Ultrafast Meets Ultrasmall: Laser Writing at the Quantum Frontier

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Tsinghua University

Acknowledgements

Our Group @ Tsinghua University

Nanophotonics M3 Manufacturing Manipulation Measurement

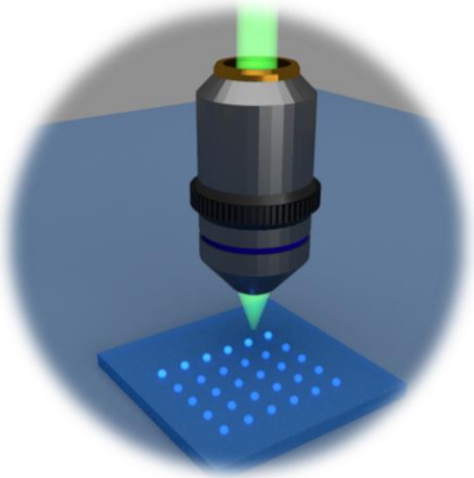
清華大學 精密仪器系
Department of Precision Instrument, Tsinghua University

Financial Support

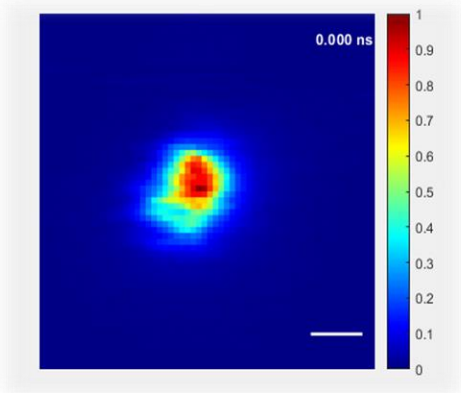
National Natural Science Foundation of China

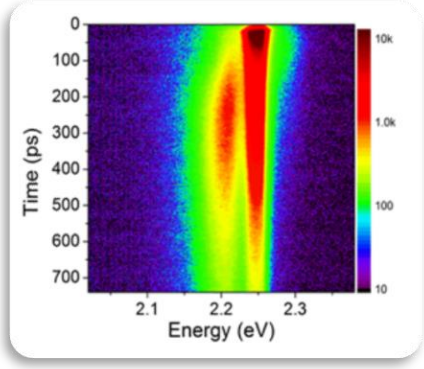
Research Activity

Light-Matter Interaction @ Nanoscales

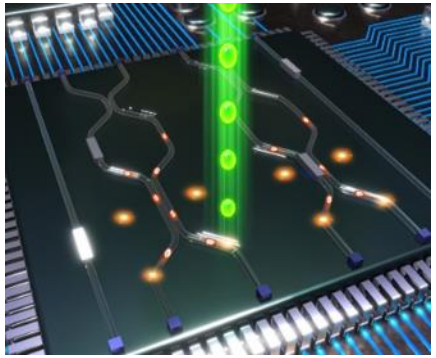


Laser Writing





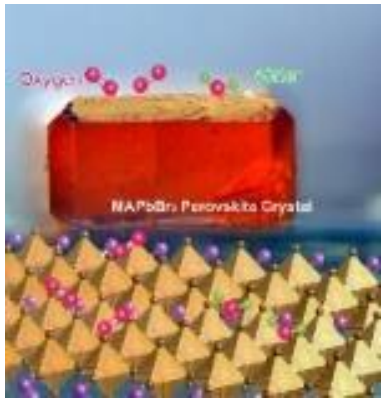
Time-resolved Image & Spectroscopy



Nanophotonics

Light: Science & Applications 13 (1), 6，2024
Applied Physics Letters 123 (13)，2023
Nano Letters 23 (7), 2743-2749，2023
Laser & Photonics Reviews, 2100029，2021

Ultrafast Science 0002， 2022
Advanced Optical Materials 11 (4), 2202038，2023
Advanced Optical Materials 11 (4), 2202038，2022
Advanced Functional Materials 30 (6), 1907979，2020
Physical review letters 123 (6), 067401，2019
Nature Communications 9 (1), 243，2018



Optoelectronics

Outline

I. Introduction to Ultrafast Laser Writing Techniques

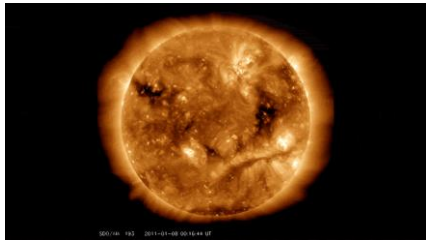
II. Laser Writing for Quantum Techniques

III. Laser Writing Color Center: Example of AlN Crystal

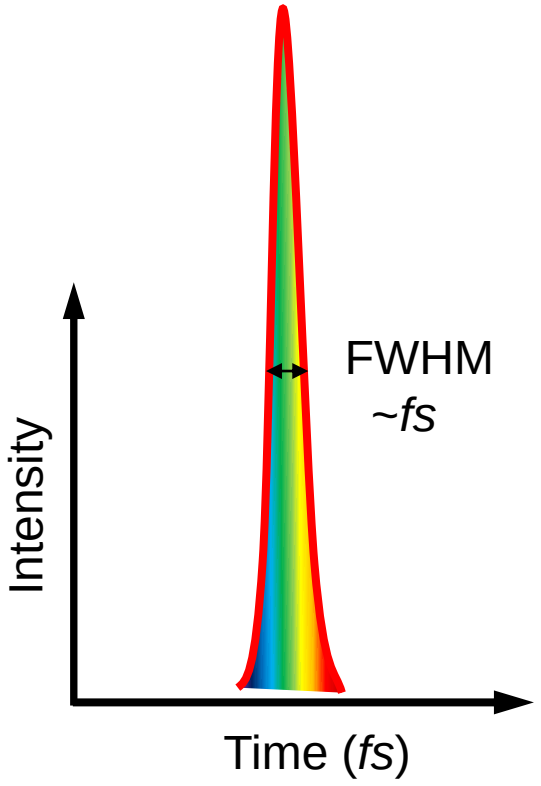
IV. Direct Laser Writing Color Centers Approaching Q-Limit

V. Laser Writing Sub-10 nm Structure in Semiconductors

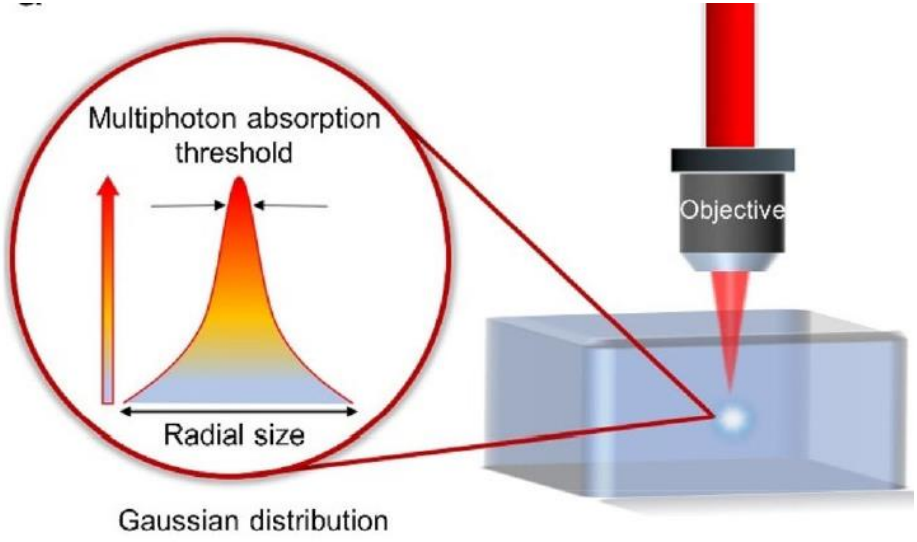
I. Introduction



Ultrafast laser matter interactions



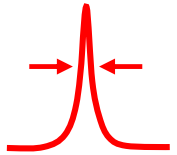
- ◆ **Ultrashort Laser Pulses**
- ◆ **Ultrahigh Peak Power**
- ◆ **Nonlinear Optical Process**
- ◆ **3D Fabrication Capability**



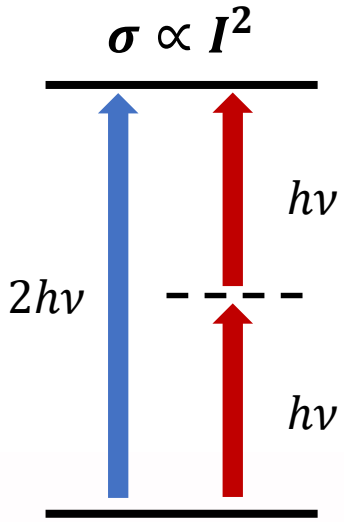
Laser & Photonics Reviews, 2100029，2021

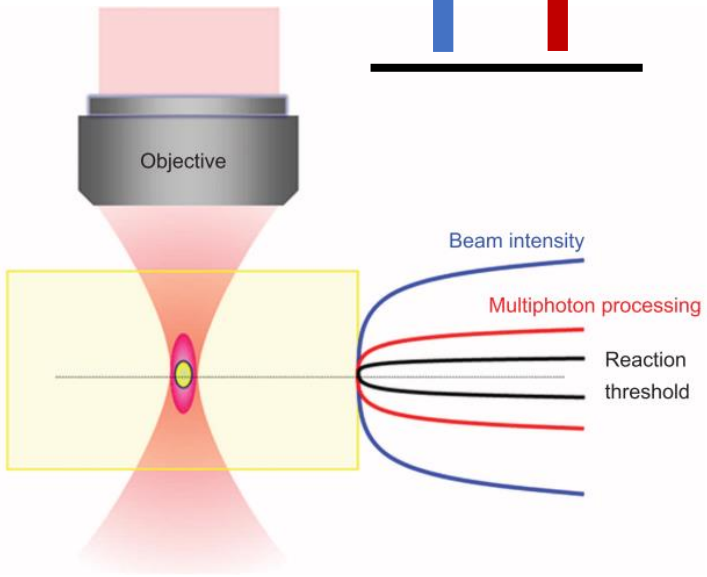
I. Introduction

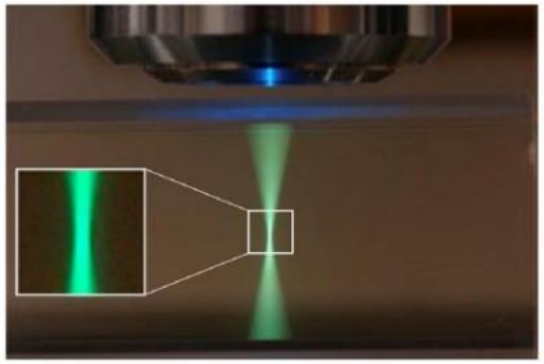
Ultrashort Pulse



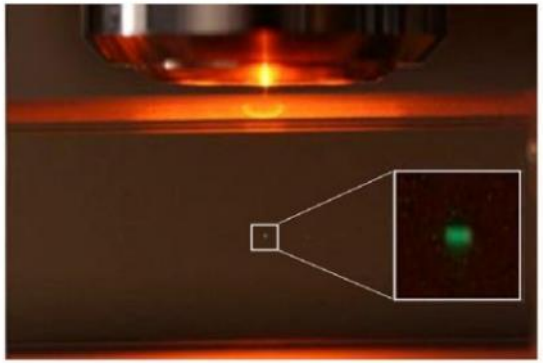
$\tau < 10^{-12}$ s



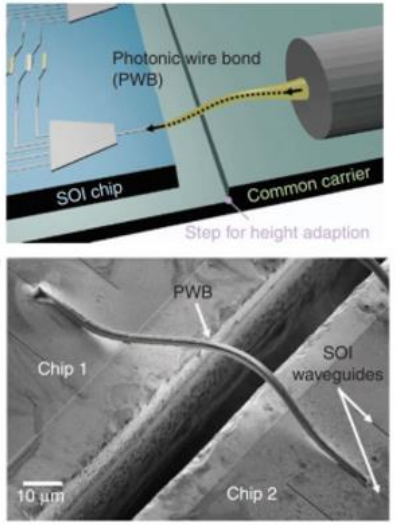


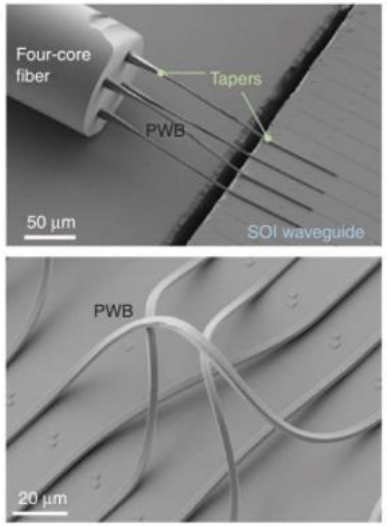


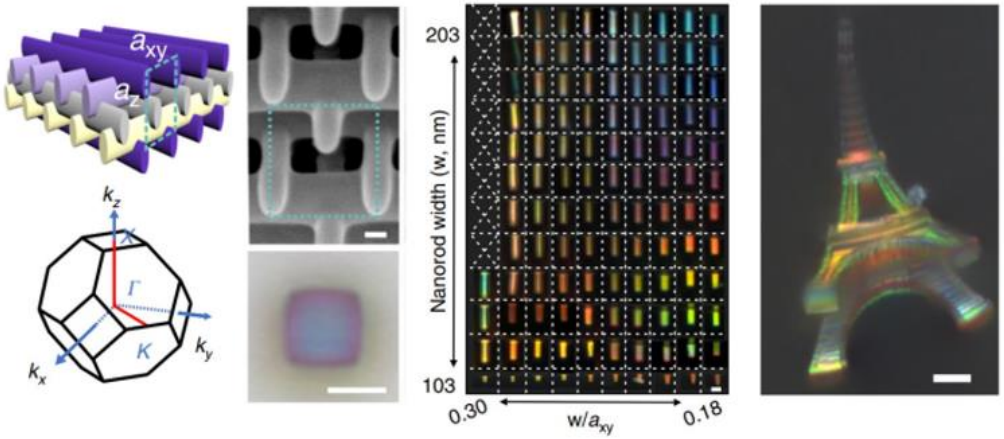
Single-photon absorption



Two-photon absorption







Ref: Light5.8 (2016): e16133-e16133.

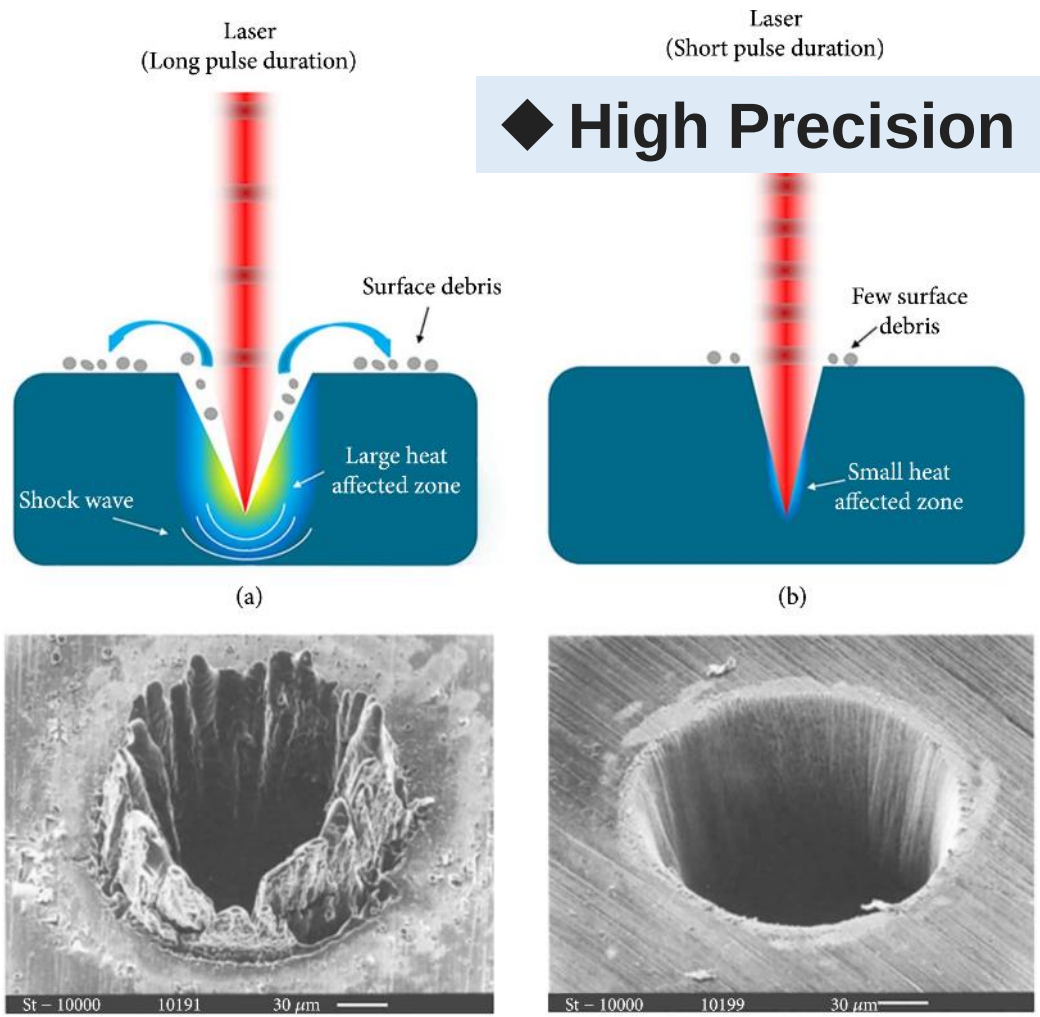
Ref: Nature，412 697 (2001)

Ref: Nat. Commun, 10, 1-8 (2019)

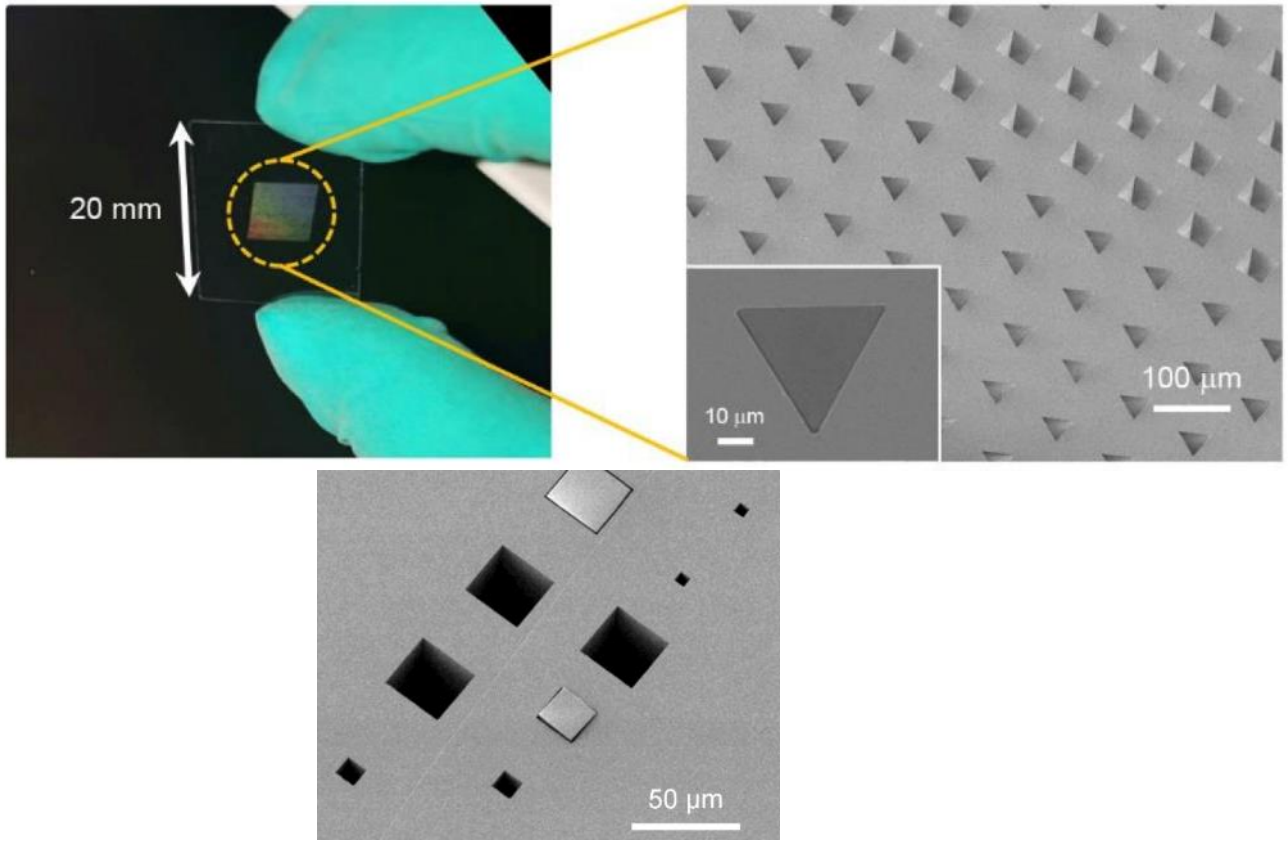
I. Introduction

Ultrafast Laser Ablation

◆ **High Precision**



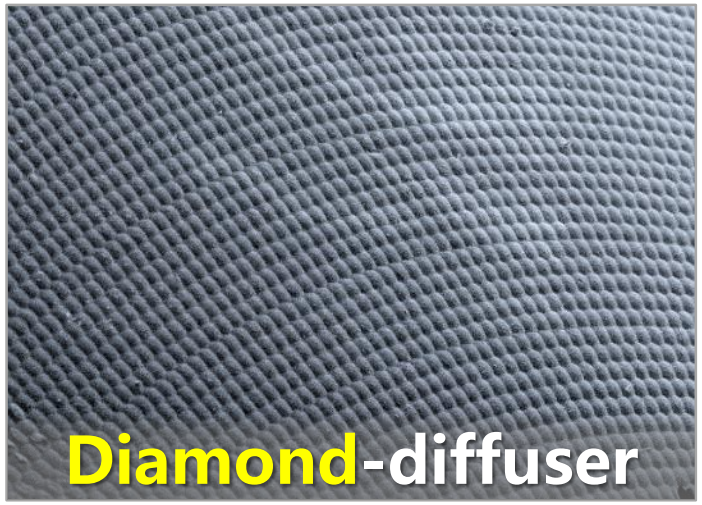
Appl. Phys. A **63**, 109–115 (1996).

