}$$

$$\eta = \frac{m_e}{e^2 n_e} (\nu_{ei} + \nu_{en}) \tag{3}$$

where ν_{ei} and ν_{en} are the electron-ion and electron-neutral collision frequencies, m_e is the mass of an electron, e is the magnitude of the electron charge, and u is the flow velocity.

Using the state equation, $p_e = n_e k T_e$, Eq. (1) may be rewritten as[8-10]

$$\frac{3}{2} \frac{1}{T_e} \frac{dT_e}{dx} - \frac{1}{n_e} \frac{dn_e}{dx} = \frac{j^2 \eta + Q_{th} + Q_{in} + Q_c + Q_{rad}}{u_e n_e k T_e} . \tag{4}$$

Bray and Pratt[9] point out that in order to explain the observed elevated electron temperatures the source terms on the right hand side of Eq. (4) must be large enough to counterbalance the term $\frac{1}{n_e} \frac{dn_e}{dx}$. In other words, a heat source term must be comparable to, and the same sign as

$$\beta \equiv - u_e n_e k T_e \frac{1}{n_e} \frac{dn_e}{dx} . \tag{5}$$

The thermal conduction term would not appear in the equation for a Maxwellian plasma, but has been added in Ref. 9 in an ad hoc manner. It is shown in Refs. 8-10 that if the electron temperature and heavy-particle temperature are initially equal, in the absence of sources they will remain equal.

The appearance of the pressure gradient term in Eq. (1) marks the most significant difference from analyses preceding Refs. 8-11. The presence of this term has been shown formally in Ref. 14. The conduction current is related to the pressure gradient by conservation of electron momentum. Neglecting inertial forces, the momentum equation for the electrons is[8]

$$e n_e (u_e - u) \eta = -E - \frac{kT_e}{e} \frac{d \ln p_e}{dx} \tag{6}$$

where E is the induced electric field which opposes charge separation.

Several authors[8-11] have discussed the interaction between the pressure gradient and the induced field which must also satisfy Poisson's equation. For the case of a nozzle expansion, the electron pressure gradient is negative and hence the field is directed against the downstream motion of the electrons. For conducting nozzle walls the situation is more complicated because a sheath will form on the walls[9,10]. However, for the data presented in Ref. 13, this complication does not arise because the measurements were made in a Fiberglas nozzle.

3. SOURCES OF ELECTRON ENERGY IN A WEAKLY IONIZED, NITROGEN NOZZLE FLOW

The experimental data for electron number density and temperature, used here to evaluate the sources of electron energy in a nitrogen nozzle flow, were obtained[13] to determine the rate coefficient for the reaction

$$e^- + N_2^+ \longrightarrow 2N$$

The technique used was to vary the rate coefficient in numerical solutions of the nonequilibrium nozzle expansion in order to match calculated and measured electron densities. Since the deionization rate to be determined depends on the electron temperature, the calculated number densities must account for this dependence in order to obtain a valid result for the rate coefficient. In Ref. 13, rather than attempt a solution of the electron energy equation, the measured values of the electron temperature were specified as inputs to the nozzle-flow calculations of the electron number density. The species N_2, N, N_2^+, N^+, and e^- were included in the calculations. The vibrational and electronic degrees of freedom were assumed to maintain thermodynamic equilibrium but the chemical reactions were allowed to proceed at finite rates.

The measurements reported in Ref. 13 were made at two reservoir conditions in a shock tunnel consisting of a pressure-driven shock tube and a conical nozzle which is constructed of Fiberglas. For a detailed description of the facility and the measuring techniques, see Refs. 3, 15, and 16. The electron-temperature data obtained with thin-wire Langmuir probes at a reservoir condition of 7200 °K, 17 atm, are shown in Fig. 1. In addition to the probe data, the calculated heavy-particle temperature is also shown. At this reservoir condition the number density of electrons is 1.88×10^{15} cm^{-3} and the total particle density is 1.74×10^{19} cm^{-3}. The source terms in the electron energy equation are evaluated below for this reservoir condition, using the results of the nozzle-flow solution which gave agreement with the measured number densities.

