Electron Heating in the Measurement of Electron Temperature by Thomson Scattering: Are Thermal Plasmas Thermal?

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Thomson scattering measurements have yielded electron temperatures T_e up to 7000 K greater than the ion temperature in 1 bar thermal plasmas. To account for laser heating of electrons, T_e was measured as a function of laser pulse energy, and an unperturbed T_e obtained by linear extrapolation to zero pulse energy. It is shown that the absorption of laser energy by the electrons, and the cooling of the electrons by collisions and radiative emission, depend strongly on T_e. Considering all these processes gives T_e values that are in much closer agreement with the ion temperature.

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It has long been debated whether atmospheric-pressure thermal plasmas, such as welding arcs and plasma jets, are in local thermodynamic equilibrium (LTE). LTE requires that the translational energy distributions of all species are Maxwellian, that the excitation energies of bound electrons follow a Boltzmann distribution, and that the temperatures defined by these distributions are the same for all species.

It has been demonstrated that deviations from LTE occur, for example, within 1 mm of the cathode of free-burning arcs due to the rapid convective influx of cold gas caused by the pinch effect [1], and in the fringes of the plasma due to resonance absorption of radiation [2] and the steep gradients [3]. However, the bulk of the theoretical [4,5] and experimental [6,7] evidence is that the regions away from the electrodes and fringes are in LTE for electron densities above about 10^{23} m^{-3}, owing to the rapid equilibration due to the high collision rates.

Over the past eight years, a number of authors have published the results of Thomson scattering measurements of electron and ion temperatures in thermal plasmas. The temperatures have been derived from the spectral profile of the scattered light [8]. The measurements indicate that all regions of the plasma are far from LTE, in particular, that the electron temperature is some thousands of kelvin higher than the ion temperature. Snyder, Lassahn, and Reynolds [9] measured an electron temperature of 20 900 $\pm$ 1700 K and an ion temperature of 14 200 $\pm$ 700 K at a position 2 mm below the cathode of a 100 A free-burning arc in argon. Bentley [10] repeated the electron temperature measurements, obtaining 20 400 $\pm$ 500 K. Spectroscopic measurements of a similar arc yielded an excitation temperature of 16 600 K, in agreement with the ion temperature given by a laser-scattering technique in which the scattered signal was integrated over a range of wavelengths [6]. Tanaka and Ushio [11] compared Thomson scattering measurements of electron and ion temperatures and spectroscopic measurements of excitation temperature of 50 and 150 A arcs in argon. They found that the electron temperature was about 5000 K higher than the ion and excitation temperatures. Measurements performed in an atmospheric-pressure plasma jet also gave electron temperatures far in excess of the ion temperatures and temperatures measured spectroscopically. For example, 2 mm downstream of the aperture for a 900 A argon jet, the electron temperature was measured to be 22 000 K, the ion temperature to be 13 000 K, and the spectroscopically measured excitation temperature to be 14 000 $\pm$ 1000 K [12].

Gregori *et al.* [13] and Snyder, Crawford, and Fincke [14] have investigated the dependence of the measured electron temperature on the scattering angle and the laser wavelength. Gregori *et al.* [15] recently suggested that the standard method of deriving the electron temperature from the spectrum of the scattered signal [8] should be replaced by a memory function method. This has wider applicability but requires an additional free parameter to be fitted to the measured frequency spectrum. They obtained an electron temperature of 15 700 $\pm$ 500 K in a plasma jet, which was within 1500 K of the excitation temperature.

