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Summaries of Discussion Sessions

Discussion session 3: Pulsed and CW LII: modeling, evaluation, and unresolved questions

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Introduction

While both pulsed and continuous-wave laser-induced incandescence (LII) are widely viewed as reliable and mature diagnostics for characterizing soot-laden aerosols, and, increasingly, non-carbonaceous nanoparticles, there remain important questions concerning the accuracy and completeness of the measurement models used to interpret the spectral incandescence data. In the context of time-resolved LII (TiRe-LII), the measurement model can be subdivided into a spectroscopic submodel that relates the observed spectral incandescence to the instantaneous temperature of the nanoparticles within the probe volume (ad also the nanoparticle volume fraction), and a heat transfer/cooling model that connects the observed spectral incandescence/pyrometric temperature decay rate to the nanoparticle size distribution and other quantities-of-interest (QoI). The accuracy of QoI inferred from LII signals is predicated on the fidelity of the measurement models. Consequently, since the first LII workshop in 2005, there have been considerable advancements in our understanding of the physics that underlie both the spectroscopic and heat transfer submodels.

Spectroscopic Submodel

Since the last LII workshop in 2016, arguably the most attention has focused on understanding the spectroscopic model, or at least defining what is not known about the model. For example, while changes in the radiative properties of soot have long been empirically connected with laser heating/graphitization (e.g. Vander Wal et al., Appl. Phys. B 1998), more recent efforts have attempted to quantify this effect through atomistic-level modeling (Moulin et al., JQSRT 2008, Fernandez et al., JQSRT 2015). There also remain questions about how the heterogeneous structure of nanoparticles (e.g. oxide layers or coating surrounding a nanosphere or primary particle) may affect the spectral absorption cross-section; the laser pulse is usually presumed to remove/destroy this coating, but this may not always be the case.

In the context of aggregates, while Rayleigh-Debye-Gans Fractal Aggregate (RDG-FA) theory is appealing due to its simplicity and computational-efficiency, in many cases the underlying assumptions needed to support its validity are violated, which can lead to large model errors, and consequently errors in the inferred aerosol properties. Yon et al. (JQSRT 2015), Doner and Liu (JQSRT 2017), Huber et al. (JQSRT 2017), and Talebi Moghaddam et al. (J. Aerosol. Sci. 2018) have attempted to quantify this model error, and, in the latter case, develop a statistically-

robust correction that can be used to enhance the accuracy of the spectroscopic models without sacrificing the efficiency and convenience of RDG-FA. More fundamentally, the conceptual model of a fractal aggregate consisting of identically-sized spheres in point contact is often far from reality (Singh et al., Appl. Phys. B 2018), but attempts to model these effects should be carefully considered in the context of overall model error and model complexity, as described below.

Increasingly LII is being applied to noncarbonaceous nanoparticles, especially metal aerosols. In principal, the radiative properties of metallic nanoparticles should be more certain than those of soot since: (i) they are "pure materials"; (ii) the size of the nanoparticles is typically much larger than the mean free electron path, so electron scattering is not typically an issue; and (iii) the bulk properties of metals (particularly liquid metals) are both well-characterized and well-understood from a theoretical basis. Nevertheless, LII measurements on metal aerosols (e.g. Fe, Mo, Ag, Cu) reveal many unexpected features. At least some of these arise from the universal (and very often wrong) application of Rayleigh theory to calculate the absorption cross-section of metal nanoparticles. In order for Rayleigh theory to apply, the nanoparticle must be small relative to the wavelength, as defined by the size parameter $x = \pi d_p/\lambda$, but the phase shift parameter $|m|x << 1$ must also be small in order for all the dipoles within the nanoparticle to oscillate in phase with the incident wave. While this condition is satisfied for carbonaceous nanoparticles, it is generally not satisfied for metal nanoparticles. As a consequence, a reevaluation of many LII signals collected on metal aerosols is currently underway.

