Experimental state-to-state rate coefficients of gas-phase inelastic molecular collisions

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Reliable data on state-to-state rate coefficients are essential for accurately modelling rarefied gases. In fact, the necessity of this information has been claimed in order to develop chemical models of gas phase environments, such as the interstellar medium or the planetary atmospheres. Thus, from the over 300 of molecules already observed in the interstellar medium, the study of rotational collisional energy transfer processes has been only theoretically calculated for over 60 species. In the case of the experimental studies this amount is dramatically lower, counting only a few of specific studies, such as ammonia, because they are quite limited and remained sometimes unaffordable.

Thus, one example is the experimental method proposed to measure state-to-state rate coefficients of rotational inelastic ammonia collisions [1]. This method is based on pump-probe experiments using Fourier transform rotational spectroscopy techniques. They used the inversion doublets of the ammonia molecule to pump the system and so alter the thermal equilibrium population of these levels, and then probe how collisional relaxation processes affect to different transitions by means of chirp-pulses of the correlated transitions. After changing the timing between the pump and probe pulses, the state-to-state rate coefficients were determined, and thanks to the chirped pulse technique the propensity rules associated to these collisional processes were also characterized. Nevertheless, the high specificity of the method means that only a few of laboratories around the world can use it.

Here we present the first steps in the development of a pump-probe procedure for general molecular systems, based on the previous mentioned one, to obtain experimental state-to-state rate coefficients for inelastic rotational energy transfer of molecules of astrochemical interest by the use of the rotational spectroscopy setup in Rennes. This procedure aims to unravel the propensity rules for inelastic energy transfer of polyatomic molecules such as acrylonitrile and propose experimental measurements of state-to-state collisional rate coefficients in order to do comparison with theory. The first results obtained from acrylonitrile and methyl cyanide collisional relaxations with Argon will be also displayed.

[1] C. P. Endres, P. Caselli, S. Schlemmer, *J. Phys. Chem. Letts.*, **10**, p. 4836-4841 (2019).

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