SPECTROSCOPY OF ATMOSPHERIC GASES IN EXTREME CONDITIONS BY INELASTIC X-RAY SCATTERING

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Non-resonant inelastic x-ray scattering (NRIXS) by electronic excitations is a versatile technique for studying the electronic structure of matter. In NRIXS a hard x-ray photon scatters off an electron system, transferring energy and momentum to it. The energy and momentum transfers can be varied independently which allows the study of various single and many electron excitations and their momentum dependence.

Historically NRIXS has been mostly applied to solids and liquids, since the combination of small interaction cross section and low density had rendered experiments on gaseous samples difficult. This however is changing due to developements in synchrotron storage rings, x-ray optics and spectrometers as demonstrated by recent experiments on gaseous N_2 and He [1, 2]. In this work we extend these studies to extreme conditions, namely high temperature, opening new possibilities for *in situ* spectroscopy. We have performed NRIXS experiments on gaseous N_2 and CO_2 at room temperature and $\sim$900 K. Study of gas phase chemical reactions at high temperatures is of obvious relevance to e.g. atmospheric science and various branches of chemistry. Furthermore N_2 and CO_2 are common model systems for di- and triatomic molecules and suitable testing ground for theoretical and computational studies of molecular properties.

The experiments were perfomed at the European Synchrotron Radiation Facility (Grenoble, France). The gases were pumped to a quartz capillary to pressures of $\sim$40 bar. Samples were heated with a gas blower. Our measurements for CO_2 at large energy losses, corresponding to C and O K-edges, show clear temperature effects due to increasing population of molecules in vibrationally excited states. The spectral evolution with temperature is reproduced with density functional theory (DFT) calculations and Franck-Condon approximation. Valence IXS spectra recorded at energy losses of 1-50 eV show less pronounced temperature effects. Time dependent DFT calculations within the framework of Ref. [3] have been performed to interpret the valence spectra.

[1] J. A. Bradley et al., Physical Review Letters **105** (2010) 053202.
[2] B. P. Xie et al., Physical Review A **82** (2010) 032501.
[3] A. Sakko et al., Journal of Chemical Physics **133** (2010) 174111