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Way to increase the used part of radiation of color centers in diamond

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A way to increase the extracted and collected part of the radiation of color centers in diamond is suggested. It is based on the concentration of this radiation in modes, formed in a plane waveguide consisting of the upper surface of a diamond sample (from which they are reflected due to the total internal reflection effect) and a periodic sequence of low and highly boron doped layers with slightly different refraction indices (from which they are reflected due to the Bragg effect) lying below. The radiation contained in these modes are partly extracted through the upper surface of a diamond sample by means of the frustrated total internal reflection method. For a particular example of NV-centers by numerical modelling it is shown that the suggested way allows to increase the part of their radiation extracted from diamond by more than 2 times. In the same time it does not lead to the deterioration of the NV-center parameters important for applications and remains efficient for any coordinates of these centers in the diamond sample plane and for their high concentrations.

Keywords: Bragg effect, diamond, NV-center.

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1. Introduction

Color centers (NV, SiV, GeV, etc.) in diamond are defects of its crystal lattice. They are formed by the N, Si, Ge or other atoms that substitute the carbon atoms in the lattice and by the neighboring empty lattice sites, i.e. vacancies V. Over the past few decades, researchers have made an increased focus on these centers due to their potential application for quantum computations, data storage and measurement of various physical quantities with high sensitivity and resolution [1,2]. The NV-centers (Figure 1) are the best-understood and very interesting in terms of these applications, and will be discussed in this work. However, all qualitative conclusions made below are also valid for other color centers. The NV-centers in diamond were discovered using the electron paramagnetic resonance more than 40 years ago [3]. Due to very long lifetimes (several milliseconds) and times of the loss of mutual coherence (more than $1\,\mu$s) of their spin states at room temperature [4,5], and to the possibility to control these states by electromagnetic radiation using optically detected magnetic resonance [1], they are widely used in magnetometry [6], quantum computations and data processing [7].

However, due to a high refractive index of diamond n (about 2.4 [8]) in the optical range that contains the luminescence wavelength of the NV-centers 637 nm [1], just a very small portion of their radiation is extracted from a diamond specimen (generally in a form of a plate about 0.5 mm in thickness and a few millimeters long and wide) through its upper surface to air and after collection by the optical system is used for the above-mentioned applications. This portion may be evaluated as the relation of the solid angle $2\pi(1-\sqrt{1-1/n^2})$ at the apex, situated in the NV-center point, of the right circular cone with the apex angle $2\mathrm{ArcSin}(1/n) \approx 49.2^\circ$ and the axis along

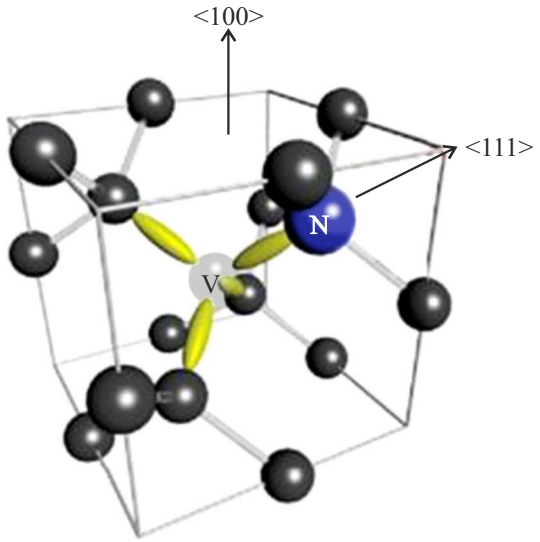

Figure 1. Schematic view of the NV-center in the diamond lattice: black circles — carbon atoms, blue circle N — nitrogen atom, grey circle V — vacancy, covalent bonds between the carbon atoms (grey thick lines), orbitals of electrons belonging to the NV-center (yellow), and ⟨100⟩ and ⟨111⟩ crystallographic directions.

the normal to the top surface of the specimen, within which the radiation wave vector direction shall rest to avoid full internal reflection on this surface, to the full solid angle 4π, within which this wave vector may be oriented, i.e. $(1-\sqrt{1-1/n^2})/2 \approx 0.045$. The remaining radiation portion (95.5%) of the NV-centers in these applications is not used because it either has multiple full internal reflections on the specimen surface and is finally absorbed in it, or is extracted to air through the bottom or side surfaces of the specimen and, therefore, is not collected by the optical system. Thus, it is important to increase the radiation portion of the NV-centers that is used in devices based on the NV-centers.

