Hybrid nanodiamond and titanium dioxide nanobeam cavity design

KELVIN CHUNG,1,* TIMOTHY J. KARLE,2 CHENG WANG,3 MARKO LONČAR,3 AND SNJEZANA TOMLJENOVIC-HANIC1

1School of Physics, The University of Melbourne, VIC 3010, Australia
2The Florey Institute of Neuroscience and Mental Health, VIC 3052, Australia
3John A. Paulson School of Engineering and Applied Sciences, Harvard University, MA 02138, USA
*kelvinc@student.unimelb.edu.au

Abstract: Titanium dioxide is an emerging optical material, with a relatively high refractive index ($n \sim 2$), which allows for high confinement of the electromagnetic field. Extensive research has been conducted on the negatively charged nitrogen vacancy centre in diamond due to its robust electronic and optical properties. In particular, its stable room-temperature photoluminescence properties have been considered for quantum optical applications. Nanobeam cavities are a geometry that offer exceptionally large-$Q$ values given their minimal footprint, a must for high density and compact optical architecture. This paper presents an ultrahigh-$Q$ nanobeam cavity within titanium dioxide in a low-refractive index environment operating at the negatively charged nitrogen vacancy centre of diamond. This research opens the possibility of hybrid optical devices utilising titanium dioxide and diamond.

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1. Introduction

Titanium dioxide (TiO\textsubscript{2}) has recently emerged as a promising optical material in which waveguide and resonator structures have been fabricated [1–13]. It is a wide bandgap semiconductor with a bandgap energy range of 3–3.3 eV which is dependent on the crystalline phase [14]. It has a high refractive index for visible wavelengths ($n = 2.3$ at 550 nm [15]), allowing for strong light confinement, and is also optically transparent. Amorphous and polycrystalline TiO\textsubscript{2} thin films can be deposited using conventional methods, such as electron-beam evaporation and ion sputtering [16], allowing for on-chip integration with other materials.

Diamond has garnered interest as a future photonic material [17] and has a similar refractive index to TiO\textsubscript{2}. Research has focused on enhancing the emission of the zero-phonon line (ZPL) of the negatively charged nitrogen vacancy (NV\textsuperscript{−}) colour centre [18–24]. The ZPL emission has a narrow linewidth which constitutes 3–5% of the total NV\textsuperscript{−} photoluminescence and enhancement can be achieved by coupling this emission to a high-$Q$ optical cavity through the Purcell effect. The magnitude of enhancement is quantified by the Purcell factor which is dependent on the ratio between quality factor and modal volume ($\sim Q/V$); the larger this ratio, the greater the enhancement of the spontaneous emission rate.

Diamond substrates for photonic applications can be classified as either: individual micro- and nano-crystalline diamond (ND); polycrystalline diamond films and single-crystal diamond. The last classification is the leading candidate for diamond photonics, however it requires an intensive processing effort to engineer devices into the single crystal morphology. Individual ND have also attracted interest and the intrinsic fluorescence for the NV colour centre has been used for bioimaging applications [25]. Given the similar refractive indices of TiO\textsubscript{2} and diamond, it is a natural progression to combine both materials for hybrid applications where the former has already been shown to be an emerging visible photonic material [1, 2, 7, 10–13].

An optical cavity geometry that offers high-$Q$ with minimal footprint is known as the nanobeam cavity; it is a photonic crystal cavity with periodic refractive index change in one-dimension. High-$Q$ designs have been theoretically proposed (on the order of $10^7$) [26–30] and experimentally demonstrated (in the order of $10^5$) [27, 31, 32]. Previous high-$Q$ designs generally considered high refractive index contrast environments ensuring the light was concentrated in the cavity region.

In this article, we present an ultrahigh-$Q$ TiO\textsubscript{2} nanobeam cavity design with a quality factor in the order of $10^7$ operating at the ZPL of NV\textsuperscript{−} of diamond. 3D FDTD simulations show that an ultrahigh-$Q$ cavity is possible in this low refractive index contrast environment using a relatively simple design recipe.