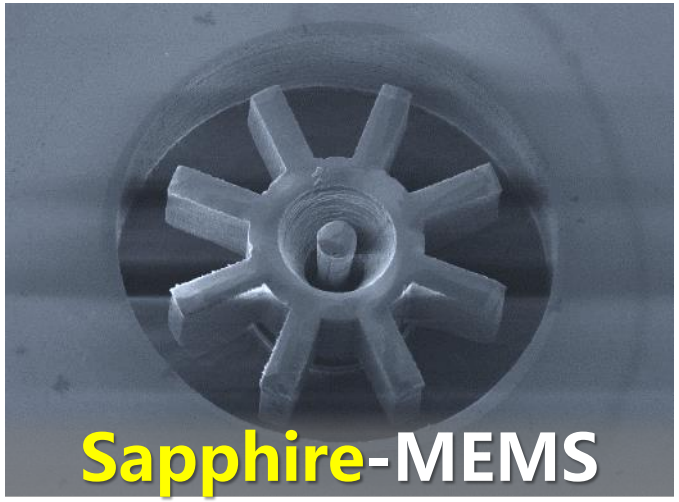


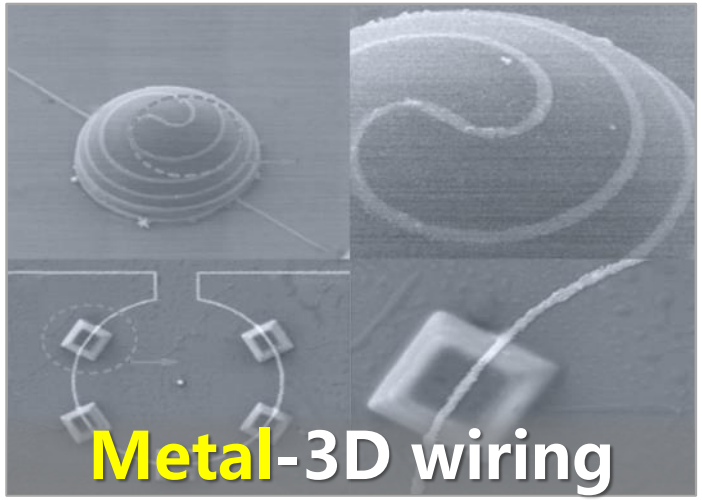
Super-stealth dicing of transparent solids with nanometric precision. *Nat. Photon.* (2024).

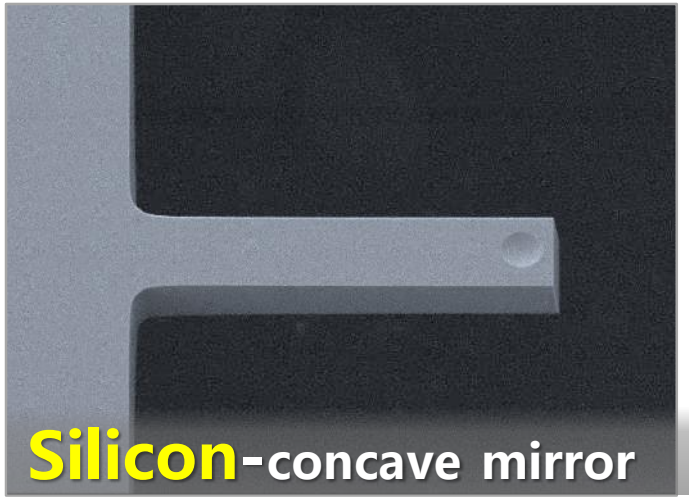
I. Introduction

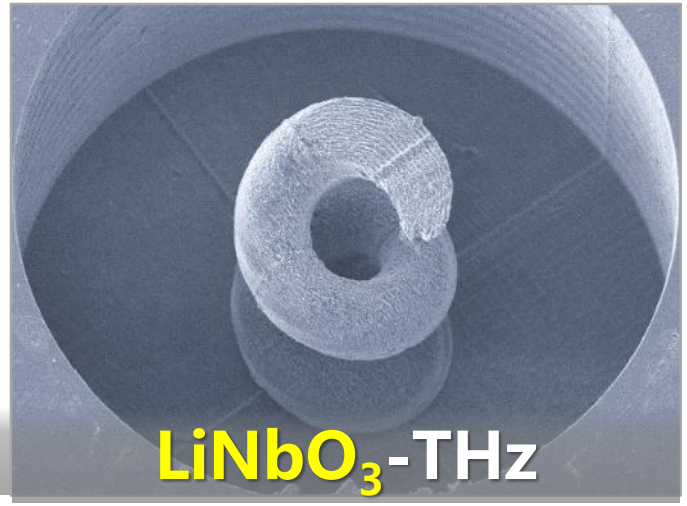
Ultrafast Laser as High Precision 3D Micro/Nano- Fabrication Tools





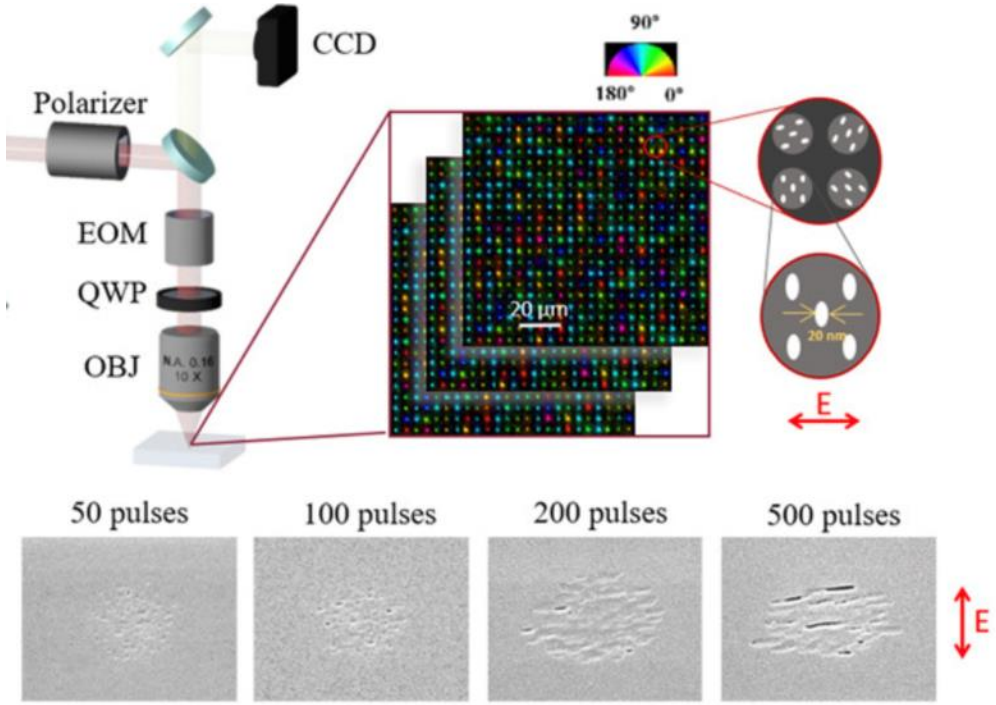






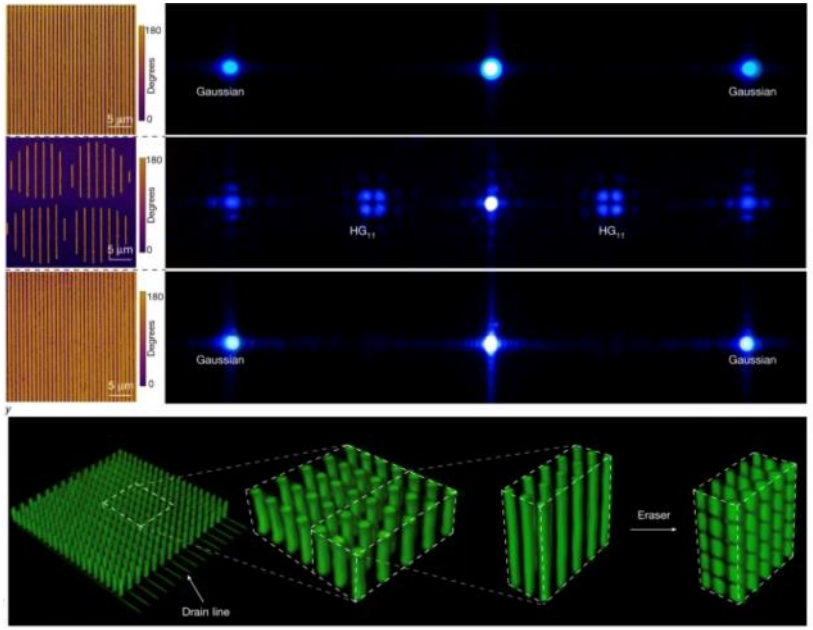
I. Introduction

Ultrafast Laser as Powerful Tools for Materials Modification



Data Storage by Ultrafast Laser Nanostructuring

Laser Photonics Rev. 2022, 16, 2100563



Femtosecond laser write ferroelectric nanodomains

Nature, 609, 496–501 (2022)

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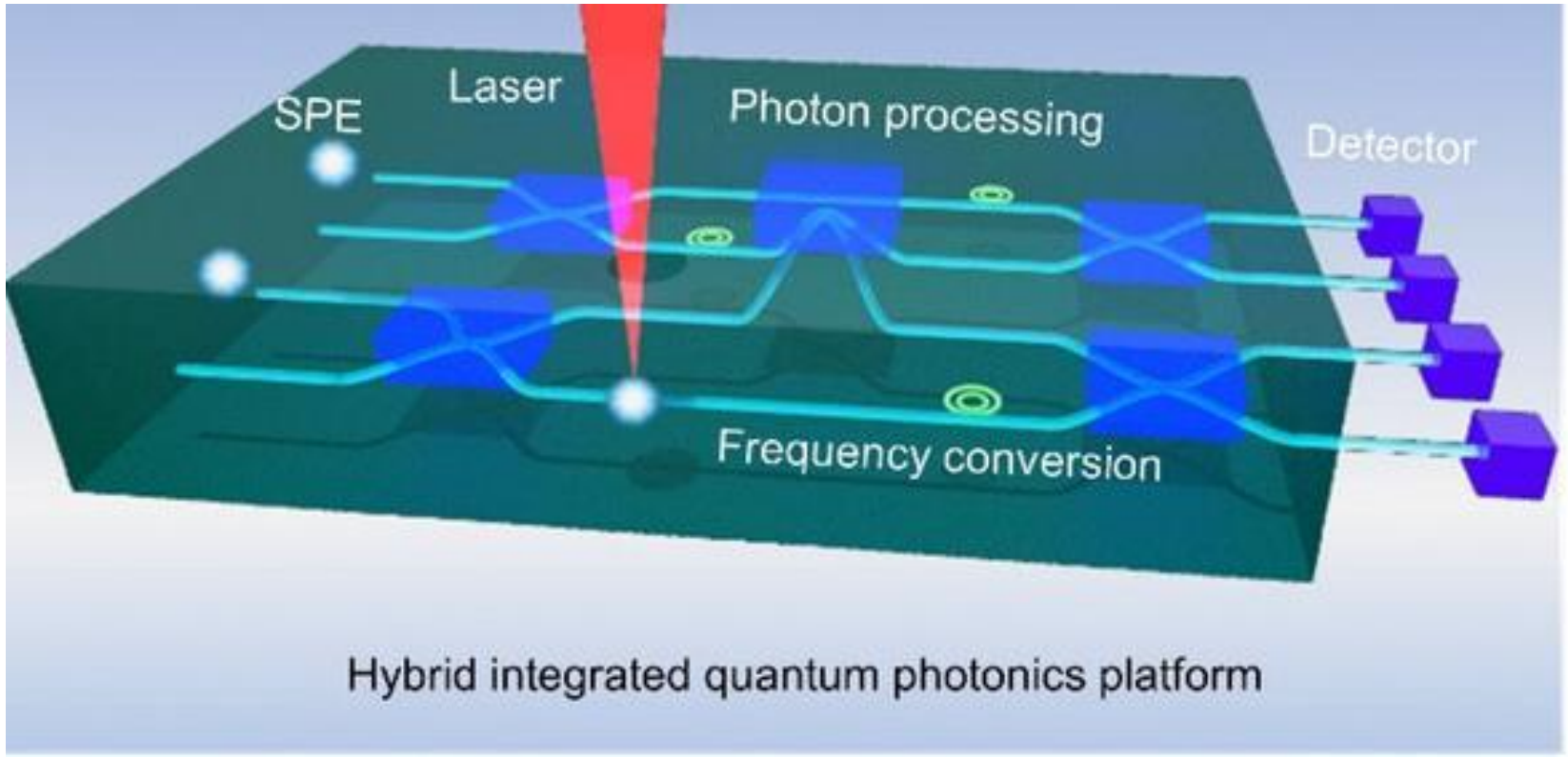
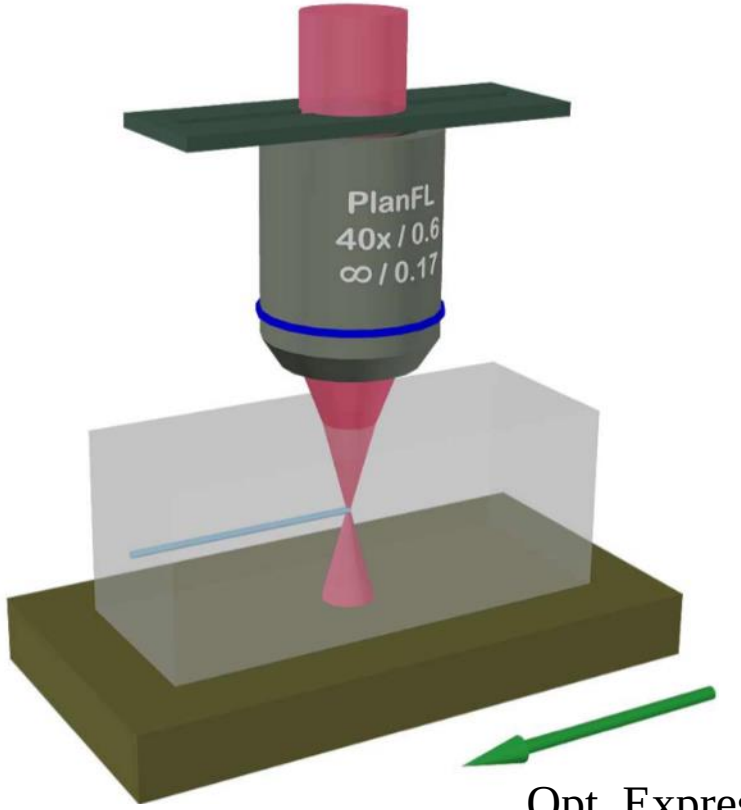
Laser Writing for Quantum Techniques

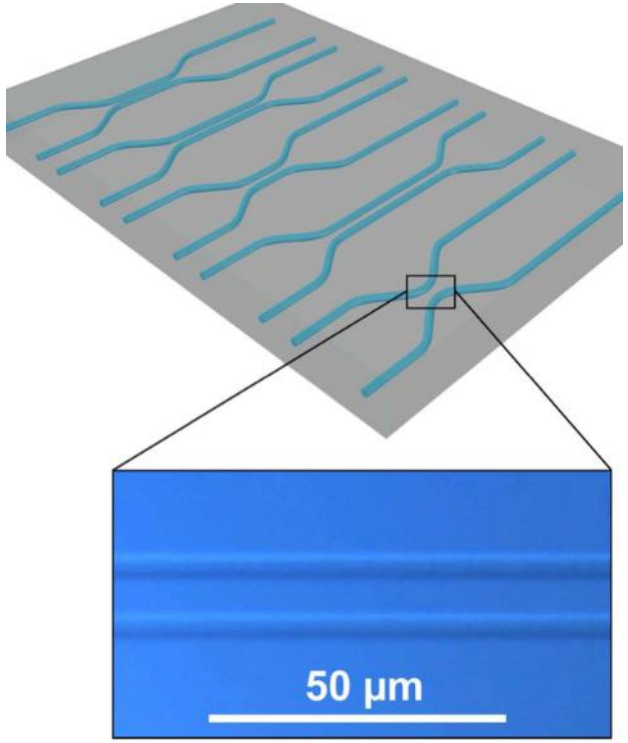


Laser & Photonics Reviews, 2100029， 2021

Laser Writing for Quantum Techniques

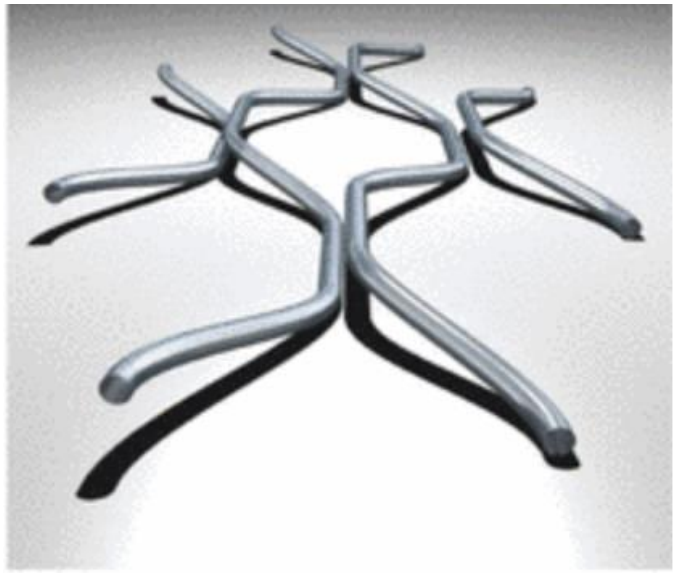
Laser Writing Waveguide

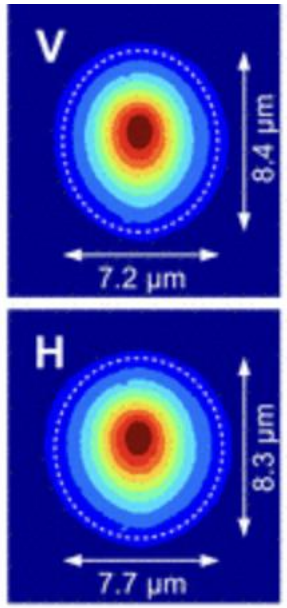




Opt. Express，2009，17(15):12546

Macquarie University, Australia





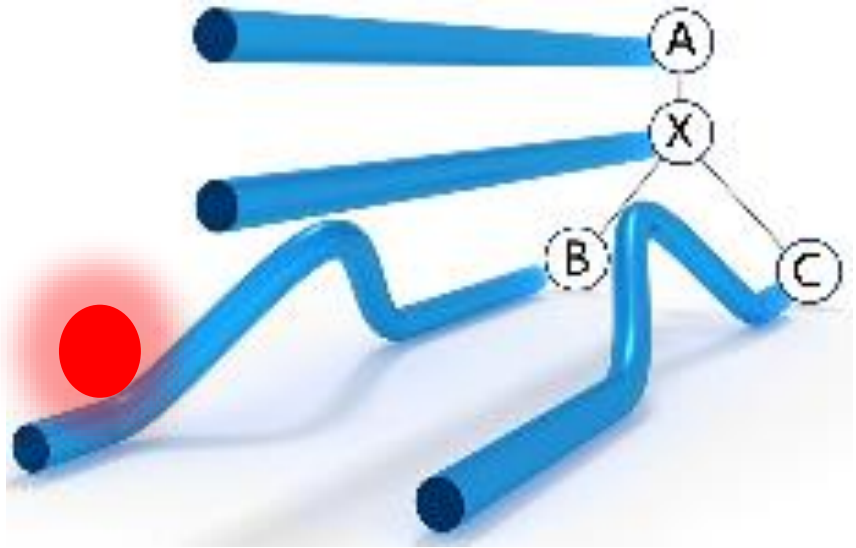
Two-Particle Bosonic-Fermionic Quantum Walk via Integrated Photonics

Phys. Rev. Lett. 2012，108, 010502

Sapienza Universita` di Roma, Italy

Laser Writing for Quantum Techniques

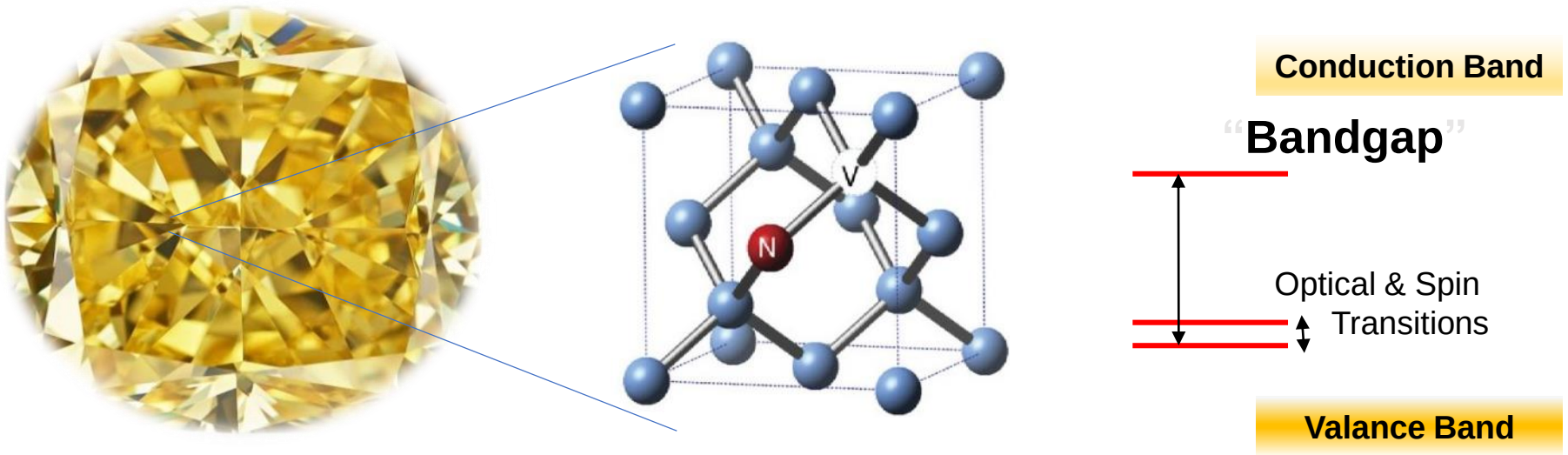
Laser writing technique — an enabler for quantum photonic integration



