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**Quantum Optics in Diamond Nanophotonic Chips**

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PROGRESS REPORT

**Program Manager: Dr. Gernot Pomrenke**

# QUANTUM OPTICS IN DIAMOND NANOPHOTONIC CHIPS

**AFOSR Directorate: Physics and Electronics**

**Research Areas:**

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## Abstract

This report summarizes our recent progress on the development of photonic nanostructures for sensing and information processing. We have developed a nitrogen vacancy (NV) spin qubit-cavity system in the strong Purcell regime in which the spin qubit predominantly interacts with the cavity mode. This system is realized in a single-crystal diamond photonic crystal nanocavity with quality factors (Q) up to  $10^4$ . Furthermore, we measure electron spin coherence times of cavity-coupled NVs of 200  $\mu$ s. We have also developed techniques for NV-based super-resolution imaging and precision sensing, using high-speed parallel readout on a microscope camera; and we have developed techniques for precision clocks based on collective oscillation of ensembles of NVs. We also report on graphene electro-optic devices and sensors.

# 1 Progress towards on-chip quantum network in diamond

The field of quantum information processing (QIP) takes advantage of the properties of quantum mechanics to perform tasks that are classically impossible, including exponentially faster computational algorithms, unconditionally secure communication, and efficient simulation of complex physical and biological models [1, 2]. A number of recent theoretical and experimental advances have identified promising candidate systems for the implementation of quantum information technologies. A key requirement of such a system is the ability to create shared quantum entanglement among a large number of quantum memories that are individually addressable. This entanglement can be created through atom-photon interactions, allowing the establishment of quantum networks for the implementation of theoretical protocols, including the teleportation of distant stationary qubits via photons [3], the conversion of quantum states of light and motional states of atoms [4], and quantum computation through cavity-assisted interactions [5]. The quantum network requires long-lived quantum memories, such as particle spins, that can be efficiently mapped to photons through high-fidelity quantum interfaces. While several atomic systems, such as trapped ions, now fulfill these requirements, there is strong interest in solid-state implementations for scalability, stability, and device integration.

Among solid-state qubits, the negatively charged nitrogen vacancy ( $\text{NV}^-$ ) center in diamond has in recent years emerged as one of the most promising systems. The NV center consists of a nitrogen atom adjacent to a vacancy in the diamond lattice. This lattice point defect creates an atom-like electronic wavefunction with well-defined spin and orbital angular momentum. Spin-selective optical transitions allow individual NV electron spins to be easily observed using standard confocal microscopy. The NV has two unpaired electrons that naturally provide a ground state with three electron spin sublevels. Two sublevels are employed to encode a quantum bit (qubit). Because of the nearly nuclear spin-free carbon lattice and weak spin-lattice interactions, these electronic ground states have extremely long coherence: milliseconds-scale phase coherence times have recently been reported [6] at room temperature, longer than any other solid state electron spin qubit. Furthermore, the electronic spin state may be coupled to proximal nuclear spins, providing additional degrees of freedom that have been employed as quantum logic gates [7]. A primary goal of this