The measurement of electron temperature from the spectral profile of the Thomson scattered signal requires the use of high-powered pulsed lasers; generally a frequency-doubled Nd-YAG laser (wavelength = 532 nm) is used. The interaction of the laser beam with the plasma rapidly heats the electrons by linear inverse bremsstrahlung [16]. To take this effect into account, workers have measured electron temperature as a function of laser pulse energy, fitted a straight line to the results, and extrapolated the line to zero pulse energy to obtain the electron temperature free of influence from laser heating. The use of a straight line fit has been justified [9,11] by reference to Hughes [16]. Hughes, however, gives the following expression for absorption of laser light by a thermal plasma:

$$\alpha = \frac{n_e n_i Z^2 e^6 [1 - \exp(-\hbar\omega/k_B T_e)]}{6\pi\epsilon_0^3 c\hbar\omega^3 m_e^2} \times \left(\frac{m_e}{2\pi k_B T_e}\right)^{1/2} \frac{\pi}{3}\,\overline{g}, \qquad (1)$$

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where α is the absorption coefficient, n_e and n_i are, respectively, the electron and ion number densities, T_e is the electron temperature, Z is the average ionization level of the plasma, ω is the laser frequency, and the constants e, m_e, $\hbar$, k_B, and c are, respectively, the electron charge and mass, Planck's constant, Boltzmann's constant, and the speed of light. Equation (1) shows that absorption of the laser light depends on T_e, n_e, and n_i, both directly, and through the average Gaunt factor $\overline{g}$, which is the sum of the free-free Gaunt factor [17] and the free-bound Gaunt factor [18], both of which depend on T_e. The absorption will therefore vary with time during a laser pulse, and the absorbed energy will have a nonlinear dependence on the laser pulse energy.

The electrons heated by the laser pulse are cooled by four processes: electron thermal conduction, energy transfer to heavy particles by elastic collisions and by electron-impact ionization, and radiative emission. Each depends strongly on the electron temperature. Hence, both the electron cooling and heating processes have a nonlinear dependence on the laser pulse energy.

We can write a one-dimensional equation in polar geometry to describe the effects of heating of the electrons due to absorption of laser radiation, and cooling due to each of the four processes [1]:

$$\frac{\partial}{\partial t}\left(\frac{5}{2}k_B n_e T_e\right) = \frac{1}{r}\frac{\partial}{\partial r}\left(r k_e \frac{\partial T_e}{\partial r}\right) + \frac{\alpha E_p}{A\tau_p} - W_{eh} - \sum_{i=1}^{2} R_i E_i - U\,, \tag{2}$$

where E_p and τ_p are, respectively, the laser pulse energy and duration, A is the cross-sectional area of the laser beam, r is the radial coordinate ($r = 0$ corresponds to the center of the laser beam cross section) and t is time, k_e and U are, respectively, the electron thermal conductivity and the radiative emission coefficient, and E_i is the ith ionization energy of argon. The rate of transfer of energy from electrons to heavy particles through inelastic collisions, W_{eh}, is calculated using the expression of Lelevkin *et al.* [1,19], extended to take into account doubly ionized atoms. The rates of electron-impact ionization of neutral and singly ionized argon atoms, R_1 and R_2, respectively, are given by

$$R_i = k_i\left[n_e n_{i-1} - \left(\frac{n_{i-1}}{n_e n_i}\right)_{\text{eq}} n_e^2 n_i\right], \tag{3}$$

where n_0, n_1, and n_2 are, respectively, the number densities of the neutral, singly ionized, and doubly ionized atoms. The data of Almeida *et al.* [20] are used for k_1 and k_2, respectively the rate constants for first and second ionization of argon atoms. The subscript *eq* denotes the values calculated for a plasma in LTE.

I have solved Eq. (2) numerically, using the method described by Patankar [21]. It is assumed that the laser beam profile is Gaussian, the pulse shape is square with time, and that initially the heavy particle and electron temperatures are equal. I further assume that the heavy particle temperature is constant during the pulse. This is consistent with Thomson scattering measurements of ion temperature, which show that the ion temperature is independent of laser pulse energy [9]. The values of n_0, n_e, n_1, and n_2 are calculated as a function of time using the ionization rates given by Eq. (3), with the initial values calculated assuming LTE.

