Analytical calculation of the gas temperature and measurement of the electron temperature for gas discharges in Ne and He mixtures with copper, bromine, hydrogen and strontium

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Abstract. Thermal conductivities of binary gas systems are calculated on the basis of 12-6 Lennard-Jones and rigid sphere inter-atomic interaction approximations for the case of gas discharges in He and Ne with small admixtures of copper, bromine, hydrogen and strontium. Assuming that the gas temperature varies only in the radial direction and using the calculated thermal conductivities, analytical solutions of the steady-state heat conduction equation are found for two cases of uniform and non-uniform power input, respectively. For both cases the average gas temperature is found by averaging the radial gas temperature distribution over the radius. Measurement of the relative intensities of some He and Ne spectral lines originating from different upper levels has enabled us to determine the average electron temperature.

1. Introduction

One of the main problems in plasma physics is the determination of characteristic constants for basic processes in plasma, such as asymmetric charge transfer, Penning ionization, electron-heavy particle collisions, diffusion and heat conduction, which are fundamentally important for gaseous discharges, laser physics, plasma technologies, gas-discharge mass spectroscopy, absorption and emission spectroscopy and plasma in general. It is well known that characteristic constants for the heavy particle interaction depend on the gas temperature. The experimental technique for gas temperature determination using measurement of Doppler broadening of spectral lines is definitely imprecise. Electron impact excitation, de-excitation and ionization, as well as three-body recombination, depend on the electron temperature. In particular, in laser physics these processes directly influence the creation of population inversion, or indirectly through excitation transfer by the above-mentioned heavy particle collisions. Measurements of the electron temperature by a Langmuir probe are not suitable for pressures of helium or neon above 10 Torr, particularly in high-voltage and high-current nanosecond pulsed longitudinal discharges. In the literature there are several models of varying degrees of complexity, which predict, among other parameters, the electron temperature values with considerable variation without any overlap.

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2. Theoretical and experimental results

In order to obtain the gas temperature distribution, the following steady-state heat conduction equation is solved:

\[ \text{div}(k \ \text{grad}T_g) + q_v = 0, \]  

where \( k \) is the thermal conductivity, \( T_g \) is the gas temperature and \( q_v \) is the power deposited into the discharge zone per unit volume. Considering cylindrical geometry and assuming that the gas temperature varies only in the radial direction, equation (1) assumes the following form:

\[ \frac{1}{r} \frac{d}{dr} \left( rk \frac{dT_g}{dr} \right) + q_v = 0. \]  

The dependence of the thermal conductivity \( k \) of gases and gas mixtures on the gas temperature has the form \( k = B \cdot T_g^a \), where \( B \) and \( a \) are constants (within a certain temperature range) specific for each gas or gas mixture. The constants \( B \) and \( a \) could be obtained through fitting the existing experimental data taken from the Handbook-tables of Physical Constants. Unfortunately, except for rarified gases, data for the thermal conductivities of the chemical elements of our interest are either very scarce in the literature or are within a narrow temperature range. This is why it is necessary to calculate the thermal conductivities to obtain the gas temperature distribution via solving the steady-state heat conduction equation. There are two widely used theoretical approaches for thermal conductivity determination – rigid, i.e. hard, sphere (R Sph) and 12-6 Lennard–Jones (L–J) approximations, which consider different interactions between the particles. Following [1] the thermal conductivities \( k_1 \) and \( k_2 \) for R Sph and L-J approximations are expressed as follows:

\[ k_1 = 0.083264 \frac{T_g^{\frac{1}{2}}}{\mu^2 d^2}, \quad k_2 = 0.083264 \frac{T_g^{\frac{1}{2}}}{\mu^2 \sigma^2 \Omega \left( \frac{k_b T_g}{e_0} \right)}, \]  

