Optics and Spectroscopy, 2025, Vol. 133, No. 10

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Increasing the Efficiency of Silicon-Vacancy Color Center Radiation Collection from Diamond Microspheres Using a Glass Microsphere

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Received April 30, 2025
Revised June 27, 2025
Accepted October 24, 2025

The results of the study of the angular dependence of the photoluminescence of silicon-vacancy color centers in diamond microscopic hemispheres are presented. It is experimentally demonstrated that the use of a glass microsphere with a diameter of $200\,\mu$m and a microlens with a numerical aperture of NA=0.9 in the scheme of recording the photoluminescence signal allows achieving a tenfold increase in the intensity of the photoluminescence signal of the zero-phonon line of the silicon-vacancy color center. Due to the effective collimation of radiation by the microsphere, it became possible to narrow the angular dependence of the color center photoluminescence many times.

Keywords: diamond microparticles, color centers, microsphere, radiation collimation.

DOI: 10.61011/EOS.2025.10.62557.7965-25

1. Introduction

Luminescent isolated diamond microparticles with embedded color centers attract attention as solid-state single-photon sources [1]. Their biocompatibility, chemical inertness, and low cytotoxicity make them promising for biomedical applications [2]. The silicon-vacancy (SiV) color center in diamond is a point defect consisting of an interstitial Si atom and two adjacent vacancies in neighboring lattice sites [3]. At room temperature, the photoluminescence (PL) spectrum of SiV color centers consists of a strong zero-phonon line at a wavelength of 738 nm and a weak vibronic band in the 760 nm region. The full width at half maximum (FWHM) of the SiV zero-phonon line (ZPL) at room temperature is $1-5$ nm [4].

One significant obstacle to the practical application of SiV centers in quantum information systems is the low efficiency of radiation extraction, associated with total internal reflection of radiation from the centers due to the high refractive index of diamond ($n \sim 2.4$) corresponding to a total internal reflection angle of 24.5°. In the case of a flat diamond boundary, a substantial fraction (up to 91% [5]) of the SiV centers' PL signal undergoes total internal reflection.

Methods proposed in the literature for increasing radiation extraction efficiency using waveguide modes are complex and costly [5–7]. Another approach to enhancing PL collection efficiency involves applying hemispherical microlenses onto diamonds with emitting color centers. However, methods for their fabrication via lithography followed by etching or electron beam formation are also extremely labor-intensive [8,9] and require precise positioning of the emitting center at the focus of these microlenses.

A widely used approach to improving radiation extraction efficiency involves modifying the surface shape of diamond crystals, specifically forming diamond hemispheres [6,9]. For such hemispheres, the reflection coefficient at the diamond/external medium interface and the numerical aperture of the collection optics determine the radiation collection efficiency. In isolated microscopic diamond hemispheres, the PL signal propagates over a wide solid angle [9], necessitating the use of high-numerical-aperture microlenses, which are expensive and have short working distances to achieve the required collection efficiency [10]. The present work proposes and demonstrates a simpler method for collimating radiation from diamond microscopic hemispheres using a glass microsphere.

2. Experimental Method

Hemispherical diamond particles were synthesized on a sapphire substrate using microwave plasma chemical vapor deposition (MWCVD) [11]. Formation of SiV color centers occurred during the growth of diamond particles via their doping with Si atoms, sourced from silane. MWCVD process parameters: microwave power 600 W (2.45 GHz), substrate temperature — 700 °C, hydrogen flow rate 500 sccm, methane concentration in the gas mixture 4%, reactor working pressure 15 Torr, diamond particle growth time 1 h. In the MWCVD processes, silane was added to the reactor from a silane-argon mixture ($SiH_4/(Ar+SiH_4) = 1\%$), with the relative silane concentration being $SiH_4/(SiH_4 + CH_4) = 0.1\%$ [12]. Detonation-synthesized nanodiamonds with a characteristic size of ~ 4 nm were applied as diamond nucleation centers via aerosol spraying. The density of detonation-synthesized nanodiamonds applied to the substrate surfaces was $\sim 10^7\,cm^{-2}$. The size of the diamond microparticles was $2-3\,\mu$m (inset to Fig. 1).

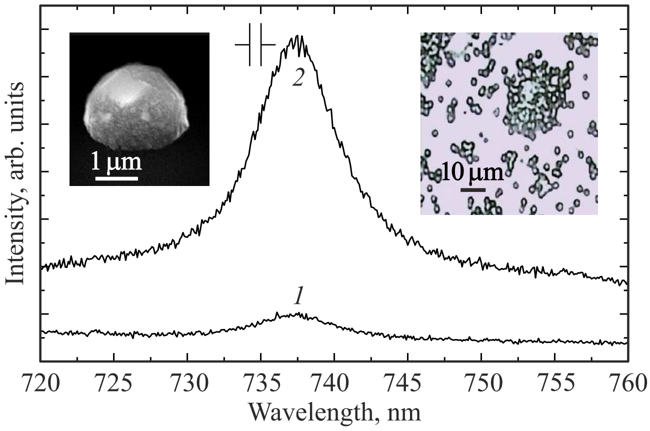

Figure 1. PL spectra of the SiV color center in diamond microspheres, recorded: without a glass microsphere in front of the microobjective (*1*), with a microsphere (*2*). Insets show — image of a single hemispherical diamond particle obtained using a scanning electron microscope; image of the substrate surface with diamond particles obtained using an optical microscope.

