PTB

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EUMETRISPEC Workshop

on “Traceability of Spectral Reference Line Data”

9 – 10 October 2014

EURAMET

EUMETRISPEC

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6	High accuracy intensity calculations of H_2O, CO and CO_2 *Oleg Polyanski, University Colleg of London*

Abstracts

Speed-dependent effects and Dicke narrowing in spectral line shapes

Roman Ciuryło[1]

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The recent development of the Doppler limited spectroscopy as well as theory of spectral line shapes clearly demonstrated limitation of Voigt profile commonly use in the experimental data analysis. It is now well established that the analysis of accurate spectra requires to go beyond the standard Voigt profile. Beside the Doppler and collisional broadening and shifting, the other effects such as: the speed dependence of collisional broadening and shifting, the Dicke narrowing, the correlation between velocity-changing and phase or state changing collisions, and finally the dispersions line asymmetry should be taken under consideration. To properly describe spectral line shapes the analytical models as well as *ab initio* approaches were developed. Properties of analytical line shape models were analysed and confronted with *ab initio* approaches to line shape calculations.

Precision laser spectroscopy of water isotopologues in the near-IR

Livio Gianfrani

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I will make an overview of precision measurements that are being performed at the Second University of Naples on the vibration-rotation spectrum of water isotopologues, by using a pair of phase-locked extended cavity diode lasers at 1.39 μm. In particular, the observation of spectral line profiles with unprecedented fidelity is exploited to retrieve a number of physical quantities and spectroscopic parameters of interest for a variety of reasons in different research fields. In particular, I will mention the spectroscopic determination of the thermodynamic temperature and the accurate measurement of line centre frequencies and intensity factors.

“SI-traceable line parameters of greenhouse gases measured using cavity ring-down spectroscopy”

Dr. Joseph T. Hodges

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I will present cavity ring-down spectroscopy (CRDS) gas concentration measurements for air- and self-broadened samples of water vapor, carbon dioxide, methane, and oxygen. This laser technique gives an SI-traceable measure of sample absorption coefficient which can be expressed in terms of time (ring-down time constant) and frequency (laser detuning about line center) observables. Typically absolute line intensities are obtained by integrating CRDS absorption spectra acquired on gravimetrically prepared samples of known molar fraction. Once known, these line intensities, which constitute an intrinsic and conserved property of the molecule, serve as the molecular ruler by which measurements of integrated sample absorption coefficient yield analyte concentration in an unknown sample. Realization of this spectroscopic technique also involves the application of advanced line shape models, which are used to accurately determine the integrated spectrum area from discretely sampled spectra. Finally, I will also discuss an alternative to gravimetrically prepared samples which involves quantum-chemistry calculations of line intensities. Recent experiments show that these *ab initio* calculations have the potential to yield line intensities with relative standard uncertainties as low as 0.3 %.

GEISA-2014 spectroscopic database: context, contents, quality requirements, evolution

Nicole Jacquinet, Chérif Boutammine, Laurent Crépeau, Anis Boudahoui, Raymond Armante, Cyril Crevoisier, Virginie Capelle, Noëlle Scott, Alain Chédin

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The role of molecular spectroscopy in modern atmospheric research has entered a new promising perspectives phase for remote sensing applications (meteorology, climatology, chemistry) with the advent of highly sophisticated spectroscopic instruments. With the launch of high spectral resolution instruments like AIRS on board EOS-Aqua (2002), IASI on Metop-A (2006), and now on Metop-B (2012), GOSAT (2009) and Suomi NPP (2011) spectroscopic data appear to be at the root of the investigation of climate change providing an improved understanding of the different phenomena driving the atmospheric system.

Since 1974 - celebrating its 40th birthday (*) this year! - the LMD GEISA database (Gestion et Etude des Informations Spectroscopiques Atmosphériques: Management and Study of Atmospheric Spectroscopic Information; Jacquinet-Husson et al, JQSRT 112 (2011) has been at the heart of state-of-the-art developments in *spectroscopy* and *radiative transfer modeling* to meet the needs of the international *space agencies*, by collecting, archiving and distributing all the necessary inputs for atmospheric radiative transfer models. GEISA is constantly evolving, taking into account the best available spectroscopic data. It comprises not only the line-by-line parameters database in the spectral range from 10^{-6} to 35,877.031 cm^{-1}, but also two additional sub-databases: infrared and ultraviolet absorption cross-sections, microphysical and optical properties of atmospheric aerosols.

