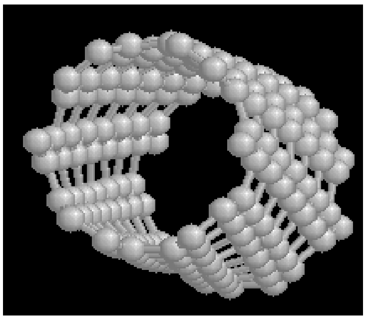
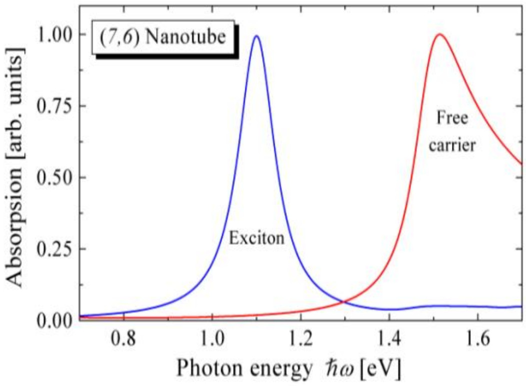
Optical properties of carbon nanotubes

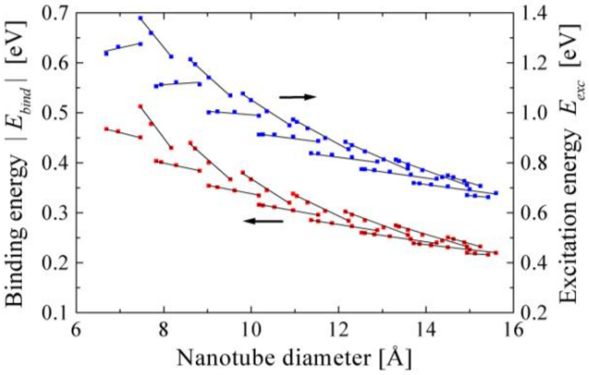


Carbon nanotubes are hollow tubes made entirely from carbon with diameters in the nanometer range such as the one shown in the picture below. Nanotubes are promising candidates for nanoscale light-emitters and many other optical applications, especially in the infrared wavelength region of interest for e.g. optical communication. Because nanotubes can be both semiconductors and metallic depending on the diameter and "twist" they display highly interesting features.



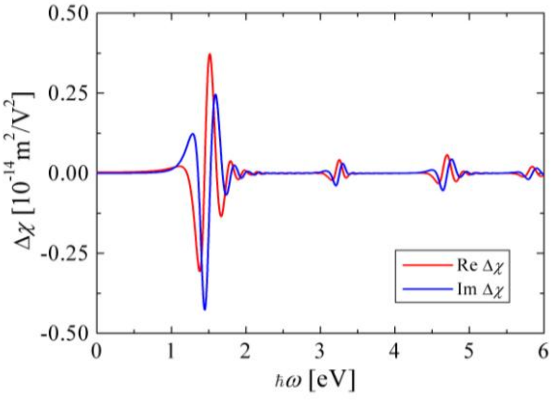
We have studied in detail the unique optical properties of carbon nanotubes using a variety of theoretical tools including excitonic effects in semiconducting nanotubes. A demonstration of the importance of excitons is found in the figure here comparing the absorption spectrum with and without excitonic effects.

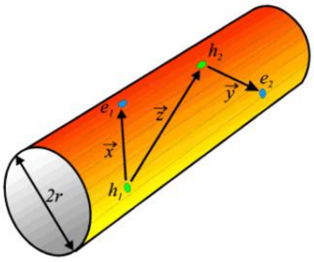
Also, as shown in this figure, the exciton binding energy is extremely large and correlates very well with experimentally measured excitation energies for a wide range of nanotubes.



As a separate investigation the *nonlinear* optical properties and electro-optic are calculated. These are very important for applications such as lasers, high-field emitters, saturable absorbers and electro-optic modulators.

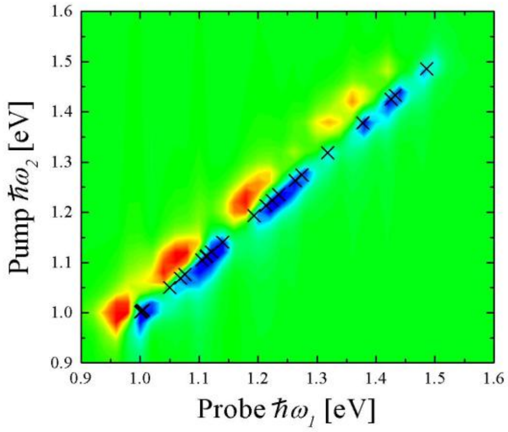
The plot to the right illustrates a non-perturbative calculation of the electro-optic response of a (7,0) carbon nanotube. The resonances coincide with the band gaps of the nanotube. In addition, completely analytical approximations have been obtained.

