Improving photon pair generation in silica nanofibers through PMMA/DR1 nonlinear coating optimization

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Abstract. We report on the use of PMMA/DR1 coating to enhance the efficiency of photon pair generation in silica nanofibers. The coating improves the second-order nonlinear susceptibility of the nanofibers, leading to improved photon pair generation efficiency. We investigate the effect of varying the nonlinear optical properties of the composite material, and we characterize the photon pair generation efficiency of the coated silica nanofibers. Our modelling results show a significant enhancement in photon pair generation efficiency by a factor of 1000 compared to a bare silica nanofiber.

1 Introduction

Photon pair generation is an important process for various quantum technologies such as quantum cryptography, quantum communication, and quantum computing. The generation of correlated photon pairs by Spontaneous Parametric Down-Conversion (SPDC) can be performed in bulk or periodically polarized crystals such as lithium niobate [1] or in semiconductor waveguides. One of the disadvantages of these techniques is the difficulty of insertion into fiber networks due to coupling losses. The use of silica optical fibers allows to get rid of these coupling problems. However, since silica is a centrosymmetric material, it does not exhibit second order nonlinearity in its solid form, so it must be artificially created. All-fiber sources based on the SPDC process in periodically polarized silica fibers have been proposed [2] but due to technical difficulties, the interaction lengths are limited to a few centimeters. Silica nanofibers have the advantage that they can be inserted into fiber networks with a minimum of coupling losses, and the control of their geometry offers many possibilities for controlling light propagation. They are identified as promising platforms for efficient photon pair generation due to their high surface area, low loss, and compatibility with a wide range of optical fibers. However, their inherently low second-order nonlinear susceptibility limits their efficiency for photon pair generation. PMMA/DR1 coated silica nanofibers have been studied for enhancing the second-order nonlinear susceptibility of the composite material. The PMMA/DR1 composite material has been shown to exhibit a second-order nonlinear susceptibility of 27±5 pm/V [3]. The combination of the high nonlinear optical properties of the DR1 chromophore and the strong adhesion of PMMA to silica surfaces makes the PMMA/DR1 composite an attractive coating material for silica nanofibers. The enhancement in second-order nonlinear susceptibility is attributed to the strong electric field confinement and the high surface-to-volume ratio of the nanofibers, which can increase the effective nonlinear coefficient of the composite material. The nonlinear optical properties of PMMA/DR1 coated silica nanofibers can also be tuned by adjusting the thickness of the coating, the concentration of the DR1 chromophore, and the fiber dimensions.

In this work, we demonstrate that coating silica nanofibers with a thin layer of PMMA/DR1 composite material significantly enhances their second-order nonlinear susceptibility and photon pair generation efficiency. We investigate the effect of composite material's nonlinear optical properties and show that PMMA/DR1-coated silica nanofibers exhibit significant improvement in photon pair generation efficiency compared to bare silica nanofibers, mainly due to the higher nonlinearity of the PMMA/DR1 composite material.

2 Theory and modeling

We present a novel source of correlated photon pairs based on SPDC in a silica nanofiber. The correlated photons are generated through surface dipole and bulk multipole nonlinearities. Its principle is illustrated in Figure 1. Since silica is an isotropic material, phase matching by birefringence is not allowed in a silica nanofiber. Therefore, we opted for modal phase matching. It is verified using a pump photon in the TM_{01} mode (or the HE_{21} mode, depending on the phase matching) at wavelength λ_p (i.e., frequency ω_p) to generate photon

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pairs at both wavelengths λ_s and λ_i (i.e., frequencies ω_s and ω_i) in the fundamental HE_{11} mode such that $\omega_p = \omega_s + \omega_i$ for energy conservation. The $TM_{01} - HE_{11}$ phase matching is more efficient than the $HE_{21} - HE_{11}$ interaction due to the efficient nonlinear overlap between the TM_{01} and HE_{11} modes. Additionally, the TM_{01} mode is easier to generate due to its simpler spatial transverse structure. Therefore, we exclusively consider the $TM_{01} - HE_{11}$ interaction for photon pair generation, utilizing a TM_{01} pump mode at 775nm to generate photon pairs in the HE_{11} mode around 1550nm. We focus solely on the photon pair generation initiated by quantum vacuum fluctuations in the absence of an initial wave at ω_s or ω_i.

