Discussion session 4: Non-soot LII

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Motivation:

There is a wide variety of gas-borne non-carbon particulates of interest that require the development of possibilities for determining their properties. These include:

- Atmospheric aerosols other than black carbon (which are expected to be unstable at LII conditions)
- Manufactured nanoparticles (metals and metal oxides) where
(a) it is important to gain fundamental understanding of reaction processes. Here, it is of interest to measure particle sizes and particle volume fraction both *in situ* to observe spatial / temporal variation in particle size and to develop a model understanding of the underlying complex processes.
(b) Because synthesis is of commercial interest, process control (inline, potentially via sampling: faster than conventional methods) is a second topic of interest
(c) For health and safety the detection of nanoparticles in the (workplace) atmosphere is of interest
- Welding fumes generate unwanted nanoparticles at the workplace
- Additionally, the investigation of LII provides information for basic sciences to further understand particle–gas-phase interaction

Challenges:

In contrast to LII on carbonaceous materials, there is a very small number of investigations for non-soot materials reported in literature. Therefore, there is a

- Severe lack in fundamental information including
    - Optical properties of many materials (which might additional be different for different phases reached during laser heating)
    - Thermal accommodation coefficients
    - Phase-transition temperatures and thermodynamics unknown (which can include non-metal / metal transitions with resulting strong variation in optical properties)
    - Evaporating species (that can potentially lead to additional emission once electronically excited)
    - Sources for interfering signal (Raman, Plasma emission, ...) when increasing fluence

There are additional fundamental challenges for:

- Elemental materials (Fe, Si, Cu, Mo, ...)
    - Many materials have comparably low boiling points that lead to weak LII signals, which makes LII potentially hard to detect in high-temperature environments
    - Low melting points and other phase transitions occur in the temperature range that is reached during the LII process. These might (a) change the optical properties and (b) contribute to the heating and cooling behavior because of the contribution of the latent heat during phase transitions. As a further complication, it is not clear if these processes happen under thermodynamic control or if they are kinetically controlled on the timescale of interest
- Oxidic materials
    - High-temperature chemistry (i.e., loss of oxygen) happens at high temperature. Therefore, additional heat-loss paths (with the implicit question of they are happening under kinetic or thermodynamic control) are opening up
    - Many oxidic materials show low absorption in the NIR and visible (e.g., SiO_2, TiO_2)

Advantages:

In comparison to soot, non-soot materials also provide advantages.

- The particle morphology is usually simpler compared to soot and therefore, the “maturity issues” observed for soot are less important
- During the synthesis of tailored materials, the reaction conditions are usually well controlled. Therefore, there is a much better chance to determine the boundary conditions (such as gas-phase temperature, size-distribution and aggregation). These conditions even provide the possibility to generate isolated particles under well-defined conditions that can be used for fundamental investigations of the interaction between heated particles and their environment

Current status / Publications for non-soot particle LII

Iron nanoparticles

Study	Year	Materials	Experimental notes	Model	QoI
Vander Wal et al.	1999	Fe, W, Mo, Ti	Laser ablation, examined fluence dependencies and emission spectra	-	-
Starke et al.	2003	Fe, C	Shock wave reactor, ~10 nm	Conduction (transition regime) submodel, temperature independent properties	d_p
Kock et al.	2005	Fe	Hot wall flow reactor, ~30 nm	Conduction, evaporation, and radiation submodels	d_p, α
Eremin et al.	2008	Fe	UV laser photolysis, a range of nanoparticle diameters	-	d_p, α
Eremin et al.	2011	Fe, C	Shock wave reactor, a range of nanoparticle diameters	Conduction, evaporation, and radiation submodels	d_p, $E(m_\lambda)$
Eremin et al.	2013	Fe, C	Shock wave reactor, simultaneous extinction measurements, focus on evaporation model	Conduction, evaporation (with Kelvin equation), and radiation submodels	d_p, f_v, T_{peak}
Sipkens et al.	2015	Fe	Aerosolized colloid, 30-70 nm	Conduction and evaporation (with Kelvin, Tolman, and Watson equations) submodels	d_p, α
Sipkens et al.	2017	Fe, Ag, Mo	Aerosolized colloid, 30-70 nm, comparative study	Conduction and evaporation (with Kelvin and Watson equations) submodels	d_p, α
Sipkens et al.	2018	Fe	Aerosolized colloid, 30-70 nm, focus of model selection	Conduction and evaporation (with Kelvin equation) submodels	Δh_v

Metal nanoparticles (other than iron)

Study	Year	Materials	Experimental notes	Model	QoI
Vander Wal et al.	1999	Fe, W, Mo, Ti	Laser ablation, examined fluence dependencies and emission spectra	-	-
Filippov et al.	1999	C, Ag, TiN	Spark generator, powder in a shock tube	Conduction, evaporation, and absorption submodels	d_p
Murakami et al.	2005	Mo	UV laser photolysis, fluence curves are included	Conduction submodel (temperature independent properties)	d_p
Reimann et al.	2010	Ni			
Sipkens et al.	2013	Mo	UV laser photolysis, reanalysis of data from Murakami et al.	Conduction and evaporation (negligible) submodels	d_p/α, σ_g
Eremin and Gurentsov	2015	Mo	UV laser photolysis, study of nanoparticle formation	Conduction, evaporation, and radiation submodels	d_p, T_{peak}

Sipkens et al.	2017	Fe, Ag, Mo	Aerosolized colloid, 30–70 nm, comparative study	Conduction and evaporation (with Kelvin and Watson equations) submodels	d_p, α
Daun et al.	2016	Cu, Si	Plasma reactor (Si), arc discharge reactor (Cu), LOSA measurements	Spectroscopic model	-

Metal oxide nanoparticles

Study	Year	Materials	Experimental notes	Model	QoI
Weeks and Duley	1974	Al_2O_3, C	Powders in a drift tube, 300 nm	Conduction, radiation, and absorption submodels	T_{peak}
Altman et al.	2001	SiO_2	Flame reactor	-	-
Lehre et al.	2005	MnO	Powders in an evaporation chamber, ~40 nm	Conduction, evaporation, radiation, and absorption submodels	Clausius-Clapeyron equation parameters
Maffi et al.	2008	TiO_2	Flame reactor	-	-
Cignoli et al.	2009	TiO_2	Flame reactor, 17–48 nm	-	-
Tribalet et al.	2012	Fe_2O_3	Low-pressure flame reactor	At least a evaporation submodel	d_p, α

Seminconductor nanoparticles

Study	Year	Materials	Experimental notes	Model	QoI
Eom et al.	2003	Si	Low-pressure plasma reactor	Melton model (conduction, evaporation, radiation, and absorption submodels)	d_p
Eom et al.	2004	Si	Low-pressure plasma reactor	Melton/Holfeldt model (conduction, evaporation, radiation, and absorption submodels)	d_p
Sipkens et al.	2014	Si	Low-pressure plasma reactor	Conduction and evaporation (Kelvin, Tolman, and Watson equations) submodels	d_p, σ_g
Menser et al.	2016	Si	Low-pressure plasma reactor, considered uncertainties in vapor pressure, LOSA measurements	Conduction, evaporation (Kelvin equation) submodels, and absorption submodels	d_p, α
Daun et al.	2016	Cu, Si	Low-pressure plasma reactor (Si), arc discharge reactor (Cu), LOSA measurements	Spectroscopic model	-
Menser et al.	2018	Ge	-	-	-