3.1 Ohmic Heating

The ohmic heating term in Eq. (4) can be estimated without a detailed solution to the electron conservation equations and Poisson's equation[9]. The maximum electron drift velocity is realized when the induced field is zero. Then, setting $E = 0$ in Eq. (6),

$$(u_e - u)_{max} = - \frac{kT_e}{e^2 n_e \eta} \frac{d \ln p_e}{dx} \qquad (7)$$

Letting $(j^2 \eta)_{max}$ denote the ohmic heating term evaluated for $(u_e - u)_{max}$ then from Eqs. (2) and (3)

$$(j^2 \eta)_{max} = m_e n_e (\nu_{ie} + \nu_{en}) (u_e - u)^2_{max} \qquad (8)$$

The quantity $(j^2 \eta)_{max}/\beta$ has been evaluated and is listed in Table 1 for area ratios of 10, 100, and 1000. The Langmuir-probe and microwave-interferometer measuring stations were all between area ratios of 80 and 800. A value of T_e of 4000 °K, which represents the mean of the electron temperature data, was used, and u_e was taken equal to u in evaluating β. In the present case, $\frac{d \ln p_e}{dx} \cong \frac{d \ln n_e}{dx}$. When these approximations are made, then $(u_e - u)_{max}/u$ is identically equal to $(j^2 \eta)_{max}/\beta$.

The values of $(j^2 \eta)_{max}/\beta$ given in the table are much less than one, showing that ohmic heating is not an important source of electron heating for the conditions of these experiments. Notice that at large area ratios this effect is beginning to become important. This behavior would be expected at very low densities, as may be seen from Eq. (7).

In the above calculations, the collision frequencies were obtained from the expressions

$$\nu_{ei} = n_e \bar{C} Q_{ei}$$

where

$$Q_{ei} = \frac{2.51 \times 10^{-6}}{T_e^2} \ln \left[\frac{8.77 \times 10^3 T_e^{1.5}}{\sqrt{n_e}} \right] + \frac{9.74 \times 10^{-7}}{T_e^2} \ (cm^2)$$

with T_e in °K and $\bar{C}$ is the thermal speed of the electrons. Since N_2 is the dominant neutral species,

$$\nu_{en} = n_{N_2} \bar{C} Q_{e-N_2}$$

where

$$Q_{e-N_2} = 5.26 \times 10^{-16} + 2.17 \times 10^{-19} T_e - 1.44 \times 10^{-23} T_e^2$$

$$+ 2.35 \times 10^{-28} T_e^3 + 1.01 \times 10^{-33} T_e^4 \ (cm^2)$$

The electron-ion collision cross-section formula is that given by Spitzer [17] with the addition of the last term as suggested by Lin [18]. The electron-nitrogen molecule cross-section formula is a polynomial fit to thermal averages of the monoenergetic values as given by Shkarofsky et al. [19]

3.2 Thermalizing Collisions

The rate of cooling of the electrons due to elastic collisions with the heavy particles is given by the expression [8]

$$Q_{th} = -3 n_e \frac{m_e}{m_n} (\nu_{ei} + \nu_{en}) k (T_e - T) \tag{9}$$

Using the expressions for the collision frequencies given above, Q_{th} has also been computed and is compared with β in Table 1. As can be seen from the tabulated results, thermalizing collisions play an important role in the subject experiments. The electron energy source responsible for the high value of T_e must more than compensate for this cooling effect.

3.3 Inelastic Collisions

One source of electron heating by inelastic collisions is due to energy exchange with the N_2 vibrational degree of freedom. Schulz[20] has measured the cross section for the excitation of the first excited vibrational level of N_2 by electrons. Recent theoretical studies have shown that this excitation occurs through the formation of a short-lived negative ion complex[21]. Using Schulz's data, Hurle[12] has obtained expressions for the thermally averaged cross-section for the excitation of the first vibrational level of N_2. Hurle presents results for the quantity $n_{t,r} / n_v$ where n_v is the number of collisions required to transfer one quantum of N_2 vibrational energy and $n_{t,r}$ is the number of collisions required to exchange an equal amount of energy with the N_2 translational and rotational degrees of freedom. For an electron temperature of 4000 °K, Hurle gives $n_{t,r} / n_v \cong 8$, and hence the energy exchange with vibration is a much more efficient process than the exchange with N_2 translation and rotation for electron temperatures in the range of interest.