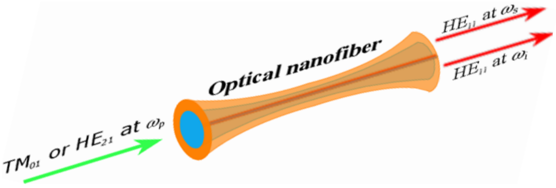

Fig. 1. SPDC principle in a PMMA/DR1-coated silica nanofiber. Blue indicates the silica core material, while orange represents the PMMA/DR1 nonlinear coating at the surface of the nanofiber.

The photon pair generation spectral density per pump mode in a 100µm long silica nanofiber can be calculated for the non-degenerate case as follows,

$$G(L, \omega_s) = g^2 L^2 \mathrm{sinc}^2\left[\sqrt{\left(\frac{\Delta\beta}{2}\right)^2 - g^2 L}\right] \quad (1)$$

Here, $g^2 \approx \rho^2 a^2(\omega_p)$, where $a(\omega_p)$ represents the square root of the power at the pump frequency ω_p, and $\Delta\beta$ is the phase mismatch. We assume that ρ, which is the efficiency of the sum frequency generation process (i.e., the reverse process of SPDC), is constant over the wavelength range considered [4].

3 Results and discussions

We estimate photon pair generation spectral densities for two configurations: bare silica nanofibers (silica-air) and PMMA/DR1-coated silica nanofibers (silica-PMMA/DR1). Figure 2a and 2b show the respective spectral densities. Using a narrow spectral width 775nm laser as a pump in the TM_{01} mode, we generate two pairs of photons near 1550nm in the fundamental HE_{11} mode. Photon pair generation efficiency depends on the uniformity of the nanofiber's diameter after fabrication, which is less critical in the silica-PMMA/DR1 configuration, making it the optimal choice for maximizing photon pair generation rates. For this configuration, we use a 100µm long silica nanofiber with a diameter uniformity of around 50nm. The optimal PMMA/DR1 coating thickness in the simulations was 140nm. Photon pair generation spectral density increases with the square of the nanofiber length but narrows the radius tolerance, varying inversely with the length L. We integrate over half the emission spectrum (the medium being defined for a 1550nm wavelength). Filtering at the output reduces the number of detected pairs. The silica-PMMA/DR1 configuration generates more photon pairs due to its higher spectral density and wider phase matching curves.

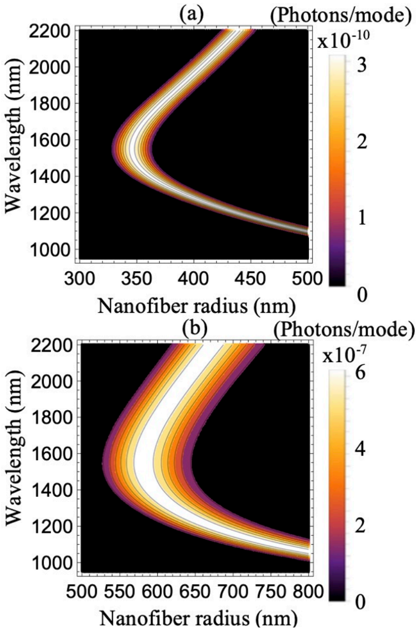

Fig. 2. Photon pair generation spectral densities in the HE_{11} mode for a 100µm long silica nanofiber in (a) silica-air and (b) silica-PMMA/DR1 configurations.

We have indeed experimentally deposited PMMA/DR1 on µm range diameter nanofibers with a thickness of ~100nm. The coating thickness was found to decrease linearly with fiber diameter, which may limit the thickness reachable on smaller fibers with a single layer coating. Thicker deposits can however be made on smaller fibers with multilayer coatings.

4 Conclusion and perspectives

Our work proposes a promising approach to enhance the nonlinear optical properties of PMMA/DR1 coated silica nanofibers for efficient photon pair sources in quantum technologies. Other coatings can improve the diameter tolerance for longer nanofibers and maximize correlated photon pair generation. We achieved over 1000-fold enhancement in photon pair generation efficiency compared to bare silica nanofibers.

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