For this, it has been proposed to change the optical collecting system in such a way that it could collect the NV-center radiation extracted from the diamond specimen through its side faces, rather than through the top surface [9]. This approach makes it possible to collect approximately a half of radiation of a single NV-center [9]. However, this approach to increasing the collected radiation portion of the NV-centers, as pointed out by the authors of [9], may be used only at quite low concentration of these centers in the diamond specimen because, radiation from each NV-center goes (generally repeatedly) through the whole diamond specimen before collection by the optical system, and, therefore, may be resonantly absorbed on the same other NV-centers. Therefore, when the NV-center concentration is high, efficiency of such method for their radiation extraction from the diamond specimen is low.

Another way to increase the collected radiation portion of the NV-centers is to create a hemispherical diamond lens with its center near the examined NV-center and diameter of about 1 mm on the top surface of the diamond specimen using a combination of laser processing and machining [10]. As a result, radiation of this NV-center in each point on the top surface of the diamond specimen is oriented along the normal to this surface and, therefore, when the radiation is transmitted through the top surface, it is not subjected to full internal reflection. Thus, the authors of [10] managed to collect 20−30% of radiation of the NV-center situated near the center of the hemispherical lens. However, as the NV-center moves away from the center of this lens at distances exceeding approx. $30\,\mu$m, the radiation portion collected in such way is reduced dramatically and becomes equal to approx. 4%, i.e. the same value as without the lens. Therefore this method for increasing the collected radiation portion of the NV-centers is only applicable for those of them that are situated near the center of the hemispherical lens, which is its main disadvantage.

The collected radiation portion of the NV-centers may be also increased, if they are integrated in diamond nanocrystals with subwavelength sizes [11,12], due to failure to meet the geometrical optics applicability conditions. Thus, in diamond nanowires, the NV-centers emit mainly along their axes, therefore the collectedd radiation portion achieves 40% [12]. However, the application-critical parameters of the NV-enters in the diamond nanocrystals appear to be considerably worse that those of bulk specimens: they have longer lifetimes of excited states and therefore lower photon emission rates [11] and shorter coherent times of spin states [13]. This makes their use in the above-mentioned applications difficult, if not impossible, which is a serious advantage of this method for increasing the collected radiation portion of the NV-centers.

This study proposes a new approach to increasing the collected radiation portion of the NV-centers in diamond. It is applicable to bulk diamond specimens and, therefore, does not lead to degradation of the NV-center parameters typical or the diamond nanocrystals. At the same time, in contrast with the diamond lens method, it remains effective at any coordinates of the NV-centers in the diamond specimen plane and, in contrast with the method for radiation extraction through the side faces of the diamond specimen, may be also used at higher concentration of the NV-centers. The main idea of the method is described in the next section. Section 3 provides the mathematical model parameters of the proposed method and describes its computation scheme. Section 4 provides and discusses the calculation results. Conclusion sets out recommendations for practical application of the proposed method for increasing the collected radiation portion of the NV-centers in the diamond.

2. Concentration of NV-center radiation in waveguide modes and their extraction from the diamond specimen by the attenuated total reflection method

The main idea (Figure 2) of the proposed method for increasing the collected radiation portion of the NV-centers in the diamond is in concentration of this radiation in the waveguide modes whose wave vectors are parallel to the top surface of the diamond specimen (xy plane) and have the same values. These modes are formed in a planar waveguide formed by the top surface of the specimen (reflection from which is caused by the full internal reflection effect) and the underlying periodic sequence of layers with different refractive indices (reflection from which is caused by the Bragg effect). Minor change (i.e. decrease) in the refractive index of diamond needed to create such sequence may be achieved by heavy p-type boron doping that produces free charge carriers (holes) whose vibrations in a variable electric field of an electromagnetic wave cause the reduction of polarization and, thus, reduction of the refractive index of diamond.

Radiation contained in these modes may be extracted into the environment using the attenuated total reflection method [14]. It is based on the fact that these modes partially penetrate through the top surface of the diamond specimen in air where they attenuate exponentially when moving away from this surface because the refractive index