where \( \mu \) is the particle mass in amu, \( T_g \) is the gas temperature in K, \( d \) is the R Sph diameter (sum of atomic radii of the interacting particles) in Å, \( \sigma \) is the inter-atomic distance in Å at which the potential energy is zero, \( e_0 \) is the potential well depth, \( \Omega \) is the collision integral, which depends on the interaction potential. \( \Omega \) for the R Sph model is unity, while for the L–J potential the following expression is used: \( \frac{1}{\Omega} = 0.697 \left[ 1 + 0.323 \ln \left( \frac{k_b T_g}{e_0} \right) \right] \). Thermal conductivities of binary gas systems are calculated on the basis of the empirical method of Brokaw [1] for the case of gas discharges in He (10 Torr) and Ne (16.7 Torr) with small admixtures of hydrogen (0.03 Torr), copper (0.3 Torr), bromine (0.3 Torr and 1.4 Torr) and strontium (0.7 Torr). The binary gas mixtures considered in this paper are optimal for laser operation in the deep ultraviolet - Cu – Ne-H\(_2\)-CuBr, and in the middle infrared - He-SrBr\(_2\) lasers. The results for the thermal conductivities are presented in table 1.

For the case of uniform power input, i.e. \( q_V = \text{constant} \), in a gas discharge with radius \( R \) and taking into account the following boundary conditions \( T_g|_{r=R} = T_w \) experimentally measured and \( \frac{dT_g}{dr}|_{r=0} = 0 \), the solution of equation (2) has the form:

\[ T_g(r) = \left( T_w^{\frac{1}{1+a}} + \frac{1+a}{4B} q_v (R^2 - r^2) \right)^{\frac{1}{1+a}}. \]  

For calculation of characteristic constants it is convenient to use the average gas temperature in the discharge zone, which is found by averaging (4) over the radius \(T_0 = T_g(r=0)\):

\[
\langle T_g \rangle = \frac{4B}{(2 + a)qR^2} \left( q_0^{2+a} - T_w^{2+a} \right).
\]

(5)

**Table 1.** \(B\) and \(a\) – constants, which are for \(k\) in W.m\(^{-1}\).K\(^{-1}\), determining thermal conductivity, RSph – Rigid Sphere approximation, L–J – Lennard–Jones approximation.

| gas or gas mixture | \(B\) (exp. fit) | \(a\) (exp. fit) | \(B\) (RSph) | \(a\) (RSph) | \(B\) (L–J) | \(a\) (L–J) |
|-------------------|-----------------|-----------------|--------------|--------------|-------------|-------------|
| Ne                | 9.7x10\(^{-4}\) | 0.685           | 38.3x10\(^{-4}\) | 0.500        | 12.3x10\(^{-4}\) | 0.648       |
| He                | 34.9x10\(^{-4}\) | 0.670           | 90.9x10\(^{-4}\) | 0.500        | 46.2x10\(^{-4}\) | 0.626       |
| H [800K, 1600K]   | 4.4x10\(^{-4}\)  | 1               |              |              |              |             |
| H [1600K, 2600K]  | 4.4x10\(^{-25}\) | 7.56            | 130x10\(^{-4}\) | 0.500        | 31x10\(^{-4}\) | 0.649       |
| H [80K, 2000K]    | 11.3x10\(^{-4}\) | 0.873           |              |              |              |             |
| Cu                | -                | -               | 58.6x10\(^{-4}\) | 0.500        | 0.9x10\(^{-4}\) | 0.982       |
| Br                | -                | -               | 3.3x10\(^{-4}\)  | 0.500        | 0.6x10\(^{-4}\) | 0.740       |
| Ne-H [800K, 1600K]| 9.7x10\(^{-4}\)  | 0.686           | -             | -            | -            | -           |
| Ne-H [1600K, 2600K]| 0.7x10\(^{-4}\) | 1.042           | -             | -            | -            | -           |
| Ne-Cu             | -                | -               | 10.1x10\(^{-4}\) | 0.680        | 9.3x10\(^{-4}\) | 0.690       |
| Ne-Br             | -                | -               | 9.7x10\(^{-4}\)  | 0.679        | 9.1x10\(^{-4}\) | 0.678       |
| He-Cu             | -                | -               | 35.3x10\(^{-4}\) | 0.666        | 31.4x10\(^{-4}\) | 0.679       |
| He-Br             | -                | -               | 32.7x10\(^{-4}\) | 0.658        | 29.0x10\(^{-4}\) | 0.675       |
| He-Sr             | -                | -               | 31.9x10\(^{-4}\) | 0.660        | 26.8x10\(^{-4}\) | 0.680       |
| He-Br\(_2\)       | -                | -               | 32.6x10\(^{-4}\) | 0.658        | 28.9x10\(^{-4}\) | 0.675       |

