EPJ Web of Conferences **103**, 01013 (2015)
DOI: 10.1051/epjconf/201510301013

Nanodiamond Emitters of Single Photons

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Abstract. Luminescence properties of single color centers were studied in nanodiamonds of different origin. It was found that single photon emitters could be realized even in molecular-sized diamond (less than 2 nm) capable of housing stable luminescent center "silicon-vacancy." First results on incorporation of single-photon emitters based on luminescent nanodiamonds in plasmonic nanoantennas to enhance the photon count rate and directionality, diminish the fluorescence decay time, and provide polarization selectivity are presented.

Keywords: Single photon emitter, color center, nanodiamond, plasmonic nanoantennas.

Single-photon sources (SPSs), producing nonclassical light, are pivotal components in quantum communication technology. They produce individual photons with no chance for time overlaping the photons. For common light sources (bulbs, sun light) and in lasers, multiple (nonseparated) photons cannot be avoided even in very faint sources. For today the most popular single-photon emitters, which operate at room temperature, are dye molecules, colloidal nanocrystal quantum dots, and various color-centers in nanodiamonds. Because it possesses a longest photostability and a high quantum yield of luminescence, diamond material with different color centers are the best candidates for practical implementation of single-photon devices.

Detonation Nanodiamonds (NDs) demonstrate a high potential for the production of highly desirable SPS based on luminescent “nitrogen-vacancy” (NV) centers, as these NDs contain large amount of N impurity (2-3% mass) and are produced at an industrial scale. However, till now the NDs synthesized from explosives was not among the candidates for SPS application owing to lack of optically active particles containing NV centers. In our work we have systematically studied representative classes of NDs produced by detonation shock wave conversion of different carbon precursor materials, namely, graphite and a graphite/hexogen mixture into ND, as well as ND produced from different combinations of explosives using different cooling methods (wet or dry cooling). We demonstrate that (i) the N content in nanodiamond particles can be controlled through a correct selection of the carbon precursor material (addition of graphite, explosives composition); (ii) particles larger than approximately 20 nm may contain in situ produced optically active NV centers; (iii) in ND produced from explosives, NV centers are detected only in ND produced by wet synthesis; (iiii) ND synthesized from a mixture of graphite/explosive contains the largest amount of NV centers formed during synthesis. It was shown that the single photon emitters can be produced on a base of NV centers in detonation nanodiamond synthesized from graphite-hexogen mixture [1].

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Production of SPS was seen elusive for very small diamond nanoparticles because of their limited availability and a lack of fundamental understanding of impurity stability in such nanostructures. We have shown that isolated diamond nanoparticles as small as 1.6 nm, comprising only ~400 carbon atoms, are capable of housing stable photoluminescent colour centres, namely the "silicon-vacancy" (SiV) [2]. Surprisingly, fluorescence from SiVs was stable (no bleaching) over time, and few or only single colour centres were found per nanocrystal. We also observed size-dependent SiV emission supported by quantum-chemical simulation of SiV energy levels in small nanodiamonds.

A minimum size of diamond nanoparticles, in which intense single photon emitters could be formed, was evaluated. The studied diamond nanoparticles presented a composite structure of "core-outer layer" type. The core was 20-nm HPHT diamond particle containing NV centers, whereas outer diamond layer containing SiV centers was formed by CVD synthesis. The proximity of the surface suppresses the NV and SiV luminescence. We have shown that the interaction of the luminescent centers with ND surface becomes insignificant at a distance to its surface exceeding 12 nm for NV centers, and 4 nm for SiV centers [3]. Our finding determines minimum sizes of nanodiamonds, in which stable single photon emitters could be formed, namely, 24 nm for NV emitters and 8 nm for SiV emitters.

To enhance the single-photon count rate (the bit rate of future quantum information systems) and improve SPS directionality, coupling efficiency into a fiber system and polarization selectivity, single emitters should be embedded in microcavities, photonic crystals [4] and other micro- or nano-structures (e.g., plasmonic nanoantennas, metamaterials, etc.). Using high-precision electron-beam lithography, we fabricated gold bowtie nanoantennas [5]. To place nanodiamonds in the gap between the two triangular arms of this nanoantenna we developed a technique to manipulate nanodiamonds using an atomic force microscope (AFM) tip (lithography option of an AFM) [5]. Our next step will be to use this technique to place diamonds nanocrystals containing NV(SiV) centers in a gap between the arms of the nanoantenna to enhance the photon count rate and directionality, diminish the fluorescence decay time, and provide polarization selectivity.

This work was supported in part by a grant from the US Department of State. The opinions, findings and conclusions stated herein are those of the authors and do not necessarily reflect those of the US Department of State.

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