2. TiO\textsubscript{2} nanobeam geometry and model

The design for this optical cavity was based on previous work by Quan et al. [33,34] which has a design philosophy following three main rules: (1) the cavity length should be zero, i.e. no defect is introduced into the periodicity; (2) the periodicity was kept constant; and (3) the requirement for a modulated Bragg mirror (referred in the rest of this letter as the Gaussian mirror). Rule (1) ensures the modal volume ($V$) is kept to a minimum alongside a high-$Q$ cavity will result in a large $Q/V$ ratio necessary for strong cavity interactions. Rule (2) guarantees a constant phase velocity between the periodic elements, i.e. the airholes. Finally, rule (3) results in a gentle modulation of the electric field between the Bragg mirrors which was achieved by a Gaussian envelope function [35]. The modulation maximises the $Q$ for the nanobeam by minimising the Fourier components that exist above the light line, i.e. radiation loss into free space. It must be mentioned, another loss mechanism within our system was due to the additional mirror section which tapers into a feeding waveguide. If there are not a sufficient number of airholes in this section, leakage of light from the modulated Bragg mirror section can occur. This leakage was
minimised by simply adding more airholes within the additional mirror section. Figure 1 shows a cross-sectional view at the cavity centre with a TiO$_2$ trapezium profile resultant of the etching recipe by Bradley et al. [6], and the top-view schematic of the nanobeam showing different sections: additional mirror (A), where the airhole size tapers down and becomes a waveguide; Gaussian mirror (G) and the centre (C).

The nanobeam’s periodicity consisted of airholes perforating the TiO$_2$ layer which sits upon an SiO$_2$ layer. This oxide layer was grown by oxidation of the silicon wafer and we choose to keep this layer as to demonstrate a high-$Q$ cavity is possible in a low refractive index environment. The silicon substrate was not included in the simulations. The resonant wavelength of the nanobeam is chosen to be at the ZPL of the NV$^-$ centre which is 637 nm.

The original design recipe by Quan et al. [33, 34] assumed vertical side walls only. Angled side walls was an appropriate consideration since we based the design on the etching recipe in Ref. [6]. The thicknesses of the TiO$_2$ and SiO$_2$ were set to $t_{\text{TiO}_2} = 0.27 \mu$m and $t_{\text{SiO}_2} = 0.99 \mu$m. Finally, the refractive indices of TiO$_2$ and SiO$_2$ were set to 2.27 and 1.46 respectively.

Three-dimensional FDTD simulations (Lumerical) were performed to determine the $Q$ of the cavity. An electric dipole was placed closed to the centre of the nanobeam cavity at coordinates of $(x, y, z) = (0, 0.2, 0) \mu$m. The spatial grid size was 22 nm; the PML thicknesses were different at each boundary: 2.51 $\mu$m (top of nanobeam), 2.37 $\mu$m (sides of the nanobeam), and 400 nm (longitudinal directions of the nanobeam). Vertical airholes were considered for all the simulations.

### 3. Results and discussion

Before exploring the significant enhancement achieved from the nanobeam, it is worth examining the electrodynamics for a simple system first. The emission rate of an emitter in a homogenous refractive index environment can be modified via simple encapsulation in a planar interface environment. The NV$^-$ centre can be approximated as a classical oscillating dipole radiating within a dielectric sphere near an interface [36, 37]. The power radiated by the dipole is inversely proportional to the emitter’s fluorescence lifetime. Three-dimensional FDTD simulations (RSoft) were considered for three unstructured environment scenarios: (1) ND in air,
(2) ND in TiO$_2$ and (3) ND on TiO$_2$ in air. The refractive indices were taken to be: $n_{ND} = 2.4$, $n_{air} = 1$ and $n_{TiO_2}$. The dipole resides at the centre of the spherical ND with a diameter of 60 nm and has an emission wavelength of 637 nm. For scenarios (1) and (2), the dipole is emitting into a homogenous environment. For scenario (3), there is a planar interface between the TiO$_2$ and the surrounding air. This interface acts as the reference point for parallel ($\parallel$) and orthogonal ($\perp$) dipole polarisations. The total radiated power by the dipole ($P_S$) was calculated by summation over the time-averaged Poynting vector of six faces of a cubed power monitor when the simulation had reached a steady state solution. The computational domain size was $1 \times 1 \times 1$ $\mu$m$^3$, the spatial grid size was 3 nm and PML was 1 $\mu$m for all simulations. The enhancement ($\zeta$) was calculated as the ratio between $P_S$ and the total power radiated from the ND in air (i.e. scenario (1)) for both parallel and orthogonal dipole polarisations. Table 1 summarises these numerical FDTD results.