Color center chips for Quantum Simulation

Laser Writing for Quantum Techniques

Color Centers, atomic defects in crystals

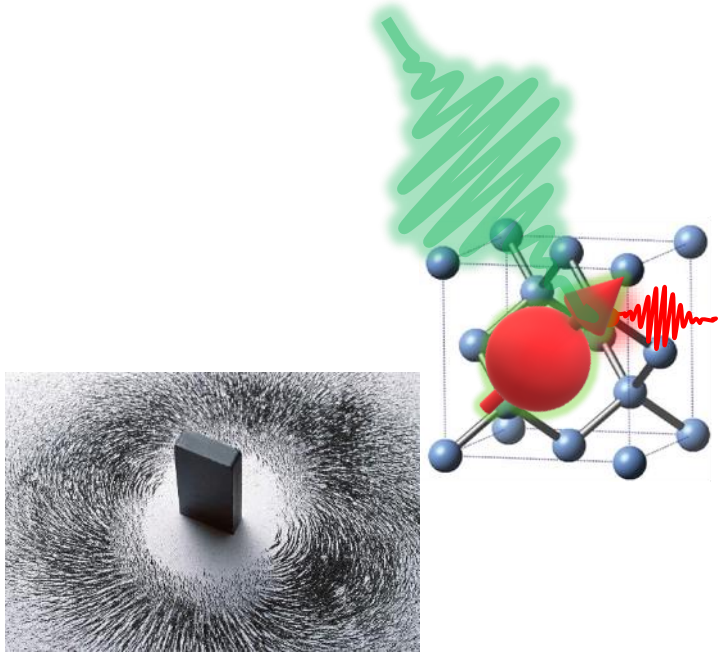


1. Single-photon emission

2. Spin

Laser Writing for Quantum Techniques

Properties of color cennters



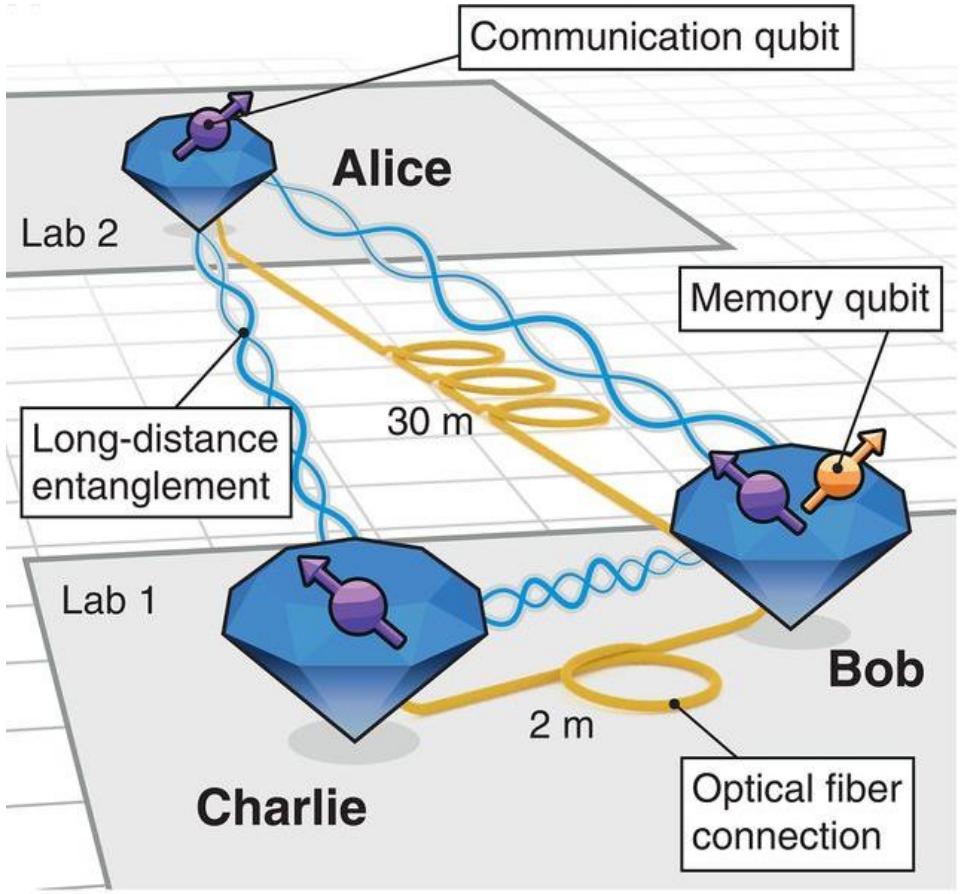
1. Quantum Light Source

2. Spin

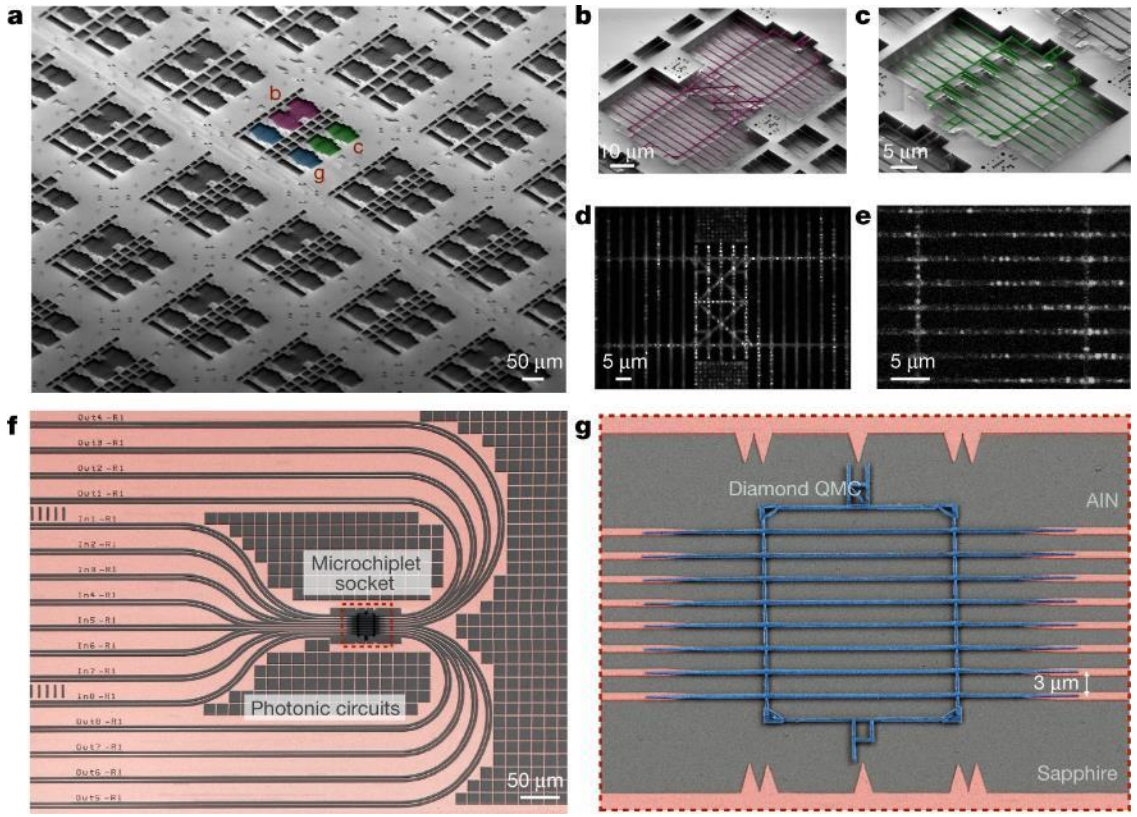
Quantum Information Process
Nanoscale Sensor

Laser Writing for Quantum Techniques

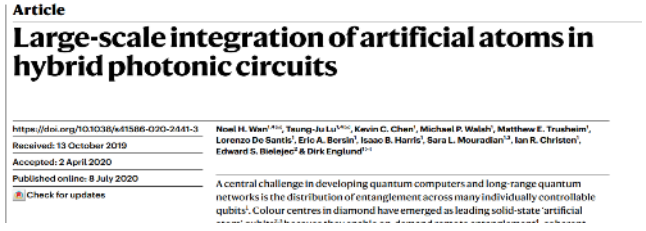
Color Center as Quantum Nodes



SCIENCE, 2021, 372, 259-264



Integrated quantum photonics with color centers



Article

Large-scale integration of artificial atoms in hybrid photonic circuits

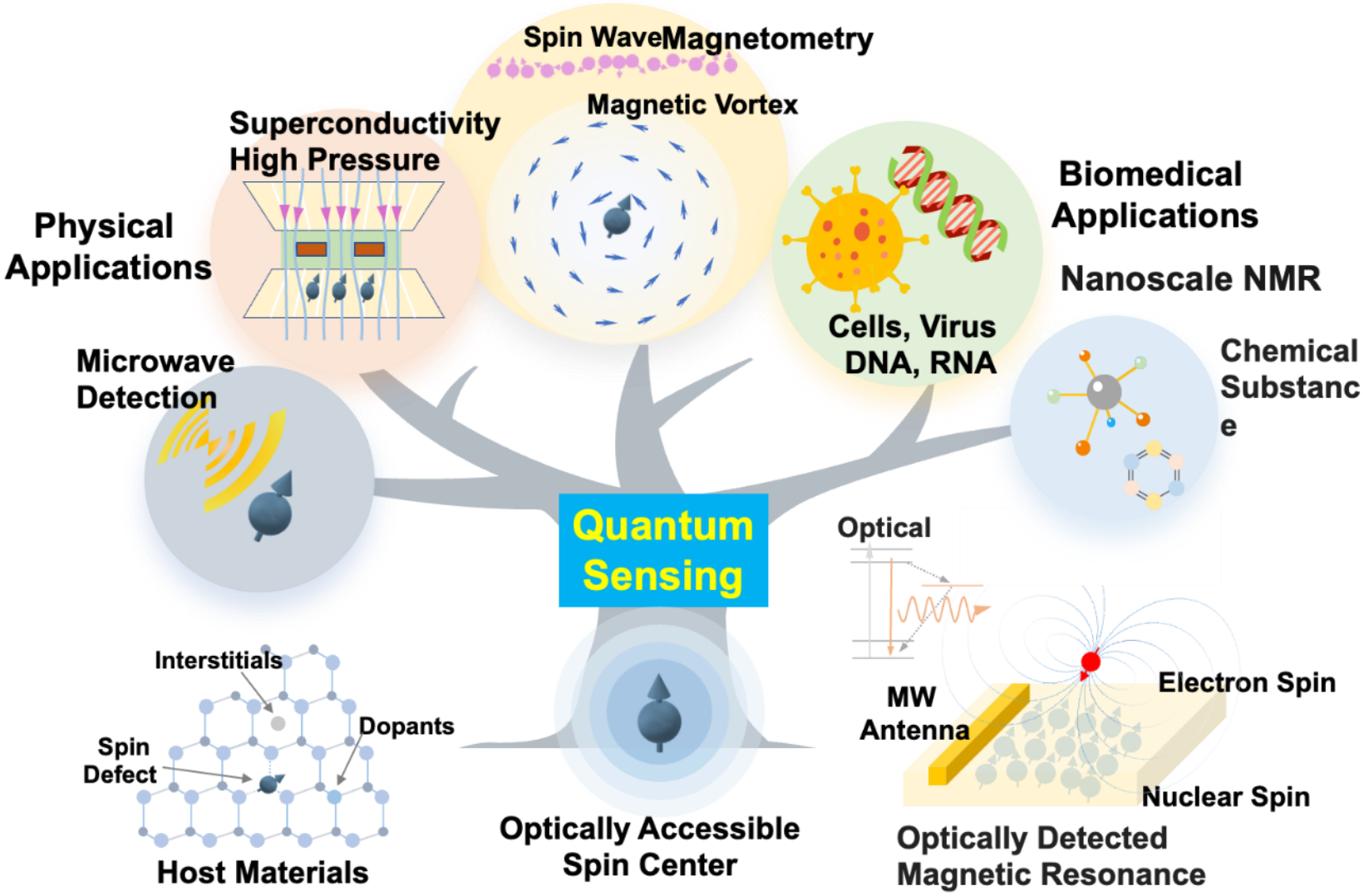
https://doi.org/10.1038/s41586-020-2441-3
Received: 13 October 2019
Accepted: 2 April 2020
Published online: 8 July 2020
Check for updates

Noel H. Wan[1,4], Tsung-Ju Lu[1,4], Kevin C. Chen[1], Michael P. Walsh[1], Matthew E. Trusheim[1], Lorenzo De Santis[1], Eric A. Bersin[1], Isaac B. Harris[1], Sara L. Mouradian[1,3], Ian R. Christen[1], Edward S. Bielejec[2] & Dirk Englund[1,3]

A central challenge in developing quantum computers and long-range quantum networks is the distribution of entanglement across many individually controllable qubits[1]. Colour centres in diamond have emerged as leading solid-state 'artificial

Laser Writing for Quantum Techniques

Quantum Sensing with color center



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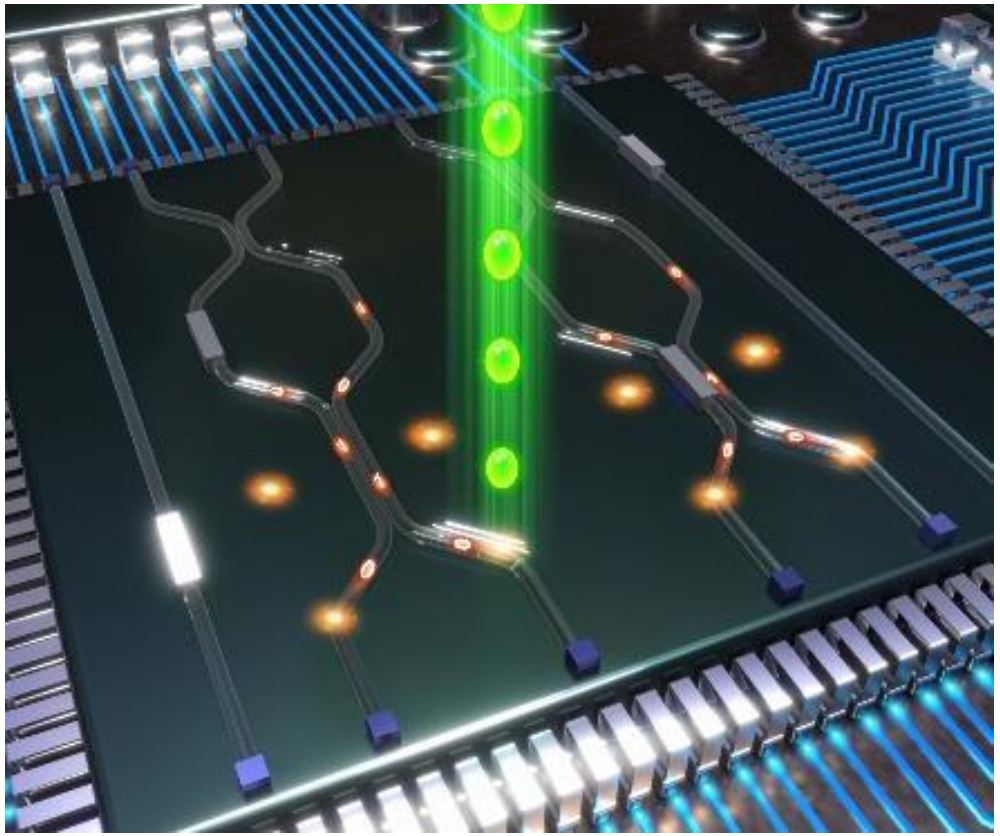
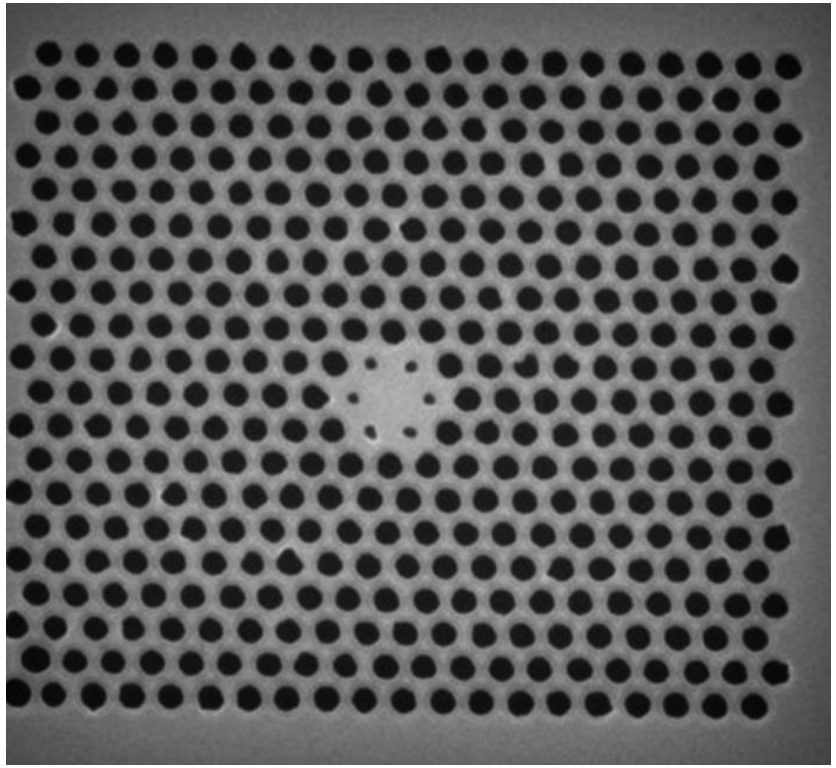
Color Center in AlN

AlN-based integrated photonics





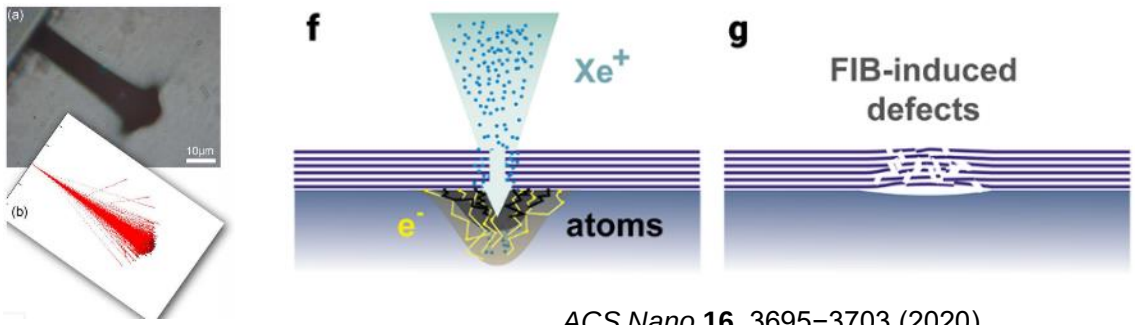
AlN photonic crystal membranes.

Int. J. Nanotechnol., Vol. 9, Nos. 3–7, 2012

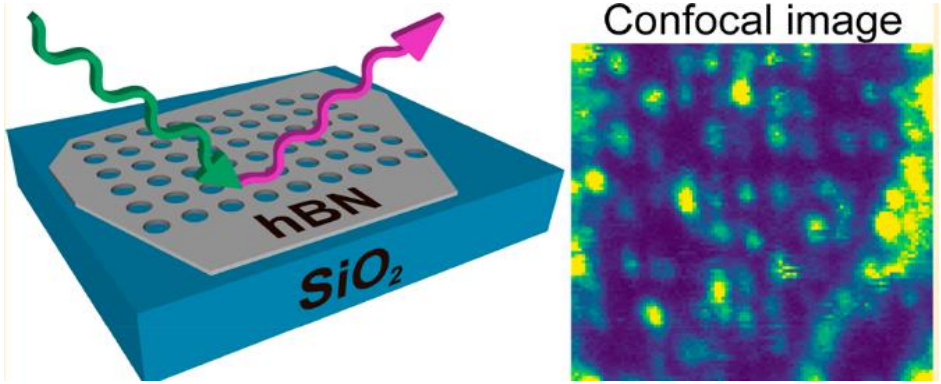
Color centers Engineering

High Precision Engineering Color-Center

Engineering NV spatial patterns.



ACS Nano **16**, 3695−3703 (2020)



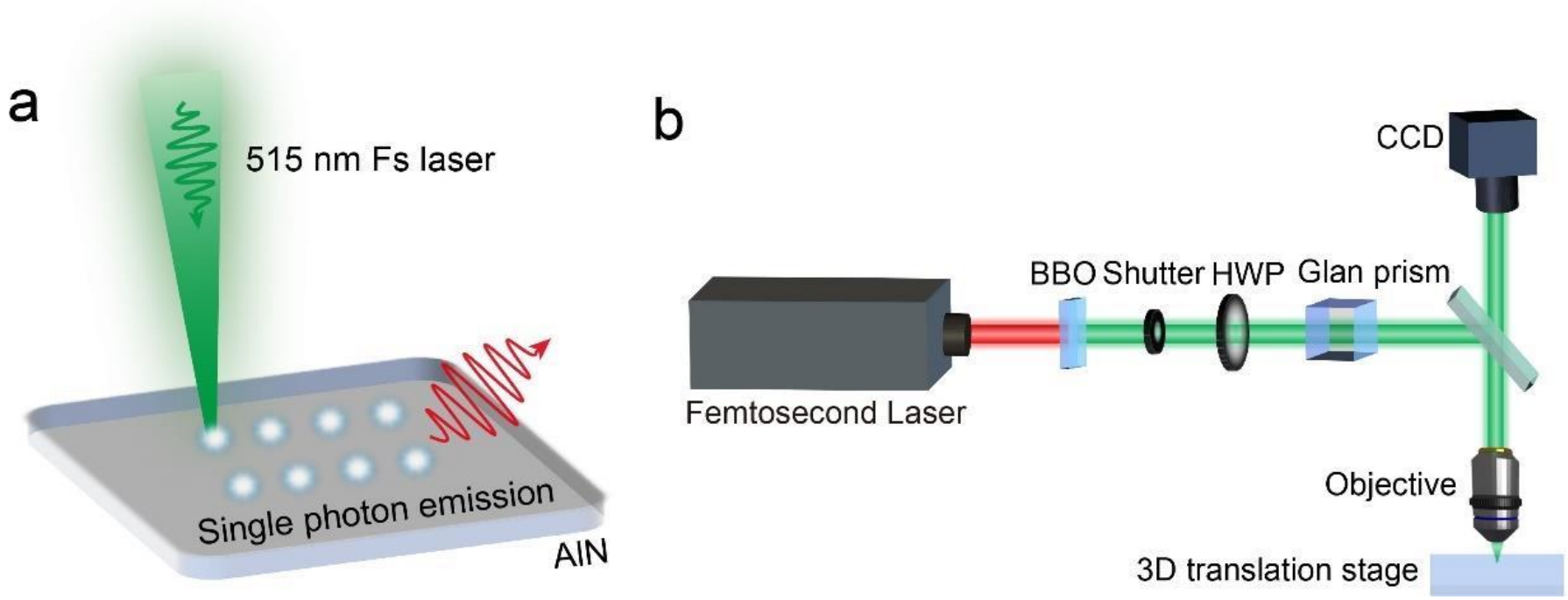
Nano Lett. **19, 2121−2127 (2019)**

Focused ion beam

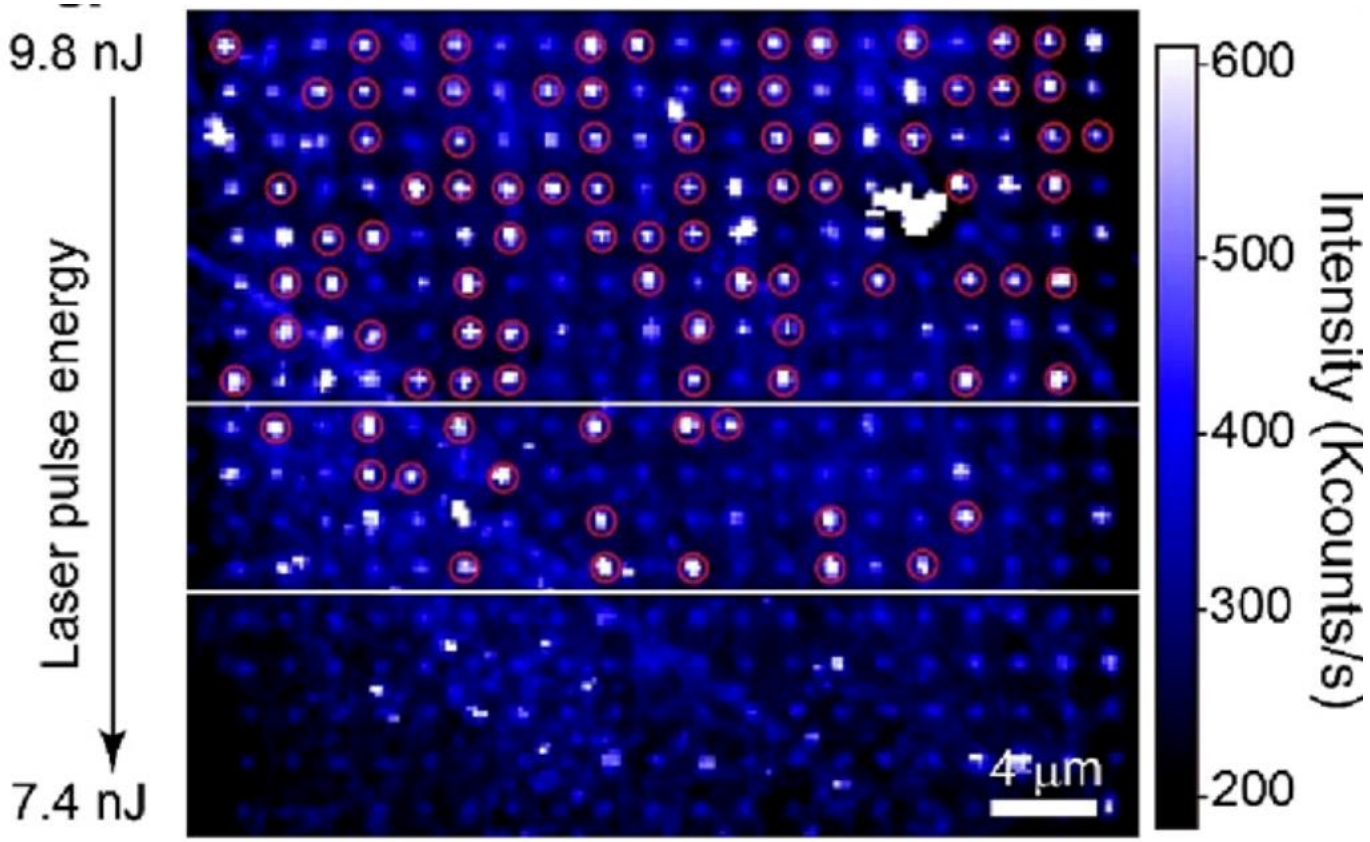
- ◆High energy, strong scattering
- ◆Lattice damage
- ◆Degraded optical properties
- ◆Low produce yield