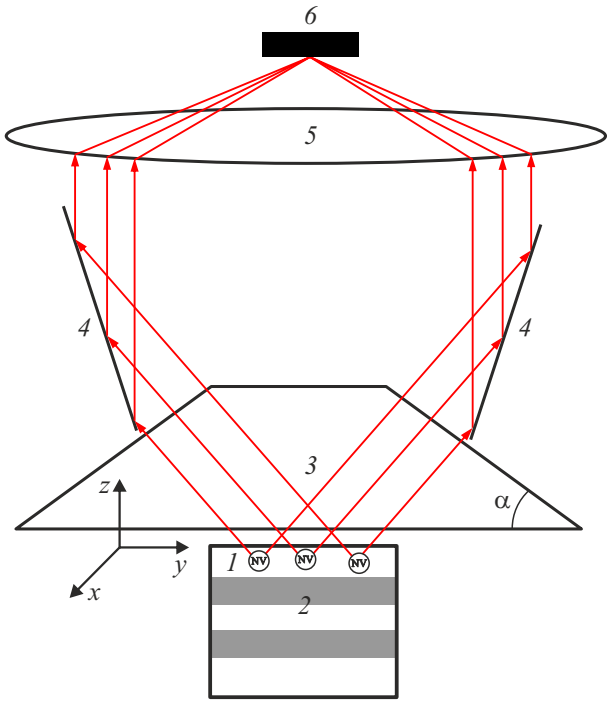

Figure 2. Schematic (not to scale) illustration of the proposed method for increasing the collected radiation portion of the NV-centers in the diamond (sectional elevation) and the coordinate system used herein: *1* — NV-centers near the diamond specimen surface, *2* — reflecting periodic sequence of layers with unreduced (white, low boron-doping) and reduced (grey, high boron-doping) refractive indices (only 2 periods are shown), *3* — glass circular truncated cone with the base angle α, *4* — conical mirror, *5* — collecting lens, *6* — photodetector. Red arrows show the wave vector directions of the collected NV-center radiation.

of air is lower than that of diamond. Therefore, if a glass circular truncated cone is placed at a distance from the specimen surface on the order of the scale of exponential electromagnetic field attenuation of these modes, then these modes will penetrate the cone. Due to the fact that the refractive index of this cone is higher than the refractive index of air, electromagnetic fields of these modes in the cone will be no longer attenuated when moving away from the top surface of the diamond specimen and will propagate in it without attenuation at a particular angle to the bottom of the cone that is defined by the wave vectors of these modes and refractive index of the cone. Then, if the angle α between the cone generatrixes and base has been chosen in such a way that the propagation directions of these modes in the cone are orthogonal to its face, then these modes come from the cone into the environment without full internal reflection on the cone face. Thus, extraction of the waveguide mode radiation from the diamond is achieved.

For effective implementation of the attenuated total reflection method, it is important that the NV-center radiation is concentrated in the waveguide modes with the same wave vector values. This is due to the fact that, when there were no waveguide, the wave vectors of this radiation would have different values of their components in the diamond specimen plane and, therefore, at any chosen angle α between the cone generatrixes and base, the wave vectors would be orthogonal to the cone face only for a small portion of this radiation.

3. Mathematical model and computation scheme

Let's consider the radiation of the NV-center situated at 20 nm from the top surface of the diamond. The top layer of the diamond is low-boron-doped (boron atom concentration is $5 \cdot 10^{18}\,\mathrm{cm}^{-3}$) and is 75 nm thick. Its is underlaid by a 78 nm high-boron-doped layer (boron atom concentration is $4 \cdot 10^{20}\,\mathrm{cm}^{-3}$). Then (i.e. deep into the diamond) this pair of layers is repeated 49 times in the same sequence, i.e. the reflecting lattice contains 50 periods. Then it is followed by low-boron-doped (boron atom concentration is $5 \cdot 10^{18}\,\mathrm{cm}^{-3}$) diamond substrate. The model assumes a lattice of 50 periods because our computer simulation has shown that for such (or thicker) lattice attenuation of the waveguide modes due to their escape into the substrate, i.e. due to extraction from the diamond specimen not in the truncated cone, is negligibly low. Thus, the lattice with 50 and more periods will actually serve as an endless lattice ensuring negligibly low attenuation of the waveguide modes due to their undesired escape in the substrate. Refractive index of the low-doped layers was taken equal to the refractive index of the undoped diamond $n = 2.4$ due to high activation energy of boron atoms 370 meV that is much higher than the thermal energy 26 meV at room temperature and, therefore, negligibly low concentration of free holes in these layers [8]. Refractive index of the high-doped layers was simulated by the Drude equation [15] $\sqrt{n^2 + 4\pi i e^2 N\tau/[m\omega(1 - i\omega\tau)]}$, where i is the imaginary unit, e is the hole charge equal to the elementary charge, N is the concentration of free holes taken equal to the boron atom concentration in these layers $4 \cdot 10^{20}\,\mathrm{cm}^{-3}$ due to their full ionization as a result of insulator-conductor phase transition taking place at such high concentration [16], $\tau = 1$ fs is the relaxation time of hole quasi-momenta [17], m is the typical effective hole mass taken equal to 0.366 from the free electron mass [8], ω is the angular frequency of the electromagnetic field that depends in the complex form on the time t as $\exp(-i\omega t)$, the branch of the two-valued square root of the complex number shall be chosen in such a way that the real part of the refractive index is positive. At a frequency responsible for the NV-center radiation wavelength 637 nm (see above), this equation gives $2.33 + 0.03i$. Round base of the truncated cone with the refractive index equal to 2 and the base angle $\alpha = 33°$ is at 100 nm from the top surface of the diamond. Its side surface has antireflection coating.

