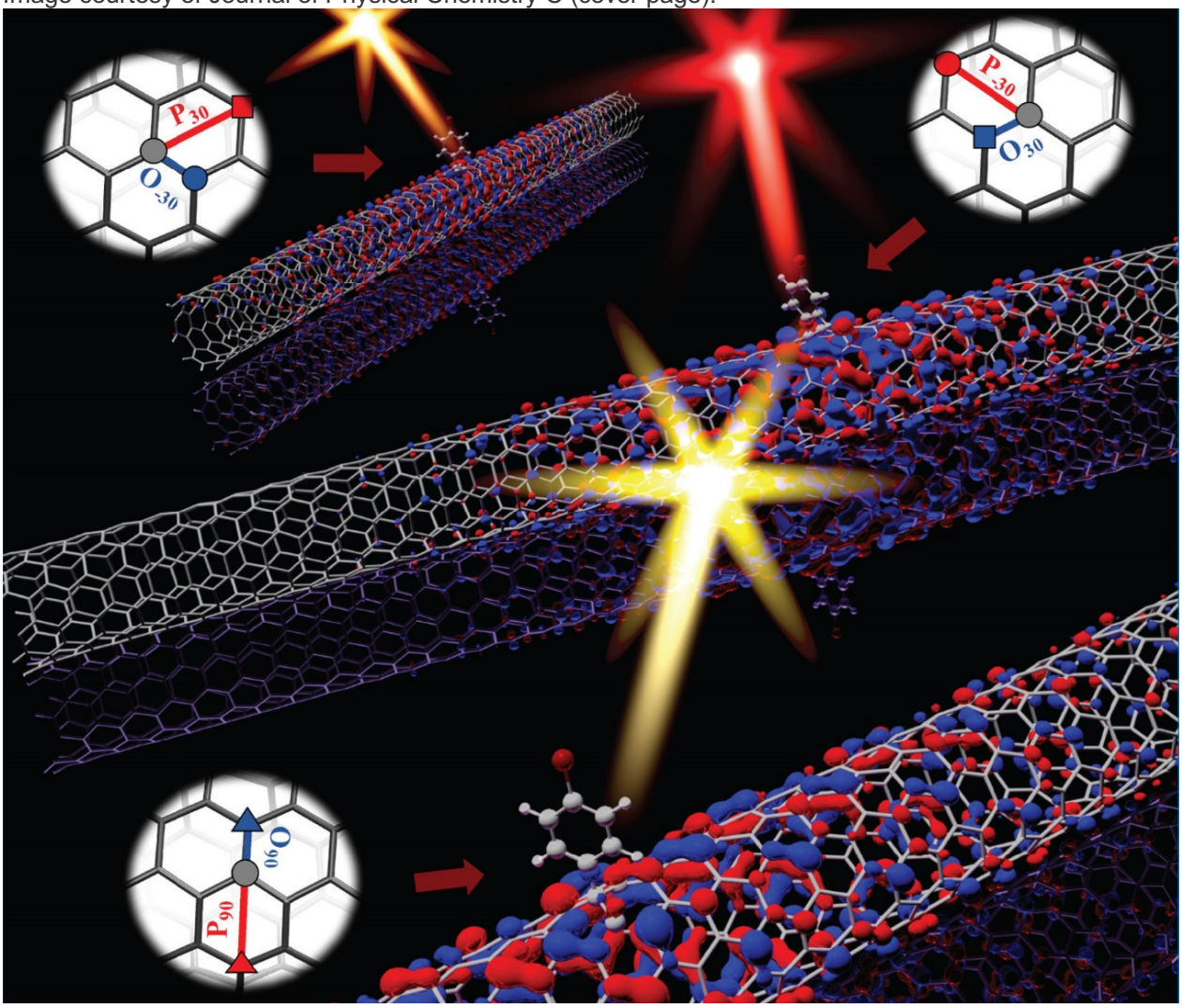
February 2018

Shining Light on Functionalized Carbon Nanotube Emission

Functionalized carbon nanotubes emit light with wavelengths dependent on the topological position of chemical adduct

Image courtesy of Journal of Physical Chemistry C (cover page).



Spatial extent of the electron-hole pairs (excitons) around the defect site in functionalized carbon nanotubes. Three defect configurations are synthetically possible. The topological position of defects defines emission characteristics.

