Enhanced Multi-Photon Emission from Single NV Center Coupled to Graphene by Laser-Shaping

Jing Liu[1,2], Yaowu Hu[3], Prashant Kumar[3], Mikhail Y. Shalaginov[2,4], Alexei S. Lagutchev[2,4], Vladimir M. Shalaev[2,4], Gary J. Cheng[3], and Joseph MK. Irudayaraj[1,2,*]

[1]Department of Agricultural and Biological Engineering, Bindley Bioscience Center,
[2]Birck Nanotechnology Center,
[3]Schoold of Industrial Engineering,
[4]School of Electrical and Computer Engineering,
Purdue University, West Lafayette, IN 47907, USA
Author e-mail address: liu456@purdue.edu

Abstract: We experimentally demonstrated the enhanced multi-photon emission from single NV center coupled to 2D and 3D graphene by a nanoscale laser-shaping technique. Our results provide new non-classical light source for quantum optics and light harvesting.
OCIS codes: (270.0270) Quantum optics; (160.4236) Nanomaterials.

1. Introduction

Nanodiamonds (NDs) with single nitrogen-vacancy (NV) center recently draw tremendous attractions due to its complex excited state dynamics.[1] Its spintronic property make it successfully at nanoscale magnetic and electronic sensing, and the realization of the multispectral super resolution imaging.[2] The bio-compatibility of the NDs and non-blinking, non-bleaching fluorescent signals from the NV center embedded NDs make NDs ideal potential fluorescent probes for biology. [3] Furthermore, single NV center in the ND can emit stable single photons at the room temperature, which makes the NDs pronouncing single-photon source for integrated quantum computation and communication. However, quantum optics not only requires a single photon source, but also needs a multi-photon source which can generate entangled photon pairs. Coupling the single NV center to graphene allows for the generation of on-demand multi-photons from the single NV center. This unconventional phenomenon is the result of the enhanced nonradiative decay rate from the NV center due to the presence of graphene. Our efforts provide new insights on understanding the ND-graphene interactions, which is essential in the generation of non-classical light source for quantum optics and in the inspiration of developing a new class of devices for light-harvesting.

2. Experiments and Fabrication of Graphene-NV center

The coupling of NV center to single layer graphene was realized by imbed the host nanodiamond of single NV centers to the graphene with atomic contact *via* a newly reported nanoscale laser-shaping approach.[4] As shown in Figure 1(a), nanodiamonds (Microdiamant AG) with the size around 50 nm were spincoated on the coverslip, and then covered with a single layer graphene by transformation. The 3D graphene-nanodiamond hybrid structure was fabricated by being explored to a Q-switch Nd-YAG laser (Continuum Surelite III, 1064 nm) with a 5 ns duration time. The generated plasma results in a laser shock pressure (~GPa) directed downwards and forces the single layer graphene to be conformally shaped with a 3D configuration over nanodiamonds. Atomic-scale integration of the graphene and nanodiamonds can be realized by this unique opto-mechanical approach, which is vital for engineering the excited states of emitters close to the graphene.

Emission behavior of the NV center was investigated by the home-built time-resolved spectroscopy. a picosecond-pulsed laser beam (LDH-FA, Picoquant Inc. 532 nm) with tunable repetition frequency (10/20 MHz) was delivered onto the sample by a water-immersion objective (Olympus, NA 1.2) as excitation. Fluorescence emission was collected by the same objective, and filtered by a 50 μm pinhole as well as a band pass filter (685-70, Chroma). Then photons from NDs were split via a 50/50 beam-splitter into two identical single photon avalanche photodiodes (SPAD) (SPCM-AQR-14, PerkinElmer Inc.). The photon was recorded in the time correlated single photon counting (TCSPC) module (TimeHarp 200, Picoquant Inc.). Fluorescence lifetime of the NV center was obtained by fitting their TCSPC histogram with a multi exponential equation. The single photon emission property was investigated by a measurement, a Hanbury Brown-Twiss (HBT) geometry.

3. Results

Different from the signature of single photon emission of single NV centers on the coverslip, individual NV centers coupled to the graphene yield a significantly enhanced possibility of multi-photon emission, i.e., a peak appears at the zero time delay. In addition, the fluorescence intensity and the lifetime from the NV centers were also reduced.