There is also growing speculation that many of the "mysterious phenomena" in LII experiments may be attributed, at least in part, to non-incandescent laser-induced emission (LIE). These include: anomalous cooling; enhanced absorption and emission cross-sections during combined LII/LOSA experiments; enhanced absorption cross-sections based on the laser fluence vs. peak pyrometric temperature; and, most recently, an unexplained "double peak" in spectral incandescence observed from LII measurements on soot using 532 nm excitation (Bauer et al., 8th Int'l Workshop on LII) One possible explanation for some of these phenomena is broadband neutral bremsstrahlung caused by laser-induced electron emission from the nanoparticle. Since the translational energy of electrons is not quantized, they emit radiation over a broad range of wavelengths as they scatter from neutral gas atoms, and absorb over a broad range of wavelengths as they accelerate in an oscillating E-M field. The broadband nature of bremsstrahlung emission makes it difficult to discern from incandescence, although one would expect a "blue shift" in the observed signal due to the high electron temperature compared to that of the nanoparticle. A critical re-analysis of the LII measurements on silver nanoparticles reported by Sipkens et al. (Appl. Phys. B 2017) strongly suggests that the laser-induced incandescence signal reported in that study was, in actuality, bremsstrahlung emission. While silver nanoparticles are particularly susceptible to non-incandescent LIE due to their low absorption cross-sections, this phenomenon may also affect LII measurements on soot, although presumably to a lesser extent. Neutral bremsstrahlung emission during LII is the focus of ongoing research.

Non-modeled experimental artifacts can also profoundly affect interpretation of LII data. Mansmann et al. presented a slide about recent LII measurements on an ethylene laminar diffusion flame using three different detection wavelengths. The measurements were compared

to recently published three-color data from a similar burner from Goulay et al. (Appl. Phys. B, 2013). Two-color pyrometry and spectral fitting were applied on both datasets using constant *E(m)*. It was demonstrated that temperatures for multiple two-color ratios and spectral fitting differed by 300 K for both datasets.

Musikhin et al. presented a slide about LII measurements performed on the same laminar diffusion flame used by Mansmann acquired simultaneously with streak camera and an array of four PMTs. Measurements revealed a discrepancy in the timing between the LII peak and the maximum temperature of 8 ns for the PMTs and 20 ns for the streak camera, causing a difference between the two temperatures of 40 K for the PMTs and 530 K for the streak camera. This result highlights the fact that the "maximum temperature" and "temperature at peak incandescence" may not be the same due to measurement artifacts.

Spatial filtering using a pinhole, aperture, or optical fiber is a common technique in LII to form a "probe volume". Mansmann et al. presented a slide about recent measurements showing the difference in LII peak temperature when using normal lenses vs. achromatic lenses. The measurements revealed differences in LII peak temperatures of more than 1000 K. The error introduced by normal lenses decreased with decreasing temperatures and was negligible for particle temperatures around the gas temperature.

Finally, at least one of the workshop presenters (KD) feels **very strongly** that the term "emissivity" is inappropriate to describe absorption and emission by nanoparticles. Some LII practitioners have the erroneous picture that the wave only interacts with the atoms in the outermost layer of the nanoparticle (as is the case when a wave interacts with an opaque macroscopic surface), when, in fact, Rayleigh theory assumes that all the dipoles within the nanoparticle respond to the E-M field simultaneously and oscillate in phase. While one may argue that emissivity can be used as an "effective" parameter that abandons any physical meaning, this leads to some pretty strange results. For example, if $Q_{\mathrm{abs},\lambda} > 1$ (which often happens *cf.* Bohren and Huffman), the particle emissivity would also be greater than unity, which, in the traditional interpretation of emissivity, implies a violation of the 2nd Law, a negative reflectivity according to Kirchhoff's law, etc. There is no need to do this, it is a bad idea, please stop doing it.