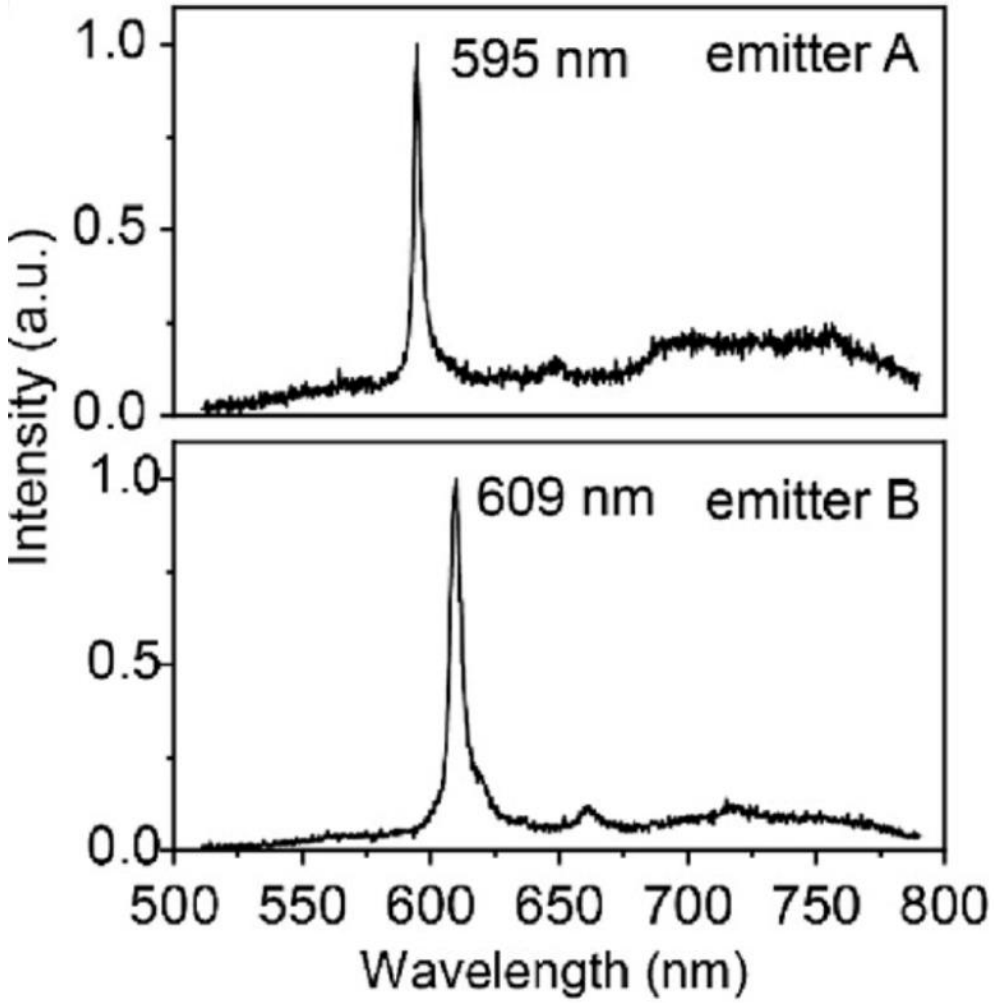
Color Center in AlN

Laser writing color center in AlN Crystal



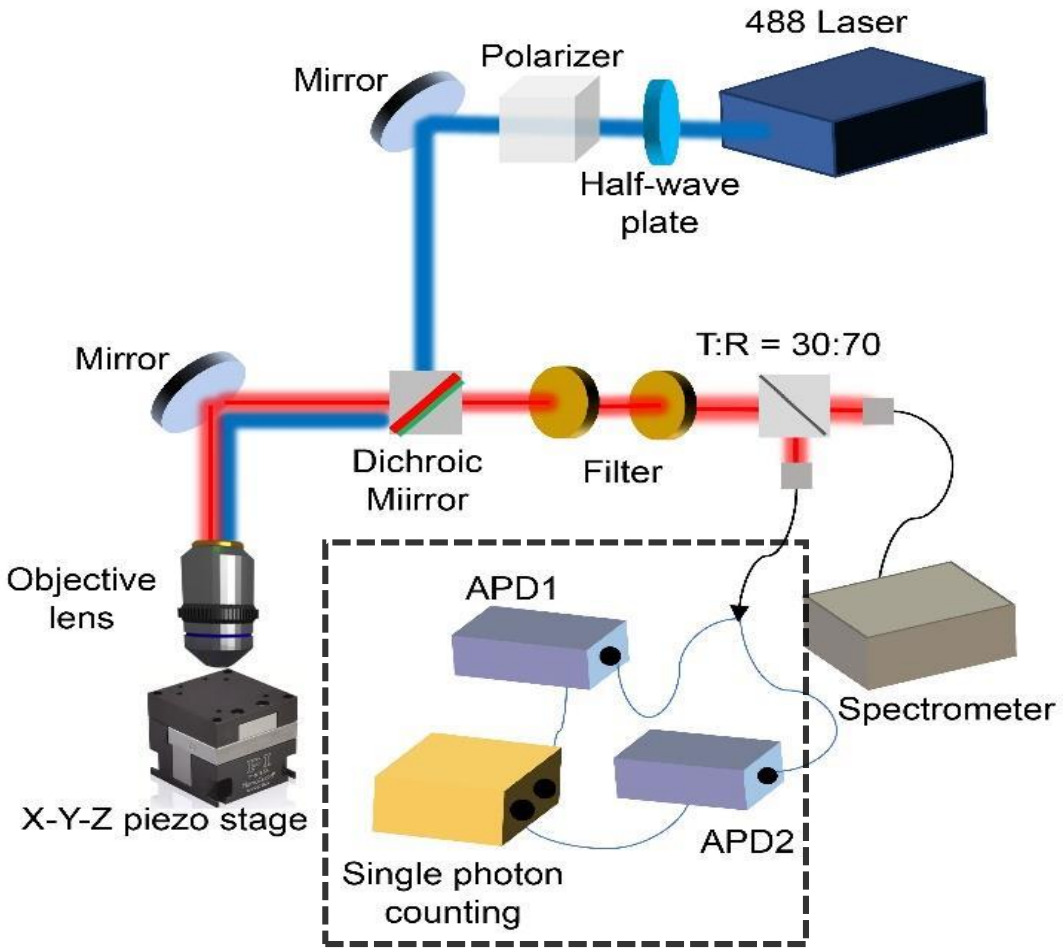
Color Center in AlN

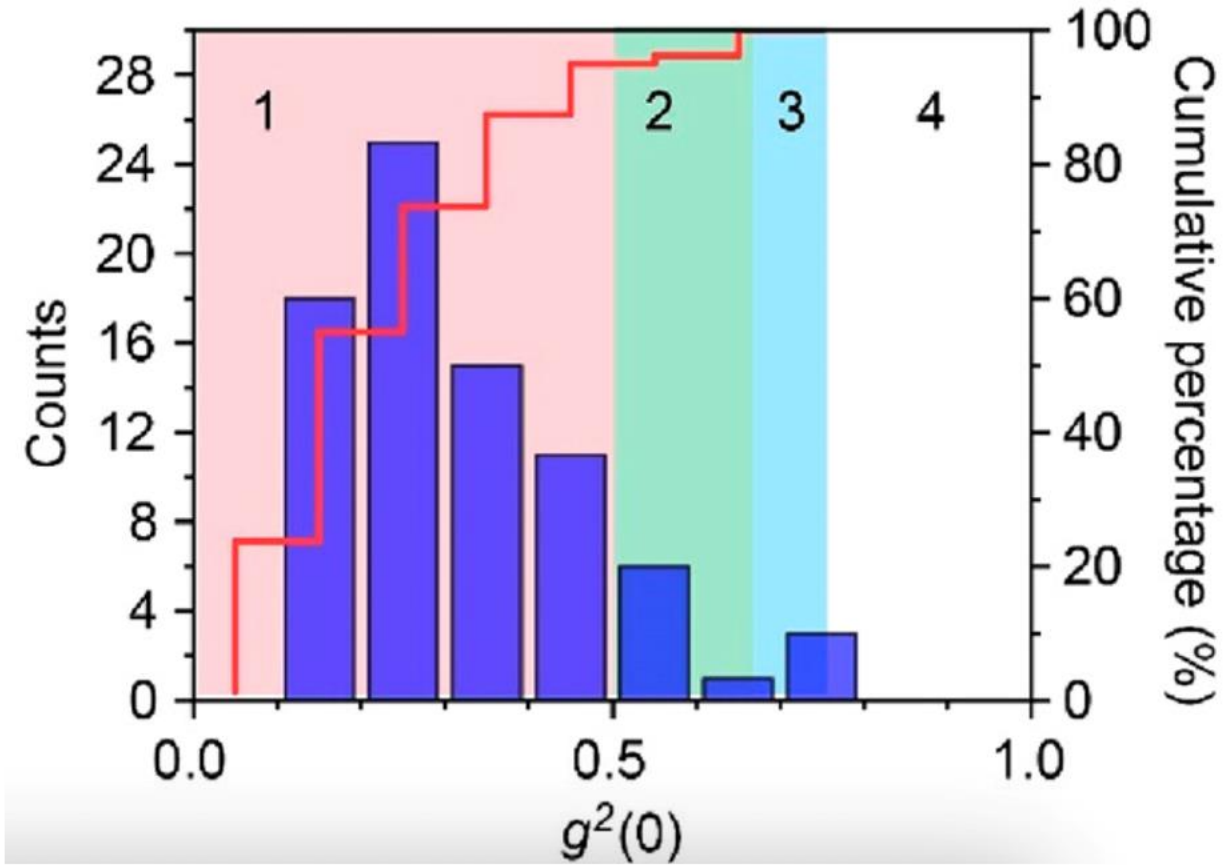




Nano Lett. 2023, 23, 7, 2743–2749

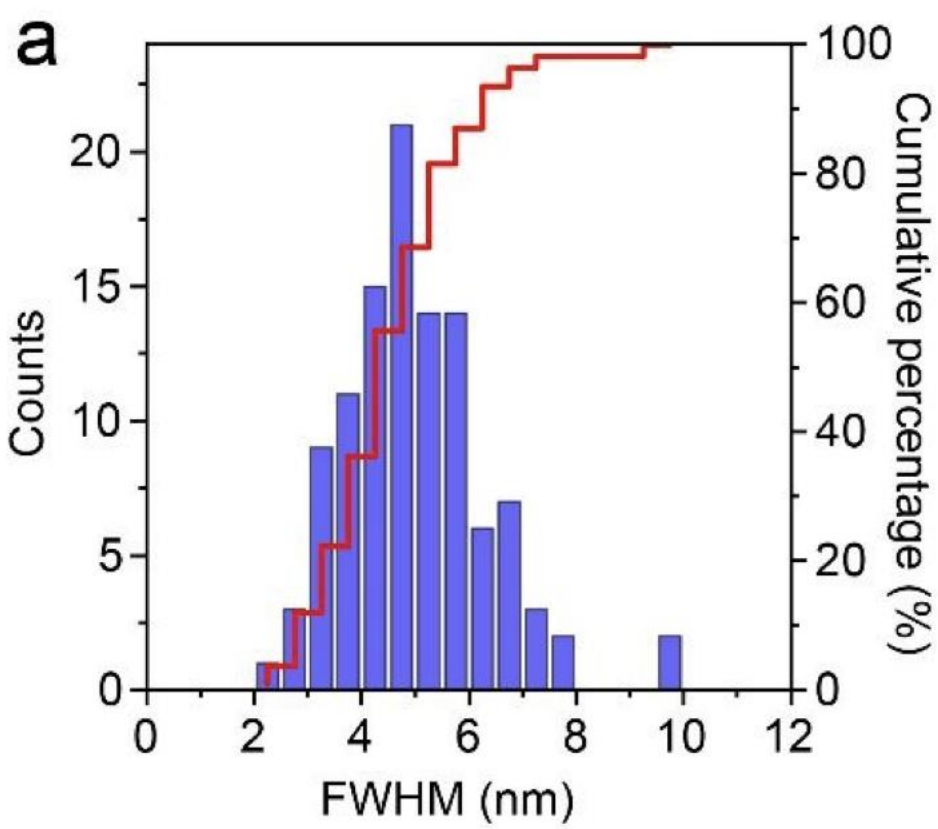
Color Center in AlN



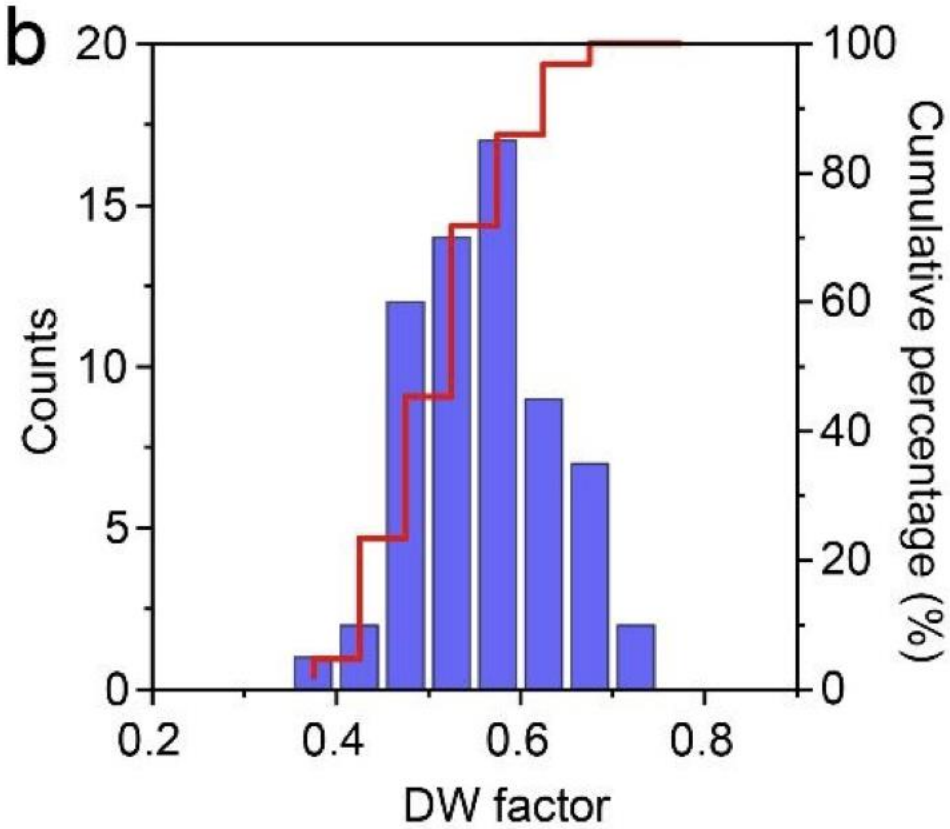


Single-photon emitter

Color Center in AlN



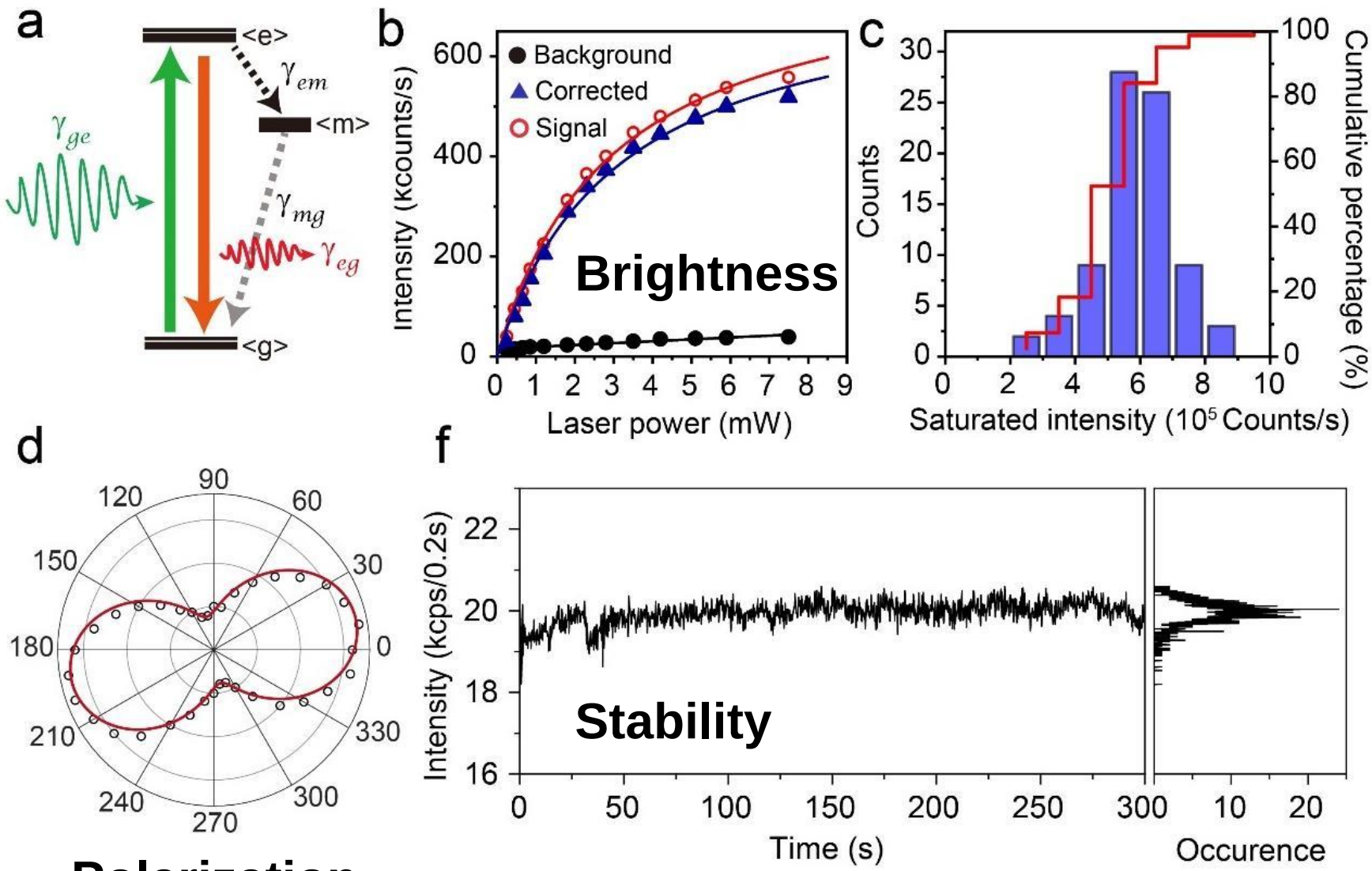
Line width at room temperature



High Debye-Waller factor

Nano Lett. 2023, 23, 7, 2743–2749

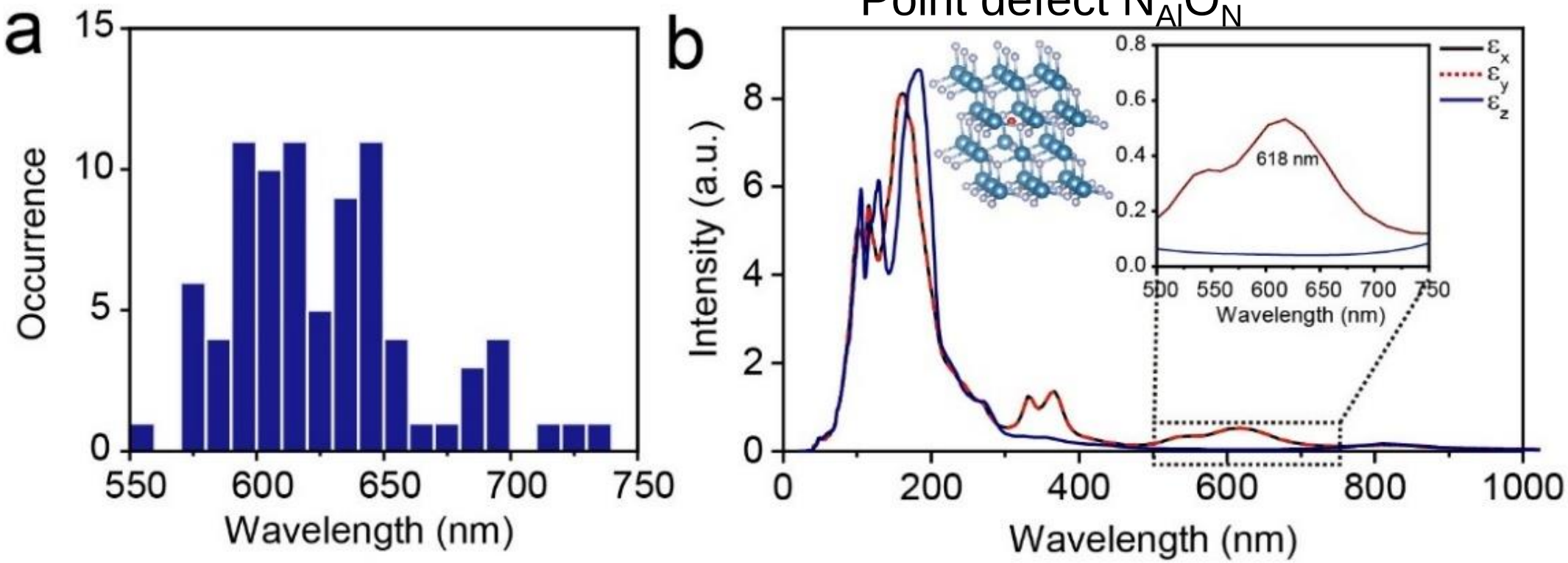
Color Center in AlN



Polarization

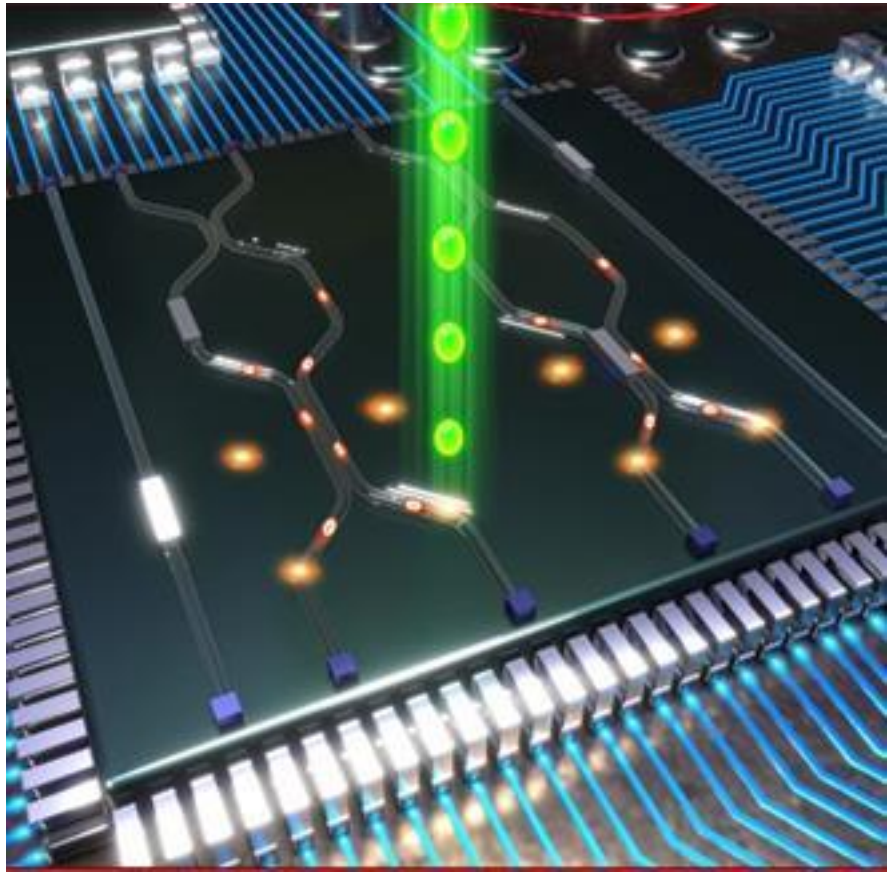
Color Center in AlN

Point defect $N_{Al}O_N$



Nano Lett. 2023, 23, 7, 2743–2749

Color Center in AlN



AlN quantum emitter

- **Bright and robust emission with a strong zero phonon line and narrow line width**
- **High Debye-Waller factor of more than 65% at room temperature**
- **Low phonon side bands (PSBs)**
- **AlN as a new quantum chip**