Waveguide modes and relation between their frequencies and wave vectors (for TE-polarization when the electric

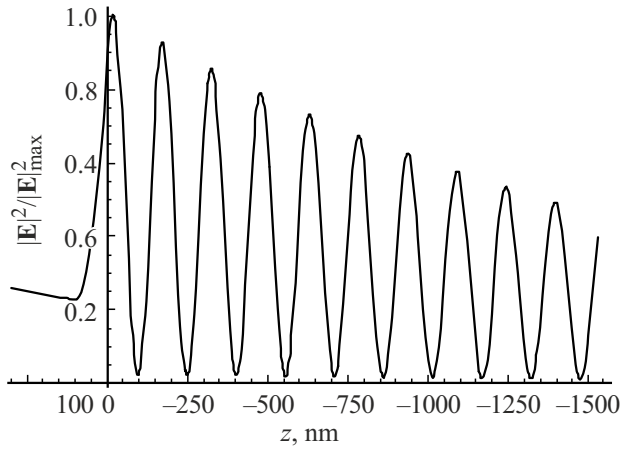

Figure 3. Squared absolute value of the complex electric field amplitude $|\mathbf{E}|^2$ of the waveguide modes normalized to its maximum value $|\mathbf{E}|^2_{\max}$ achieved at a depth of 20 nm as function of the vertical coordinate z.

field is oriented parallel to the diamond specimen surface) were calculated using the transfer matrix method [18]. This method uses the Fresnel equations to establish the relation between the amplitudes of the plane waves propagating up and down directly before and after each boundary between the layers and before and after passing each layer (whereby the air gap between the top surface of the diamond and cone base are considered as a layer with the refractive index of 1). Then, radiation boundary conditions, i.e. conditions of the absence of waves falling from the cone and substrate, are established immediately below the lower boundary of the last (from top to bottom) layer of the reflecting lattice and immediately above the truncated cone base. They lead to relation between the frequency and value of the wave vectors of the waveguide modes. Moreover, at the real frequency, the wave vectors appear to be complex, which describes the attenuation of these modes due to ohmic dissipation in the high-boron-doped diamond layers and partial escape into the truncated cone. Then, using the standard quantum electrodynamics equations in dipole approximation and the Wigner-Weisskopf method [19], probability of photon emission by the NV-center per time unit into a complex of waveguide modes is calculated and compared with the similar quantity for a complex of all other (non-waveguide) modes. The latter is calculated using the standard quantum-electrodynamic equation for radiation lifetime of the excited state of the NV-center in the undoped diamond neglecting the lattice of low-boron-doped and high-boron-doped layers.

4. Results and discussion

Computations conducted according to the above-mentioned scheme at the waveguide mode frequency ω corresponding to the NV-center radiation wavelength 637 nm show that the typical spatial field variation period of these modes in the vertical coordinate z is approximately equal to 300 nm (of squared fields — twice as lows, i.e. 150 nm, Figure 3), i.e. approximately 4 times as high as the thickness of low-boron-doped or high-boron-doped layers of the lattice (see above), which ensures effective reflection from the lattice [20]. The real part of the refractive index of the waveguide modes (i.e. relations of their complex wave vectors k to ω/c, where c is the speed of light in vacuum) is equal to 1.08. This means that, in the truncated cone with the refractive index of 2, they propagate at the angle $\mathrm{ArcCos}(1.08/2) \approx 57^\circ$ to the base, i.e. approach the cone face (forming and angle of 33° with the cone base) orthogonally, and, therefore, they are not subjected to full internal reflection on it, but almost completely (due to the antireflection coating) come out into air. As a result of computation conducted according to the above-mentioned scheme, the probability of photon emission by the NV-center per unit time into a complex of waveguide modes is approx. 4 times as low as the probability of photon emission by the NV-center per unit time into a complex of all other (non-waveguide) modes. This means that the NV-center emits approx. 20% of photons into the waveguide modes, and the rest 80% — into the other (non-waveguide) modes. Such sufficiently large radiation portion emitted into the waveguide modes is attributed to the fact that the periodic sequence of layers with various refractive indices governs the field concentration of these modes near the top surface of the diamond specimen, where the NV-centers, that excite these modes, are located, and fast attenuation of the modes when moving away from the top surface deep into the diamond. At the same time, the fields of other (non-waveguide) modes are distributed over the specimen almost evenly. The imaginary part of the wave vectors of the waveguide modes is about $2\,\mu\mathrm{m}^{-1}$. Therefore, these modes may come out through the side faces of the diamond specimen only if they are emitted by the NV-center located at a distance not more than several microns from these faces. The number of such NV-centers in a standard diamond specimen with a typical surface area of $1\,\mathrm{cm}^2$ is relatively very low, therefore, attenuation of the waveguide modes due to their escape through the side faces may be omitted. The analysis shows that the ohmic dissipation in the high-boron-doped layers and escape into the truncated cone give approximately the same contributions to the waveguide mode attenuation. This means that approximately a half of their energy escapes into the truncated cone and, therefore, is extracted from the diamond. Thus, using the waveguide modes, $20\%/2 = 10\%$ of the NV-center radiation power is extracted from the diamond, which is more than twice as high as without using the waveguide modes when the extracted radiation portion is 4.5% (Introduction).