Heat Transfer Submodel

The fundamental physical processes underlying the heat transfer submodel (laser heating and cooling by evaporation and conduction) are generally better-understood compared to the spectroscopic submodel, and most of the associated uncertainties arise from uncertain model parameters rather than the physical processes themselves. The current state-of-the-art is summarized in the review article by Michelsen et al. (PECS 2015).

If the nanoparticle is much smaller than the molecular mean free path of the gas, both evaporation and conduction occur within the free molecular regime. This means that molecules travel ballistically between the nanoparticle surface and the equilibrium gas without undergoing intermolecular collisions at an intermediate thermodynamic state. Under these conditions evaporative cooling is modeled using the Clausius-Clapeyron relation, which implicitly assumes phase equilibrium at the nanoparticle surface. In the case of soot, the main uncertainty in free-molecular evaporation concerns the latent heat of vaporization and molecular mass of the

evaporated species (e.g. Smallwood et al., JHT 2001). There has been some speculation in the literature that anomalous cooling may be related to evaporation associated with PAHs, enhanced by photoabsorption, or a temperature-dependent thermal accommodation coefficient, although it could also be attributed to non-incandescent emission as described above. This is an important topic of ongoing research.

Conduction depends on the incident gas molecular flux and the thermal accommodation coefficient (TAC). While at the time of the first LII workshop the TAC was treated as a "tuning parameter" that was freely varied to match cooling models to experimental data, comparative experimental studies (Daun et al, JHT 2008) and molecular dynamics (Daun, IJHMT 2009) have highlighted the physics underlying this parameter. This treatment has also been applied to metallic aerosols (e.g. Sipkens et al., Appl. Phys. B 2017, Daun et al., Appl. Phys. B 2013) and silicon nanoparticles (Sipkens et al., Appl. Phys. B 2014, Sipkens and Daun, 8th Int'l Workshop on LII). Further insights into the gas-surface scattering physics that underlie this parameter have been obtained using cube models, which treat the surface atoms in an ensemble way (Sipkens et al., IJHMT 2017). These and more recent MD simulations (Sipkens and Daun, 8th Int'l Workshop on LII) also suggest that, under LII conditions, the TAC is nearly independent of the surface temperature.

The picture becomes more complicated when mean free molecular path approaches the same scale as the nanoparticle diameter, in which case intermolecular collisions in the gas next to the nanoparticle become important. In this scenario both conduction and evaporation occur within the transition regime (i.e. the transition between free-molecular and continuum). In the context of conduction, a range of transition-regime schemes have been used by LII practitioners, as summarized by Fillipov and Rosner (IJHMT 2000), Liu et al. (Appl. Phys. B 2006) and Daun and Huberman (IJHMT 2012). These schemes interpolate between the continuum and free molecular regimes based on the Knudsen number, which is the ratio of mean free molecular path and particle diameter. The most successful, and recommended approach, is the Fuchs boundary sphere method with temperature-dependent properties.

This treatment is not "exact" since it assumes that the gas is stationary, when, in fact, there will be a bulk gas molecular flux away from the nanoparticle as the gas expands over a duration of approximately 50 ns. To date there is no analytical treatment that captures this phenomenon, nor any transition-regime effects associated with evaporation. Instead, to the best of our knowledge, evaporation has always been modeled as a free molecular phenomenon, even if conduction occurs in the transition regime. These phenomena have been explored through transient DSMC simulations (Memarian et al., Num. Het Trans. A 2014, Appl. Phys. B 2015) and, under most conditions, have a negligible impact on heat transfer from laser-heated nanoparticles.