Nano Lett. 2023, 23, 7, 2743–2749

Outline

I. Introduction to Ultrafast Laser Writing Techniques

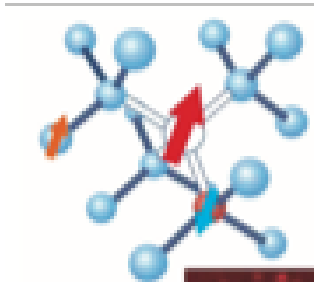
II. Laser Writing for Quantum Techniques

III. Laser Writing Color Center: Example of AlN

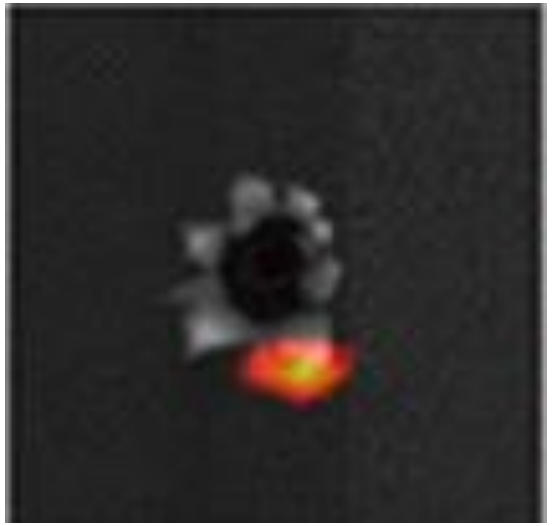
IV. Direct Laser Writing Color Centers Approaching Q-Limit

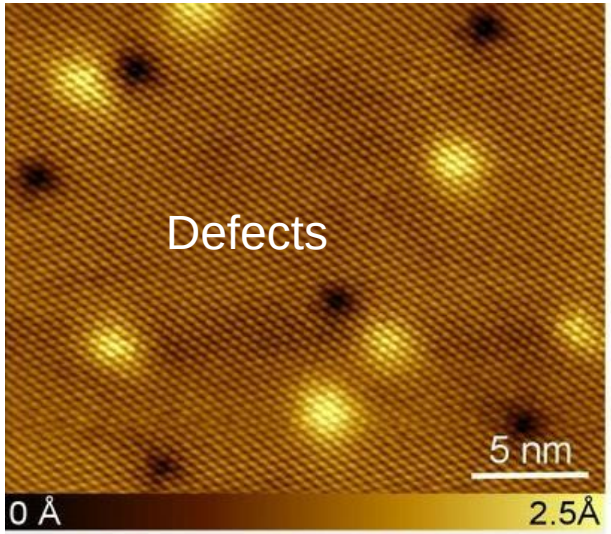
V. Laser Writing Sub-10 nm Structure in Semiconductors

Direct Laser Writing Color Centers Approaching Q-Limit



Color centers are atomic defects.
How to minimize the lattice damage?

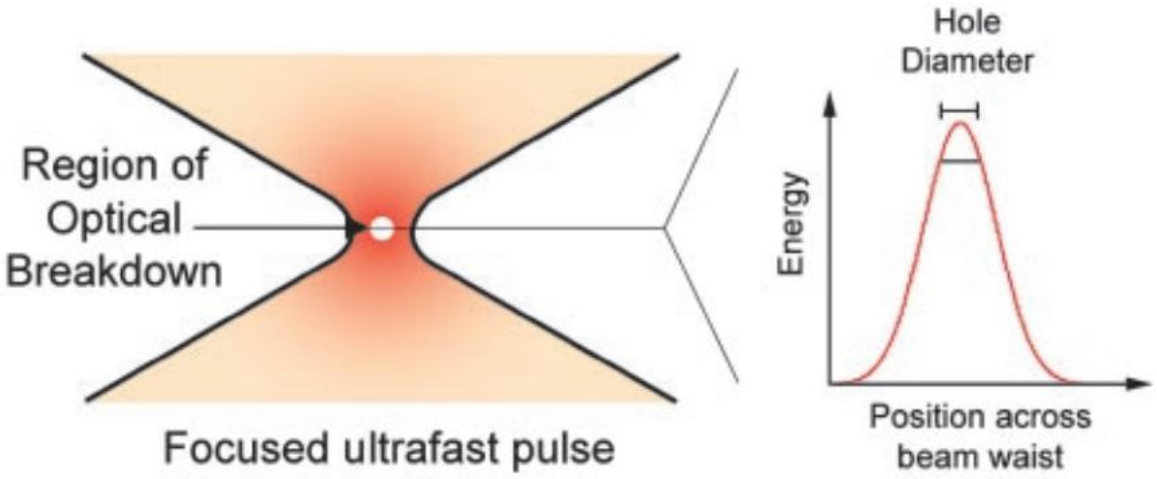


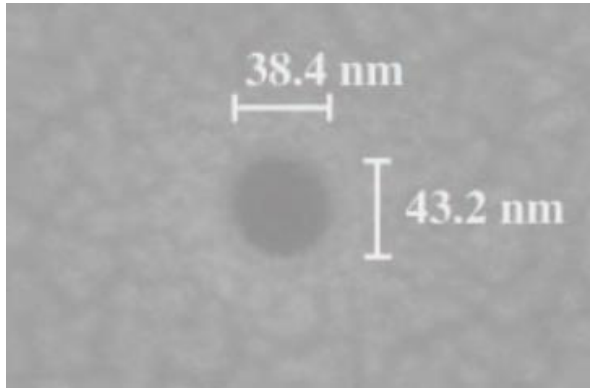


Nano Lett. 2019, 19, 7, 4371–4379

I. Introduction

Optics at critical intensity

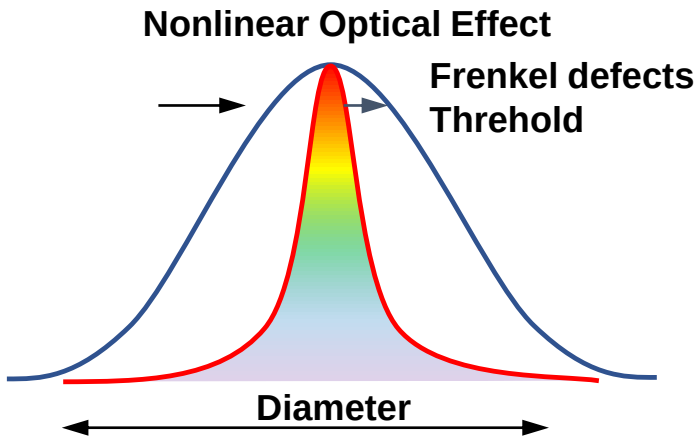


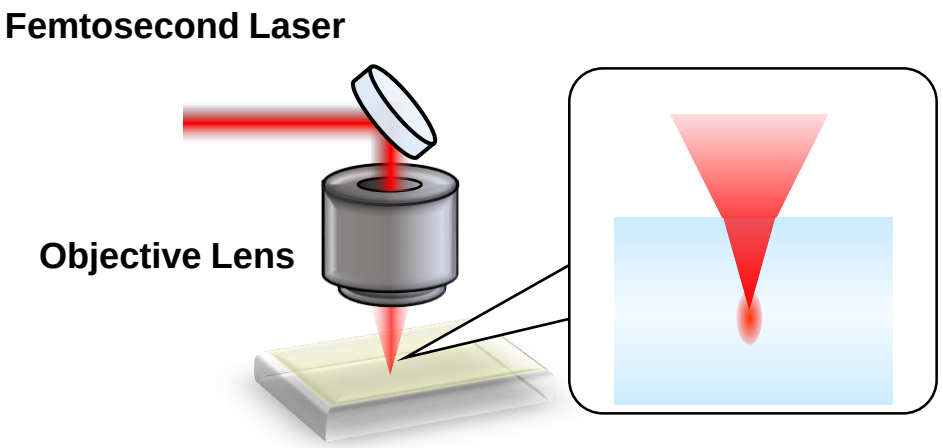


Ref: P.N.A.S., 101, 5856-5861 (2004)

University of Michigan

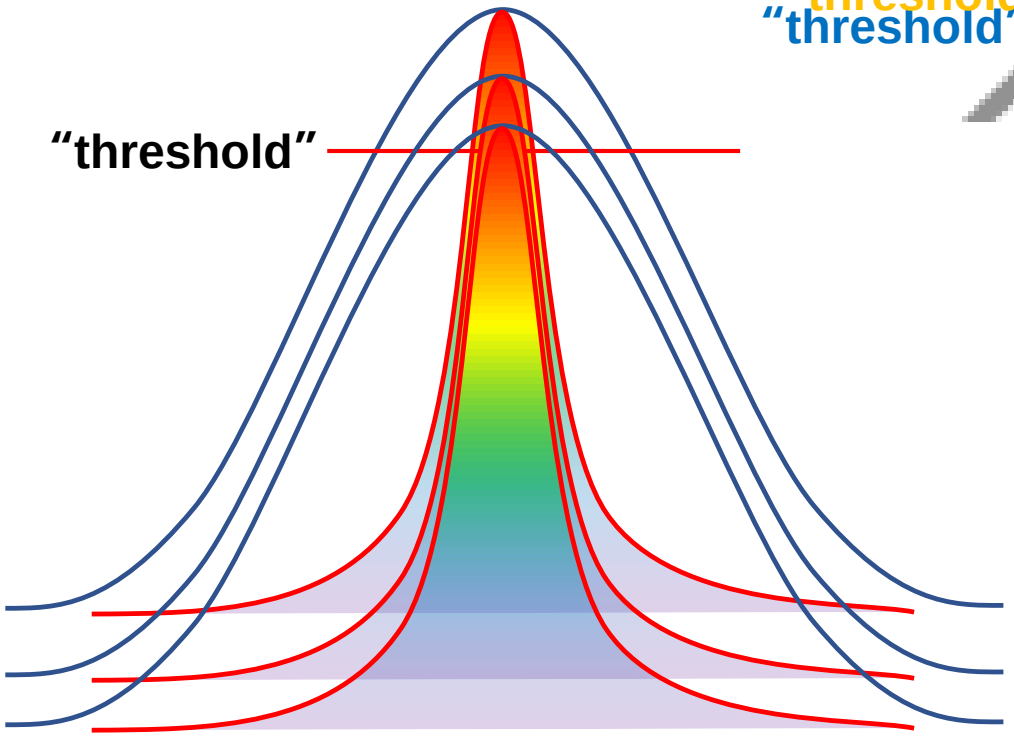
Direct Laser Writing Color Centers Approaching Q-Limit

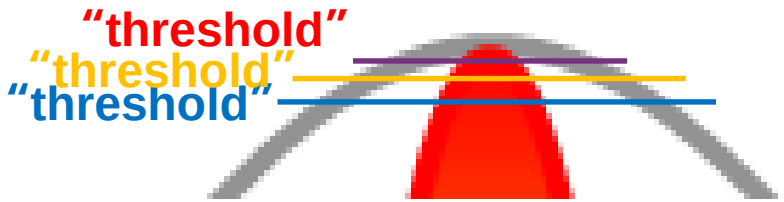




Can direct laser writing reach higher precision, atomic or close-to-atomic scale range?

Direct Laser Writing Color Centers Approaching Q-Limit





How to determine the intrinsic "threshold" ?

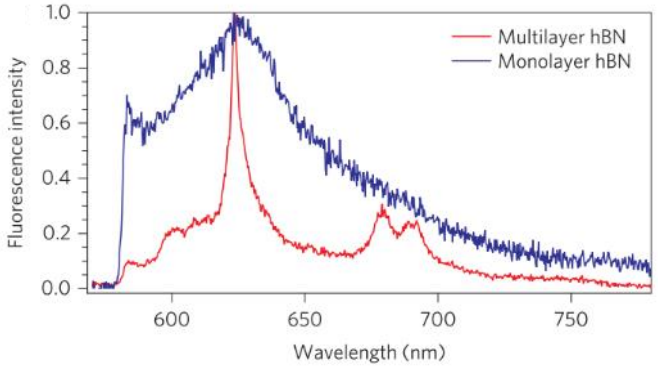
Experimentally determined "threshold" depending on characterizing methods

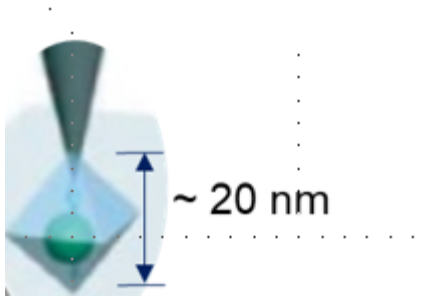
Direct Laser Writing Color Centers Approaching Q-Limit

Room Temperature Single Photon Emission from hBN

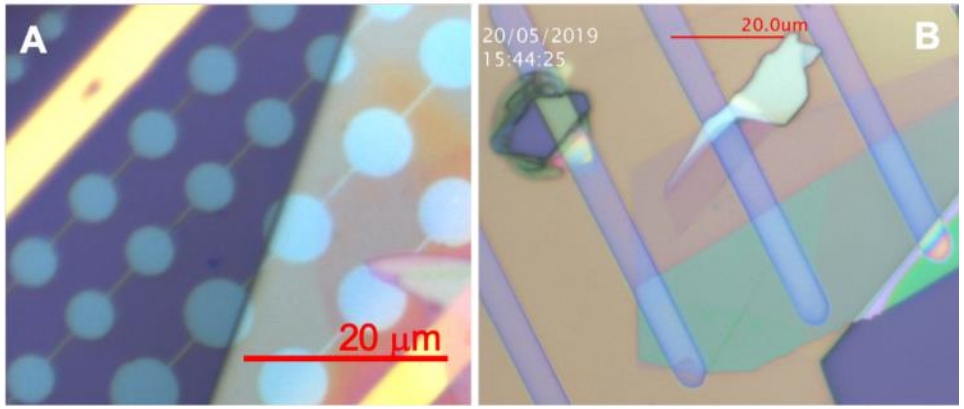
Hexagonal Boron Nitride:

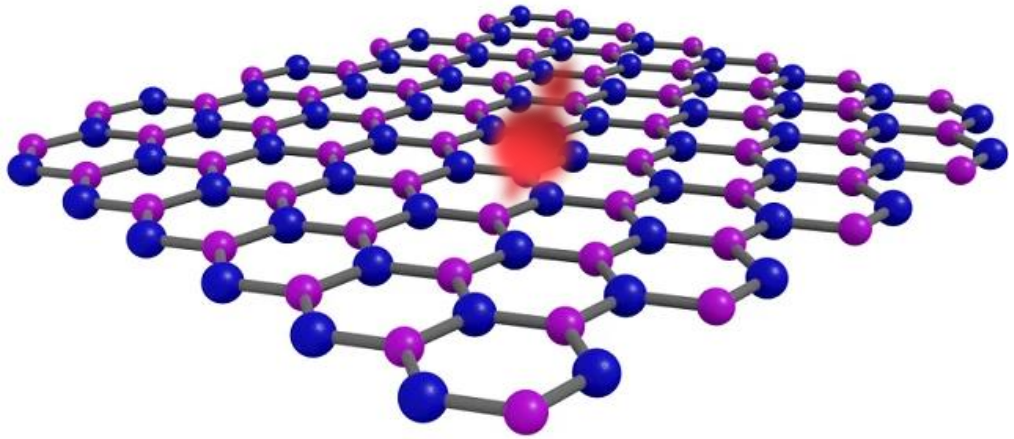
- ✓ van der Waals material
- ✓ High bandgap semiconductor (E_g ~ 6 eV)
- ✓ Atomically flat
- ✓ Reduce dielectric disorder
- ✓ Charge free





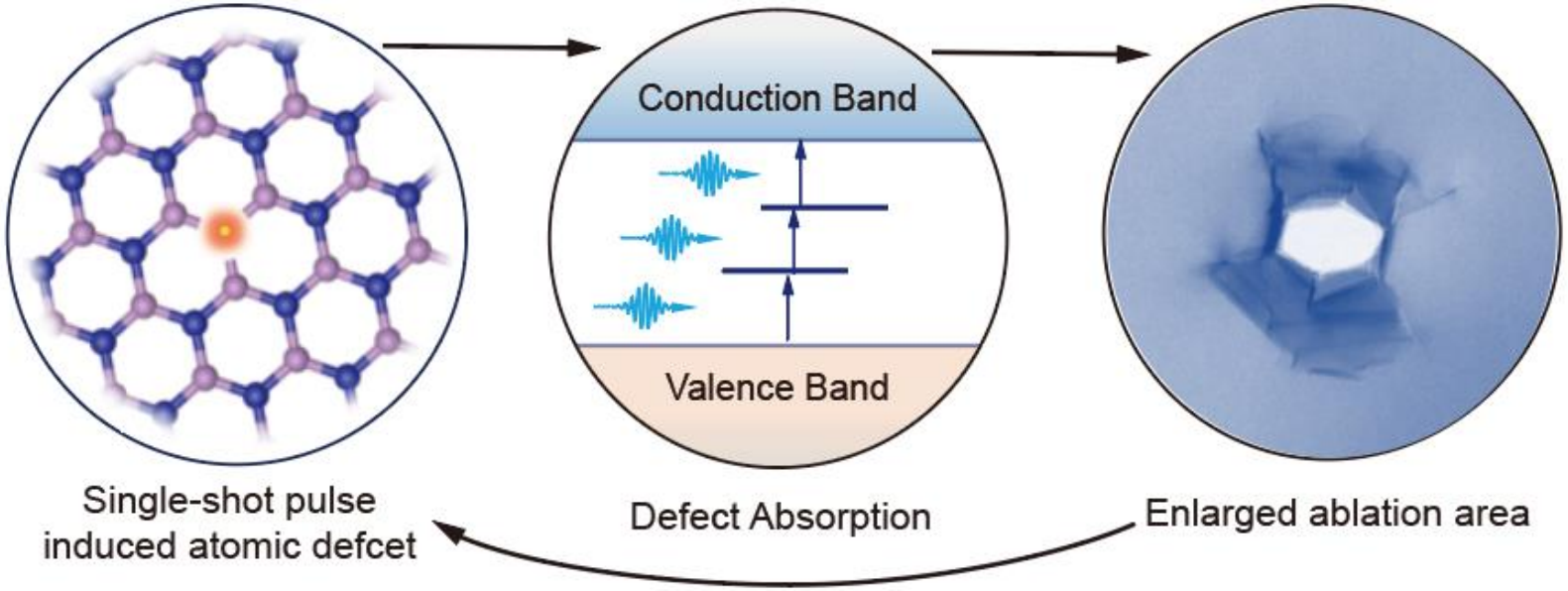
T. Tran et al., Nat. NanoTechnology. (2016)



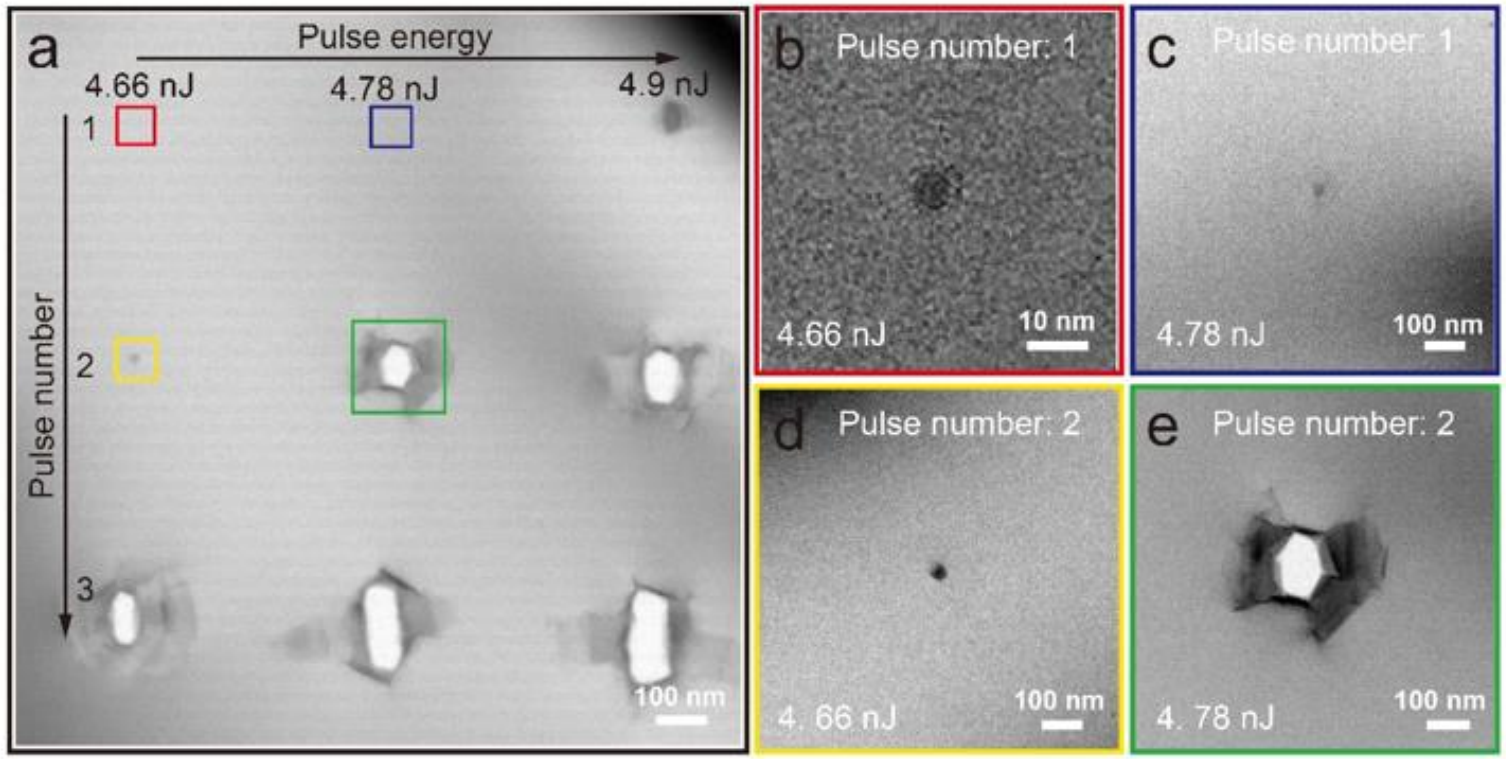


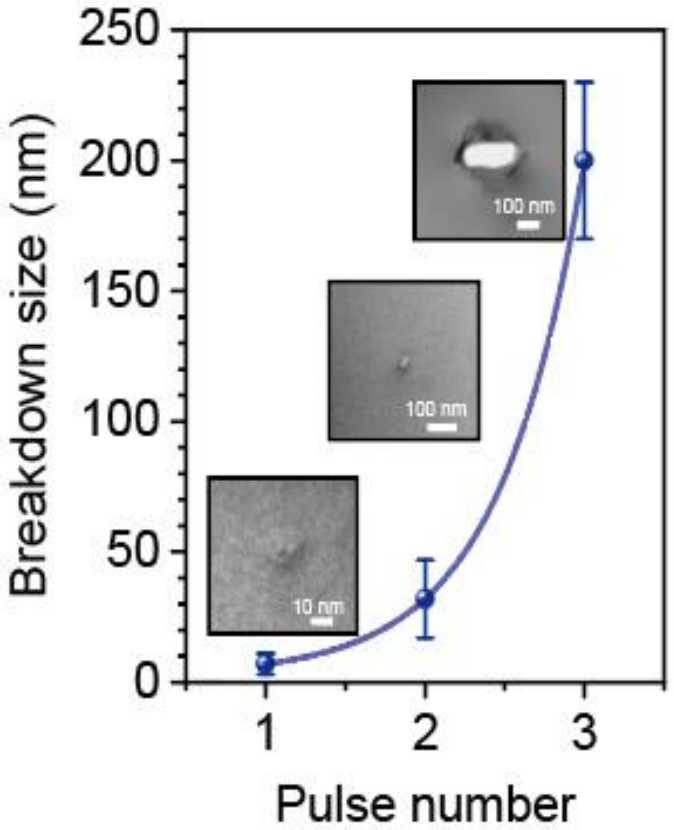
Direct Laser Writing Color Centers Approaching Q-Limit

If there is a defect, the absorption of the femtosecond laser will be largely increased.