The laser beam diameter (full width at half maximum) and pulse duration are chosen to be 200 μm and 7 ns, respectively, in accordance with the parameters used in the experiments [9,10]. The calculation region extends over a radius of 350 μm, with an evenly spaced 1 μm grid. The time step is 0.1 ns. Doubling the time and grid resolution and the size of the calculation region results in a less than 0.1% change in the calculated electron temperatures.

Figure 1 shows typical results for the evolution of the electron temperature and the species number densities during the laser pulse. It is clear that the electron temperature increases more rapidly in the early stages of the laser pulse. The 5% increase in the electron density is consistent with that measured by Bentley [10] for the same temperature increase. Snyder *et al.* [9], however, found no electron density increase during the laser pulse.

Figure 2 shows that the rate of absorption of laser energy decreases, while the rate of energy loss through each of the four cooling processes increases, during the pulse. Electron thermal conduction and energy transfer to heavy particles through electron-impact ionization are the dominant cooling processes.

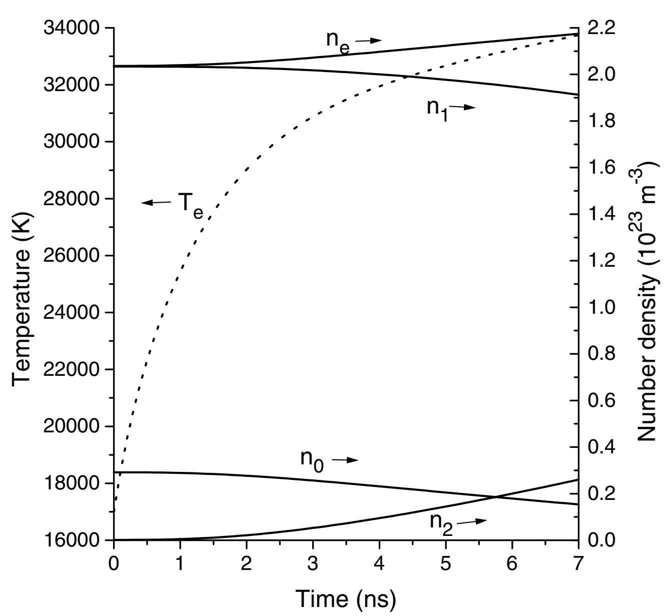

FIG. 1. Evolution of the electron temperature and species number densities at the center point during a 7 ns laser pulse. The laser pulse energy is 100 mJ, and the initial electron temperature is 17 000 K.

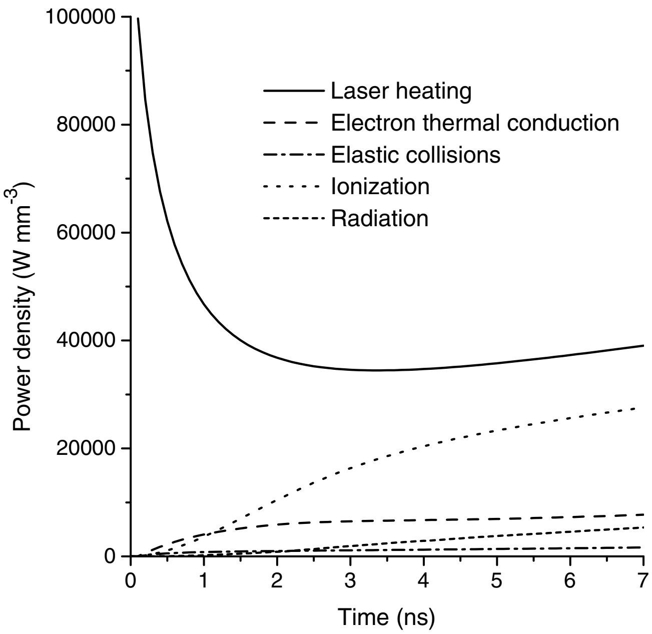

FIG. 2. Evolution of the power density of the laser heating and the different cooling processes at the center point. Parameters are as in Fig. 1.