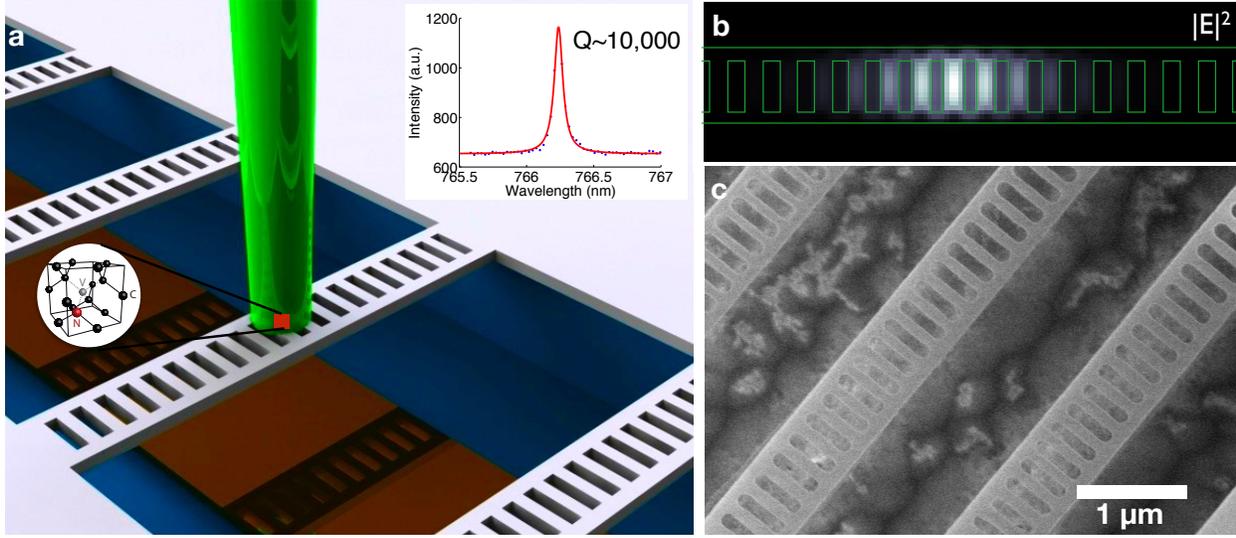


Figure 1: (a) The single-crystal diamond cavities are suspended over a microwave wire for spin manipulation. Fluorescence excitation and collection is from top through a confocal microscope. (b) The cavity design results in a maximum electric field in the diamond at the center to optimize coupling to the NV. (c) Scanning electron microscopy images of the cavities.

program is to develop techniques to entangle distant NV spin qubits by optical interactions. To this end, we have developing techniques to incorporate NV centers with long spin lifetime into optical resonators, and to efficiently interface these with photons. However, the lack of thin film technologies for crystalline diamond with low impurity levels hampers development of photonic interfaces to such diamond-based qubits. Over the course of this program, we have developed several methods for manufacturing slabs of diamond of 200 nm thickness and several microns in extent from high purity single crystal CVD diamond [8, 9, 10].

## 1.1 On-chip Quantum Control of a Cavity-Coupled NV system in Diamond

In recent months, we have also succeeded in a primary goal of this program – the fabrication of high-quality photonic crystal nanocavities in single crystal diamond. These cavities and related structures, such as waveguides, were fabricated by first thinning 10-um thick diamond membranes (produced by mechanical polishing by Element 6) and then thinned using Cl and oxygen reactive ion etching chemistries to  $\sim 200$  nm.

We employ a one-dimensional ladder PhC cavity design for maximal emitter enhancement and increased collection efficiency of cavity-coupled ZPL photons. The ladder PhC device consists of a suspended diamond waveguide patterned with a one-dimensional lattice of rectangular air gaps that defines a periodic dielectric profile, as shown in Fig.1. Low-temperature measurements indicate a Purcell enhancement of the zero-phonon-line (ZPL) of cavity-coupled NVs in excess of 60. An example of such a spectrum is shown in Fig. 2, where the spectrally selective enhancement of the ZPL is estimated at 122. This results in