GEISA, together with the LMD radiative transfer model 4A(**) in its operational version 4A/OP, co-developed by LMD and **Noveltis** (***), have become CNES references for MetOp-A and MetOp-B and, as such, are involved, at CNES, Noveltis and LMD, in the spectral calibration activities of the IASI instruments -currently recognized as a radiometric reference by the Global Space-based Inter-Calibration System (GSICS).

In the frame of the 2014 GEISA release and the work in progress in the CNES-MENINGE scientific group for the future of the IASI instruments (IASI-NG), we will present the current content and planned evolution of each of the three sub-databases specifically emphasizing the quality requirements for spectroscopic parameters, i.e.: -high and very high spectral resolution laboratory measurements,-absorption cross sections of atmospheric trace constituents (in particular for gases with greenhouse gas potential), -atmospheric aerosol precursors, -theoretical spectroscopy (non LTE conditions), line shape models and assessment of associated line parameters, -feedback from Metrology groups and from groups developing remote sensing instrumentation on latest needs to improve the precision/accuracy of spectroscopic data.

GEISA is implemented on the CNES/CNRS/IPSL “Ether” Products and Services Centre WEB site: http://www.pole-ether.fr, where all the archived spectroscopic data and related information can be handled through user friendly associated management software facilities. It is used on-line by more than 300 laboratories working in various domains like atmospheric physics, planetology, astronomy, astrophysics.

(*) : http://www.lmd.jussieu.fr/geisa2014/
(**) : http://ara.abct.lmd.polytechnique.fr/index.php?page=4a
(***) : http://4aop.noveltis.com/

Accurate and Precise FT Spectroscopy and the Complete Eigenenergy List of HCN

Georg Mellau[1,2]

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[2]Department of Chemistry, MIT, Cambridge, MA, USA.

Due to its accurate and constant ILS the FT spectrometry has a relative wavenumber precision of better than 10^{-10} (1). To retrieve line positions with the same accuracy one has to take into account a series of possible pitfalls regarding the FT instrument and the data analysis of the recorded spectra. I will point out some of the problems and present the solutions to them.
For polyatomic molecules with dense molecular spectra the limiting factor of the accuracy and precision of the measured line positions and intensities is the overlap of the studied lines with weak hotbands, overtones and isotopolgue bands. The solution for this problem is the compilation of the experimental "*complete molecular eigenenergies*" (2,3). Beside the fundamental importance to quantum mechanics (4) this concept can be used to improve the accuracy and precision of line positions and intensities retrieved from dense molecular spectra. To accomplish this task the high precision experimental eigenenergy data must be combined (5) with the at least 1000 times less precise ab initio eigenenergy lists to get access to the relatively accurate and precise ab initio line intensity information between all molecular states.

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"High accuracy *Ab intio* line intensities calculations of small molecules"

Oleg L. Polyansky,[1;2]
[1]Department of Physics and Astonomy, University College of London, London WC1E 6BT, United Kingdom, 2Russian Academy of Sciences, Nizhny Novgorod, Russia
(Dated: October 1, 2014)

The high accuracy calculation of line intensities of small molecules from the _rst principles is presented. The comparison of the calculated line intensities with the experimental measurements of subpercent accuracy is given. In particular the ab initio calculations for H^2O, CO_2, CO and NH3 potential energy surfaces (PES) and equivalent dipole moment surfaces (DMS) are presented, as well as the use of these PES and DMS for the line centers and line intensities in variational calculations.
The high accuracy of the *ab initio* intensity calculations was achieved based on 12 components of quantum chemical calculations, presented in the paper [1]. All electrons multi reference con_guration interaction (MRCI) method is used with complete basis set (CBS) extrapolation of Aug-cc-pc6z to produce the Born-Oppenheimer (BO) PES and DMS. The optimisation of complete active space (CAS) is also used.
Non-BO relativistic, quantum electrodynamic (QED) and adiabatic as well as non-adiabatic corrections are also necessary in order to achieve subpercent accuracy of intensities. The comparison of the results of the calculations with the published [2], [3], [4] and not yet published intensity [5] measurements are presented.

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Poster Session

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