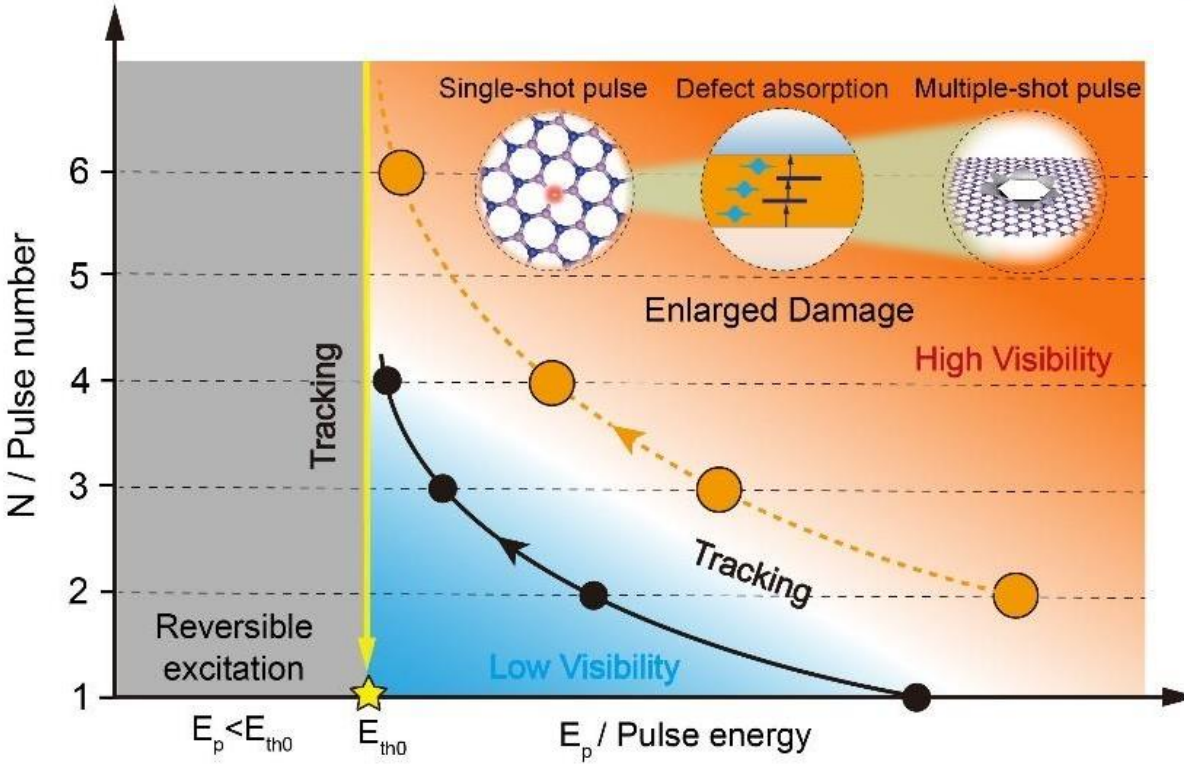
Direct Laser Writing Color Centers Approaching Q-Limit

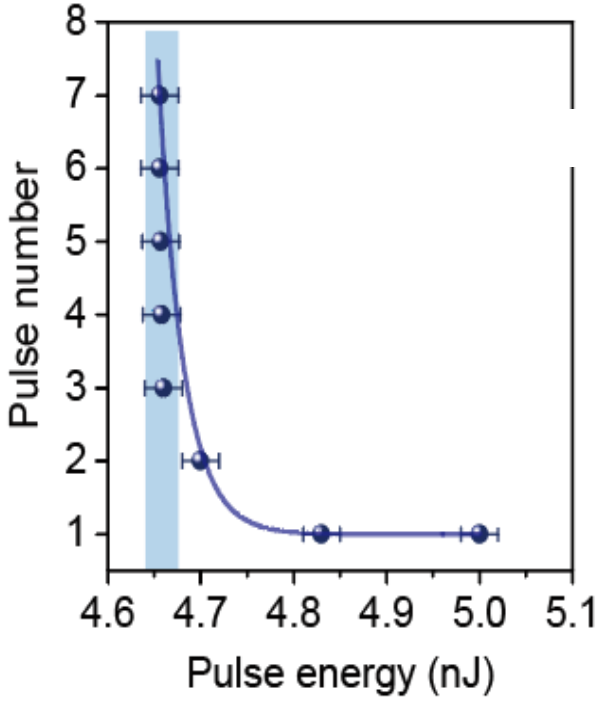




Direct Laser Writing Color Centers Approaching Q-Limit

Tracks the "intrinsic threshold" using the additional-laser-exposure dose



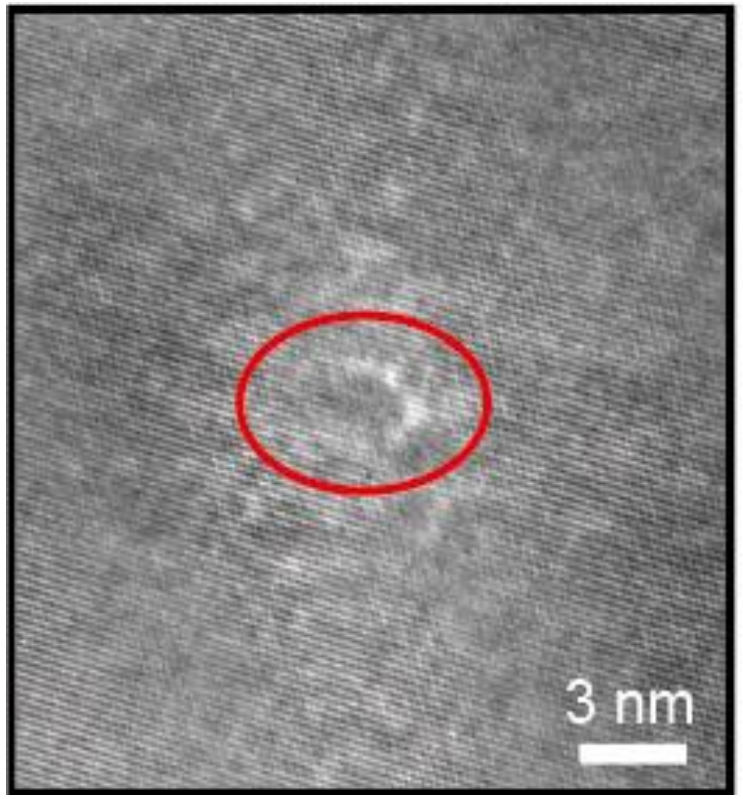


How to reach the intrinsic threshold?

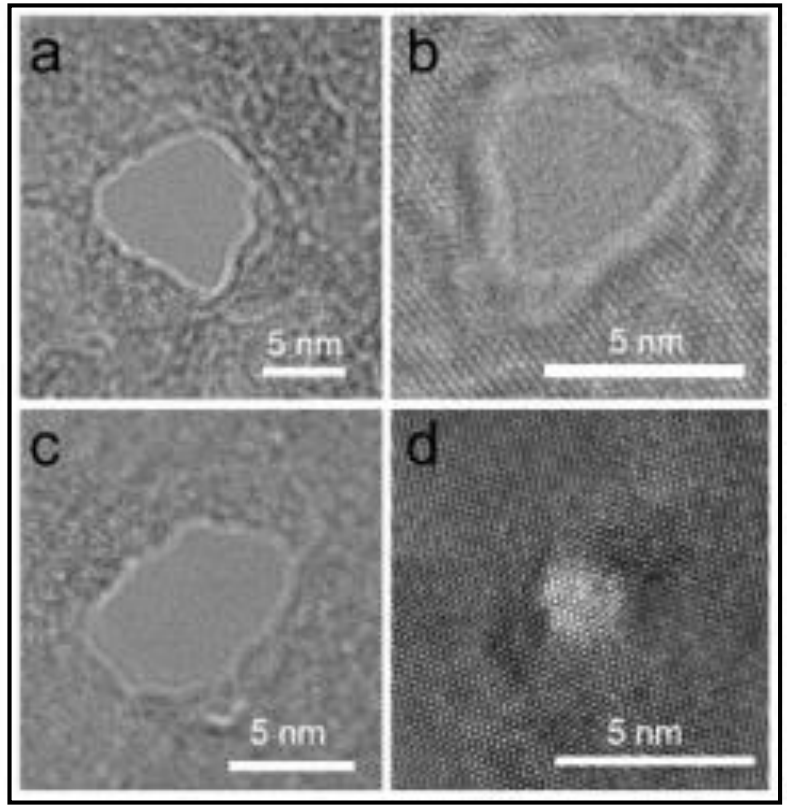
With multiple shots amplification, the experimental determination of "intrinsic threshold" becomes independent either on observation methods (by imaging or by spectroscopy, optically or electronically) or on their sensitivity

Direct Laser Writing Color Centers Approaching Q-Limit

Nanoscale modification

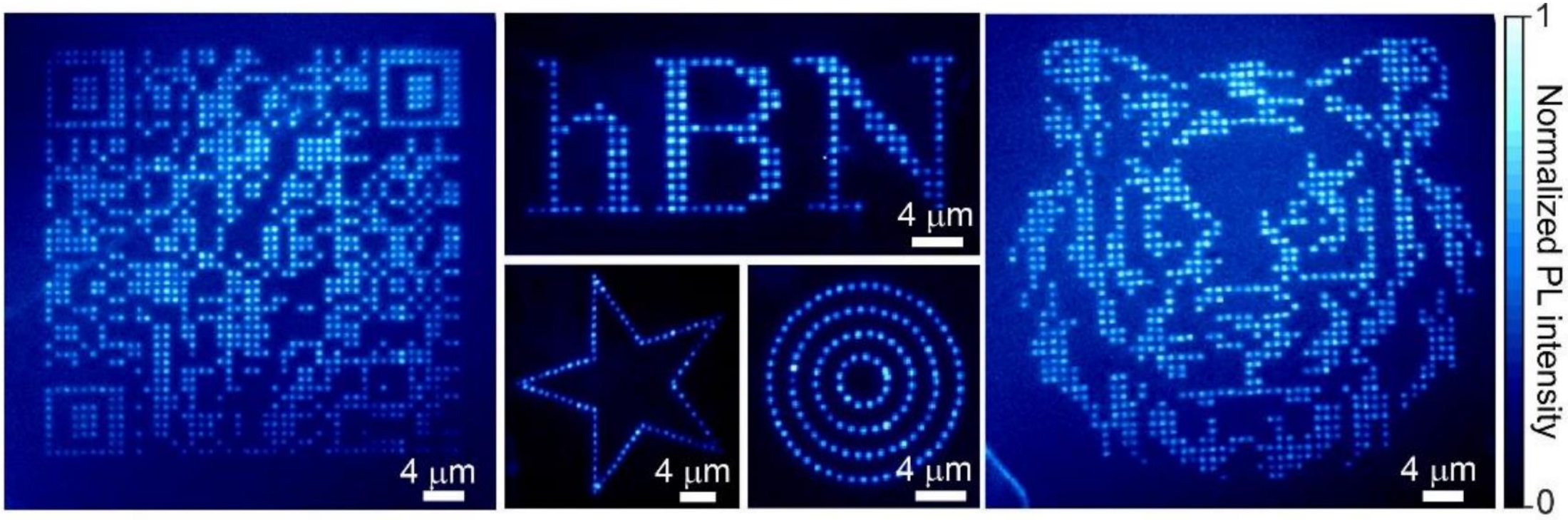


High-resolution TEM



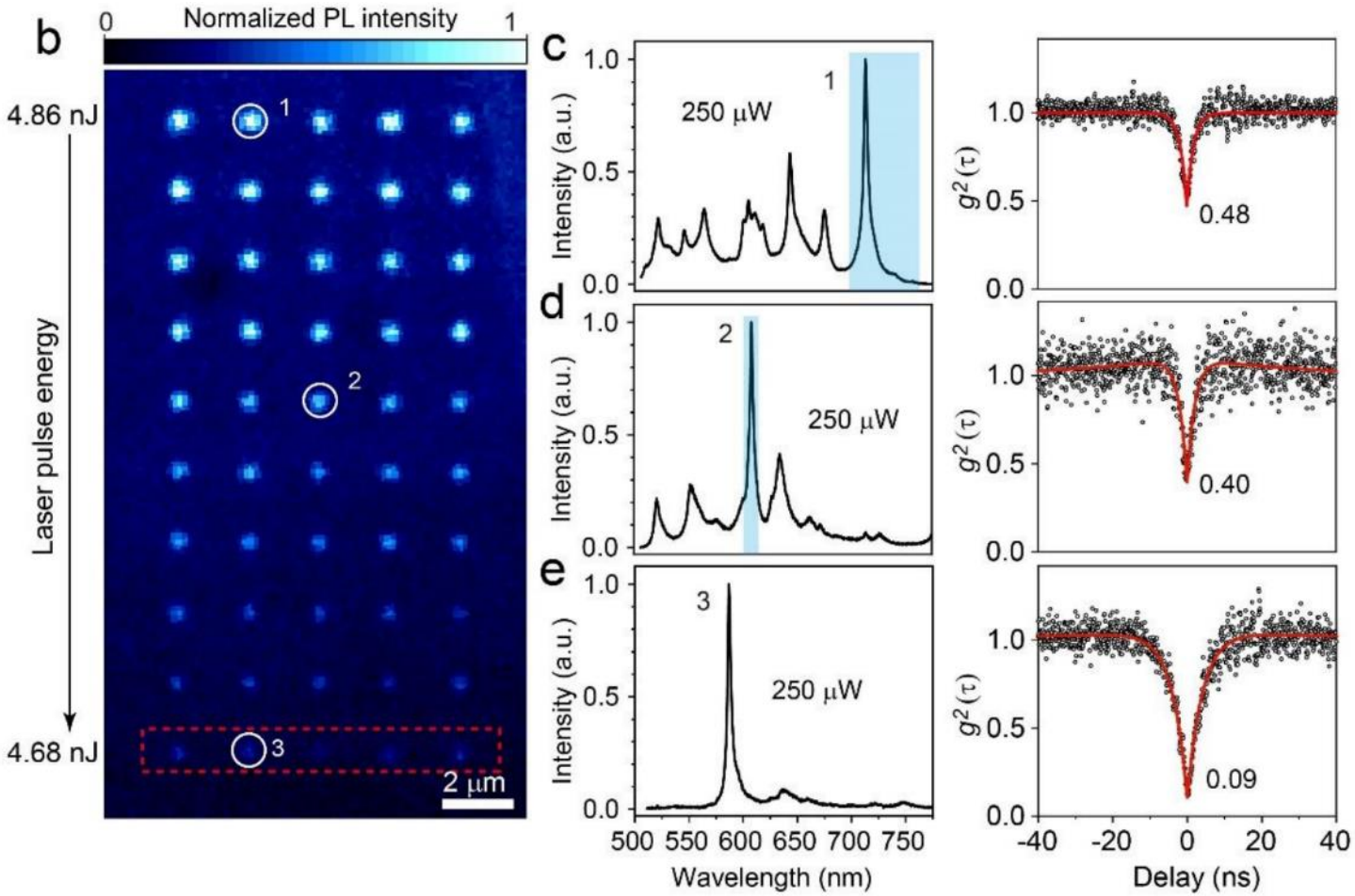
Feature size less than 5 nm

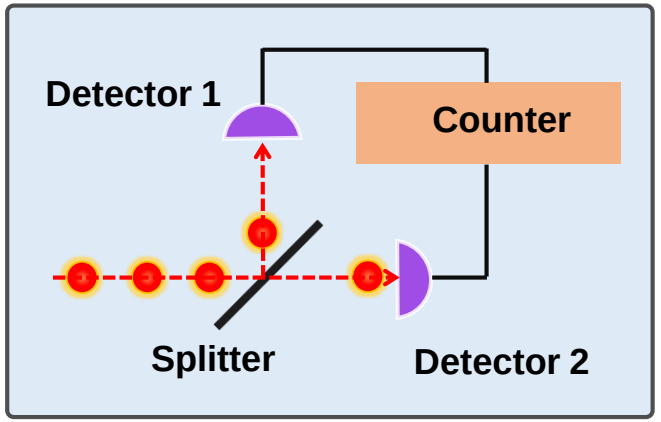
Direct Laser Writing Color Centers Approaching Q-Limit



Programmed pattern of color centers in hBN

Direct Laser Writing Color Centers Approaching Q-Limit



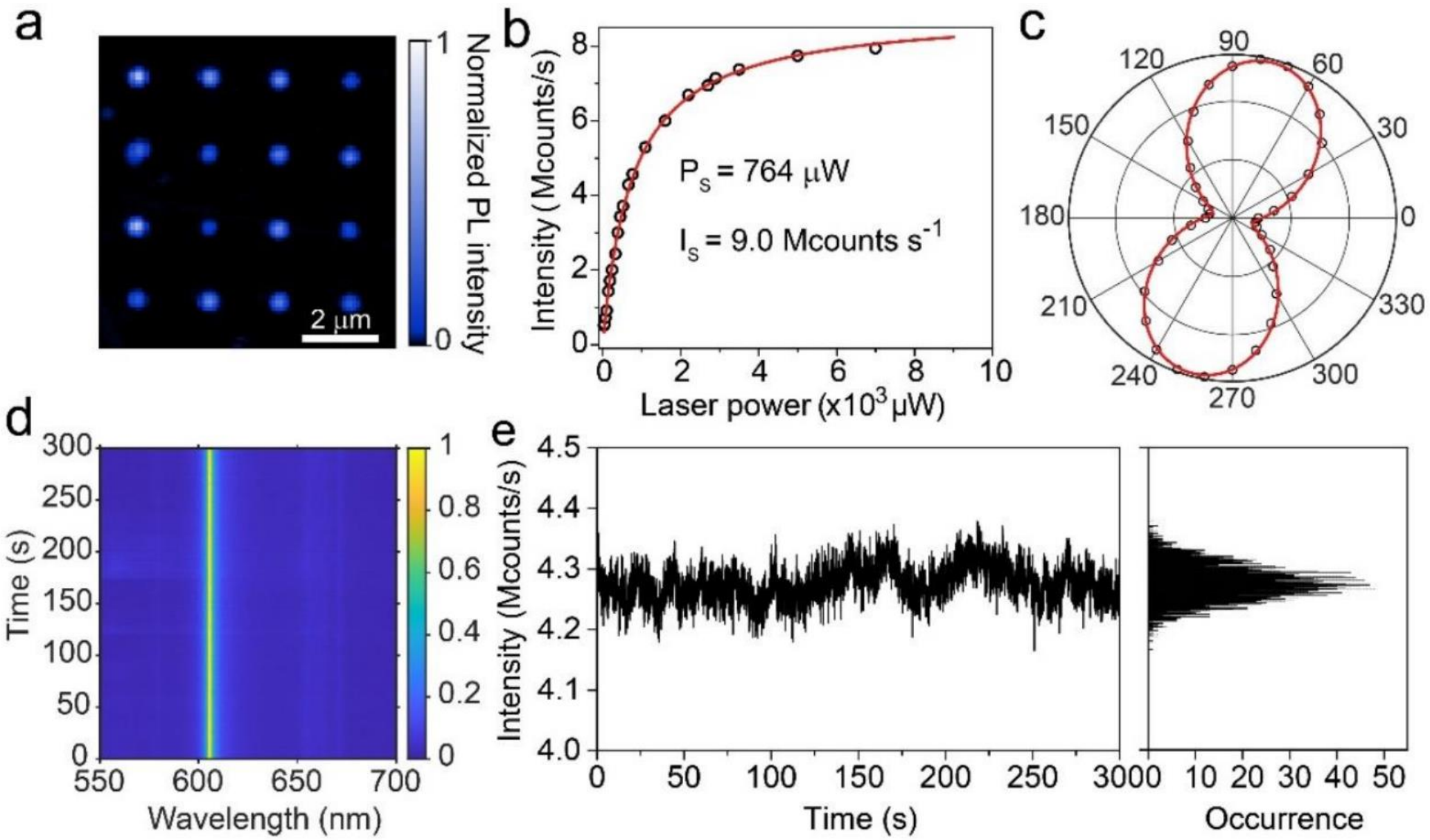


$$g^{(2)}(\tau) = \frac{\langle I(t+\tau)I(t)\rangle}{\langle I(t)\rangle^2}$$

$g^{(2)}(0)$<0.5 single photon emitter

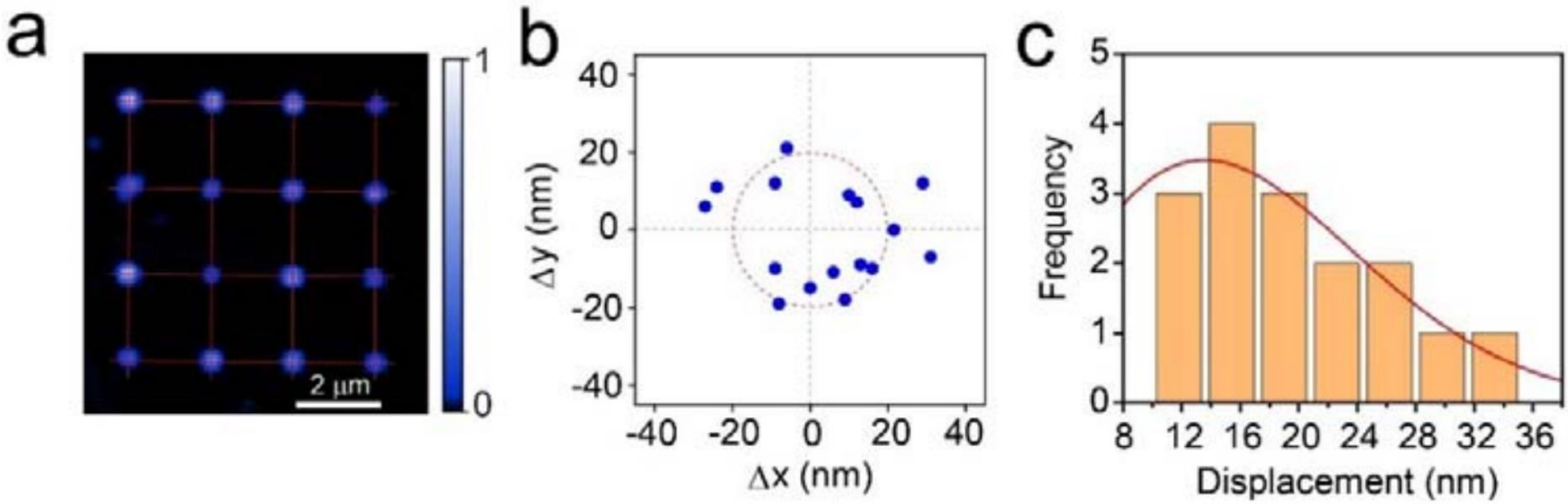
The emission from individual sites becomes polychromatic to monochromatic.