Table 1. The enhancement of parallel and orthogonal dipole polarisations for 637 nm emission in various refractive index distributions. The polarisations are measured with respect to the TiO$_2$–air interface (scenario 3).

| Scenario | $\zeta_\parallel$ | $\zeta_\perp$ |
|----------|-------------------|----------------|
| ND in air | 1                 | 1              |
| ND in TiO$_2$ | 18.29           | 18.27          |
| ND on TiO$_2$ in air | 3.83            | 1.58           |

The simulation results show that preferential enhancement in the parallel orientation of the dipole. It shows that if a ND is any high refractive index environment there will be an enhancement of the emission rate without any structured environment such as an optical cavity.

To obtain a sufficiently high-$Q$ cavity, it was found that the Gaussian mirror section should contain fifty airholes (see Figure 1(b), G sections). In addition, to best reflect an experimental situation where the nanobeam can be characterised, an additional mirror (A, see Figure 1(b)) section was also included in the simulations which was tapered down to a feeding waveguide [33]. The additional mirror section consisted of ten airholes. The total length of the nanobeam was then determined to be 20.26 $\mu$m, measured from the outer edges of the tenth airholes of the additional mirror sections.

Figure 2 shows the variation in the calculated $Q$ values and their associated resonance wavelengths for vertical sidewalls for the TiO$_2$ nanobeam cavity. The trend shows that by increasing the number of airholes in the Gaussian mirror, the $Q$ value increases with a blue shifting resonant wavelength. This means that the value chosen for G is arbitrary, any value greater than 40 was sufficient in obtaining a high-$Q$ value. With more airholes this will impart more partial reflections on the light and results in stronger enhancement without sacrificing a large shift in a resonance wavelength. A value of G = 50 was chosen as an appropriate starting point in accomplishing an ultrahigh-$Q$ cavity. The resonance wavelength for G = 50 mirror was found to be 635.15 nm, approximately a 2 nm difference with respect to the ZPL NV$^-$ wavelength. This can be shifted to 637 nm by either: changing the starting or ending filling fractions of the Gaussian mirror (equivalent to changing the radii of the airholes) or adjusting the width of the nanobeam. The latter can drastically shift the resonance wavelength as more modes fall below the substrate light line.

As mentioned in the previous subsection, to best reflect an experimental situation when the TiO$_2$ is etched, a sidewall angle was considered for the waveguide; this results in the resonance wavelength of the vertical sidewall nanobeam shifting from 635.15 nm. A parameter search of the top and bottom widths of the TiO$_2$ was conducted to target the 637 nm wavelength with the trapezium profile of the nanobeam. At widths of $w = 0.38$ $\mu$m and $s = 0.52$ $\mu$m, the L0 TiO$_2$ nanobeam cavity yielded a $Q = 1.04 \times 10^7$ with a resonance wavelength of 637.37 nm and
Fig. 2. $Q$ values with their respective resonance wavelengths for the TiO$_2$ nanobeam as a function of the number of airholes that constitute both Gaussian mirrors (G). The results shown in this figure were for vertical sidewalls with a width of $w = 0.44$ μm and a periodicity for the airholes of 0.17 μm.

$V = 3.7(\lambda/n)^3$. This results in a Purcell factor of $F = (3/4\pi^2)(\lambda/n)^3(Q/V) \approx 2.14 \times 10^5$, a considerably large value.

This represents a substantially large $Q$ for an optical cavity in a low refractive index contrast environment compared to previous work in polymeric nanobeam cavities [38]. However, the modal volume is large due to the presence of the SiO$_2$ substrate which results low refractive index contrast between ($n_{TiO_2}/n_{SiO_2} = 2.27/1.46 = 1.55$). The refractive index difference between TiO$_2$ and the SiO$_2$ substrate is smaller than with TiO$_2$–air and therefore the mode penetrates into the SiO$_2$ rather than being reflected back for a high refractive index difference which would be the case for a free-standing nanobeam, i.e. an all air environment.

