Nonlinear properties and collisional spectra in hydrogen-(heavy) noble-gas-atom mixtures

Titre	Nonlinear properties and collisional spectra in hydrogen-(heavy) noble-gas-atom mixtures
Type de publication	Communication
Type	Communication avec actes dans un congrès
Année	2012
Langue	Anglais
Date du colloque	2012
Titre du colloque	XXI International Conference on Spectral Line Shapes (ICSLS 2012)
Volume	Journal of Physics: Conference Series 397
Pagination	012044
Auteur	Głaz, Waldemar [1], Bancewicz, Tadeusz [2], Maroulis, George [3], Haskopoulos, Anastasios [4], Godet, Jean-Luc [5]
Pays	Royaume-Uni
Editeur	IOP Publishing Ltd
Ville	Bristol
Mots-clés	collision induced nonlinear hyper-Rayleigh scattering [6]

Résumé en anglais	In a series of previously published works the phenomenon of collision induced nonlinear hyper-Rayleigh scattering of light (CIHRS) was studied on the basis of the numerical quantum chemistry (QC) methods confronted with a theoretical approach. A class of super-molecular systems composed of hydrogen (H 2) and lighter inert gas atoms (Rg) was considered. In this report a development of this research work is presented with more massive and highly polarizable, Kr and Xe, perturbers involved. The collision-induced hyperpolarizability tensorial values, Δβ obtained by means of the QC ab initio methods are applied in order to produce the CIHRS spectral distributions; the influence of Δβ features on the line shapes is compared with the earlier theoretical predictions. Namely, the validity of the multipole-induced-multipole (MIM) mechanism is assessed. In particular, its relevance to reproduce long-range functional behavior of the so-called symmetry adapted (SA) components of the hyperpolarizability tensor Δβ(R) is discussed. A thorough analysis of the translational CIHRS spectra is then performed to identify the role of the hyperpolarizability spatial distribution in forming particular sections of the line shapes. An extension of the dipole-induced-quadrupole analytical model is suggested and tested with regard to its ability to reproduce more accurate profiles.
URL de la notice	http://okina.univ-angers.fr/publications/ua8060 [7]
DOI	10.1088/1742-6596/397/1/012044 [8]
Lien vers le document en ligne	http://iopscience.iop.org/1742-6596/397/1/012044?fromSearchPage=true [9]

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