

# MOLECULES PROBED WITH A SLOW CHIRPED-PULSE EXCITATION: ANALYTICAL MODEL OF THE FREE-INDUCTION-DECAY SIGNAL

GUILLAUME DHONT, DANIELE FONTANARI, CÉDRIC BRAY, GAËL MOURET, ARNAUD CUISSET, FRANCIS HINDLE, ROBIN BOCQUET, *Laboratoire de Physico-Chimie de l'Atmosphère, Université du Littoral Côte d'Opale, Dunkerque, France*; KEVIN HICKSON, *Institut des Sciences Moléculaires, Université de Bordeaux, Talence, France*.

A chirped pulse experiment is a powerful means to rapidly obtain an high-resolution spectrum of molecules on a large frequency band. The theoretical paper from McGurk *et al.*<sup>a</sup> is the main reference paper to describe the polarization induced by fast chirped pulses generated with microwave sources.

We built a chirped pulse spectrometer operating at 200 GHz for astrophysical applications.<sup>b</sup> It works in the millimeter domain with slower chirped pulses. In such a situation, the paper of McGurk *et al.* does not capture all the physics involved in the polarization step. In particular, the intensity of a molecular transition is dependent on its temporal position inside the chirped pulse, as discovered by Abeysekera *et al.*<sup>c</sup>

A theoretical study of the polarization of molecules subjected to a slow chirped pulse is presented for three typical cases: the cell, the uniform flow and the molecular beam. Analytical expressions are proposed alongside the numerical solution and are used in the expression of the free induction decay signal. We test the analytical expression on the rotational emission spectra of OCS molecules. In the thermalized case, a relation between the pulse duration, the line position in the chirped pulse, and the signal amplitude is proposed to correct the line intensities.<sup>d</sup>

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<sup>a</sup>J. C. McGurk, T. G. Schmalz, and W. H. Flygare, J. Chem. Phys. **60**, 4181 (1974).

<sup>b</sup>F. Hindle, C. Bray, K. Hickson, D. Fontanari, M. Mouelhi, A. Cuisset, G. Mouret and R. Bocquet, J. Infrared Millim. Te. **39**, 105 (2018).

<sup>c</sup>C. Abeysekera, L. N. Zack, G. B. Park, B. Joalland, J. M. Oldham, K. Prozument, N. M. Ariyasingha, I. R. Sims, R. W. Field, and A. G. Suits, J. Chem. Phys. **141**, 214203 (2014).

<sup>d</sup>D. Fontanari, C. Bray, G. Dhont, G. Mouret, A. Cuisset, F. Hindle, R. Bocquet, and K. M. Hickson, Phys. Rev. A **100**, 043407 (2019).