Direct Laser Writing Color Centers Approaching Q-Limit



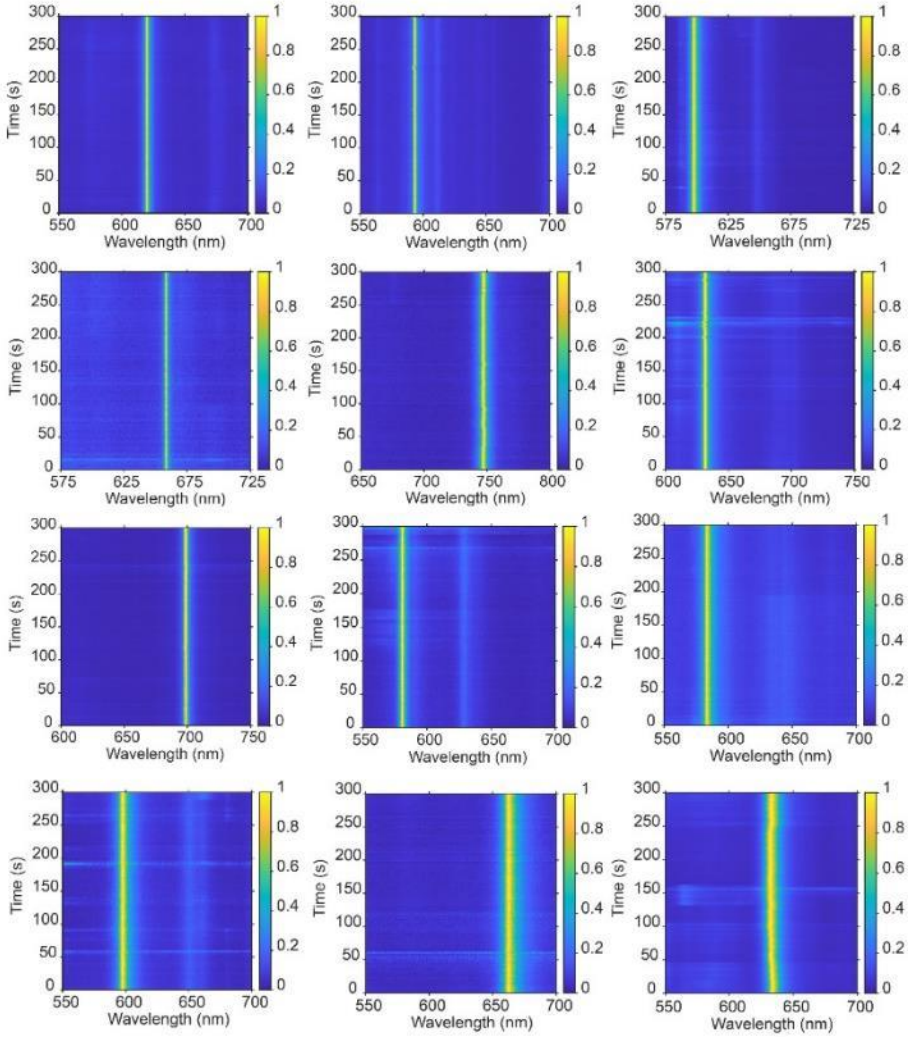
Direct Laser Writing Color Centers Approaching Q-Limit

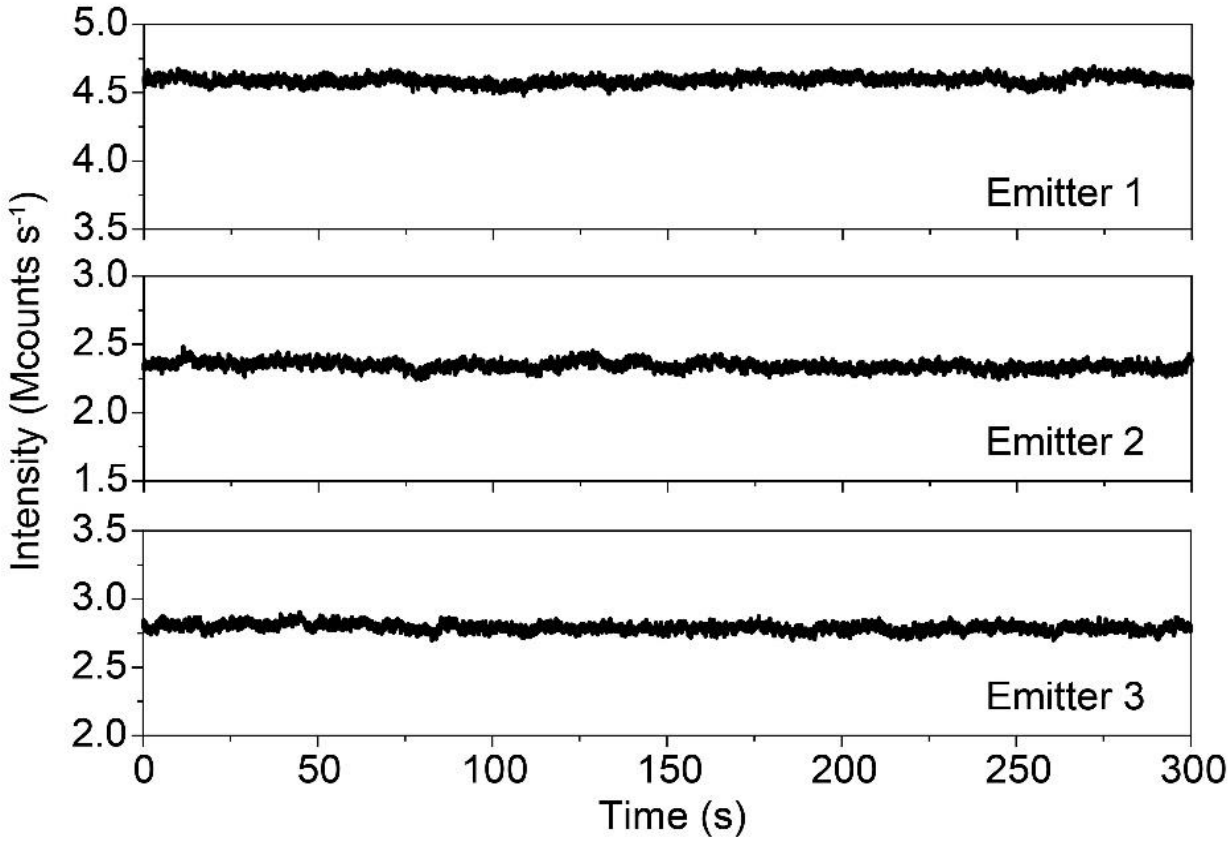
Position dispersion



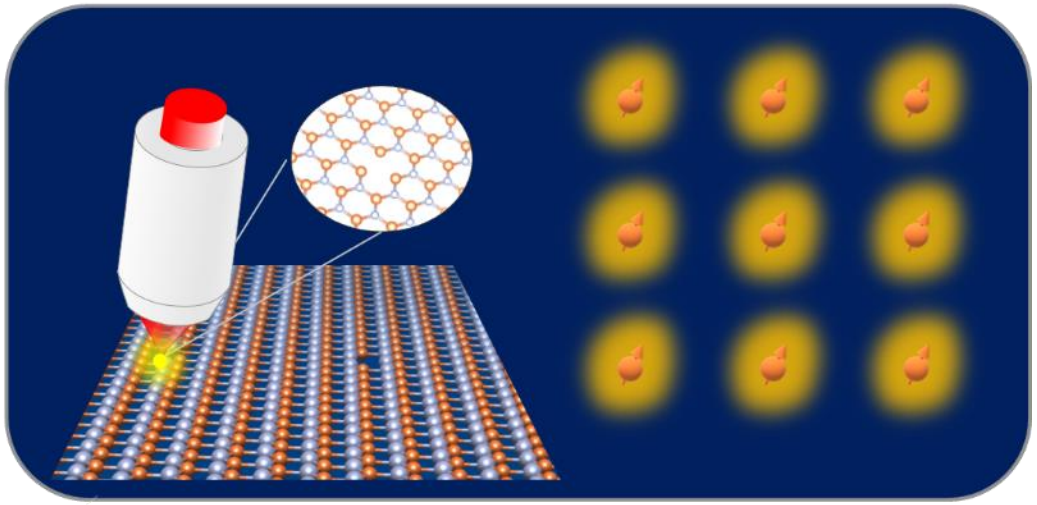
Direct Laser Writing Color Centers Approaching Q-Limit

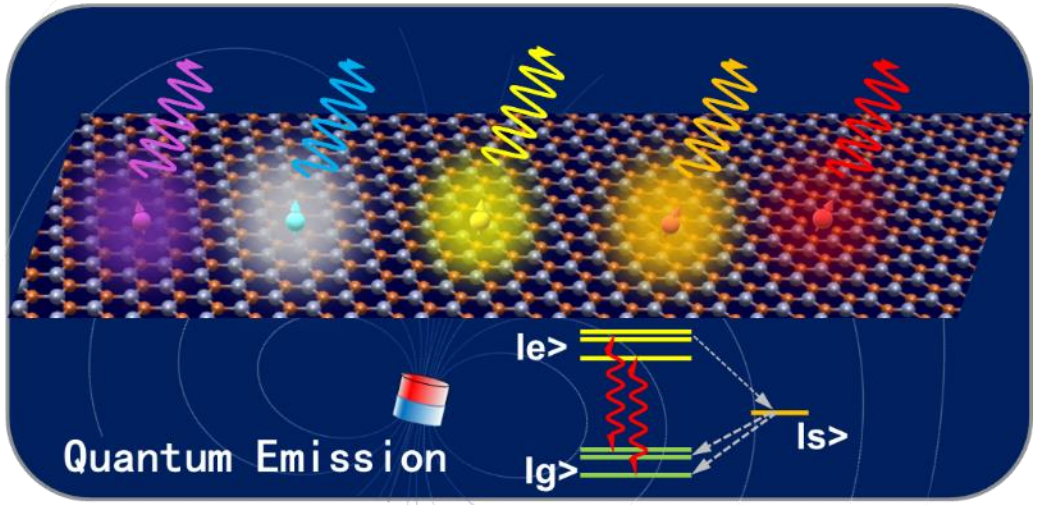
- **Highly photostable single photon emitter**

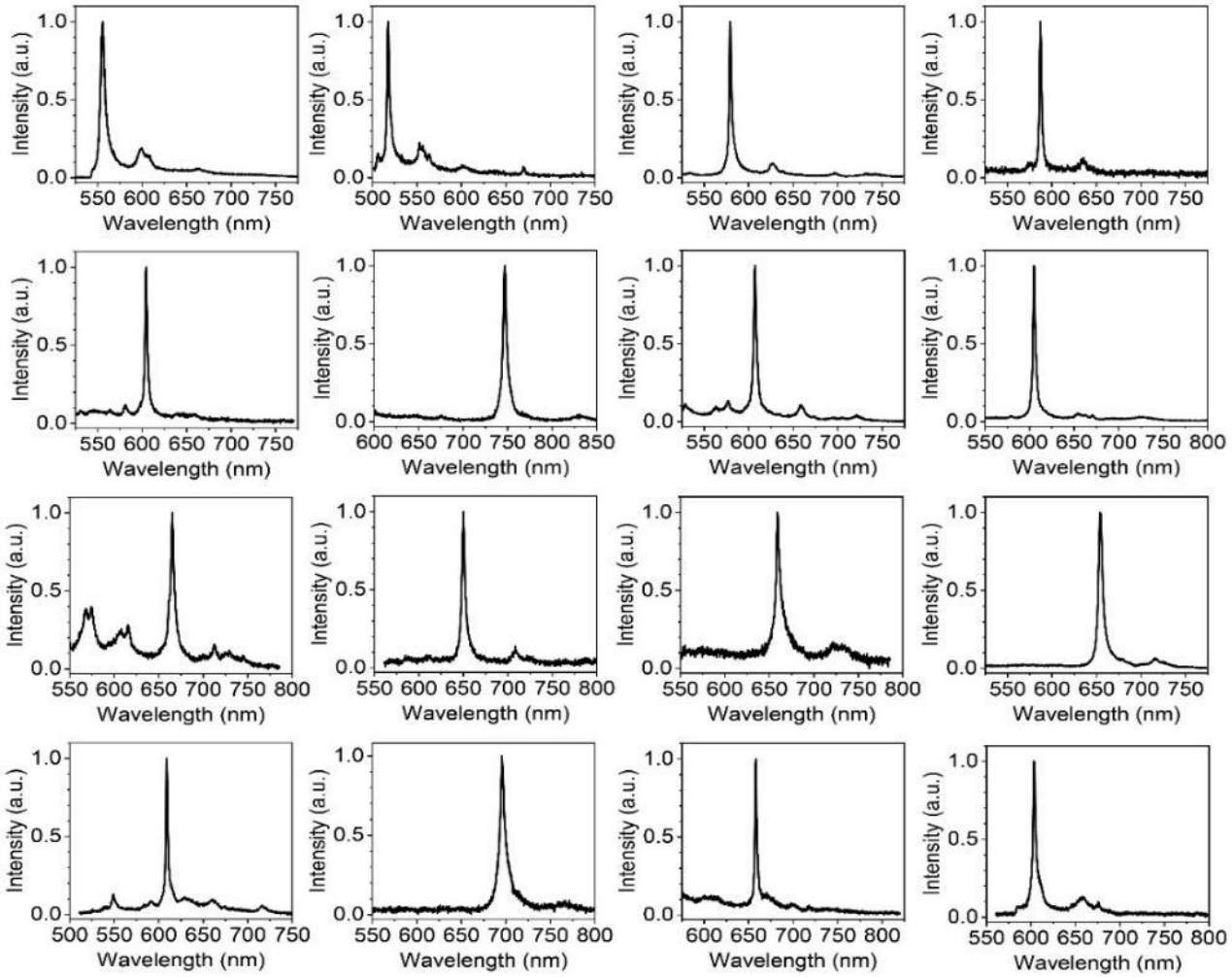




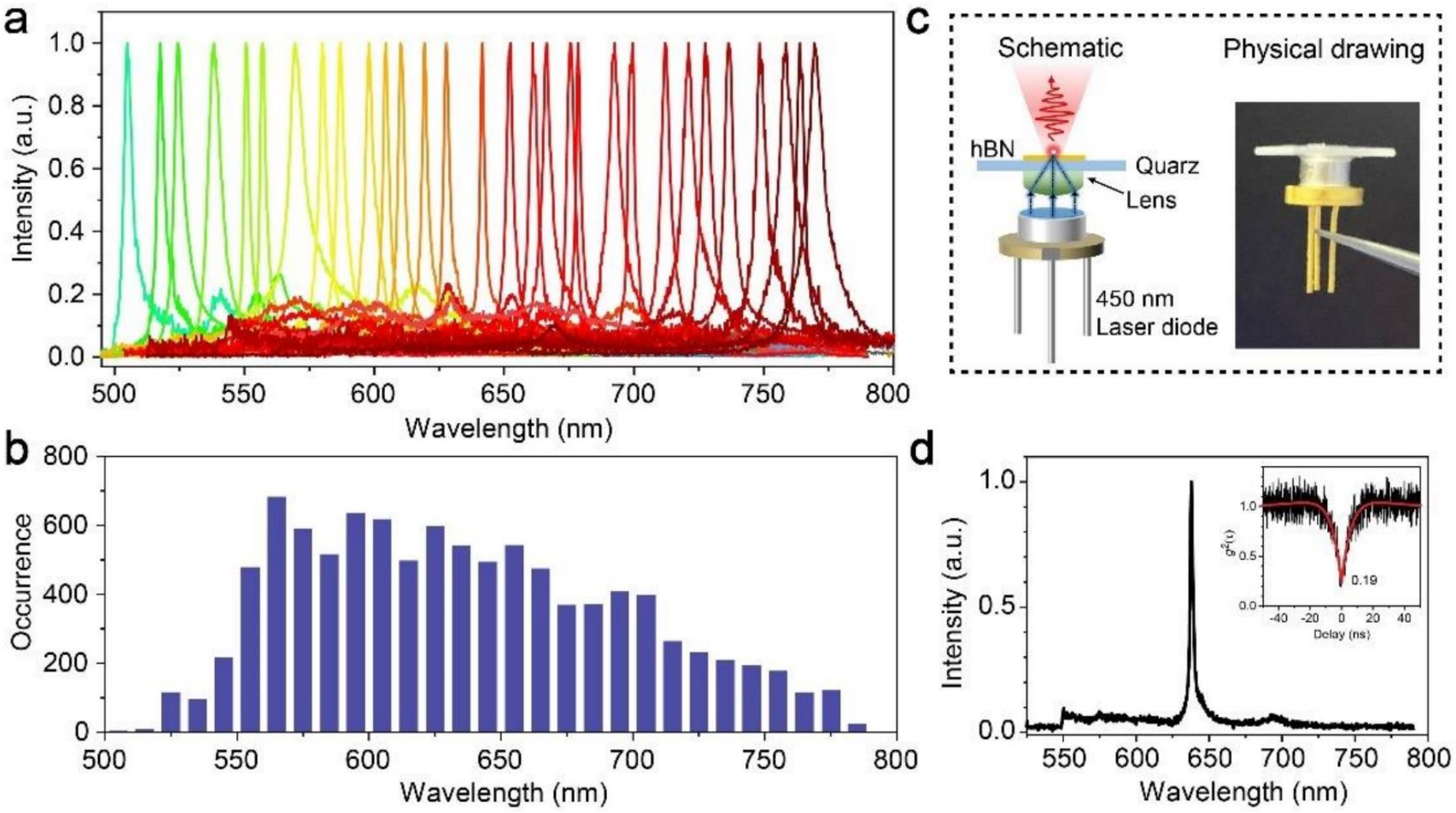
Direct Laser Writing Color Centers Approaching Q-Limit





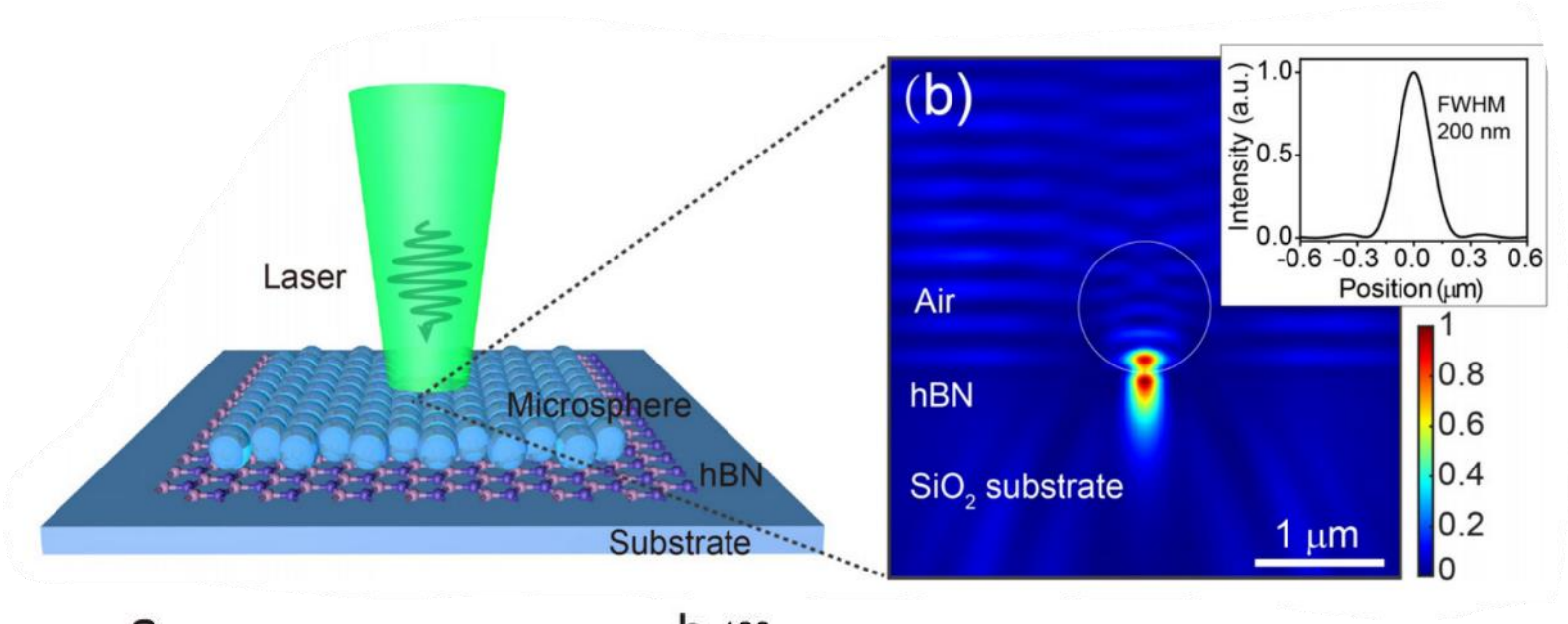


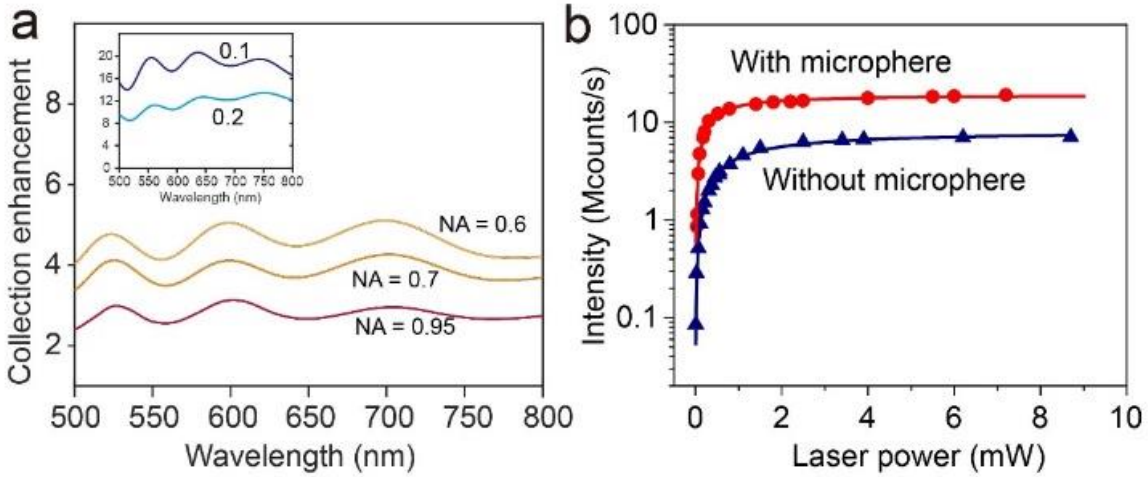
Direct Laser Writing Color Centers Approaching Q-Limit



