

# Molecular dynamics simulations of the collision-induced absorption coefficient in a CH<sub>4</sub> and N<sub>2</sub> mixture

## SCIENTIFIC REPORT

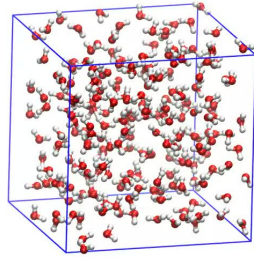
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### Subject: Short Term Scientific Mission

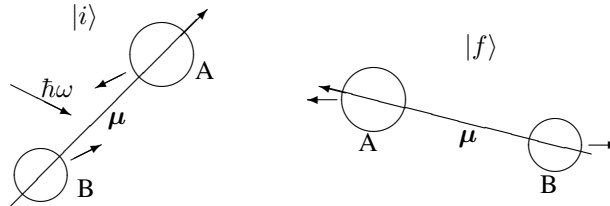
Reference: COST Action CM1405

Host institution: Jean-Michel Hartmann - Laboratoire Inter-universitaire des Systèmes Atmosphériques

Period: 21/03/2016 to 25/03/2016



(a) Colliding particles in an imaginary box



(b) Photon absorbed by two colliding particles

The goal of this STSM was to understand and master the numerical method used by Dr. Jean-Michel Hartmann and Dr. Ha Tran in order to compute absorption coefficients in a CH<sub>4</sub> – N<sub>2</sub> mixture. We recall that the use of quantum Physics method is forbiddingly CPU-intensive and therefore brings us to a classical approach.

In addition to this useful work, our interaction led to a very interesting idea that I will elaborate on below.

First, I have focused on the method itself by studying the molecular dynamics simulation code used to provide the CO<sub>2</sub> spectra in [1]. Accompanied by Dr. Jean-Michel Hartmann, I reviewed his code step by step in order to well understand each stage of the process. The principle of this method is to compute the positions of each particle in an imaginary box at each time step. At this level, one can easily get the induced dipole function value for each pair of particles at any time (as long as we have a function of the dipole depending on the distance between the particles). After computing the auto-correlation function of the dipole  $\phi(t)$ , one can obtain the absorption coefficient by,

$$\alpha(\omega) = A \frac{4\pi^2}{3\hbar c} e^{\hbar\omega/2k_B T} \omega [1 - e^{-\hbar\omega/k_B T}] \frac{1}{V} C(\omega) \quad C(\omega) = \frac{1}{\pi} \int_0^{+\infty} \cos(\omega t) \phi(t) dt \quad (1)$$

In order to compute the positions and therefore the motions of the particles, one needs to have a function of the potential describing the interaction between the atoms and/or molecules.

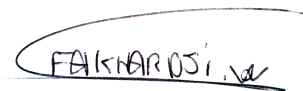
A variety of time efficient techniques are used in the code and a big part of my work was to well understand these manipulations which are the key for getting a reasonable computing time.

During the second part of my stay, with Dr. Ha Tran, I have seen the adaptation of this code to the CH<sub>4</sub> – N<sub>2</sub> system. Here this method is exactly the same except for the geometry which goes from a linear system to more complex shape. Such a system is more conveniently described using quaternion instead of the Euler angles. A potential is already implemented in the code and then allows to compute the positions of the molecules along the time. From a molecular dynamics point of view, the code is fully functional. However in order to relate the dynamics of the particles to the absorption coefficient, one needs to describe the dipole existing between CH<sub>4</sub> and N<sub>2</sub>.

For the future, we plan to do a benchmark comparison of quantum and classical molecular dynamics calculations of collision-induced absorption. Our idea is to study an atomic system, such as Ar – Xe for which full quantum treatment is feasible. By using the same input parameters (potential and dipole functions) for the two codes (quantum and classical) we will be able to estimate the accuracy of the classical approach. This study can be made with different pressure and temperature conditions to see the limitation of the technique.

### Reference

- [1] Molecular dynamics simulations for CO<sub>2</sub> spectra. II. The far infrared collision-induced absorption band, J-M Hartmann, C. Boulet, D. Jacquemart, The Journal of Chemical Physics **134**, 094316 (2011)



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