Proceedings Paper:

Houver, S, Lebreton, A, Pereira, TAS et al. (10 more authors) (2020) Giant optical nonlinearity interferences in Terahertz quantum structures. In: 2020 45th International Conference on Infrared, Millimeter, and Terahertz Waves (IRMMW-THz). 45th International Conference on Infrared, Millimeter and Terahertz Waves (IRMMW-THz), 08-13 Nov 2020, Buffalo, NY, USA. IEEE , p. 1. ISBN 978-1-7281-6620-9

https://doi.org/10.1109/irmmw-thz46771.2020.9370368

Giant optical nonlinearity interferences in Terahertz quantum structures

S. Houver[1], A. Lebreton[1], T. A. S. Pereira[2], G. Xu[3], R. Colombelli[3], I. Kundu[4], L. H. Li[4], E. H. Linfield[4], A. G. Davies[4], J. Mangeney[1], J. Tignon[1], R. Ferreira[1], S. S. Dhillon[1]

[1]Laboratoire de Physique de l'Ecole normale supérieure, ENS, Université PSL, CNRS, Sorbonne Université, Université de Paris, F-75005 Paris, France

[2]Instituto de Física, Universidade Federal de Mato Grosso, 78060-900 Cuiabá, Mato Grosso, Brazil

[3]Institut d'Electronique Fondamentale, Université Paris-Sud, CNRS UMR 8622, 91405 Orsay, France

[4] 4School of Electronic and Electrical Engineering, University of Leeds, Leeds LS2 9JT, UK.

***Abstract*— Second-order optical nonlinearities can be greatly enhanced by orders of magnitude in resonantly excited nanostructures. However, they are frequently not as heightened as predicted, limiting their exploitation in nanostructured THz nonlinear optics. Here, we show that the second-order nonlinear susceptibility can vary by orders of magnitude as a result of giant destructive interference effects. Using THz quantum-cascade-lasers as a model source to investigate interband and intersubband nonlinearities, we show that these giant interferences are a result of an interplay of the second-order nonlinear contributions of multiple light and heavy hole states. As well as of importance to engineer the resonant optical properties of nanostructures, this framework can be employed as a novel, sensitive tool to elucidate the bandstructure properties of complex materials.**

INTRODUCTION

NONLINEAR frequency generation is a ubiquitous technique with applications ranging from frequency conversion to quantum optics, and depends critically on the magnitude of the nonlinear susceptibility. Strongly enhanced second-order optical nonlinearities, that are orders of magnitude larger than in bulk, have been theoretically investigated since the early 90's and experimentally demonstrated in various semiconductor nanostructures under resonant interband or intersubband excitation (for example [1,2]). These resonant nonlinearities are of perpetual interest in nonlinear optics, especially whenever new materials are discovered [3]. However, resonant nonlinear interactions involving transitions between semiconductor bands tend to be less efficient than predicted, limiting their applications in nonlinear physics, as well as for frequency conversion in atomically-thin materials.

In this paper, we theoretically and experimentally show the strong and unexpected interplay of resonant nonlinearities that occur when multiple intersubband and interband transitions are combined through excitation by terahertz (THz) frequency (photons of energy ETHz ~ 10 meV) and near infrared (NIR, photons of energy ENIR ~ 1.5 eV) pumps, respectively. The second-order nonlinearity $\chi^{(2)}$ is thus doubly enhanced, permitting efficient sideband generation on an optical carrier (Sum frequency generation (SFG) $E_{NIR} + E_{QCL}$) [2]. Recently it has been shown that quantum cascade lasers (QCLs) can be used as both the source for THz radiation (and intersubband excitation) and the nonlinear medium with an external NIR excitation coupled into the QCL cavity [4,5]. This renders the emission of QCLs ideal to probe low energy excitations. Here, we exploit this nonlinear response within the QCL and demonstrate that the second-order susceptibility, in a complex quantum well nanostructure containing many conduction and valence quantum confined states (figure 1), presents giant variations with the excitation energy i.e. cancellations and enhancements of the susceptibility. We highlight for the first time [6] an order of magnitude reduction (figure 2) of the second order nonlinear susceptibility at specific frequencies, particularly important when studying THz (i.e. low energy) transitions. Our theoretical model shows that these effects result from pronounced susceptibility interferences between the numerous nonlinear contributions from the many light (LH) and heavy (HH) hole states. We also show that the nonlinear conversion can be used to probe the complex QCL bandstructure to attain information on the wavefunction alignment.

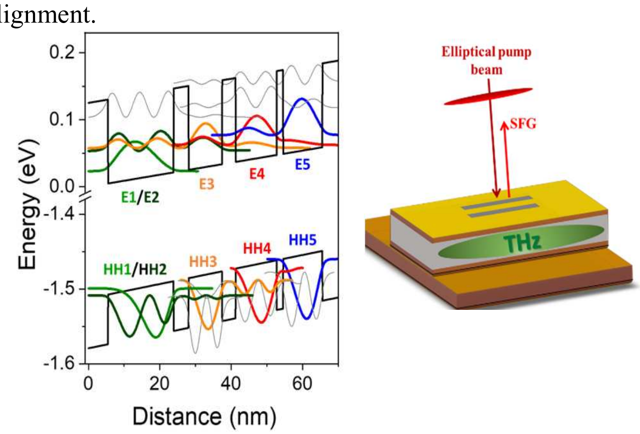

Fig 1. Left: QCL bandstructure showing the large number of states involved (heavy holes shown). Right. Geometry for short interaction distance.

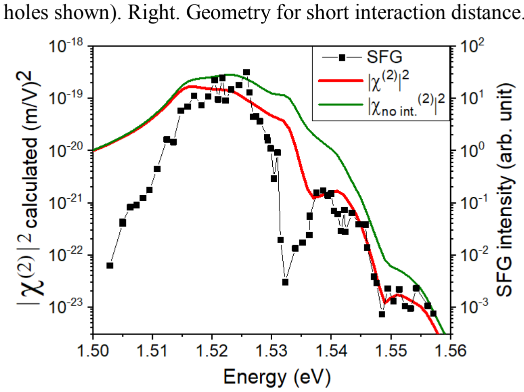

Fig 2. SFG intensity from experimental data (black squares) and theoretical model. For the latter modulus squared |χ(2)| with (red) and without (green line) quantum state interference.

REFERENCES

[1] E. Rosencher et al, Science 271, 168-173 (1996).
[2] J. Kono et al, Phys Rev Lett 79, 1758-1761 (1997)
[3] C. Poellmann et al, Nat Mater 14, 889-+ (2015)
[4] J. Madeo et al, Nat Photonics 6, 519-524 (2012).
[5] S. Houver et al ACS Photonics 5, 890-896 (2018).
[6] S. Houver et *al.* Science Advances, 5 (10), eaaw7554 (2019)