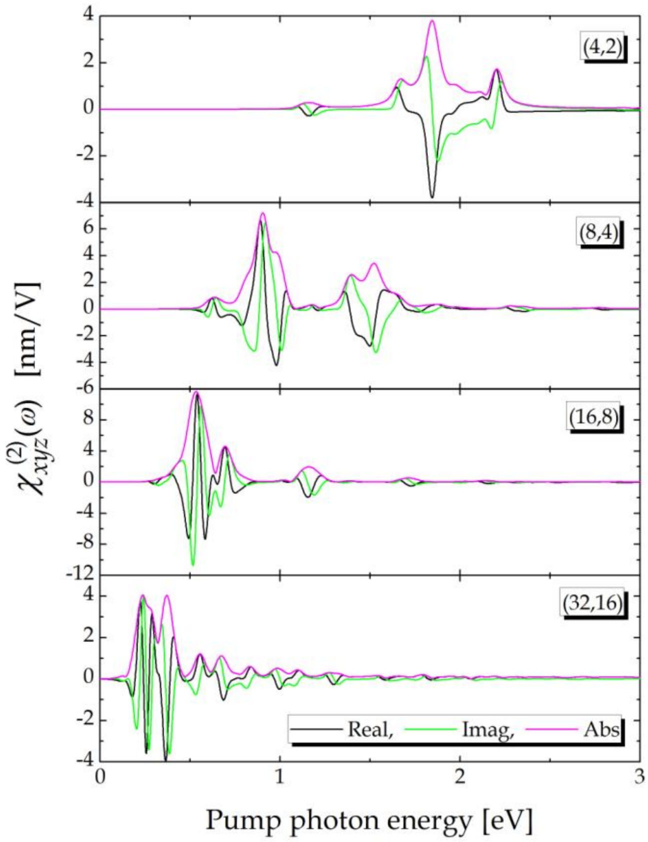




Biexcitons are four-particle complexes comprised of two electrons and two holes. They are produced under intense excitation by high-power lasers. We have demonstrated that such complexes are highly stable in nanotubes and should be clearly detectable in experiments.

To guide experiments, we have computed the response of nanotubes pumped by an intense laser and probed by a weaker beam. The plot to the right shows that the probe beam will experience either increased absorption (red areas) or reduced absorption (blue areas) depending on the probe photon energy. Subsequent to our work, biexcitons have indeed been seen in measurements based on pump-probe spectroscopy (Y.Z. Ma *et al.*, Molecular Physics 104, 1179, 2006).





In a separate project, we investigate the nonlinear optical properties of carbon nanotubes. In particular, optical second harmonic generation has been analyzed theoretically and it is hoped that experiments will follow soon. Our predictions are that nanotubes possess very large optical nonlinearities, which peak for diameters around 15 Å. Moreover, semiconducting species are found to show larger response than metallic ones. So far, excitonic effects have not been considered in the nonlinear response but we aim to correct this in the future. In the spectra to the left, the nonlinear response of (2*n*,*n*) nanotubes are compared.

The following is a list of publications, in which the above mentioned results have been published.

1. T. Garm Pedersen, "Variational approach to excitons in carbon nanotubes", Phys. Rev. B67, 073401 (2003).
2. T. Garm Pedersen, "Exciton effects in carbon nanotubes", Carbon 42, 1007 (2004).
3. H. Cornean, P. Duclos and T.Garm Pedersen, “One dimensional models of excitons in carbon nanotubes”, Few Body Systems 34, 155 (2004).
4. T. Garm Pedersen, "Biexcitons in carbon nanotubes", Fullerenes, Nanotubes and Carbon Nanostructures, 13, 33 (2005).
5. T. Garm Pedersen, K. Pedersen, H. Cornean and P. Duclos, "Stability and signatures of biexcitons in carbon nanotubes", Nano Lett. 5, 291 (2005).
6. A. Zarifi and T. Garm Pedersen "Analytic approach to the linear susceptibility of zigzag carbon nanotubes”, Phys. Rev. B. 74, 155434 (2006).
7. T. Garm Pedersen "Exact polarizability of low-dimensional excitons", Solid State Commun. 141, 569 (2007)
8. A. Zarifi, C. Fisker and T. Garm Pedersen "Theoretical study of the quadratic electrooptic effect in semiconducting zigzag carbon nanotubes”, Phys. Rev. B76, 45403 (2007).
9. B. Ricaud, T. Garm Pedersen and H.D. Cornean, “Ground and excited states in semiconductor carbon nanotubes: Perturbation and variational approach”, Cont. Math. 447, 45 (2007).
10. A. Zarifi and T. Garm Pedersen "Theoretical analysis of the Faraday effect in semiconducting zigzag carbon nanotubes”, Phys. Rev. B. 77, 85409 (2008).
11. A. Zarifi and T. Garm Pedersen “Linear optical and quadratic electro-optic response of carbon nanotubes: universal analytic expressions for arbitrary chirality”, J. Phys.: Condens. Matter. 20, 275211 (2008).
12. T. Garm Pedersen and K. Pedersen, ”Systematic tight-binding study of optical second harmonic generation in carbon nanotubes”, Phys. Rev. B. 79, 035422 (2009).
13. T.F. Rønnow, T. Garm Pedersen and H. Cornean “Stability of singlet and triplet trions in carbon nanotubes”, Phys. Lett. A. 373, 1478 (2009).
14. A. Zarifi and T. Garm Pedersen “Universal analytic expression of electric dipole matrix elements for carbon nanotubes”, Phys. Rev. B. 80, 195422 (2009).
15. S.V.Goupalov, A. Zarifi, and T. Garm Pedersen ”Calculation of optical matrix elements in carbon nanotubes”, Phys. Rev. B. 81, 153402 (2010).
16. T.F. Rønnow, T. Garm Pedersen and H. Cornean “Correlation and dimensional effects of trions in carbon nanotubes”, Phys. Rev. B. 81, 205446 (2010).
17. T.F. Rønnow, T. Garm Pedersen and H. Cornean “Dimensional and correlation effects of charged excitons in low-dimensional semiconductors”, J. Phys. A. 43, 474031 (2010).