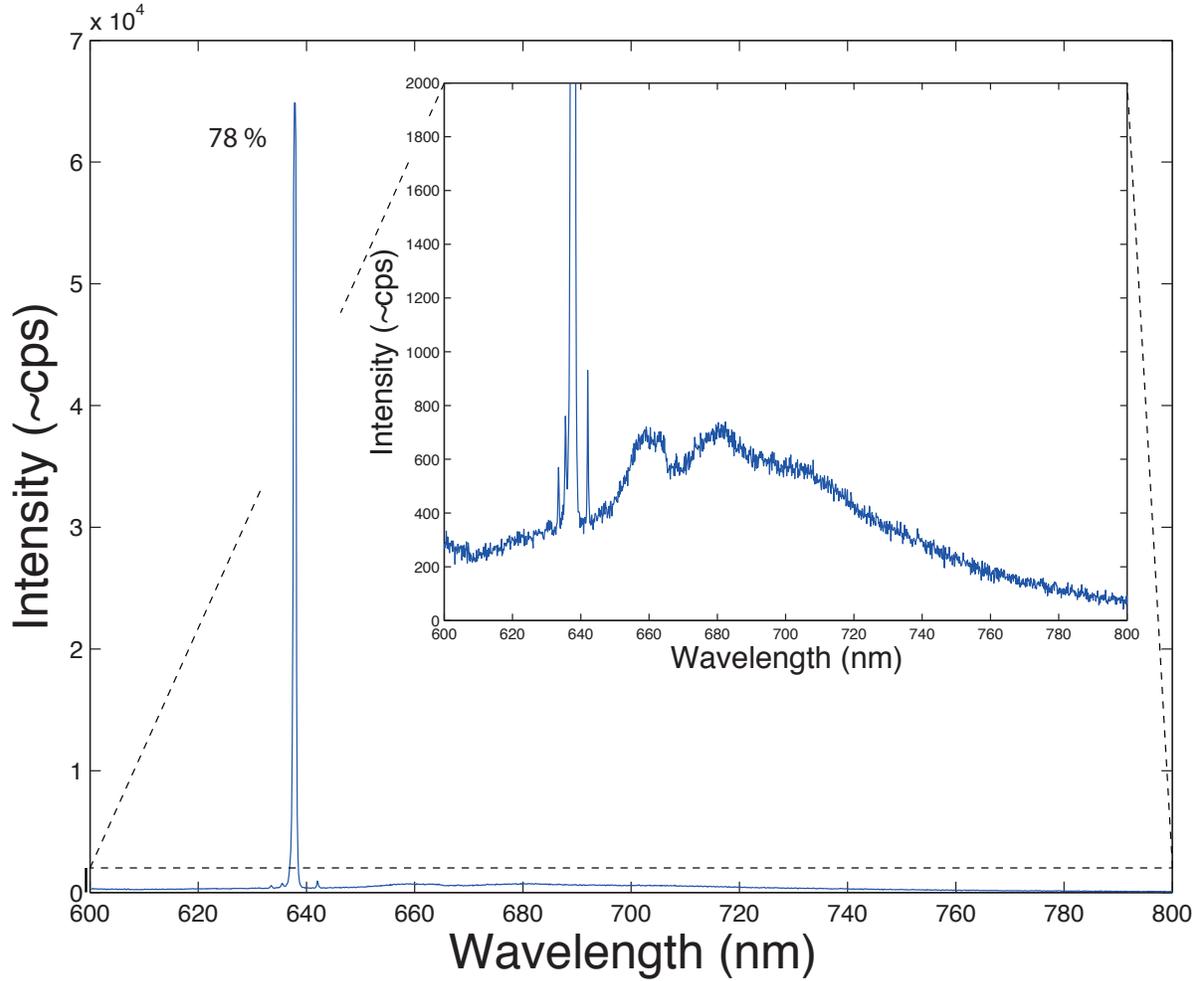


Figure 2: Coupled NV-cavity spectrum, showing an enhancement of the NV ZPL transition rate of  $\sim 62$ .

more than 80% emission into the ZPL, compared to just  $\sim 3\%$  for naturally occurring NVs.

The spin coherence times of NVs coupled to such cavities is similar to that observed in high-purity bulk diamond. We measured the coherence times using a confocal microscope setup and a microwave strip line integrated directly underneath the diamond cavities, as illustrated in Fig. 1. Our measurements indicate a phase coherence time in excess of  $200\mu\text{s}$ , as evaluated using a Hahn-echo protocol [10]. This represents a two order of magnitude improvement in the coherence time of solid state quantum memories coupled to cavity-based spin-photon interfaces. We are currently implementing dynamic decoupling spin protocols, which puts the spin coherence times of such emitters beyond 1 ms.

