Theoretical considerations in modeling LII at low pressures

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This study presents some fundamental considerations in modeling LII at low-pressures and identifies conditions under which particle radiation heat loss dominants over sublimation and conduction.

Introduction

Low-pressure LII has received some attention in the literature. The rationale of conducting LII experiments in this regime is to minimize particle heat conduction loss and to extend the signal lifetime. If the particle temperature is sufficiently low to neglect sublimation heat loss, particle cooling occurs primarily through radiation and consequently low-pressure LII can be used to study radiative properties of nano-sized particles.

Several incorrect expressions for particle radiation heat loss rate exist in the LII literature, however, which have led some authors to erroneously claim that near-vacuum LII can be used as a nano-particle sizing technique. One of the main objectives of this study is to establish the correct expression for radiation heat loss rate from a spherical particle. We also investigate the error inherent in using the Rayleigh approximation to calculate particle laser energy absorption and the implication for two-color LII. Conditions under which particle radiation heat loss is dominant are identified in terms of particle temperature, pressure, and particle diameter, and a brief discussion is given on absorption and emission by aggregated particles.

LII Model and Theoretical Considerations

We employ the conventional single-particle based LII model given by

$$\frac{\pi d_p^3}{6}\rho_s c_s \frac{dT}{dt} = Q_{abs}\pi d_p^2 F_0 q(t) + \dot{q}_{rad} + \dot{q}_{cond} + \dot{q}_{sub} \quad (1)$$

$$\frac{dM}{dt} = \frac{\rho_s \pi d_p^2}{2}\frac{dd_p}{dt} = -\pi d_p^2\, \beta\, p_v \sqrt{\frac{M_v}{2\pi RT}}. \quad (2)$$

Radiant loss rate from a spherical particle is written as

$$q_{rad} = -\pi d_p^2 \int_0^\infty Q_{abs} E_{b,\lambda}\, d\lambda = -\pi d_p^2 \int_0^\infty \varepsilon_\lambda (d_p) E_{b,\lambda}\, d\lambda. \quad (3)$$

The particle absorption efficiency, Q_{abs}, obtained using both Rayleigh and Mie theories over size parameters relevant to LII is plotted in Fig. 1. Rayleigh theory underestimates Q_{abs} for size parameters greater than 0.15.

Figure 2 shows the conditions under which the ratios q_{rad}/q_{cond} and q_{rad}/q_{sub} are greater than 10 for a particle of 30 nm in diameter. For radiation heat loss to dominate, the particle temperature and gas pressure must be below 2760 K and 3×10^{-4} atm respectively.

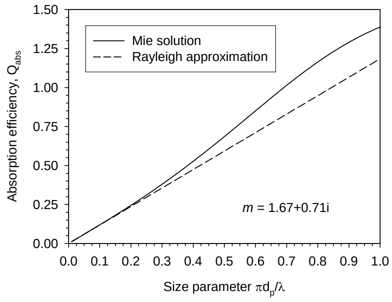

Fig.1: Absorption efficiencies calculated using Rayleigh and Mie theories.

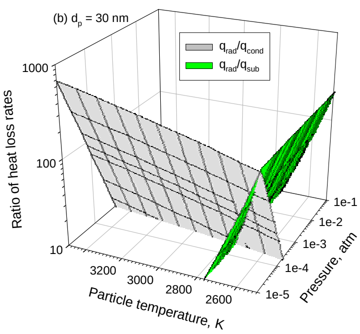

Fig.2 Variation of q_{rad}/q_{cond} and q_{rad}/q_{sub} with particle temperature and pressure for d_p = 30 nm.

The effect of using the Rayleigh approximation when predicting the effective temperature, T_e, of a polydisperse particle ensemble is also investigated. Rayleigh approximation could predict significantly lower peak particle temperature, especially when the green laser (λ = 532 nm) is used. The decay of T_e^{-4} under conditions where radiation heat loss is dominant is proportional to $\overline{E}(m)$, the mean particle absorption function over the visible and IR, for particles within the Raleigh regime.

Conclusions

Using the Rayleigh approximation can potentially induce considerable error in LII modeling for both laser energy absorption and the particle temperature analysis using two-color LII. Near-vacuum LII experiments can only be used to determine the mean value of E(m) provided that the particles are in the Rayleigh limit. Only under conditions of sufficiently low pressure and sufficiently low particle temperature radiation heat loss is the dominant particle cooling mechanism.

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