Hurle[12] also points out that the exchange of electron energy with the N_2 rotational mode is faster than the exchange with translation. The average energy lost to rotation per collision is given by $10\left(\frac{2m_e}{m_{N_2}}\right)\left(\frac{3}{2} k T_e\right)$ as opposed to $\frac{2m_e}{m_{N_2}}\left(\frac{3}{2} k T_e\right)$ lost to translation. Hence, the rate of cooling of the electrons due to exchange with rotation, Q_{ROT}, is given by

$$\frac{Q_{ROT}}{\beta} \cong 10 \frac{Q_{th}}{\beta} \tag{10}$$

the latter quantity being given in Table 1. Since the ratio of energy exchange with vibration to that with translation plus rotation is inversely proportional to $n_{t,r}/n_v$, then $\frac{Q_{vib}}{\beta} >> 1$ and the energy exchange with N_2 vibration is a dominant source of electron energy for these nitrogen experiments.

Another source of electron energy from inelastic collisions is provided by the mechanism of recombination heating. As discussed below, although the dissociative recombination of N_2^+ was the rate controlling deionization path in the experiments of Ref. 13, N^+ was by far the dominant ion at the measuring stations. In the calculations performed to demonstrate this point, the three-body electron-electron-ion recombination of N^+ was found to have a negligible effect on the electron number density. The effect of the small amount of recombination by this mechanism may provide a source of electron heating however. Appleton and Bray[8] give this source of electron heating as

$$Q_{in} = (\frac{5}{2} k T_e + I) \alpha_c n_e^2 \tag{11}$$

where I is the ionization energy of N^+ and α_c is the collisional recombination-rate coefficient. The results for α_c given by Bates, Kingston and McWhirter[22] have been used to evaluate Q_{in}. The resulting values, which are shown in Table 1, are much smaller than β at all of the measuring stations.

Energy exchange between electrons and the metastable state, $N_2(A^3\Sigma_u^+)$, would not be included in Eq. (11) because the collisional-radiative recombination theory of Ref. 22 is for hydrogen-like ions. However, an upper bound for the energy exchange can be given by comparing the depletion rate for $N_2(A^3\Sigma_u^+)$ with that for N_2^+. It was established in Ref. 13 that although the dissociative recombination of N_2^+ was the rate-controlling deionization path in those experiments, N^+ was by far the dominant ion at the measuring stations. This behavior is a result of the charge exchange reaction,

$$N_2^+ + N \rightleftharpoons N_2 + N^+$$

In the initial portion of the expansion N^+ is consumed by this reaction but the rate of removal of N^+ is determined by the rate of recombination of N_2^+.

The population of the $A^3\Sigma_u^+$ state can be estimated for the experimental conditions of Ref. 13. Young and Gilbert[23] have shown that N atoms are fairly efficient in deexciting the $A^3\Sigma_u^+$ state of N_2. They report a rate coefficient for this process of $k = 5 \times 10^{-11}$ cm^3/sec. For the reservoir conditions of 7200 °K and 17 atm, the population of the $(A^3\Sigma_u^+)$ state is twice the N_2^+ concentration in the reservoir. Using the above value of k and the dissociative-recombination rate coefficient reported in Ref. 13, the rate of deexcitation of the $A^3\Sigma_u^+$ state can be shown to be about 6 times greater than the rate of recombination of N_2^+ by dissociative recombination. Since N_2^+ is also produced by charge exchange between N_2 and N^+ in the initial part of the expansion, the population of the $(A^3\Sigma_u^+)$ state of N_2 will be significantly lower than the N_2^+ concentration for $A/A^* \geq 10$.

Assuming that the population of the A -state is equal to the concentration of N_2^+ and that every collision between an electron and an N_2 molecule in the A -state transfers the full energy of the state to the electron, this heating source is estimated to be less than the heat lost in thermalizing collisions. Hence, this source of electron energy can be neglected safely.