As far as the proposed method for increasing the extracted radiation portion of the NV-centers in the diamond is not associated with the use of nanocrystals or any other diamond nanostructures and is intended for large diamond specimens, it does not lead to degradation of the NV-center parameters [11,13] that are critical for practical applications.

At the same time, in contrast with the diamond lens method [10], it is applicable to the NV-centers having any coordinates in the diamond specimen plane xy and spaced away from the top surface at a distance not more than about 30 nm (where the waveguide mode field is still close to its maximum value achieved at a depth of 20 nm, Figure 3), and not only to the NV-centers spaced away from the center of this lens at a distance of not more than 30 μm.

In contrast with the method for radiation extraction through the side faces of the diamond specimen [9], the proposed method is still effective at high concentration of centers because the waveguide modes attenuate by e times in power at a distance about 1 μm (because the imaginary part of their wave vectors is about $2\,\mu\mathrm{m}^{-1}$), i.e. before exit into the environment , photons contained in the modes cover a distance of about 1 μm in the diamond. At the same time in method [9], before exit into the environment, the photons emitted by the NV-centers repeatedly pass through the diamond specimen with typical sizes of about several millimeters, i.e. have much higher probability to be absorbed resonantly on other NV-centers than in the proposed method.

5. Conclusion

Finally, it should be noted that, for practical implementation of the proposed method for increasing the portion of color center radiation extracted from the diamond, the periodic sequence of low-boron-doped and high-boron-doped layers with the required parameters may be created during the homoepitaxial growth of diamond film by the vapor-phase deposition on the single-crystal diamond substrate as, for example, in [21]. The truncated cone with the refractive index of 2 needed for implementing the attenuated total reflection method may be fabricated from optical glass made by Ohara Corporation (USA). Glass with lower refractive index may be used, for which the angle α at the truncated cone base shall be increased, respectively. However, it should be noted that glass with a lower refractive index will cause a decrease in the contribution of escape into the truncated cone compared with the contribution of the ohmic dissipation in high-boron-doped layers to the waveguide mode attenuation, i.e. decrease in the energy portion of these modes extracted from the diamond. The lower refractive index limit of the truncated cone glass is defined by the condition of applicability of the attenuated total reflection method, i.e. is equal to the real portion of the refractive index of the refractive index of the waveguide modes that is equal to 1.08 as specified in Section 4. Diameter of the truncated cone base shall exceed the length and width of the diamond specimen so that the proposed method is applicable to any color center located near the specimen surface. Diameter of the cone's top face also shall exceed the specimen dimensions in order to provide the extraction of the NV-center radiation propagating orthogonally to the top surface of the diamond. Short (100 nm) distance from the top surface of the diamond specimen to the truncated cone base may be provided by a precision positioner made, for example, by SmarAct (Germany) and having a resolution of 1 nm. Radiation coming out from the truncated cone normally to the cone face may be transformed into a tubular beam using a cone-shaped mirror surrounding the cone. This beam may be later focused on a receiving photocell (Figure 2) using the collecting lens, thus, collection of the extracted radiation will be provided. The same lens may be used to deliver the pumping radiation exciting the NV-centers to the specimen.

Thus, the proposed method may be used to increase the color center radiation portion extracted from the diamond and collected by the optical system by a factor of two independently of the color center positions in the diamond specimen plane and color center significant concentration without degradation of their parameters that are most important for applications. The experimental implementation of the method requires only standard laboratory equipment and is feasible.

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Conflict of interest

The authors declare that they have no conflict of interest.

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