Following [2], from \(q_V = j.E\) and \(E(r) = E_0 \cdot J_0((2.4/R)r)\), where \(J_0((2.4/R)r)\) is the Bessel function of the first kind of zero order, one can obtain \(q_V = Q_0 \cdot [J_0((2.4/R)r)]^2\), where the constant \(Q_0\) remains to be obtained. In order to obtain an analytical solution, a polynomial fit of the third degree is used instead of \([J_0(x)]^2\), i.e. \([J_0(x)]^2 = b + c.x + d.x^2 + e.x^3\), where \(x = (2.4.r)/R\). Through fitting, the following values of \(b, c, d\) and \(e\) are obtained: \(b = 1.005, c = -0.016, d = -0.5702\) and \(e = 0.1687\).

In this way, the following expression for \(q_V\) is obtained:

\[
q_V(r) = Q_0 \left[ b + c \frac{2.4}{R} r + d \left( \frac{2.4}{R} \right)^2 r^2 + e \left( \frac{2.4}{R} \right)^3 r^3 \right].
\]

(6)

Comparing the areas bounded between the functions \(q_0 = constant\) and \(q_V = q_V(r)\) and the variable axis \(r\), the following expression for \(Q_0\) is obtained:

\[
Q_0 = \frac{\int_0^r q_0 \cdot \left( \frac{b}{2} + \frac{c}{3} \frac{2.4}{R} + \frac{d}{4} \frac{2.4^2}{R^2} + \frac{e}{5} \frac{2.4^3}{R^3} \right) r^2 \, dr}{\int_0^r q_0 \, dr}.
\]

(7)

For this case of non-uniform power deposition and taking into account the above-mentioned boundary conditions, the solution of (2) has the following form:

\[
T_w = \left( T_w^{a+1} + \frac{(a + 1)Q_0}{B} \left[ \frac{n}{4} (R^2 - r^2) + \frac{2.4c}{9R} (R^3 - r^3) + \frac{2.4^2d}{16R} (R^4 - r^4) + \frac{2.4^3e}{25R^2} (R^5 - r^5) \right] \right)^{1/(a+1)}.
\]

(8)

In this case the average gas temperature is found by comparing the areas bounded between the gas temperature profiles for \(q_0 = constant\) and \(q_V = q_V(r)\) and the variable axis \(r\). The gas temperature
distributions (4) and (8) for uniform and non-uniform power input, respectively, could be easily
determined, because they both have analytical solutions. Equations (4) and (8) are obviously different,
but the attention must be drawn to the result that the average gas temperatures in both definitely
different cases are almost equal. The discrepancy varies from 0.2 % to 1 % - the averaged value is
about 0.6 %.

In the case of local thermodynamic equilibrium (LTE), a measurement of the relative intensity ratio
of two lines originating from different upper levels \(i\) and \(j\), usually, but not necessarily, terminating at
the same lower levels, would be sufficient for the determination of the electron temperature \(T_e\). The
following expression is used [3]:

\[
T_e = \frac{(E_i - E_j)}{k_b} \ln \left( \frac{I_i \lambda_i g_i A_i}{I_j \lambda_j g_j A_j} \right),
\]

where for the measured transitions from upper levels \(i\) and \(j\), \(\lambda\) is the wavelength, \(I\) is the measured
intensity, \(A\) is the transition probability, i.e. spontaneous rate coefficient, \(g\) is the statistical weight, i.e.
degeneracy of the corresponding upper level and \(E\) is the energy of the respective upper level.