3.4 Thermal Conduction

Since the thermal speed of the electrons is much higher than that of the neutrals, their thermal conductivity is also much higher. The thermal conduction term in the energy equation is given by

$$Q_c = \frac{d}{dx}\left(k_e \frac{dT_e}{dx}\right) \tag{12}$$

Using Spitzer's value[17] of the thermal conductivity for the electrons, this term is found to be negligible at the measuring stations, mainly because of the small electron-temperature gradient. As may be seen from Fig. 1, the heavy-particle temperature is changing most rapidly at about $x = 5$ cm., which corresponds to $A/A^* \cong 2$. Assuming $T_e = T$ at this point, the value of Q_c was found to be much less than β.

3.5 Radiation

For an optically thin plasma, Q_{RAD} represents a heat loss term whereby energy is radiated away from the hot gas. Bray and Pratt[9] have postulated a mechanism of heating electrons at high area ratios by the absorption of radiation from the high-temperature portion of the expansion. While they have argued that this "trapping" of radiation could be important in flows seeded with alkali metals, it does not appear that "radiation trapping" by electrons would be important in the pure nitrogen flows treated here. The electrons absorb radiation by the inverse Bremsstrahlung mechanism and the electron number densities are too low for any significant amount of radiation to be absorbed.

Spitzer[17] gives the following absorption coefficient for inverse Bremsstrahlung at frequency ν

$$K_\nu = 3.69 \times 10^8 \frac{Z^3 n_e^2}{T^{1/2} \nu^3} \ cm^{-1} \tag{13}$$

where Z is the number of electronic charges on the ions present and n_e is the electron number density. Defining an absorption length, l_ν, as the reciprocal of K_ν, l_ν is greater than 10^{10} cm for radiation at $1\,\mu$ wavelength and the conditions at $A/A^* = 10$. In these nitrogen experiments, radiation in this wavelength region comes from the first-positive system of N_2. Other sources of radiation from the hot part of the expansion, such as recombination radiation, are at higher frequencies which lead to even larger values of l_ν.

4. COMPARISON OF ELECTRON TEMPERATURE AND VIBRATIONAL TEMPERATURE

In the previous section the individual terms in the electron energy equation were evaluated for a nozzle expansion of weakly ionized nitrogen from reservoir conditions of 7200 °K and 17 atm. Energy exchange with the translational, rotational, and vibrational degrees of freedom of N_2 were found to be the most significant source terms. The electron temperature data shown in Fig. 1 decrease gradually with distance along the nozzle, which is consistent with this finding. In high-temperature nozzle expansions, the vibrational temperature is known to freeze at a high value while the translational and rotational temperatures decrease together throughout the expansion. Hence, the measured electron temperatures would be expected to lie between the vibrational temperature and the monotonically decreasing heavy-particle translational temperature. Since $n_{t,r}/n_v \cong 8$ for T_e = 4000 °K, the electron temperature should follow the vibrational temperature more closely than it does the translational temperature. An approximate calculation of the vibrational temperature has been made in order to compare with the measured electron temperatures.

In the nozzle-flow calculations performed in Ref. 13 to infer the dissociative-recombination rate of N_2^+, vibrational equilibrium was assumed. Since vibrational nonequilibrium effects on the gasdynamic properties are small, these computed results for the variation of the translational temperature and flow velocity along the nozzle were used to obtain an approximate soltuion to the relaxation equation for the vibrational energy, E_v ,

$$\frac{dE_v}{dx} = \frac{\bar{E}_v - E_v}{u\tau_v} \quad . \tag{14}$$

Using the calculated results for u and T , the variation of $u\,\tau_v(T)$ along the nozzle was computed and Eq. (14) was integrated using the third-order, Runge-Kutta method. While this computation does include the effect of the recombination on the translational temperature, it does not account for the variation in the total number of vibrators. However, this latter effect should not appreciably alter the population of the lower-most vibrational levels.

These lower vibrational levels are the dominant sources of electron heating by exchange with vibration. For a Boltzmann distribution at a vibrational temperature of 4000 °K, more than 95% of the vibrators are in the first four levels. It is also important to note that the number of electrons is too low to influence the vibrational relaxation.