As noted earlier, the decrease in the absorption of laser energy, and the increased rate of cooling, as the electron temperature increases, result in a nonlinear relationship between electron temperature and pulse energy. The extent of the deviation from the linear relationship used to derive unperturbed electron temperatures in previous works is shown in Figs. 3 and 4. The figures compare least-squares linear fits to the measurements of Snyder *et al.* [9] and Bentley [10] with least-square fits to solutions of Eq. (2). These latter fits were obtained using two free parameters: the initial electron temperature T_{e0} and a constant C by which the absorption coefficient α was multiplied. Allowing deviations from $C = 1$ takes into account uncertainties in the laser beam's spatial profile and diameter, in the time profile and duration of the laser pulse, and in the spatial and time averaging of the electron temperature. In the experiments, the temperature is derived from the integrated spectrum of the light scattered for the duration of the laser pulse, and from the full cross section of the laser beam. The electron temperature measured for each pulse energy is therefore both a time- and spatially averaged value. The frequency spectrum of the Thomson-scattered light is a complicated function of electron temperature and electron density, so the averaging process is a significant source of uncertainty. Here the electron temperature is calculated using a simple time average over the duration of the laser pulse and using a spatial average over the laser beam diameter, weighted according to the laser beam intensity profile.

Note that while both Snyder *et al.* and Bentley quote identical values for laser beam parameters, the electron heating shown in Fig. 4 is significantly greater than that

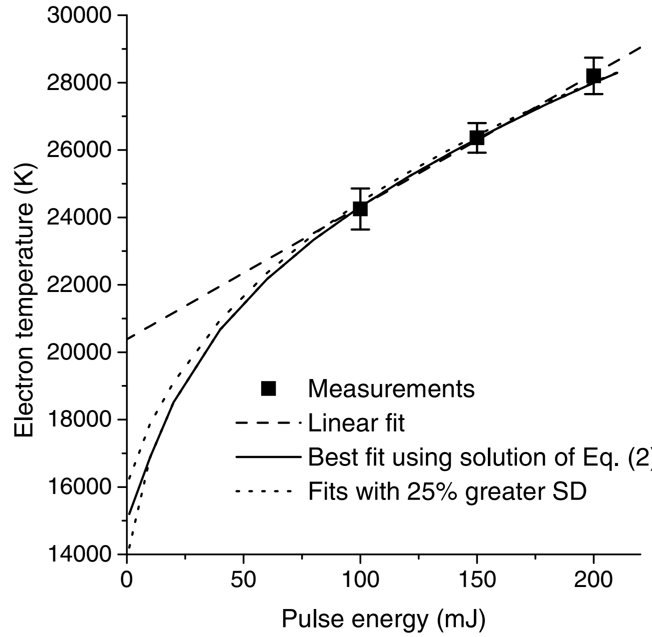

FIG. 3. Comparison between the best fit obtained using solutions to Eq. (2), and the line of best fit, to the measured data of Snyder *et al.* [9]. Also shown are fits obtained using solutions to Eq. (2) for which the standard deviation from the experimental points is 25% greater than for the best fit.

shown in Fig. 3. Hence a larger value of C is required to fit the results shown in Fig. 4.

The least-squares best fit to the measurements of Snyder *et al.* shown in Fig. 3 is obtained for $T_{e0} = 15\,200$ K and $C = 1.05$. This compares with the ion temperature measured by Thomson scattering of $14\,200 \pm 700$ K [9]. As a measure of the sensitivity of the least-squares fit, fits for

