

Non-degenerate optical four-wave mixing in single-walled carbon nanotubes

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Abstract

A theory is presented that describes the generation of radiation at the anti-Stokes frequency $2\omega_1 - \omega_2$ created by two coherent light beams, with a difference in frequency $\omega_1 - \omega_2$, injected into an array of uniformly sized and closely packed semiconducting single-walled carbon nanotubes (SWCNTs), which are oriented along the polarization vector of all involved optical waves. Both the real and imaginary parts of the third-order nonlinear susceptibility $\chi^{(3)}$, responsible for such an optical-frequency mixing, are calculated within a simple model based on the two-band approximation of the electronic structure of SWCNTs. It is shown that one can maximize the nonlinear optical response by tuning independently the frequencies ω_1 and ω_2 in resonance with the lowest interband transitions of the semiconducting SWCNTs. The highest peak value of $|\chi^{(3)}|$ under such a resonance excitation is found to be about 10^{-2} esu, which may be important for nonlinear optical applications of SWCNTs.

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1. Introduction

In the last few years there has been an upsurge of research activity in the area of nonlinear optics of carbon nanotubes, which are believed to be among the most promising materials to impact future nanotechnology (for a review see [1,2]), and now there is extensive literature on the subject [3–57]. Most of the interest has focused on the third-order nonlinear optical (NLO) properties of these materials. The

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NLO coefficient relevant to such properties, the third-order susceptibility $\chi^{(3)}$, is proportional to the fourth power of the dipole transition matrix element (which is of the order of electron charge $-e$ times the nanotube radius R), and is expected to be large, even in nonresonant conditions [3]. Various calculations of $\chi^{(3)}$ for ensembles of parallel-arranged carbon nanotubes [4–10] and of the second-order hyperpolarizability of individual nanotubes of finite length [11–18], have indicated that strong enhancement of the NLO response is quite possible, especially under a resonant excitation. Until recently, experimental efforts have mainly been concentrated on the study of the optical nonlinearity and nonlinear time-resolved spectroscopy of carbon nanotubes imbedded in solids (nanocomposites), as well as of carbon nanotubes solutions and suspensions [22–46]. The values of $\chi^{(3)}$ on the order of 10^{-10} – 10^{-12} esu obtained for such systems under off-resonant conditions by using different methods such as the optical Kerr effect, degenerate four-wave mixing and optical limiting, and simultaneously the fast response time (on the order of 1 ps) observed in time-resolved investigations indicate that the above-mentioned systems are very promising for potential applications in near-infrared signal processing. From those experiments with solutions of carbon nanotubes, the $\chi^{(3)}$ values measured for single-walled carbon nanotubes (SWCNTs) are much larger than those for multi-walled ones, which is in good agreement with the theoretical prediction of an overall suppression of the third-order NLO signal intensity from ensembles of SWCNTs with a broad diameter distribution of the tubes [8,9].

Very recently, several papers came out reporting experimental data on the third-order optical nonlinearities and femtosecond pump–probe spectroscopy of carbon nanotubes fabricated into solid-state forms such as thin films or layers deposited on various substrates (glass, silicon or quartz) [47–56], which are often required in device applications. Near-infrared saturable absorption observed by Tatsuura et al. [49] and Sakakibara et al. [50] in thin film samples of SWCNTs is of particularly great interest because of its possible applications in NLO devices such as all-optical switch in optical telecommunication systems. A very large enhancement of $\chi^{(3)}$ (by two orders of magnitude) over the previously reported $\chi^{(3)}$ values for carbon nanotubes in suspensions and nanocomposites has been observed by the Tatsuura [49] and Lauret [53] groups, and the imaginary part of $\chi^{(3)}$ in their measurements has been estimated as 10^{-7} esu under resonant conditions, which is in satisfactory agreement with the value of $\text{Im}\chi^{(3)}$ calculated for the degenerate four-wave mixing process in Ref. [4]. An appealing feature of SWCNT films explored in Refs. [49,50] is that they exhibit the very useful combinations of optical properties when studied in the infrared region: high NLO response, high linear absorption coefficient, and ultra-fast response time (on the order of 1 ps). The results obtained by Tatsuura et al. [49] also provide strong support to the simple one-electron model used in our previous analysis [3–9] of third-order NLO response of semiconducting SWCNTs, indicating that it contains a fair amount of the essential physics necessary to explain the data and the theoretical results thereof can form a reasonable basis to predict details of the frequency dispersion of the $\chi^{(3)}$ and to give the correct order of its magnitude.

A logical next step in the development of third-order nonlinear optics of SWCNTs with the prospects for their device application in new optical communication technologies is paying attention to investigating various four-frequency processes that may occur when several coherent light beams with, in general, different frequencies are incident on a SWCNT sample. In this paper, we consider non degenerate four-wave mixing (NDFWM), resulting in difference-frequency generation, as a typical process of this kind. The NDFWM is determined by a third-order nonlinear polarization of the form

$$P^{(3)}(\omega_s) = \chi^{(3)}(-\omega_s; \omega_1, \omega_1, -\omega_2) E^2(\omega_1) E^*(\omega_2), \quad (1)$$

where $\omega_s = 2\omega_1 - \omega_2$ is the frequency of a field that is generated as a result of the wave mixing, ω_1 and ω_2 are the frequencies of two incidental coherent light waves with $\omega_1 > \omega_2$, $E(\omega_1)$ and $E(\omega_2)$ are the amplitudes of the electric fields associated with those waves, and $\chi^{(3)}(-\omega_s; \omega_1, \omega_1, -\omega_2)$ is the third-order nonlinear susceptibility, responsible for the optical-frequency mixing. The effect we are concerned with here is closely related to the coherent anti-Stokes Raman scattering process, in which the two ω_1 photons are converted

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