The influence of aggregate structure on heat transfer is an ongoing area of study. In the free-molecular regime, primary particles in the interior of the aggregate are shielded from incident gas molecules by exterior primary particles. This effect is most often modeled using the equivalent sphere technique proposed by Liu et al. (JQSRT 2005), based on a Monte Carlo simulation of gas molecules travelling ballistically from a boundary sphere enveloping the soot aggregate; a fraction of these molecules scatter from one or multiple primary particles, and an effective diameter is derived from an overall energy balance over the molecules crossing the

spherical domain that is a function of aggregate size, fractal structure, and TAC. Subsequent work examined how factors like realistic scattering kernels (Daun, JHT 2010) as well as necking and sintering (Johnsson et al., Appl. Phys. B 2013) affect heat transfer rates, but these are secondary effects. Some studies, such as Bladh et al. (Appl. Phys. B 2011), have attempted to isolate these effects experimentally. Moreover, there have also been some attempts to infer the aggregate fractal parameters from the observed incandescence/pyrometric temperature decay rate by inverting the shielding parameter, but Bayesian analysis suggests that obtaining statistically-robust estimates is not possible in the context of measurement noise and model parameter uncertainty (Bauer et al., 8th Int'l Workshop on LII).

A glaring "hole" in LII modeling capabilities is the lack of a detailed transition-regime model for aggregates. The standard approach is to use the effective diameter obtained from the equivalent sphere method, which is derived under free-molecular conditions, along with the Fuchs boundary sphere method, which only applies to nanospheres. Obviously this approach is only approximate and does not represent the true gas molecule/aggregate interactions. To the best of our knowledge, no DSMC calculation of transition-regime heat conduction from an aggregate has been carried out to date due to its computational complexity, since simulations would need to be carried out for a large number of aggregate sizes, geometries, and thermal accommodation coefficients, in order to obtain a statistically-meaningful ensemble behavior.

There is also growing concern about whether the gas temperature surrounding the nanoparticle is actually the equilibrium gas temperature. This assumption presumes that heat conducted from the nanoparticles is uniformly diffused throughout the gas over the period between pulses, so that the local gas temperature surrounding the nanoparticle is the bulk gas temperature at the beginning of each pulse, but recent calculations (Snelling et al., Appl. Phys. B. 2009), observations with sequential signal detection (Mansmann et al., Opt. Express 2017), and 2D pyrometric imaging (Cenker et al., Aerosol Sci. Tech. 2017) suggest that, for high fluence measurements and high soot loading conditions, the gas temperature may increase by up to several hundreds of degrees K. This would obviously influence any parameter inferred from the spectral incandescence/pyrometric temperature decay.

In principle the cooling rate may also be influenced by radiation, oxidation, annealing, and thermionic emission, although these terms are negligible under almost all experimental conditions. Nevertheless, these effects are important in different ways, e.g. radiant emission is (hopefully) the process through which the signal is produced; annealing (which has been investigated recently using two-pulse LII, e.g. Mansmann et al., 7th Int'l Workshop on LII 2016, Cenker et al., Appl. Phys. B 2017) certainly affects the optical properties of soot (Saffaripour et al., Appl. Phys. B 2015), albeit in an uncertain way; and thermionically-emitted electrons may contribute to non-incandescent emission through neutral bremsstrahlung.

Continuous Wave LII (CW-LII)

The CW-LII community approaches data analysis in a fundamentally different way to the pulsed-LII community, for one fundamental reason: single-particle CW-LII can be directly calibrated to mass, alleviating the need for a physical model to interpret the data.

CW-LII almost exclusively use the commercial Single Particle Soot Photometer (SP2) (Droplet Measurement Technologies, USA) and all statements in this section apply to this instrument.

The SP2 measures time-resolved LII and time-resolved light-scattering as particles traverse and vaporize within a 1064 nm laser beam. The majority of modelling work on CW-LII is applied to the SP2 scattering signal, in order to retrieve information on the mixing of rBC (or other incandescing material) with non-incandescing material. Four unique approaches to mixing-state retrieval can be identified in the literature.