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## 1.2 Scalable Fabrication of High Purity Diamond Nanocrystals with Long-Spin-Coherence Nitrogen Vacancy Centers

The nitrogen vacancy (NV) center in nanodiamond has been the focus of many recent investigations across a broad range of applications, including its use as a spin qubit in a hybrid photonic architecture and as highly localized sensor of temperature and magnetic fields that can be integrated with biological systems. The performance of the NV for these applications depends crucially on its electron spin phase coherence time, which is limited to microseconds in high-pressure high-temperature (HPHT) diamond nanocrystals due to a high concentration of paramagnetic impurities. With PECASE support, we have demonstrated a top-down fabrication process using a porous metal mask and a self-guiding reactive ion etching process that enables rapid nanocrystal creation across the entirety of a high-quality chemical vapor deposited (CVD) diamond substrate. High-purity CVD nanocrystals produced in this manner exhibit NV phase coherence times in excess of 200  $\mu$ s. This record-long spin coherence time enabled record-precision magnetometry with diamond nanocrystals [11].

## 1.3 Wide-Field Multispectral Super-Resolution Imaging Using Spin-Dependent Fluorescence in Nanodiamonds

Recent advances in fluorescence microscopy have enabled spatial resolution below the diffraction limit by localizing multiple temporally or spectrally distinguishable fluorophores. With PECASE support, we introduced a super-resolution technique that deterministically controls the brightness of uniquely addressable, photostable NV centers, whose brightness is modulated deterministically by optically detected magnetic resonance techniques [12]. Using a CCD camera, this “deterministic emitter switch microscopy” (DESM) technique enables super-resolution imaging with localization down to 12 nm across a  $35 \times 35 \mu\text{m}^2$  area. DESM is particularly well suited for biological applications such as multispectral particle tracking since fluorescent nanodiamonds are not only cytocompatible but also nonbleaching and bright. We observe fluorescence count rates exceeding  $1.5 \times 10^6$  photons per second from single NV centers at saturation. When combined with emerging NV-based techniques for sensing magnetic and electric fields, DESM opens the door to rapid, super-resolution imaging for tracking and sensing applications in the life and physical sciences.

## 2 Graphene Photonics

Graphene features unique optical properties. Strong light-matter coupling yields an unexpectedly high opacity for an atomic monolayer with a startlingly simple value: it absorbs  $\pi\alpha \approx 2.3\%$  of white light, where  $\alpha = e^2/\hbar c \approx 1/137$  is the fine-structure constant. This is “a consequence of the unusual low-energy electronic structure of monolayer graphene that features electron and hole conical bands meeting each other at the Dirac point.” [13] Graphene’s optical absorption, and generally the light-graphene interaction, can be further increased by enhancing the light-matter interaction using waveguides or cavities. With support of this PECASE program, we have developed high-contrast graphene opto-electronic modulators [14, 15] based on graphene integrated in photonic crystal nanocavities. We have reported

operating speed in excess of 1 GHz [16], and are currently working on a new generation of devices that indicate substantially faster modulation speed.

Graphene photodetectors have recently attracted much attention because of the material’s fast carrier dynamics and the possibility of photodetection as fast as 500 GHz, as well as intrinsic carrier multiplication effects in graphene. A remaining problem concerns the limited optical absorption in graphene, which results in a low optical responsivity. We addressed this problem by coupling a graphene photodetector to a silicon-on-insulator waveguide, see Fig. 4. The extended interaction between graphene and the silicon waveguide enables a high responsivity of photodetection of 0.108 A/W, competitive with current non-avalanche Ge detectors for silicon photonics integration [17, 18]. Owing to the high mobility of both electrons and holes, the photodetector displays a high frequency response (up to 20 GHz) even under zero bias operation with a responsivity of 16 mA/W. We have also demonstrated data transmission at up to 12.5 Gbps [19]. We recently also demonstrated a photonic cavity-integrated high-responsivity graphene photodetector [20], which enables wavelength-selective receivers with ultra-small footprint on photonic integrated circuits.