The Science

Through a close collaboration between experimentalists and theorists in the Center for Nonlinear Studies (CNLS) and Center for Integrated Nanotechnology (CINT), researchers have unearthed the origin of diverse emission features in single-walled carbon nanotubes (SWCNTs) with functional groups bonded to the sidewalls. Absorption of a photon in such system generates an electron-hole pair (exciton) on the unmodified portion of the SWCNT. The exciton then moves to the defect site, from which emission occurs. Since the defect site forms a quantum well and lowers the energy of the exciton, emission energies are significantly lower than absorption and can be manipulated by controlling the geometry of the defect.

The Impact

The emission features of functionalized SWCNTs are brighter and at lower energies than for pristine systems, increasing their utility as single-photon sources (SPSs) in fiber optical telecommunications. The discovery that geometry at the defect site heavily influences the energies of emission features suggests a strategy for tuning their optical properties for such applications.

Summary

Single-walled carbon nanotubes (SWCNTs) are pseudo one-dimensional nanostructures that exhibit diameter-dependent electronic and optical properties. Taking advantage of these properties in a number of applications from photovoltaic devices to light emitting diodes has been considered for the past decade. However, the lowest energy electronic transitions in pristine systems are optically dark, and therefore their emission is inefficient. Adding chemical groups to the sidewall of carbon nanotubes results in brightening of emission features. While this makes functionalization an attractive strategy for manipulating the optical properties of SWCNTs, it is accompanied by the generation of multiple emission peaks with an unexplained origin. To approach this issue, scientists at CINT and CNLS performed a joint study in which aryl-functionalized SWCNTS were both synthesized and modeled computationally. Taking cues from synthetic chemistry, they identified the positions at the surface of the SWCNT that could react with chemical groups during functionalization. The energies of emission features were shown to be significantly influenced by the positions of functionalization. These findings justify the existence of multiple emission features and suggest a strategy for further experimental tuning of the optical properties in functionalized SWCNTs. This is a major step toward using such materials for fiber optical applications.

Contact

Brendan Gifford giff@lanl.gov

Steven Doorn skdoorn@lanl.gov

Sergei Tretiak tretiak@lanl.gov
Los Alamos National Laboratory

Funding

The work at Los Alamos National Laboratory (LANL) was supported by LANL Laboratory Directed Research and Development (LDRD) program. Work at North Dakota State University (NDSU) was supported by grant CHE1413614 from the National Science Foundation (NSF). This research used resources of the Center for Functional Nanomaterials, which is a U.S. DOE Office of Science facility, at Brookhaven National Laboratory under contract no. DE-SC0012704. Computational resources were provided by the NDSU Center for Computationally Assisted Science and Technology (CCAST), LANL Institutional Computing (IC), and National Energy Research Scientific Computing Center (NERSC) allocation awards 86678, supported by the Office of Science of the DOE under Contract No. DE-AC02-05CH11231.

Publications

B. J. Gifford, S. Kilina, H. Htoon, S. K. Doorn, S. Tretiak, Exciton Localization and Optical Emission in Aryl-Functionalized Carbon Nanotubes. *J. Phys. Chem. C*. **122**, 1828–1838 (2018). *Editor Choice and Cover Page*

X. He, B. J. Gifford, N. F. Hartmann, *et al.*, Low-Temperature Single Carbon Nanotube Spectroscopy of sp^3 Quantum Defects. *ACS Nano*. **11**, 10785–10796 (2017).

Related Links

ACS Liveslides of Gifford *et al.*

https://pubs.acs.org/doi/suppl/10.1021/acs.jpcc.7b09558/suppl_file/jp7b09558_liveslides.mp4

Science Web Highlights
February 2018

Additional information that will not be publically displayed on the webpage:

Image usage rights:
Click here to enter text.

Program Contact(s):
Click here to enter text.

Originating Reviewer(s) (if applicable):
Click here to enter text.

Internal Reviewer(s):
Click here to enter text.

Science Web Highlights
February 2018

Metadata Tags: highlight all tags that are appropriate for a given highlight

PROGAM ***Select only those that contributed funding or resources***	**PERFORMER/FACILITY** ***Select at least one (or more than one) of the elements below***	**ADDITIONAL** ***Select as appropriate***
ASCR	**University** *(includes non-profit academic institutions)*	**Technology Impact**
BER	**DOE Laboratory**	**Collaboration**
BES	**Industry**	
FES	**User Facilities**	
HEP		
NP		

Science Web Highlights

February 2018

Collaborators:

Los Alamos National Laboratory

Center for Integrated Nanotechnology

Center for Nonlinear Studies

North Dakota State University

National Renewable Energy Laboratory

Chemical and Materials Science Center

Stevens Institute of Technology