First, so-called delay-time analysis compares the scattering and incandescence signals and identifies 'thickly-coated' particles as those whose scattering signal decreases prior to the onset of incandescence. This decrease in scattering indicates evaporation of a substantial fraction of particle volume (internally-mixed, non-refractory material), and the technique typically requires over 30% of the particle volume to be non-refractory.

Second, the time-resolved scattering signal can be combined with a known laser beam fluence profile (measured using scattering by non-absorbing particles) and the assumptions of a core-shell particle morphology, plus coating refractive indices, to obtain time-resolved scattering cross-sections. This data is used to infer the effective (core-shell) coating thickness.

Third, a detailed spectroscopic, heat, and mass transfer model for the soot aggregate during CW laser heating may be used to interpret the CW-LII signal. Only one publication has performed such an analysis (Bambha and Michelsen, J. Aerosol Sci. 2015). The results were highly informative, and showed how varying soot morphology affected the SP2 signals. There was some discussion at the 8th LII Workshop about the potential utility of continuing and expanding this work.

Fourth, experimental control rather than modelling has been used to infer particle mixing state using CW-LII. The experimental configuration involves mass-classifying particles using a technique such as the centrifugal particle mass analyzer, followed by measurement with CW-LII. The result is a direct measurement with no assumptions, at the cost of experimental and analysis complexity, and has been highly successful (Liu et al., Nature Geosci. 2017, Broda et al., Aerosol Sci. Technol. 2018.)

In personal discussions following this session, the discussion leaders had the impression that the consensus opinion of the SP2 user community holds that direct empirical calibration is superior to numerical modelling, when possible, according to the 'model complexity' topic raised at the end of this document. However, the possibility of 1064-nm-absorbing, but non-incandescing particles being detected by SP2 was also demonstrated during this session. These particles demonstrate a unique usefulness of single-particle optical modelling, and may motivate future studies similar to Bambha and Michelsen (J. Aerosol Sci. 2015).

LII Data Analysis

In the context of metrology, it is just as important to quantify the uncertainty attached to an LII-derived quantity-of-interest, as it is to report the quantity itself. A major challenge lies in the fact that obtaining QoIs from LII data constitutes an inverse problem, broadly defined as a problem in which quantities or values are inferred from indirect measurements. Inverse problems are often mathematically ill-posed, which, in the case of LII, is because multiple candidate solutions (e.g. nanoparticle size distributions) can explain the observed incandescence traces within the

bounds of uncertainty (*cf.* Roth and Filippov, J. Aerosol Sci. 1996, Daun et al. Appl. Phys. B. 2007). Ill-posedness arises from an information deficit in the measurement data (i.e. there is not enough information to specify a unique solution) so the antidote is to impose additional information known before the measurement, a.k.a. "prior" information. For example, TiRe-LII nanoparticle sizing almost always assumes a low-order parameter form for the size distribution, e.g. log-normal. This prior information reduces the statistical degrees-of-freedom (DOF) of the inference problem to two parameters, making it less ill-posed. On the other hand, including uncertain model parameters in the inference process makes the problem more ill-posed and sensitizes the problem to uncertainties, so it is important to include as much prior information as possible to counterbalance any increase in degrees-of-freedom as long as the prior information is reliable. Treating these parameters as "fixed" is not appropriate, since this would cause the QoI to be understated.

Uncertainties in the measurement process are amplified by the ill-posedness of the problem into large variations in the recovered parameters. Uncertainties come from four main sources: uncertain aspects in the experiment (e.g. spatially-nonuniform laser fluence, temporal misalignment of LII traces); an incomplete understanding of the physics underlying the spectroscopic and heat transfer models (i.e. model errors); measurement noise (e.g. photonic shot noise); and uncertain model parameters (e.g. ρ, c_p, α, etc.). Examining the consistency of temperatures inferred from four-color LII, and quantifying RDG-FA model error by comparing it to higher-order model approximations, are examples addressing the first two sources. Uncertainty arising from measurement noise can be captured through sample-based approaches (e.g. the standard-deviation of results obtained through shot-averaging), while uncertain model parameters are historically accounted for using univariate sensitivity studies (e.g. the Kline-McClintock procedure).