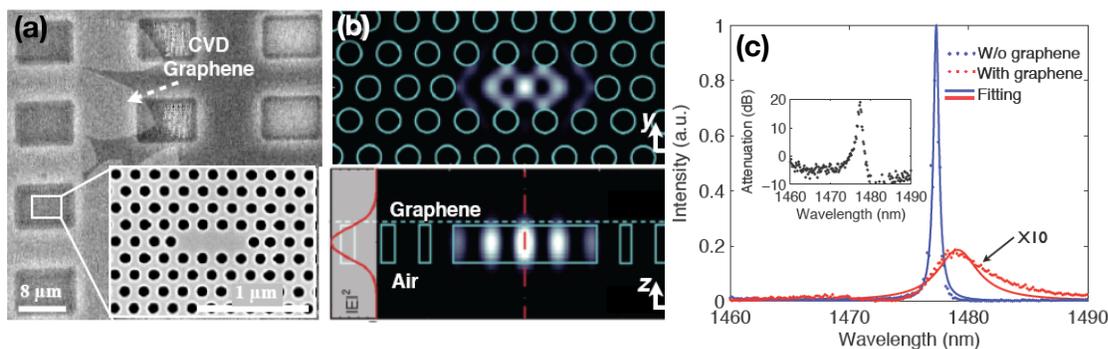


Figure 3: Enhancement of coupling from free space modes to graphene. (a) Scanning electron microscope image of PPC cavities integrated with single-layer graphene. The darker shade defines the edges of the graphene monolayer. (b) Simulated energy distribution of fundamental resonant mode of the L3 cavity, shown in plane (top) and in cross-section (bottom). The graphene layer interacts with the evanescent field. (c) The single atomic layer of graphene couples strongly to the cavity field, sharply reducing the cavity quality factor by over  $100\times$ . The strong interaction allows for cavity-enhanced Raman spectroscopy on sub-wavelength regions of a graphene sample [14] as well as high-contrast electro-optic modulation [15].

### 3 Polymer Photonics for Information Processing and Sensing

Polymers have appealing optical, biochemical, and mechanical qualities, including broad-band transparency, ease of functionalization, and biocompatibility. However, their low refractive indices have precluded wavelength-scale optical confinement and nanophotonic applications in polymers. With PECASE support, we developed a suspended polymer photonic crystal (SPPC) architecture that enables the implementation of nanophotonic structures typically limited to high-index materials [21]. We demonstrated nanophotonic band-edge filters, waveguides, and nanocavities featuring quality (Q) factors exceeding 2,300 and mode

