# Investigations of the mechanisms involved in LII particle detection

H.A. Michelsen*, M.Y. Gershenzon, P.O. Witze
Sandia National Laboratories, Livermore, CA 94551

We have made measurements of the temporal response of Laser-Induced Incandescence to pulsed excitation of soot in a flame and developed a corresponding LII model that accounts for particle heating by laser absorption, oxidation, and annealing and cooling by sublimation, radiation, and conduction to the surrounding atmosphere. The model also includes mass loss by oxidation, sublimation, and nonthermal photodesorption of carbon clusters. The results of this study allow us to identify the largest uncertainties associated with the understanding of LII and predict the influence of measurement parameters on LII signal under varying conditions.

## Experimental Studies

To test current models and investigate the influence of experimental conditions on LII behavior, we have measured time-resolved LII signals from soot in a nonsmoking coflow ethylene diffusion flame over a wide range of laser fluences as high as 4 J/cm$^2$ [1]. A Nd:YAG laser was injection seeded to provide a smooth laser temporal profile with a pulse duration of 7 ns, the output was doubled to generate 532-nm light, and the beam was passed through an aperture and relay-imaged into the flame to produce a smooth laser spatial profile. LII temporal profiles were recorded with a fast photodiode with adequate temporal resolution to capture signal evolution during the laser pulse. We used these results to aid in the development of a model that predicts the temporal behavior of LII from soot on a nanosecond time scale. The model accounts for particle heating by laser absorption, oxidation, and annealing and cooling by sublimation, conduction, and radiation. The model also includes mechanisms for convective heat and mass transfer, melting, and nonthermal photodesorption of carbon clusters [2].

Another set of experiments was performed to investigate the fast photodesorption mechanism in more detail. In these experiments the particles were heated with 532-nm pulses of ~70 ps duration from a regeneratively amplified modelocked Nd:YAG laser over a range of fluences as high as 0.6 J/cm$^2$. The signal was collected with a streak camera with a temporal resolution of ~15 ps.

## Model Development

Models typically used to describe LII are based on a model initially developed by Melton[3] in which energy- and mass-balance equations are solved to account for particle heating by laser absorption and cooling by conduction to the surrounding atmosphere, radiative emission, and sublimation. Particle size reduction during sublimation is also calculated. LII signal is derived from calculated temperatures and sizes using the Planck function weighted by the emissivity and the detector's wavelength response. The Melton model [3] uses (1) temperature-independent values for density and specific heat to determine the internal energy of the particle, (2) a Rayleigh approximation for laser absorption and radiative emission rates, (3) the approximation of kinetic control of $C_3$ only from the surface for sublimation rates, (4) a thermal accommodation coefficient and heat capacity associated with temperatures near room temperature, and (5) a formulation for conductive cooling appropriate for a transition regime between Knudsen and continuum flow.

We have developed a model [1,2] that similarly solves the energy- and mass-balance equations but also includes (1) temperature-dependent thermodynamic parameters for calculating sublimation, conduction, and internal energy storage by the particle, (2) wavelength-dependent optical parameters to describe absorption and emission of radiation based on a Rayleigh-Debye-Gans approximation to account for aggregation, (3) convective heat and mass flow (Stefan flow) during the sublimation of multiple cluster species (C, $C_2$, $C_3$, $C_4$, and $C_5$) from the surface, (4) a thermal accommodation coefficient appropriate for high temperature conductive cooling, (5) a conductive cooling mechanism assuming free molecular flow at low pressure and a transition regime at high pressure, (6) nonthermal photodesorption resulting in loss of heat and mass by carbon clusters leaving the particle, (7) phase changes (i.e., annealing and melting) and their effects on absorption, radiation, sublimation, and photodesorption, and (8) oxidative heating at the particle surface.

## References

1 H.A. Michelsen, P.O. Witze, D. Kayes, S. Hochgreb, Appl. Opt. 42, 5577-5590 (2003).
2 H.A. Michelsen, J. Chem. Phys. 118, 7012-7045 (2003).
3 L.A. Melton, Appl. Opt. 23, 2201-2208 (1984).

* Corresponding author: hamiche@ca.sandia.gov
Proceedings of the International Bunsen Discussion Meeting 2005: Laser-induced Incandescence, quantitative interpretation, modelling, application