Calculation suggests that the photon-induced carrier on the graphene plays an important role in the engineering of the emission of NV centers, the energy transfer efficiency from the NV center to the graphene can reach 80% under high laser excitation.

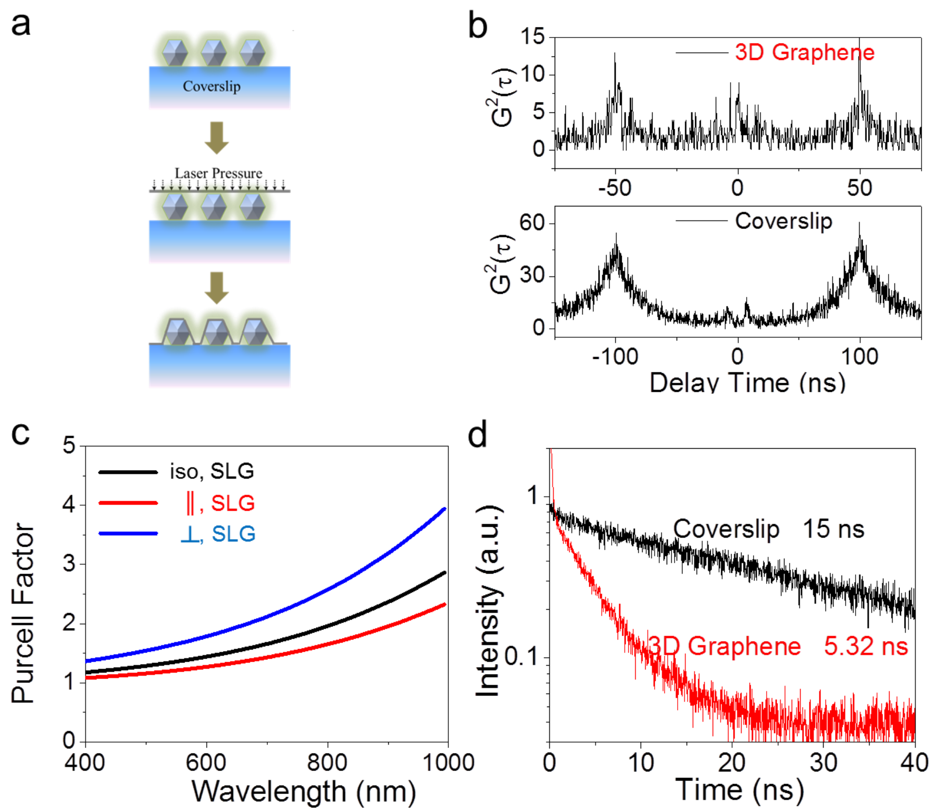

Figure 1. (a) Schematic of the sample preparation. (b). G2 measurements of the NV center on coverslip and coupled with graphene. (c). Calculated Purcell Factor. (d) Lifetime of NV center on coverslip and coupled with graphene.

4. References

1. Christian Kurtsiefer, Sonja Mayer, Parick Zarda, and Harald Weinfurter. "Stable solid-state source of single photons." *Phys. Rev. Lett.* 85, no. 2 (2000): 290-293.

2. Edward H. Chen, Ophir Gaathon, Matthew E. Trusheim, and Dirk Englund. "Wide-field multispectral super-resolution imaging using spin dependent fluorescence in nanodiamond." *Nano Lett.* 13 (2013): 2073-2077.

3. Yi-Ren Chang, Hsu-Yang Lee, Kowa Chen, Chun-Chien Chang, Dung-Sheng Tsai, Chi-Cheng Fu, Tsong-Shin Lim, Yan-Kai Tzeng, Chia-Yi Fang, Chau-Chung Han, Huan-Cheng Chang, and Wunshain Fann. "Mass producion and dynamic imaging of fluorescent nanodiamonds." *Nat. Nanotechnology* 3 (2008): 284-288.

4. H Gao, Hu Y., Xuan Y., Li J., Yang Y., Martinez R., Li C., Luo J., Qi M., and GJ Cheng, 2014, Large Scale Nanoshaping of Ultrasmooth 3D Crystalline Metallic Structures. Science, 346, 1352.