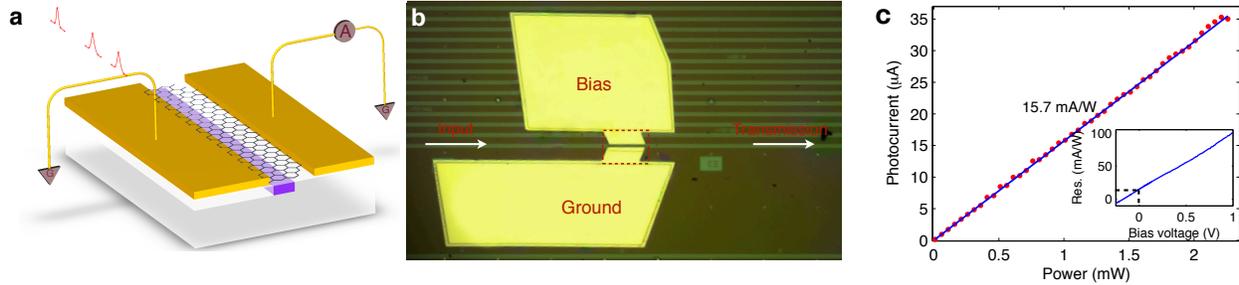


Figure 4: A waveguide-integrated graphene photodetector. (a) Illustration of the device. The silicon bus waveguide with cross-section of 220 nm by 520 nm is fabricated on a SOI wafer and then planarized using SiO<sub>2</sub>. A graphene layer is transferred onto the planarized waveguide and is separated from the waveguide by a  $\sim 10$  nm-thick SiO<sub>2</sub> layer. Two metal electrodes contact the graphene and conduct the generated photocurrent. One of the electrodes is closer to the waveguide to create a potential difference on graphene coupling with the evanescent field of the waveguide, which enables the ultrafast and efficient photodetection. **b**, Optical micrograph of the device with a bi-layer graphene covering on the waveguide. Two Ti/Au (1/40 nm) paddles are evaporated onto the graphene sheet. (c) Measured photocurrent of the device with respect to the incident power at zeros bias, showing a photoresponsivity of 15.7 mA/W. Inset: Bias dependence of the photoresponsivity. The used laser has a wavelength of 1550 nm.

volumes below  $1.7(\lambda/n)^3$ . The unprecedentedly high  $Q/V_{mode}$  ratio results in a spectrally selective enhancement of radiative transitions of embedded emitters via the cavity Purcell effect with an enhancement factor exceeding 100. Moreover, the SPCC architecture allows straightforward integration of nanophotonic networks, shown here by a waveguide-coupled cavity drop filter with sub-nanometer spectral resolution. The nanoscale optical confinement in polymer promises new applications ranging from optical communications to organic opto-electronics.

Following on our recent demonstration of high- $Q$  cavities and networks in polymer photonic crystals [21], we have also developed nanophotonic 1D polymer photonic crystal resonators that confine light at the wavelength scale with extremely low losses, enabling quality factors  $Q$  in excess of 8000 (limited by our detector resolution) at 600-700 nm. These are, to our knowledge, the highest quality factors achieved in photonic crystal cavities in this wavelength range. We have now employed these cavities for precision sensing of gases and volatile liquids [22].

## 4 Summary of Publications under this Program

Journal Papers (published and under review):

1. Hannah Clevenson, Pierre Desjardins, Xuetao Gan, and Dirk Englund. High- $Q$  suspended polymer photonic crystal cavities for gas sensing. under review (2014)
2. Scalable Fabrication of High Purity Diamond Nanocrystals with Long-Spin-Coherence Nitrogen Vacancy Centers, Matthew E. Trusheim, Luozhou Li, Edward H. Chen, Ophir Gaathon, Hassaram Bakhru, and Dirk Englund, Nano Letters 14 (2013)

