Theoretical Study of Optical Responses on Hydrogenated Graphene Structures

by

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to earn the degree of Doctor of Philosophy.

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"If you are receptive and humble, mathematics will lead you by the hand."

Paul Dirac

I dedicate this work to my wife, Paty, and my two children, Samantha and Reinaldo.

Abstract

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Acknowledgements

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1 Introduction

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1.1 Nonlinear Optics

Nonlinear optics studies the modifications of the optical properties of a material system as a result of its interaction with the electric field of light. Typically, only laser light is intense enough to produce a modification in the optical properties of a material system[1]. The nonlinearity of those phenomena came from the fact that their manifestation do not depend in a linear manner on the strength of the optical applied field.

In 1917, Albert Einstein published the theoretical possibility of amplification of radiation through stimulated emission with coherence [1] and Valentin A. Fabrikat made the calculation of the conditions needed to produce radiation amplification by stimulated emission like the population inversion; he also proposed the resonant cavity [2]. This theory and calculations was not carried out to practice until 1952 when Joseph Weber started the development of masers but the first one was demonstrated by Charles H. Townes and his working team in 1953. The principal difficulty in the development of an *optical maser*, was the transition from centimeter to nanometer waves. In 1960 Theodore H. Maiman presented the first functional ruby laser [3]. One year later, second-harmonic generation (SHG) was the the first nonlinear effect observed by Peter Franken, et al. at University of Michigan [4]. Exciting a piece of crystalline quartz with a laser beam, they produced a second beam with the double of frequency than the original but with less intensity than the original one. In 1962, Nicolaas Bloembergen et al. described the induced nonlinear electric dipole and higher moments in an atomic system using quantum-mechanical perturbation theory [5], and solved the Maxwell's equations in nonlinear dielectrics satisfying the boundary conditions at a plane interface between a linear and nonlinear medium [6]. The development of the first pulsed laser in 1964 by William

[1] Some nonlinear responses, like luminescence of dye molecules, were discovered previous to the development of the laser

B. Bridges [7], and the later works in this field, allowed the increase of laser intensities and then the induction of new nonlinear effects in nonlinear media. In 1988, Yuen-Ron Shen et al. studied the the bulk contribution to surface second-harmonic generation and presented a model for the bulk nonlinearity of molecular systems including the quadrupole bulk contribution [8]. Currently, chirped pulse amplification, developed by Gerard Morou and Donna Strickland [9], in Ti:Saphire lasers became the standard for high energy, ultrashort pulsed lasers.

1.1.1 Nonlinear polarization

To describe the optical nonlinearity, consider how the polarization of a material $P(t)$, defined as the dipole moment per unit volume, depends on the strength of an applied optical field $E(t)$. In the case of linear optics, the polarization dependence is lineal with respect to the strength of electric field and can be described as

$$P(t) = \epsilon_0 \chi^{(1)} E(t), \tag{1.1}$$

where ϵ_0 is the permittivity of free space and $\chi^{(1)}$ is the linear susceptibility. In nonlinear optics, the optical response of Eq. (1.1) is characterized by rewriting the polarization as a power series in the field strength as

$$\begin{aligned} P =& \epsilon_0[\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \cdots] \\ =& P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + \cdots . \end{aligned} \tag{1.2}$$

The $\chi^{(n)}$ expression is known as the n^{th} nonlinear optical susceptibility; the common linear effects presented in most media are described only by $P^{(1)}(t)$ that reduces the expression (1.5) to (1.1). Considering that the electric field E written here as a scalar must be considered a vector, then $\chi^{(1)}$ must be a second-rank tensor, $\chi^{(2)}$ a third-rank tensor and so on, and they depend on the frequencies of the applied fields [10].

The nonlinear polarization can also be described threating the incident electric field as a superposition of plane waves. If we assume that the electric field is written as [11]

$$\mathbf{E}(\mathbf{r}, t) = \sum_n \mathbf{E}_n(\mathbf{r}, t) \tag{1.3}$$

where

$$\mathbf{E}_n(\mathbf{r}, t) = \mathbf{E}_n(\mathbf{r}) e^{-i\omega_n t} + c.c., \tag{1.4}$$

and the, from Eq. (1.5) we can write

$$\mathbf{P}(\mathbf{r}, t) = \sum_n \mathbf{P}(\omega_n) e^{-i\omega_n t}. \tag{1.5}$$

Then, we can define the second order nonlinear polarization, that describes the second order effects in which we have interest, in terms of the second order susceptibility as

$$P^a(\omega_n + \omega_m) = \epsilon_0 \sum_{\text{bc}} \sum_{nm} \chi^{(2),\text{abc}} \left(-(\omega_n + \omega_m); \omega_n + \omega_m\right) E^{\text{b}}(\omega_n) E^{\text{c}}(\omega_m) \tag{1.6}$$

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