Increasingly, however, LII practitioners turn to Bayesian techniques to quantify uncertainty, an approach pioneered by Sipkens et al. (Appl. Phys. B 2014). In the Bayesian viewpoint, the measurement data, QoI, and ancillary model parameters are random variables that obey probability density functions (PDFs) as opposed to holding fixed, discrete values. (This does not necessarily mean that the parameters are inherently random, but reflects that our state-of-knowledge regarding the parameters is uncertain). So, in the Bayesian context it is meaningless to argue whether the TAC for soot is 0.34 or 0.35.) These PDFs are related by Bayes' equation, and the outcome of the analysis are posterior PDFs for the quantities-of-interest. The advantages of this approach over other types of uncertainty analysis are: (i) it is easy to incorporate prior information, e.g. MD-derived TACs or the range of thermophysical properties reported in the literature; and (ii) the posterior PDF width directly reflects the uncertainty in the derived quantity. It is sometimes argued that the prior PDF biases the outcome based on the analyst's viewpoint, but this can be avoided using the Principle of Maximum Entropy, in which the prior PDFs are defined so that their information entropy is maximized subject to constraints corresponding to testable (verifiable) information. It is also possible to incorporate an additional error term that accounts for generic model error, e.g. the error caused by assuming an RDG-FA model for a soot aggregate. See, e.g. Huber et al. (JQSRT 2017), Talebi Moghaddam et al. (JQSRT 2018).

Hadwin et al. used the Bayesian methodology to infer soot volume factions from LII measurements on an ethylene laminar diffusion flame (Appl. Phys. B., 2015, 2016). In the context of non-carbonaceous soot, Sipkens et al. (Appl. Phys. B 2015, 2017) derived posterior PDFs for TACs and particle sizes inferred from LII measurements on metal aerosols, and Menser et al. (Appl. Phys. B. 2016) obtained PDFs the particle size, TAC, and Antoine parameters for silicon nanoparticles. (The Antoine parameters govern the vapor pressure in the Clausius-Clapeyron equation, and are related to the Gibbs free energy of the phases.) This approach also enables fusion of different types of measurement data containing complementary information in a statistically-rigorous way, e.g. combining the peak temperature vs. fluence curves with temperature decay curves and atomic line fluorescence to obtain more robust estimates for the Antoine parameters (see Menser et al. 8th LII Workshop.)

While most applications of Bayesian inference to LII experiments treat the data as stationary, Hadwin et al. (JOSA A 2018) showed how LII data analysis can be interpreted as a nonstationary estimation problem using a Kalman filter. In this “predictor/corrector” approach, the “state variables” (temperature, volume fraction) evolve with the measurement. The heat transfer model “predicts” the next pyrometric temperature, which is then “corrected” using the observed spectral incandescence data. Hadwin et al. used this approach to show how the uncertainties of LII-derived parameters (e.g. soot volume fraction and primary particle size) evolve over the measurement signal. It can also be used to determine the optimal detection time for inferring the SVF, as a trade-off between the uncertainty caused by unexplained prompt phenomena (e.g. anomalous cooling) and the impact of polydispersity and uncertainties in the cooling model at later times.