Figure 3 cross-sectional and top-view power distributions that were calculated for the L0 TiO$_2$ nanobeam cavity with $w = 0.38$ μm and $s = 0.52$ μm.

Fig. 3. The power distribution of the TiO$_2$ nanobeam. (a) Cross-sectional and (b) top-view profiles. The dashed white lines represent the sidewalls of the nanobeam. Note: by default, Lumerical measures the Poynting vector in their power monitors.

The power profiles reveal that the majority within the cavity centre at $(x,z) = (0,0)$ in Figure 3. The results show that this nanobeam design has the potential to significantly enhance
the ZPL emission from the NV$^-$ centre for an embedded ND. In the vertical direction ($y$), the peak power distribution is slightly below the centre of the cavity and therefore this would need to be a consideration when embedding the ND into the growth process of the TiO$_2$ layer. This is only for the case that an appropriate ND size can be supported between the Gaussian mirrors at the cavity centre.

The results show that an ultrahigh-$Q$ TiO$_2$ nanobeam cavity can be realised to operate at the ZPL of the NV$^-$ centre for diamond. The $Q$ can be made arbitrarily high by simply increasing the number of airholes that constitute the Gaussian mirrors. This will inherently shift the resonance wavelength slightly, but a parameter search can be conducted on any physical aspect of the nanobeam to re-adjust the resonance wavelength. The only drawback for this short wavelength operation is the critical dimensions of the design become increasingly small which makes it more difficult to fabricate and is limited to current lithographic resolution.

In addition, incidental losses, those not included in the design, will impact the fabricated device. Unintentional refractive index changes can form scattering sites such as grain and mask inhomogeneities. Figure 4 shows a scanning electron micrograph of a the surface of a TiO$_2$ substrate created via electron-beam deposition that would be considered for the fabrication of the device.

![Fig. 4. A scanning electron micrograph of the TiO$_2$ substrate with embedded NDs.](image)

If a TiO$_2$ nanobeam was to be fabricated using this particular substrate, the optical loss would be significant, and therefore a reduced $Q$ value, due to scattering of the light because the grain boundaries are significantly smaller compared to the total length of the nanobeam (20.26 $\mu$m).

Another point of discussion is the absence of a ND encapsulating the electric dipole, i.e. a ND-TiO$_2$ interface, within our simulations and incorporating this would be a much more realistic estimation of the physical system. Since the refractive indices of TiO$_2$ ($n_{\text{TiO}_2} = 2.27$) and diamond ($n_{\text{ND}} = 2.4$) are comparable, therefore at the interface there will be minimal scattering of the light. McCutcheon et al. [39] showed that $Q$ values for silicon nitride ($n = 2.0$) nanobeams reduced by approximately 10–20% from a bare cavity when compared to an embedded 20 nm cubic ND in the cavity centre. In addition, van der Sar et al. [40] also explored the positional effects of a ND on the optical properties of gallium phosphide (GaP) S1 and L3 cavities. In this system, the refractive index contrast is much larger because GaP has a refractive index of 3.4. When a ND of 50 nm diameter is placed at the maximum mode intensity of the S1 cavity the original $Q$ value decreases by <10%. When a 60 nm ND is placed at various depths within the GaP substrate, the $Q$ value is greatest near the centre (5% of the original) and decreases near the edges because light is scattered by the ND at the surface is lost from the cavity. Therefore, previous work indicates that ND embedded within a substrate does not significantly change the $Q$ value, only when it’s near the surface with a larger refractive index contrast will there be increased losses due to the scattering.
4. Conclusions

It has been shown that TiO$_2$ can be a viable material for creating an ultrahigh-$Q$ nanobeam cavity at the ZPL of the NV$^-$ centre. Using the design recipe by Quan et al. [33, 34], a $Q$ of $1.04 \times 10^7$ at 637.37 nm can be achieved. The $Q$ value can be arbitrarily increased by additional airholes within the Gaussian mirror. This paves the way for TiO$_2$ to become a future visible photonic material.

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