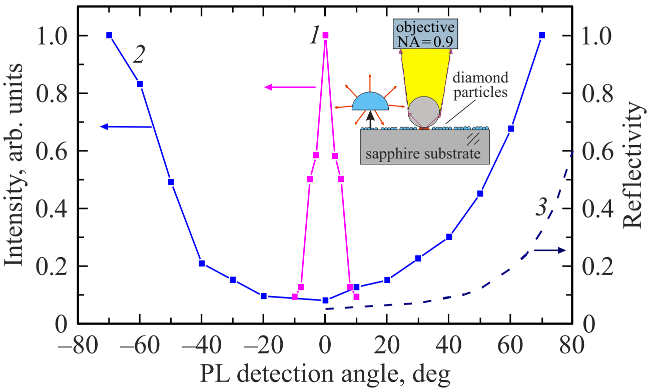

Figure 2. Angular dependence of the ZPL PL intensity of the SiV color center, obtained: using a glass microsphere in the PL measurement scheme (*1*), without a microsphere (*2*). Angular dependence of the reflection coefficient from the sapphire substrate (*3*). Inset — experimental scheme with the microsphere. The diamond particle in the inset is highlighted separately by an arrow in enlarged scale.

The experimental scheme for measuring PL of SiV color centers in diamond microscopic hemispheres is shown in the inset to Fig. 2. Excitation of SiV center PL through the microobjective and microsphere was performed using a semiconductor laser diode with emission wavelength 660 nm and power 8 mW. A photomultiplier tube FEU-79 served as the photodetector. Initially, the PL spectrum was recorded through the microobjective alone, pre-tuned to the PL signal maximum. Then, a glass microsphere with diameter 200 μm purchased from INOTEK (Russia), was introduced between the sample surface and the microobjective in the optimal position for PL signal enhancement. These microspheres are primarily used for road markings due to their retroreflective properties. The microspheres contain no air inclusions, have a refractive index ≥ 1.50 and sphericity of at least 80%. The microsphere was glued with silicone sealant to the end of a quartz fiber with thickness 125 μm. The choice of high-temperature ABRO silicone-based sealant with acetic acid prevented damage to the lightguide-microsphere contact due to heating by focused laser radiation. The selection of a lightguide with diameter 125 μm was dictated by the experimental geometry. The microsphere diameter must exceed that of the lightguide to prevent uncontrolled sealant spreading over its surface during gluing. An excessively large diameter of the glass sphere and corresponding lightguide would not fit in the narrow gap (< 0.5 mm) between the sample and the microlens used in the experiment. Positioning of the microsphere in two planes between the sample and the microlens with magnification 30× and aperture NA=0.9 was achieved using a precision micrometric mechanical stage. This ensured tight contact between the microsphere and the sample. Note that the microsphere size has negligible impact on PL collimation efficiency [13].

3. Discussion of the results

Fig. 2 shows the angular dependencies of the SiV color center ZPL PL intensity with and without the microsphere. From the obtained dependencies, it is evident that without the microsphere, the PL intensity sharply increases with registration angle up to 80°. The likely cause of such dependence is the increased reflection of PL radiation from the substrate at grazing registration angles, similar to light incidence on the sapphire substrate (Fig. 2, curve *3*). Application of the microsphere narrowed the angular PL dependence by approximately an order of magnitude. From Fig. 1, it is seen that the SiV center ZPL PL intensity using the glass microsphere also increased by about an order of magnitude. The observed PL signal enhancement arises because the microsphere, positioned close to the sample, acts as a thick focusing lens with a wide collection angle, redirecting the radiation wave vectors into a narrow collimated beam toward the microlens entrance pupil (inset to Fig. 2) [14,15]. Without the microsphere, the radiation wave vectors are directed radially in all directions from the sample. Additional contributions to radiation collimation may come from effects due to resonant light scattering by a spherical particle with diameter on the order of several wavelengths (photon nanostreams and subwavelength light focusing) [16].

4. Conclusions

Incorporating a glass microsphere as a lens in direct contact with diamond microspheres in the PL registration scheme narrowed the angular dependence of the SiV color center ZPL PL intensity and achieved nearly a tenfold increase in ZPL PL intensity. Thus, effective PL collimation by the dielectric microsphere enables enhanced radiation collection using microlenses with low aperture and cost. The proposed method is not only simple to implement but also substantially reduces the overall cost of the experimental setup, potentially enabling broader application of quantum dots and diamond color centers.

Conflict of interest

The authors declare that they have no conflict of interest.

Funding

This study was carried out under the state assignment. № FFUG-2024-0017.

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Translated by J.Savelyeva