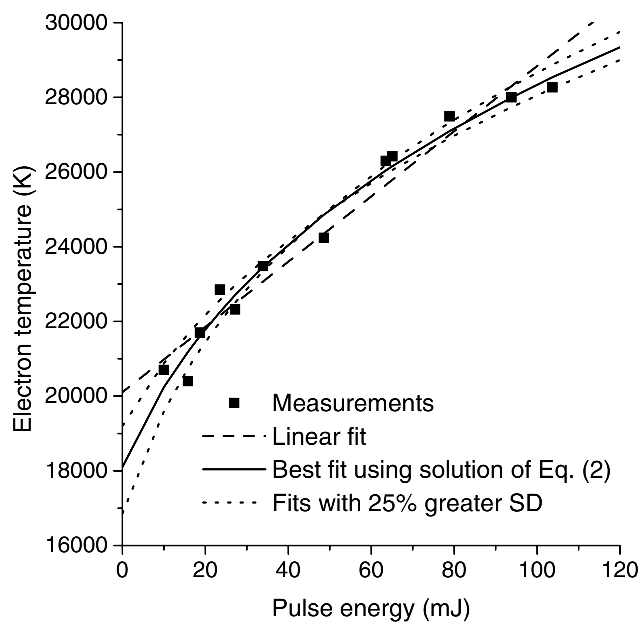

FIG. 4. As for Fig. 3, but using the measurements of Bentley [10].

which the standard deviation is 25% greater are also shown in Figs. 3 and 4. In the case of the Snyder *et al.* data, the deviation in T_{e0} is around 1000 K.

The least-squares best fit to the measurements of Bentley shown in Fig. 4 is obtained for $T_{e0} = 18\,100$ K and $C = 1.455$. The deviation in T_{e0} corresponding to a 25% increase in standard deviation is in this case 1300 K. The excitation temperature and the ion temperature were around 16 600 K for the same conditions [6]. The measured data have a nonlinear dependence on pulse energy that is consistent with that of the calculated curve.

The values of T_{e0} calculated using best fits to the solution of Eq. (2) are between 2000 and 6000 K lower than those obtained using a linear fit. They are within 1500 K of the ion temperature and excitation temperature. Since there are significant uncertainties in the transport and rate data used in the calculation, and since the results of calculations obtained using lower values of T_{e0} fit the measured data almost equally well, it is reasonable to conclude that the results admit values of electron temperature that are in agreement with ion and excitation temperatures. Hence, Thomson-scattering measurements of electron temperature, when analyzed correctly, do not provide strong evidence of deviations from LTE in thermal plasmas.

In some Thomson scattering measurements of plasma jets [13,15], an expanded laser beam diameter of 2 mm was used, with pulse energies of up to 400 mJ. The electron temperatures derived from the scattered signal using standard methods were around 20 000 K. The laser power density corresponds to around 4 mJ for the beam diameter of 200 μm used in the current study, so from Figs. 3 and 4, an electron temperature increase of around 1200 K is expected. Although the level of electron heating is decreased, the use of an expanded beam has two disadvantages. The signal-to-noise ratio is very low, and, because of the large temperature and density gradients present in arcs and jets, the scattered signal is obtained from a region in which the temperature varies by a large amount, up to 8000 K for a 2 mm beam diameter. These factors lead to large uncertainties in deriving a temperature from the scattered signal.

The results of this study have demonstrated flaws in previous measurements of electron temperature by Thomson scattering in thermal plasmas. The heating of the electrons by absorption of the laser energy means that any measurement of electron temperature is a spatial average over the cross-sectional area of the laser beam and a time average over the duration of the laser pulse. Moreover, derivation of an unperturbed electron temperature by linear extrapolation of measurements of electron temperature as a function of laser pulse energy is physically invalid. The dependence of the absorption of laser energy, and the cooling of the heated electrons by electron thermal conduction, by elastic and inelastic collisions with heavy particles, and by radiative emission, must all be taken into account. A one-dimensional analysis has indicated that these effects are substantial and has yielded electron temperatures 2000 to 6000 K lower than the linear extrapolation. These electron temperatures are comparable with heavy-particle and excitation temperatures. I conclude that this study, particularly when considered together with other recent work [15], shows that Thomson-scattering measurements of electron temperature in thermal plasmas do not provide strong evidence for significant deviations from LTE.

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