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3. Enhanced Photodetection in Graphene-Integrated Photonic Crystal Cavity, Ren-Jye Shiue, Xuetao Gan, Yuanda Gao, Luozhou Li, Xinwen Yao, Attila Szep, Dennis Walker, Jr., James Hone, and Dirk Englund, *Appl. Phys. Lett.* 103, 241109 (2013)
  4. Chip-integrated ultrafast graphene photodetector with high responsivity, X. Gan, R.J. Shiue, Y. Gao, I. Meric, T. F. Heinz, K. Shepard, J. Hone, S. Assefa, & D. Englund, *Nature Photonics AOP* (2013). In the news: MIT News, Nature, Nature Photonics News and Views, Nanowerk, Gizmodo, Photonics.com, LiveMint, IEEE Spectrum, Phys.org, RD Magazine, NanotechWeb,
  5. Controlled light-matter interaction in graphene electro-optic devices using nanophotonic cavities and waveguides, Xuetao Gan, Ren-Jye Shiue, Yuanda Gao, Solomon Assefa, James Hone and Dirk Englund, *IEEE JOURNAL OF SELECTED TOPICS IN QUANTUM ELECTRONICS on Graphene Optoelectronics* (2013)
  6. Reactive ion etching: optimized diamond membrane fabrication for transmission electron microscopy, L. Li, M. Trusheim, O. Gaathon, K. Kisslinger, C.-J. Cheng, M. Lu, D. Su, X. Yao, H.-C. Huang, I. Bayn, A. Wolcott, R. M. Osgood, Jr., and D. Englund, *Journal of Vacuum Science and Technology B*, Vol. 36 (2013). Among July 2013 Top 20 Most Downloaded articles
  7. Wide-field multispectral super-resolution imaging using spin-dependent fluorescence in nanodiamonds, E. H. Chen, O. Gaathon, M. E. Trusheim, and D. Englund, *Nano Lett.*, 2013, 13 (5), pp 20732077
  8. Nanophotonic Filters and Integrated Networks in Flexible 2D Polymer Photonic Crystals, X. Gan, H. Clevenson, C.-C. Tsai, L. Li, and D. Englund, *Nature Scientific Reports* 3, Article number: 2145(2013).
  9. High-Contrast Electro-Optic Modulation of a Photonic Crystal Nanocavity by Electrical Gating of Graphene, Xuetao Gan, Ren-Jye Shiue, Yuanda Gao, Kin Fai Mak, Xinwen Yao, Luozhou Li, Attila Szep, Dennis Walker Jr., James Hone, Tony F. Heinz, and Dirk Englund, *Nano Lett.*, 2013, 13 (2), pp 691696. In the news: NanoTech Technology Update
  10. Timekeeping with electron spin states in diamond, J. S. Hodges, N. Y. Yao, D. Maclaurin, C. Rastogi, M. D. Lukin, and D. Englund, *Physical Review A* 87, 032118 (2013). In the news: PRA Highlight, Nature research highlight
  11. Strong enhancement of light-matter interaction in graphene coupled to a photonic crystal nanocavity, X. Gan, K. F. Mak, Y. Gao, Y. You, F. Hatami, J. Hone, T. F. Heinz, and D. Englund, *Nano Letters*, 12(11):56265631 (2012). In the News: NanoTechWeb Research Update
  12. Long-lived nitrogen vacancy spin coherence in high-purity diamond membranes, Jonathan Hodges, Luozhou Li, Ming Lu, Edward H Chen, Matthew E Trusheim, S Allegri, Xinwen Yao, O Gaathon, Hassaram Bakhru, and Dirk Englund, *New Journal of Physics* 14 (2012).

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13. A high-resolution spectrometer based on a compact planar two dimensional photonic crystal cavity array, Xuetao Gan, Nadia Pervez, Ioannis Kymissis, Fariba Hatami, and Dirk Englund, Appl. Phys. Lett. 100, 23 (2012) Nature Photonics Highlight
  14. Ion-Exfoliated Single-Crystal-Diamond Membranes Exhibiting Nitrogen-Vacancy Color Centers, O. Gaathon, J. S. Hodges, E. H. Chen, L. Li, S. Bakhru, H. Bakhru, D. Englund, and R. M. Osgood, Jr., Volume 35, Issue 3, 361365 (2013)
  15. Ultrafast photon-photon interaction in a strongly coupled quantum dot-cavity system, D. Englund, A. Majumdar, M. Bajcsy, A. Faraon, P. Petroff & J. Vuckovic, Phys. Rev. Lett.108:093604 (2012).

Patent applications:

1. D. Englund et al, Ultra-high-resolution conformal lithography for patterning arbitrary samples
2. X. Gan and D.Englund, Graphene Photonics for resonator-enhanced ultrafast electro-optic processes and all-optical interactions (2012)
3. X. Gan and D. Englund, Chip-integrated Infrared Detector Employing Cavity-Enhanced Upconversion (2012)
4. X. Gan and D. Englund, Ultra-compact, high quality photonic devices in a planar polymer-on-air architecture” (2011)
5. E. Chen and D. Englund, Deterministic Emitter Switch Microscopy (DESM)” (2011)
6. D Englund, Conjugates Of Nano-Diamond And Magnetic Or Metallic Particles, Wo Patent 2,013,066,446
7. D Englund And E Chen, Systems And Methods For Deterministic Emitter Switch Microscopy, Wo Patent 2,013,059,404
8. D Englund And C Rastogi, Ultracompact Fabry-Perot Array For Ultracompact Hyperspectral Imaging, Wo Patent 2,013,059,665
9. D Englund, J Hodges, M Lukin, N Yao, High-Precision Ghz Clock Generation Using Spin States In Diamond, Wo Patent 2,013,040,446

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