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Non-incandescent emission in laser-induced incandescence measurements of metal nanoparticles

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In time-resolved laser-induced incandescence (TiRe-LII) a laser pulse energizes the nanoparticles within a sample volume of aerosol to incandescent temperatures, and the spectral incandescence from the nanoparticles is recorded as they return to the ambient gas temperature. Aerosol characteristics, including the nanoparticle size distribution, are inferred from the spectral incandescence decay rate using spectroscopic and heat transfer models. In many instances, however, LII models cannot reproduce observed experimental trends, e.g. "anomalous cooling", in which particles appear to cool faster than can be explained by known heat transfer phenomenon[1], and absorption of laser energy by metal nanoparticles that far exceeds what is predicted by Mie/Drude theory[2]. These abnormalities may indicate non-incandescent laser-induced emission sources that could corrupt the incandescence measurement, thereby biasing any aerosol parameters inferred using the measurement models.

Non-incandescent emission is particularly evident in the case of TiRe-LII measurements on silver nanoparticles with 1064 nm laser excitation, since the measured LII signal is far stronger than what could be attributed to incandescence[3]. The current work investigates the possibility that the signal is caused by neutral bremsstrahlung, in which electrons emitted by the nanoparticle through plasmon-induced hot-electron photoemission scatter from the neutral gas molecules, producing broadband emission[3]. This phenomenon is evaluated by carrying out three-color TiRe-LII measurements on Ag and Au aerosols within a range of buffer gases. The fact that the peak nanoparticle temperatures inferred from the LII signal is sensitive to the buffer gas type supports this hypothesis, since the electron-neutral cross-section, and thus the extent of the bremsstrahlung, depends on the gas molecular species.

A Monte Carlo simulation of electron emission and recombination is carried out to understand these experimental outcomes. In the simulation, the net initial charge state of the nanoparticles was adjusted to change the probability of electron emission through the photoelectric phenomenon. After the initial electron emission, the free hot carriers absorbed irradiation and emitted radiation via inverse neutral bremsstrahlung and neutral bremsstrahlung, respectively. A probability function is derived for emission and absorption based on the models. The simulations showed a buffer gas dependency on the maximum electron temperature, which is consistent with the experimental observations. This information will allow LII practitioners to derive more accurate spectroscopic models and identify operating conditions that could lead to contaminated signals.

References

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