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Tuning the nonlinear response of (6,5)-enriched single-wall carbon nanotubes dispersions

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ABSTRACT

Ultrafast nonlinear optical properties of (6,5)-enriched single-wall carbon nanotubes (SWCNTs) dispersions are investigated using the thermally managed Z-scan technique. As the (6,5) SWCNTs presented a strong resonance in the range of 895–1048 nm, the nonlinear refractive index (n_2) and the absorption coefficients (β) measurements were performed tuning the laser exactly around absorption peak of the (6,5) SWCNTs. It is observed that the nonlinear response is very sensitive to the wavelength and the spectral behavior of n_2 is strongly correlated to the tubes one-photon absorption band, presenting also a peak when the laser photon energy is near the tube resonance energy. This result suggests that a suitable selection of nanotubes types may provide optimized nonlinear optical responses in distinct regions of the electromagnetic spectrum. Analysis of the figures of merit indicated that this material is promising for ultrafast nonlinear optical applications under near infrared excitation.

1. Introduction

After the development of efficient methods of separation of single-wall carbon nanotube (SWCNT) species, new fundamental problems and applications exploring the direct influence of the SWCNTs nature in one specific physical property can now be investigated [1,2]. Among a variety of applications, SWCNTs have emerged as a unique nanostructure which may be responsible for the new generation of nonlinear optical devices. This renders SWCNTs as one of the most suitable candidate for photonic and optoelectronic nanodevices [3].

Several papers have been published exploring the study of SWCNT due to its large third-order nonlinearity and ultrafast electronic response as a consequence of delocalized π-electrons cloud along the tube axis [4–8]. In this line, a comprehensive experimental study on specific SWCNT specie is desirable to delineate the nonlinear optical properties of this particular nanometric system. Especially, aiming its use for particular applications where the electronic and optical properties can be optimized.

SWCNTs are direct-band gap materials, with a gap that depends on the diameter and chirality [9]. Many of the applications that might make use of the electronic properties of these materials require individual (n,m) nanotubes with a given chirality for optimum performance. In fact, an efficient resonant absorption occurs only in SWCNTs whose diameter corresponds to an electronic transition in resonance with the photon energy. For the sake of developing of photonic devices based on SWCNT, it is extremely important to explore its nonlinear optical properties for a specific (n,m) specie.

In the present paper we explore the nonlinear properties for a (6,5)-enriched SWCNT sample, showing that the nonlinear response is very sensitive to the wavelength when the resonance of tube corresponds to the resonance at the one-photon energy excitation.

2. Material and methods

Nanotube dispersion was prepared by adding 100 mg of

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CoMoCAT SG65i, a (6,5)-enriched SWCNT powder, to 100 mL of a 70 mM Sodium Dodecyl Sulfate (SDS) solution. This dispersion was tip sonicated at 40 W for a total of 3 h with a 5 min interval every 30 min. After sonication, the solution was centrifuged at 10000 rpm for 1 h followed by an ultracentrifugation at 40000 rpm for another 1 h to remove bundles and impurities. The resulting dispersion was then used for separation by column chromatography [10,11].

Chromatographic column were made using 10 mL plastic syringes filled with gel Sephacryl S-200 HR and a small amount of a 70 mM SDS solution. After that, 1 mL of the SWCNTs dispersion was added to the column for the separation process. As the affinity of metallic SWCNT with the gel is negligible, its interaction with gel beads is very weak, thus they flow through the column without being stuck on the beads [10,11]. Semiconducting SWCNT on the other hand has a chiral dependent interaction with the gel, being attached to the gel beads. Therefore, metallic and low affinity semiconducting SWCNT run through the column, leaving only the SWCNT that strongly interact with the medium. The (6,5) SWCNT has a strongly interaction with the gel, and as it is present in a large quantity in the sample. Thus, large quantity of this chirality is stuck on the gel beads when the nanotube dispersion runs through the column. Finally, 175 mM solutions of SDS is added to the column to remove the SWCNT stuck on the gel [11]. It is important to mention that using the column chromatography method is possible to produces samples with higher nanotube concentration as compared with other separation methods such as density gradient ultracentrifugation (DGU). Fig. 1(a) shows the optical absorption spectrum of the (6,5)-enriched sample. The spectrum is dominated by the peaks at 985 and 570 nm associated, respectively, with the E_{11} and E_{22} optical transitions of the (6,5) nanotube. Other nanotube species present in the sample are responsible for the small absorption bands observed in the spectrum.