In table 2 the data required for the experimental determination of \(T_e\), as well as average \(T_e\) obtained
experimentally and from averaging the time dependence of \(T_e\) presented in various kinetic models, are
shown for some Ne and He transitions. It should be noted that average \(T_e\) means non-time-resolved \(T_e\),
i.e. averaged over the time. The experimental error decreases as the energy gap \(E_i - E_j\) increases. This
is why the combinations are made between wavelengths given in bold and all the others. As one can
easily observe the experimentally determined \(T_e\) is in fair agreement with the calculated values. The
discrepancies are 0.01 eV and 0.02 eV, respectively, for gas discharges in Ne and He, which are in the
range in the experimental error of about 0.1 eV. The results obtained satisfy the condition for LTE
\(n_e > 1.6 \times 10^{18} (\Delta E)^{3/2} T_e^{-1/2}\), where \(n_e\) is the electron density in \(m^{-3}\), while the units of \(T_e\) and \(\Delta E\) are
eV. Using the energy gaps of 2.401 eV and 1.325 eV, respectively for Ne and He, as well as the
average \(T_e\) determined experimentally, one can easily obtain that \(n_e > 1.534 \times 10^{20} m^{-3}\) for Ne and
\(n_e > 2.859 \times 10^{19} m^{-3}\) for He, which is confirmed by the electron density of about \(3.5 \times 10^{20} m^{-3}\) for Ne
and \(7.9 \times 10^{20} m^{-3}\) for He calculated through the above-mentioned kinetic models.

**Table 2.** \(\lambda\) – wavelength at which emission intensity is measured, \(A_i\) – radiative transition
probability, \(E_i\) – energy of transition upper level, \(g_i\) – statistical weight, \(I_i\) – relative intensity,
\(T_e^{\exp}\) – electron temperature experimentally determined, \(T_e^{\text{th}}\) – electron temperature from
averaging the time dependences of \(T_e\) presented in various kinetic models.

| atom | \(\lambda\) (nm) | \(A_i\) (s) | \(E_i\) (eV) | \(g_i\) | \(I_i\) (arb. units) | \(T_e^{\exp}\) (eV) | \(T_e^{\text{th}}\) (eV) |
|------|----------------|-----------|-----------|-------|---------------------|----------------|----------------|
| Ne   | 352.047        | 9.3x10^6  | 20.369    | 1     | 4                   | 0.49 / 0.63   |                |
| Ne   | 470.886        | 4.2x10^6  | 21.014    | 3     | 5                   | 0.54          |                |
| Ne   | 534.109        | 4.2x10^6  | 21.014    | 3     | 6.3                 | 0.48 / 0.52   | 0.485          |
| Ne   | 540.056        | 9.0x10^5  | 18.966    | 1     | 6.4                 | -             |                |
| Ne   | 702.405        | 1.89x10^6 | 18.613    | 3     | 77.8                | -             |                |
| Ne   | 748.887        | 3.49x10^7 | 20.037    | 5     | 18.8                | 0.32          |                |
| Ne   | 753.577        | 4.3x10^7  | 20.026    | 3     | 24.4                | 0.36          |                |
| He   | 402.619        | 2.93x10^6 | 24.043    | 5     | 4                   | -             |                |
| He   | 587.562        | 2.94x10^7 | 23.074    | 3     | 30.1                | 0.53 / 0.66   | 0.610          |
| He   | 587.597        | 3.92x10^7 | 23.074    | 3     | 30.8                | 0.57 / 0.63   |                |
| He   | 706.519        | 1.54x10^7 | 22.718    | 3     | 47.9                | 0.53 / 0.55   |                |
| He   | 728.135        | 1.81x10^7 | 22.920    | 1     | 9.2                 | 0.61 / 0.65   |                |
3. Conclusions
Using the thermal conductivities of binary gas systems calculated on the basis of 12-6 Lennard-Jones and rigid sphere inter-atomic interaction, analytical solution of the steady-state heat conduction equation and the average gas temperature are found for uniform and non-uniform power input in gas discharges in He and Ne with small admixtures of copper, bromine, hydrogen and strontium. The average electron temperature is also determined through measurements of the relative intensities of some He and Ne spectral lines.

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