The results of three calculations for the vibrational temperature, T_v, are shown in Fig. 1 together with the measured electron temperatures. The results for T_v correspond to using the value of $(p\tau_v)$ obtained from shock-wave measurements[24] and 0.1 and 0.01 of that value. These values were used because of the faster vibrational relaxation times which have been observed[25-27] in nozzle expansions (.01 $(p\tau_v)_{sw}$ to .02 $(p\tau_v)_{sw}$). The measured values of T_e fall below the value of T_v which corresponds to the shock-tube-measured $(p\tau_v)$ but are all above that obtained using .01 $(p\tau_v)_{sw}$. Furthermore, the data at the first two measuring stations lie above the value of T_v calculated using 0.1 $(p\tau_v)_{sw}$. The estimates made here for the source terms in the electron energy equation indicate that the electron temperature must be lower than the vibrational temperature. While a precise value of τ_v cannot be obtained from the electron-temperature data presented in Ref. 13, these data appear to preclude as small a value as measured in nozzle expansions by other techniques. This comparison should not be used to infer the vibrational relaxation time because the energy exchange between electrons and upper vibrational levels has not been included.

Similar experiments to those reported in Ref. 13 for N_2^+ have been reported[28] for O_2^+. Again the electron temperature data were above the heavy-particle translational temperatures and, at the measuring stations, decreased with distance along the nozzle. In these oxygen experiments, for which the reservoir conditions were 4950 °K and 25 atm, the electron temperatures, normalized by the reservoir temperature, were closer to the heavy particle temperature than the corresponding results in nitrogen. The electron temperature decreased from about 1700 °K at $A/A^* = 80$, to about 800 °K at $A/A^* = 530$ while the corresponding values of the translational temperature were 1300 °K and 500 °K, respectively. This behavior is consistent with the electron temperature being controlled by thermal energy transfer with the translational, rotational, and vibrational degrees of freedom of O_2 because the vibrational relaxation times for O_2 are much faster than those for N_2. On the basis of the present study, the electron temperature in weakly ionized, high-temperature air plasmas appears to be governed by energy transfer with the rotational and vibrational modes of the constituent molecules.

5. SUMMARY

The individual terms in the electron energy equation have been evaluated for a shock-tunnel expansion of nitrogen from reservoir conditions of 7200 °K and 17 atm. The measured values of the nozzle-flow electron temperature and number density were used in these calculations. Energy exchange with the N_2 vibrational degree of freedom has been shown to be by far the dominant source of electron energy for this case. The electron temperature is controlled by the competing effects of heating by vibration and cooling by exchange with the translational and rotational degrees of freedom of N_2.

Since the energy exchange with vibration is more efficient, the electron temperature data were compared with calculated vibrational temperatures. While this comparison indicates that vibrational relaxation in the nozzle expansion may be somewhat faster than behind a shock wave, it is concluded that the vibrational relaxation time is greater than 0.1 of the shock-wave value for the conditions studied here. This finding disagrees with measurements of vibrational relaxation of N_2 in nozzle flows using other techniques[25-27].

Although a detailed evaluation of the terms in the electron energy equation for an oxygen expansion has not been done, electron temperature data obtained in a weakly ionized oxygen nozzle flow appear to support the conclusions reached here for nitrogen. The implications of these results to weakly ionized air plasmas is that, as suggested by previous authors[1,12], the electron temperature is closely coupled to the vibrational degrees of freedom of the molecular species.

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Table 1

SOURCES OF ELECTRON ENERGY IN NOZZLE EXPANSION OF NITROGEN

$\frac{A}{A^*}$	$n_e(\frac{\#}{cc})$	$\beta\,(\frac{ergs}{cc\text{-}sec})$	$\frac{(j^2\eta)_{max}}{\beta}$	$-\frac{Q_{th}}{\beta}$	$\frac{Q_{in}}{\beta}$†
10	9.13×10^{11}	4.40×10^{4}	1.67×10^{-2}	3.59	1.92×10^{-3}
100	7.64×10^{10}	1.06×10^{3}	4.17×10^{-2}	2.96	2.85×10^{-4}
1000	7.18×10^{9}	3.21×10	1.28×10^{-1}	1.12	5.25×10^{-5}

† DENOTES ONLY RECOMBINATION HEATING AND DOES NOT INCLUDE ENERGY EXCHANGE WITH VIBRATION.

