Operando Raman Spectroscopy of Lithium-ion Battery Electrolytes and Electrodes

Joachim Wallenstein [a], Patrik Johansson[a]
[a] *Dept. of Physics, Chalmers University of Technology, SE-41296 Göteborg, Sweden*

E-mail: joachim.wallenstein@chalmers.se

The co-optimisation of life-length and charging/discharging time, *e.g.* fast-charging dependent ageing, is a hot topic within the lithium-ion battery (LIB) research community. By characterisation at the molecular level proper understanding of various ageing mechanisms such as calendar ageing, load dependent capacity fading, and chemical degradation, etc., can be gained and ultimately rational improvements suggested.[1] In all, this will reduce overall LIB cost and environmental impact, as well as improve device energy efficiency and uptime.

Carbonaceous anode materials and electrolytes are well suited to be characterised by Raman spectroscopy, *e.g.* Li-intercalation can be followed as changes in the shape and energy of the D- and G-bands of the host, allowing us to not only monitor the amount of inserted lithium, but also structural changes including degradation.[1] Many of these phenomena are highly temperature dependent. Here we use the difference between the Stokes and anti-Stokes Raman spectra and the Boltzmann distribution to specifically monitor any non-uniform heating occurring within a small sampling volume (Figure 1). By applying *operando* Raman spectroscopy we this way can follow variations in local concentrations and interactions created by the electrochemical cell operation, and specifically the role of the local temperature for the processes.[2]

This work was funded by "Batterifondsprogrammet" of the Swedish Energy Agency.

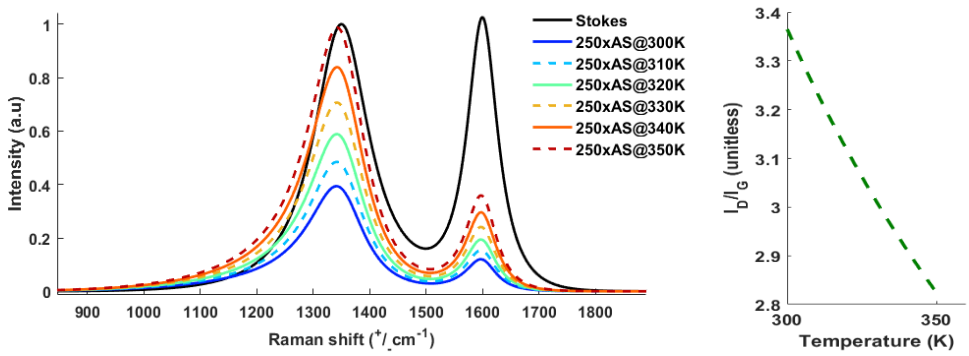

Figure 1. Stokes spectrum and simulated anti-Stokes spectra from a Boltzmann distribution at various temperatures and the ratio of the intensity of the anti-Stokes D- and G-bands (I_D/I_G) as function of temperature.

References:
[1] J. Wallenstein, P. Svens, J. Groot and P. Johansson, *"Raman signatures of aged electrolytes from fast-charged LIBs"*, in manuscript.
[2] J. Wallenstein, P. Johansson, *"Determining transient local temperature distribution in Li-ion batteries by* operando *Stokes/anti-Stokes Raman spectroscopy"*, in manuscript.