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Coherent magneto-optical polarisation dynamics in a single chiral carbon nanotube

Gabriela Slavcheva [a,*], Philippe Roussignol [b]

[a] Blackett Laboratory, Imperial College London, Prince Consort Road, London SW7 2AZ, UK

[b] Laboratoire Pierre Aigrain, Ecole Normale Supérieure, 24 rue Lhomond, 75231 Paris Cedex 5, France

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ABSTRACT

We propose a theoretical framework and a dynamical model for the description of the natural optical activity and the Faraday rotation in an individual chiral single-walled carbon nanotube in the highly nonlinear coherent regime. The model is based on a discrete-level representation of the optically active states near the band edge. Chirality is modelled by a system Hamiltonian corresponding to energy-level configurations, specific for each handedness, that are mirror reflections of each other. An axial magnetic field is introduced through the Aharonov–Bohm and Zeeman energy-level shifts.The time evolution of the quantum system following an ultrafast circularly polarised optical excitation is studied using the coherent vector Maxwell pseudospin equations. Giant natural and magneto-optical gyrotropy, exceeding the one of the artificial photonic metamaterials, is numerically demonstrated for a single (5, 4) carbon nanotube and an estimate of the magnitude of the natural and magneto-chiral circular dichroism and specific optical rotatory power is obtained. The model provides a framework for the investigation of chirality and magnetic field dependence of the ultrafast nonlinear optical response of a single carbon nanotube.

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1. Introduction

Chirality is one of the main symmetries of the carbon nanotube geometry which determines the optical properties of single-walled carbon nanotubes (SWCNTs). The interaction of polarised light with chiral materials in the absence of magnetic fields gives rise to the phenomenon of natural optical

* Corresponding author. Tel.: +44 20 759 47623; fax: +44 20 759 42007.
E-mail address: g.slavcheva@imperial.ac.uk (G. Slavcheva).

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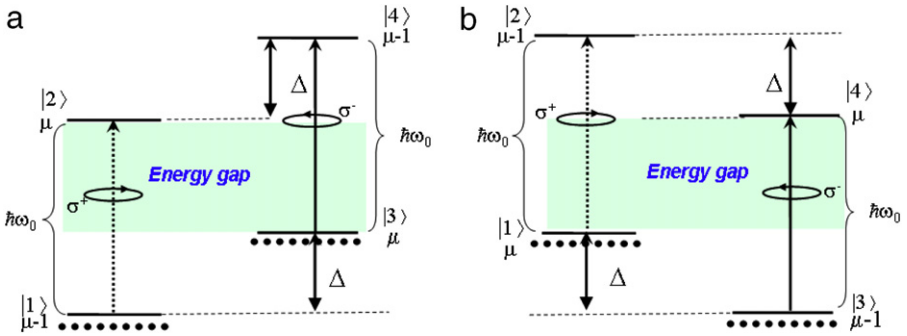

Fig. 1. Energy-level structure at the $K(K')$ point of the lowest subbands labelled by the subband index μ for an (a) AL-handed; (b) AR-handed SWCNT. Δ is the energy separation between the lowest subband and the second lowest subband near the band edge.

(rotation) activity, whereby the polarisation plane is rotated continuously during the light propagation across the medium.

Time-resolved magnetic circular dichroism and magneto-optical rotatory dispersion techniques offer spectroscopic information which is difficult or impossible to obtain by other means. A detailed understanding of the mechanisms underlying the optical and magneto-optical birefringence, circular dichroism and rotation in the nonlinear coherent regime require an adequate theoretical description. On the other hand, formulation of a theory and model of the optical activity in chiral molecules, such as individual single-walled carbon nanotubes (SWCNTs), in the high-intensity nonlinear coherent regime and under an axial magnetic field, is of special interest from a fundamental point of view. To our knowledge, no such theory has been proposed and very little is known about the polarisation dynamics of the nonlinear optical and magneto-optical response of a single carbon nanotube. Exploiting these ultrafast nonlinear effects would open up pathways for the development of a novel class of ultrafast polarisation-sensitive integrated optoelectronic devices, based on single carbon nanotubes.

2. Theoretical background

Carbon nanotubes exist in two (AL) left- ($n > m$) and (AR) right-handed ($n < m$) helical forms (enantiomers) depending on the rotation of two of the three armchair (A) chains of carbon atoms counterclockwise or clockwise when looking against the nanotube z-axis. The electronic band structure [1] is described by the quantization of the wave vector along the tube circumference, resulting in subbands in the valence and conduction band labelled by the quasi-angular momentum quantum number (μ).

We model the single chiral nanotube band-edge structure at the K point of the Brillouin zone by an ensemble of identical four-level systems, corresponding to the dipole optically allowed transitions for AL and AR nanotube enantiomers (Fig. 1). Absorption of right circularly polarised light σ^+ tuned at the resonant frequency ω_0 excites the electronic transitions $\mu - 1 \rightarrow \mu$ in AL-handed SWCNTs and $\mu \rightarrow \mu - 1$ transitions in AR-handed SWCNTs, the absorption of left circularly polarised σ^- light excites $\mu \rightarrow \mu - 1$ transitions in AL-handed and $\mu \rightarrow \mu + 1$ transitions in AR-handed SWCNTs [2] (Fig. 1). When a circularly polarised (in the x–y plane) laser pulse propagates along the z-axis of an AL or AR SWCNT (Fig. 2), only one of the two allowed transitions for a linearly polarised light (along x or y) is excited (see Fig. 1).

2.1. Coherent dynamics of the natural optical activity

We follow the semiclassical formalism developed in [3] and describe the discrete multi-level system coherently driven dynamics by Maxwell's equations in vectorial form for the polarised optical

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