While the objective of most LII data analysis is to infer the particle volume fraction from peak intensity/temperature measurements, or determine the nanoparticle size distribution, TAC, or associated parameters from the spectral incandescence/pyrometric temperature decay, increasing attention has focused on mining other types of LII data for information about the aerosol, which can reduce the ill-posedness of the inference problem. Sipkens et al. (Appl. Phys. B 2017) show how plotting the peak temperature versus the fluence can reveal information about properties including the gas temperature, absorption efficiency at the laser wavelength, and latent heat of vaporization. By defining a dimensionless peak temperature and dimensionless fluence, LII measurement data collected on different aerosols under different experimental conditions can also be collapsed onto a single fluence curve. Outliers on this curve can be used to troubleshoot issues with the experiment or models used to interpret the data. Sipkens et al. (Appl. Opt. 2017) also showed how the measurement noise, specifically the linearity between the variance and the expected value of the spectral incandescence signal, reveals how experimental conditions may vary between laser shots. This type of analysis can also be used to troubleshoot problems with the experiment (e.g. excessive shot-to-shot variation in laser fluence) as well as turbulent fluctuations in particle volume fraction.

LII Model Development

The end goal of LII model development is to derive accurate estimates of the quantities-of-interest from LII data. Model developers have pursued this goal by developing increasingly elaborate models that include second-order effects like thermionic emission and annealing, as

well as temperature dependent properties. Most often the model is developed as follows: (*i*) simulated data generated with an existing model is compared to "benchmark" experimental LII data and deficiencies are noted; (*ii*) a new, more complex model is proposed that features some additional "tuning parameters" that are fit through nonlinear regression to the benchmark data; and (*iii*) the new model is accepted based on the smaller residual between measured and modeled quantities.

Instead of improving the robustness of LII-derived quantities, this procedure often has the opposite effect due to model over-tuning. A new model may fit the data better by virtue of its increased DOF, and not because it better represents the true physics. Instead, the higher DOF model may capture other unintended phenomena, e.g. unaccounted-for-errors arising from the experimental setup. LII practitioners often report that models developed in one laboratory using one set of benchmark data cannot explain measurements made under nearly-identical experimental conditions at another laboratory; this is a classic symptom of over-tuning. In the Bayesian context, including too many DOF "diffuses" the posterior probability over a large number of dimensions, which makes the inferred parameters less robust. Most critically, over-tuning leads to false confidence about the problem physics. For example, it is reasonable to suppose that the radiative properties of soot should change during laser heating (which they probably do), and allowing this to be a DOF will inevitably improve the fit between modeled and benchmark data. It would be incorrect, however, to conclude that an improved fit, by itself, justifies the proposed innovation. A more classic example of this concerns the evolution of our understanding of planetary orbits. Tycho Brahe accepted Ptolemy's complex geocentric model over Copernicus's simpler heliocentric model based on the better goodness-of-fit between his observations and Ptolemy's model, while not accounting for the far greater DOFs in the Ptolemy model. This erroneous picture may lead researchers down false trails, confound the interpretation of experimental data, and, ultimately, erodes the credibility of LII as a reliable diagnostic.

For these reasons, the workshop discussion leaders identified **model complexity** as the single biggest "burning issue" in LII modeling. To realize further improvements in the accuracy and reliability of LII, model developers need to carefully consider whether increasing the model degrees-of-freedom is justified in the context of measurement noise, model parameter uncertainty, and model error. Sipkens et al. (J. App. Phys. 2018) showed how Bayesian model selection can be used to choose models having the "right" level of complexity. This procedure is based on calculating the posterior probability for a particular model (or, more often as a *Bayes Factor*, which is the ratio of posterior probabilities), which accounts for the goodness-of-fit, measurement nose, prior information, and uncertainty in the model parameters. Sipkens et al. used this approach to select a model that accounted for the temperature-dependence of the latent heat of vaporization for liquid iron. Model selection was first demonstrated on simulated data generated with competing models, and then used to analyze experimental LII data collected from iron nanoparticles. The LII data showed that Román's equation is preferred to Watson's equation, and both are preferred over assuming a temperature-independent value. This result also exemplifies the increasing trend towards using LII as an instrument for fundamental scientific inquiry, as opposed to a "turn-key" instrument for characterizing soot-laden aerosols. Admittedly Bayesian model selection requires some expertise and investment on

the part of the analyst, but these issues in general are something that every LII model developer and user should keep in mind.