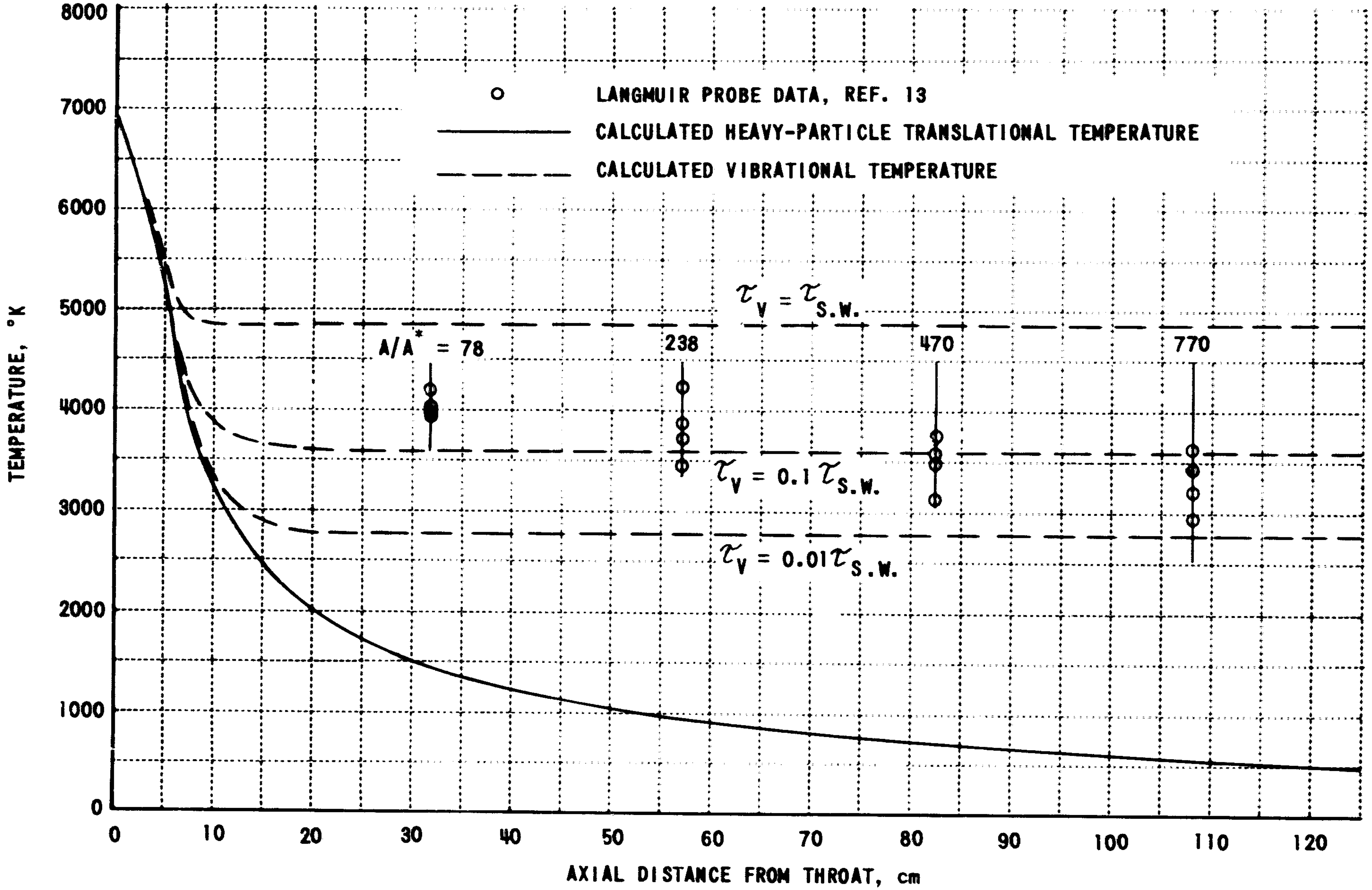

Figure 1 ELECTRON TEMPERATURE DATA FOR NITROGEN NOZZLE FLOW

$T_o = 7200°K \qquad p_o = 17.1$ atm