To completely characterize the chirality distribution of the (6,5)-enriched SWCNT dispersion, Raman spectra of the radial breathing modes (RBM) were measured in a wide range of excitation energies (2.54–1.90 eV). Fig. 1(b) presents some of the Raman spectra obtained. From the resonance Raman profiles, and the cross section corrected RBM intensity profiles, it is possible to obtain the relative concentration of the SWCNT species present in the sample, following the procedure described in Ref. [12]. The (n,m) species

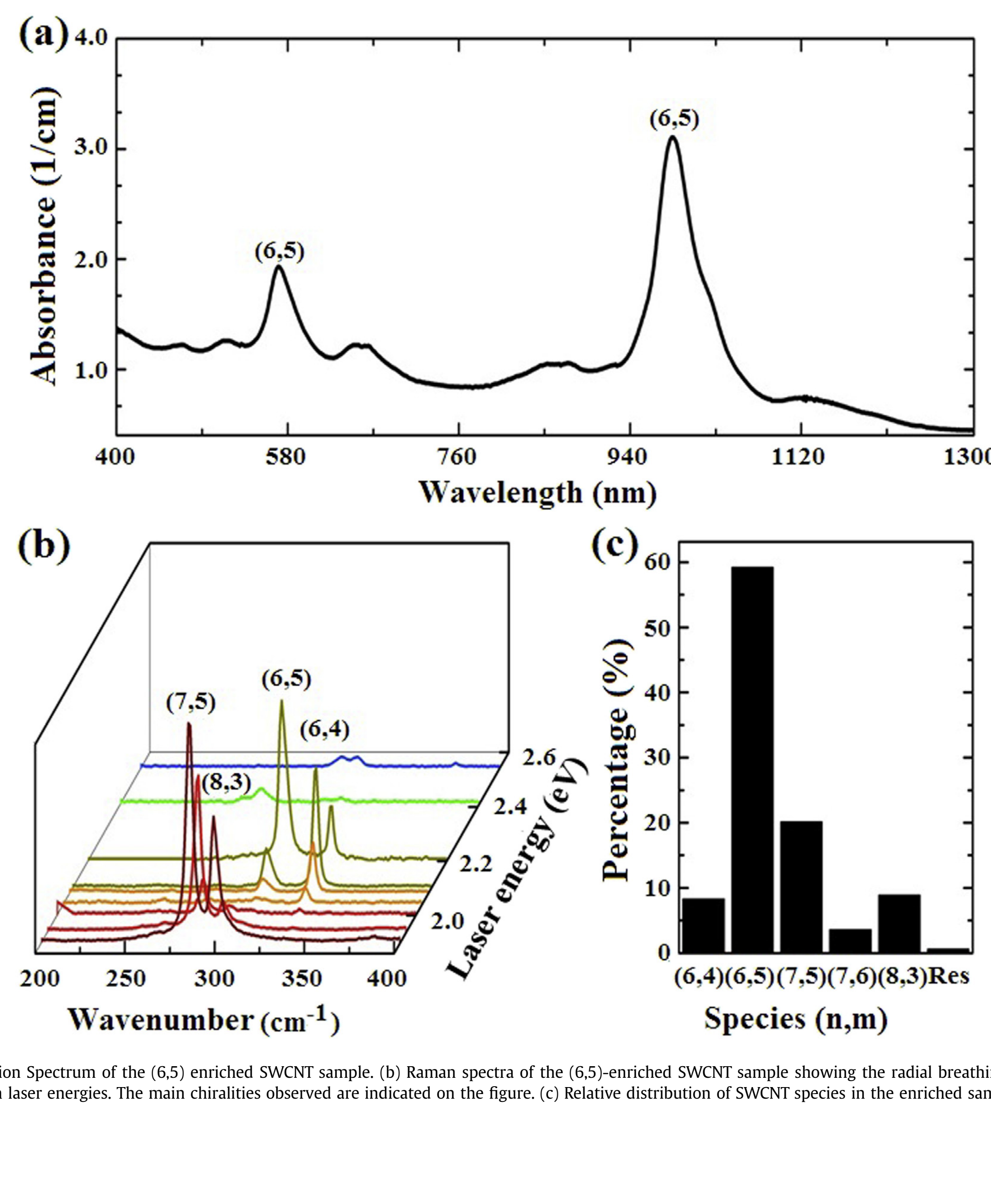

Fig. 1. (a) Absorption Spectrum of the (6,5) enriched SWCNT sample. (b) Raman spectra of the (6,5)-enriched SWCNT sample showing the radial breathing modes recorded at different excitation laser energies. The main chiralities observed are indicated on the figure. (c) Relative distribution of SWCNT species in the enriched sample.