Direct Laser Writing Color Centers Approaching Q-Limit

Coupling the color centers with photonic Structure





Enhanced brightness of quantum emitters via in situ coupling to the dielectric microsphere

Appl. Phys. Lett. 123, 133106 (2023)

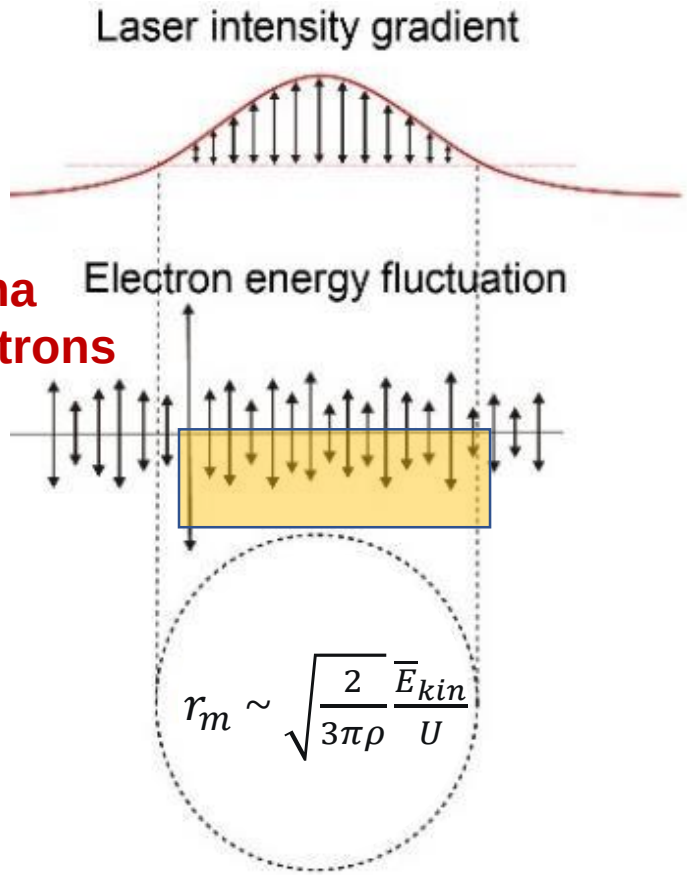
Direct Laser Writing Color Centers Approaching Q-Limit

Now, we can achieve laser writing with feature sizes as small as a few nanometers, i.e., less than 5 nm or λ/100.

Can we go further?
Can we control the type of laser-induced defect?

Direct Laser Writing Color Centers Approaching Q-Limit

Ultrafast Laser-matter Interaction



Plasma
Free electrons

Region of breakdown

~ 3 nm for hBN

The statistical (thermodynamics) description of laser-matter interaction

Free carrier could obtain quiver kinetic energy from an oscillating electric field of the light field, according to:

$$K(t) = K(t_0) - \frac{e}{\hbar}\int F(t')dt'$$

The ponderomotive energy

$$U(k) = U = \frac{e^2 E_0^2}{4\mu\omega_0^2}$$

Fluctuation of the system temperature can be expressed as:

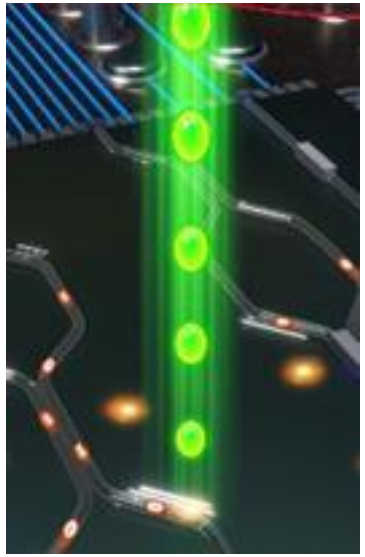
$$\Delta E_{kin} = \sqrt{\frac{2}{3N}} E_{kin}$$

Kinetic energy fluctuations ΔE_{kin} of the system comparable with the additional Energy provided by Pondermotive motion U

Direct Laser Writing Color Centers Approaching Q-Limit

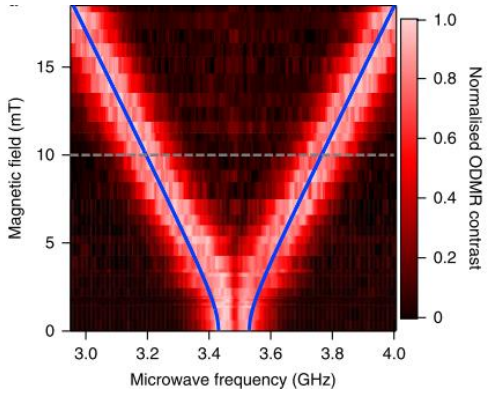
Challenge in Control Defect Type

Idea Single-photon Emitter

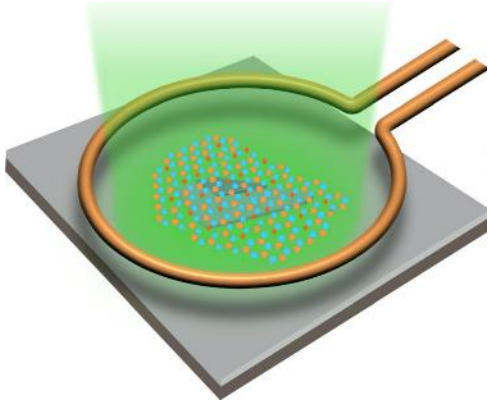


high purity, brightness
excellent stability
monochromaticity

High-sensitivity nanoscale quantum sensor



Initialization and read-out of spin defects (2020)



Quantum sensing imaging with LMs (Several groups, 2022)

high brightness
excellent stability
Long coherence

Boron vacancy

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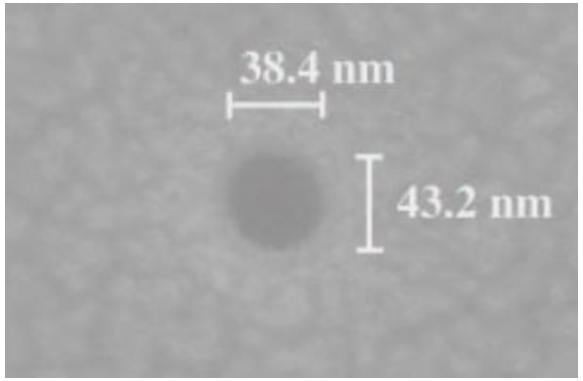
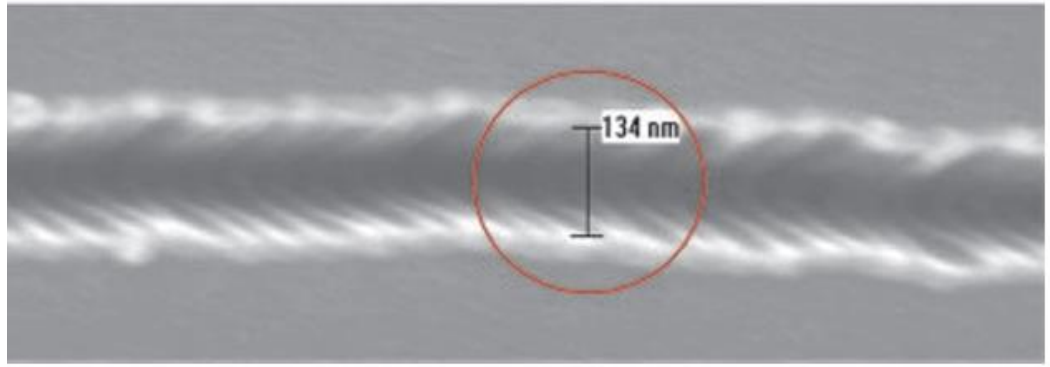
Laser Writing Sub-10 nm Structure in Semiconductors

From Dot To Line

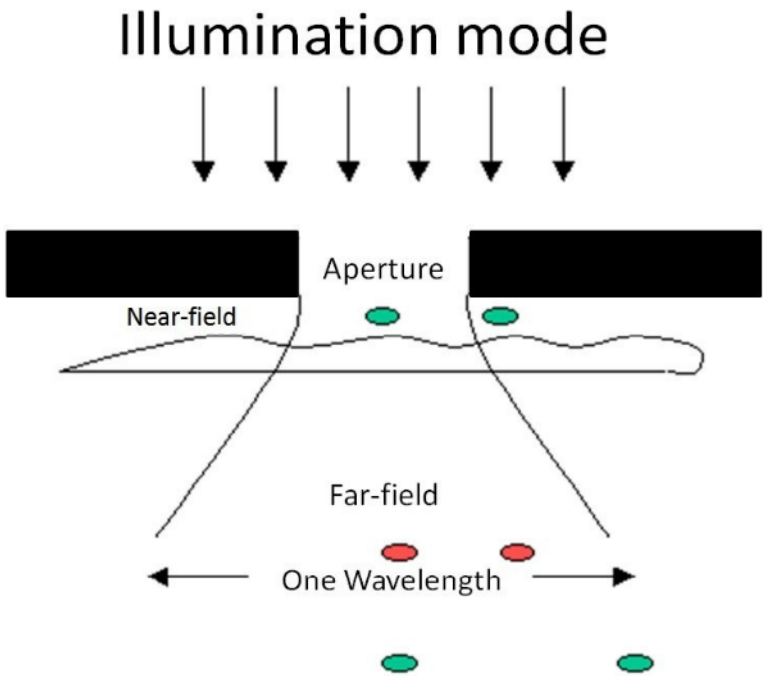


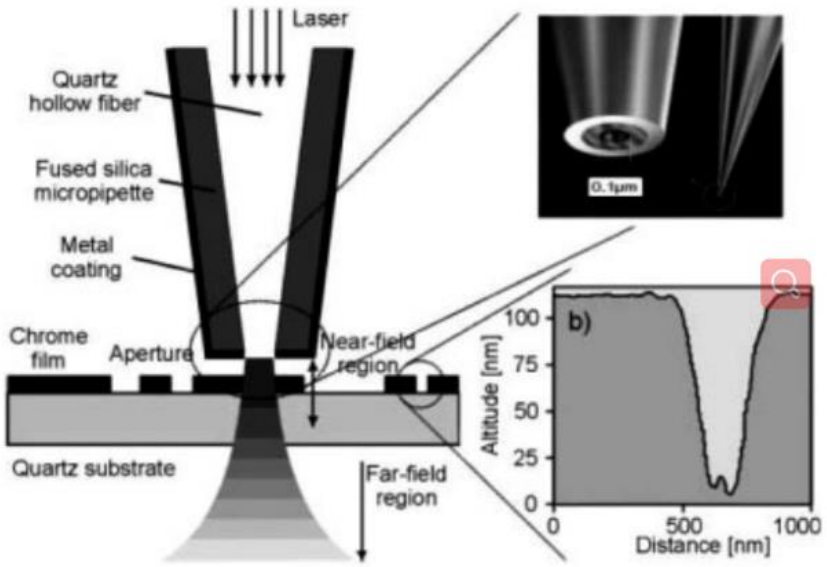


Ref: P.N.A.S., 101, 5856-5861 (2004)

Laser Writing Sub-10 nm Structure in Semiconductors

Near Field Optical Effect



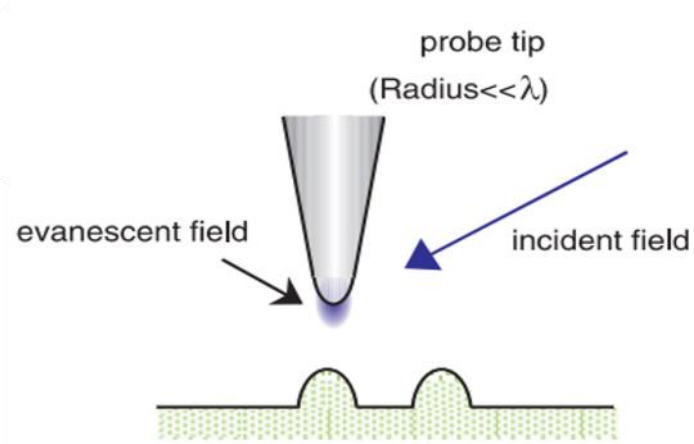


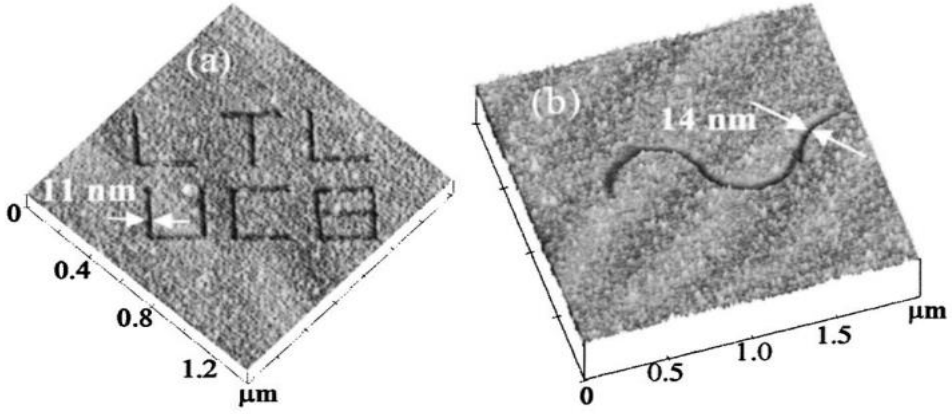
The laser NSOM combination for surface nanostructuring

Laser & Photon. Rev. 2010, 4, 123-143

Laser Writing Sub-10 nm Structure in Semiconductors

Near Field Optical Effect With AFM tip



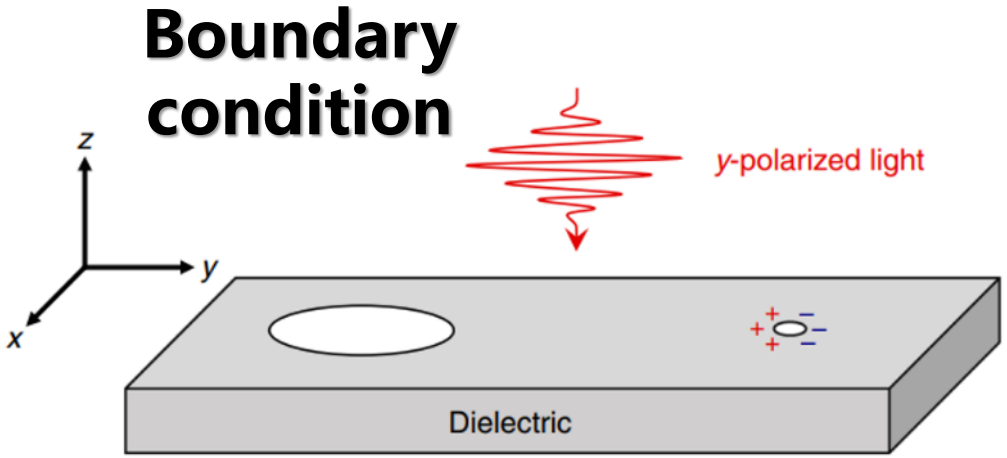


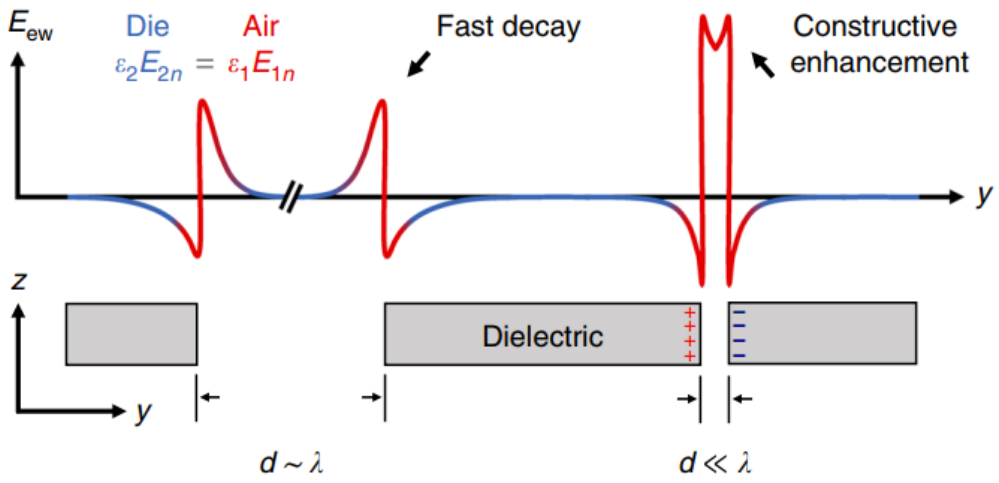
Appl. Phys. Lett. 82, 1146–1148 (2003)

Requires precise control
Easy to damage
Difficult to scan large areas

Laser Writing Sub-10 nm Structure in Semiconductors

Near Field Optical Effect in Prefabricated Structure

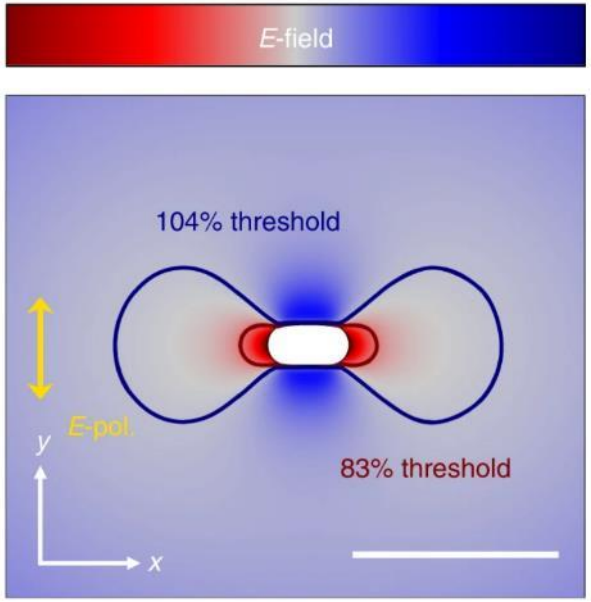


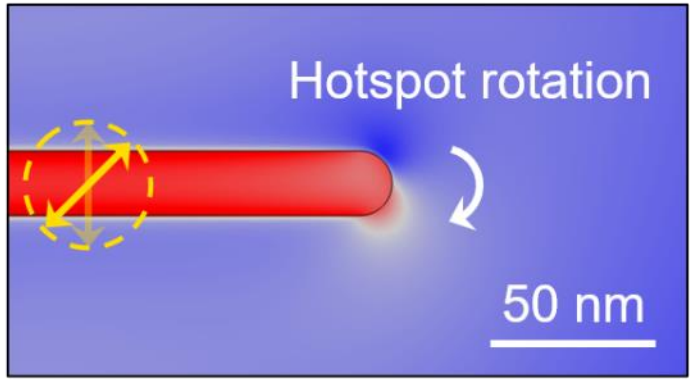


Enhanced evanescent field

Light: Science & Applications volume 9, Article number: 41 (2020)

Laser Writing Sub-10 nm Structure in Semiconductors

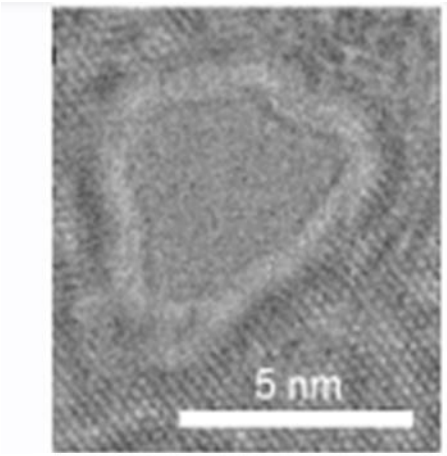


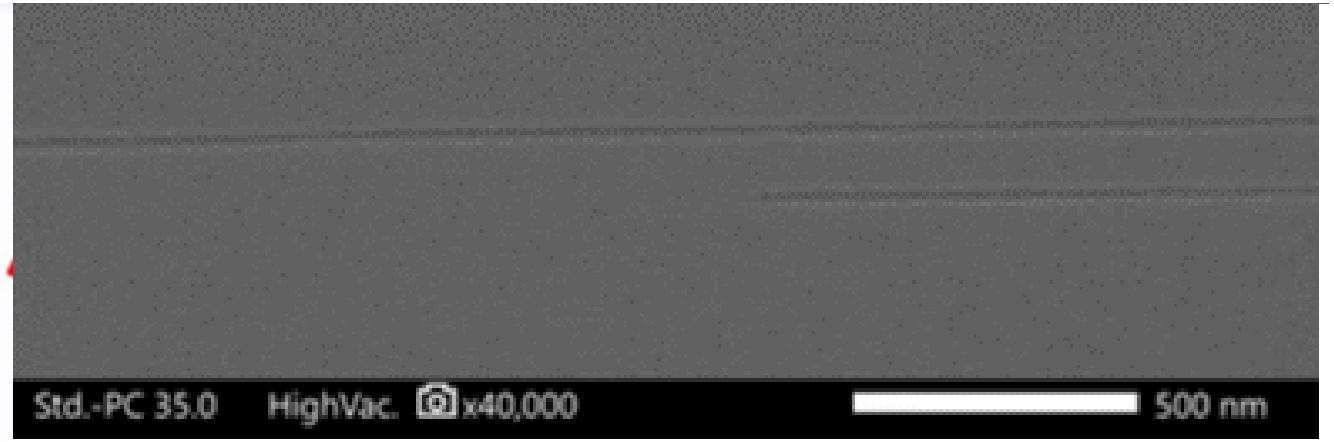


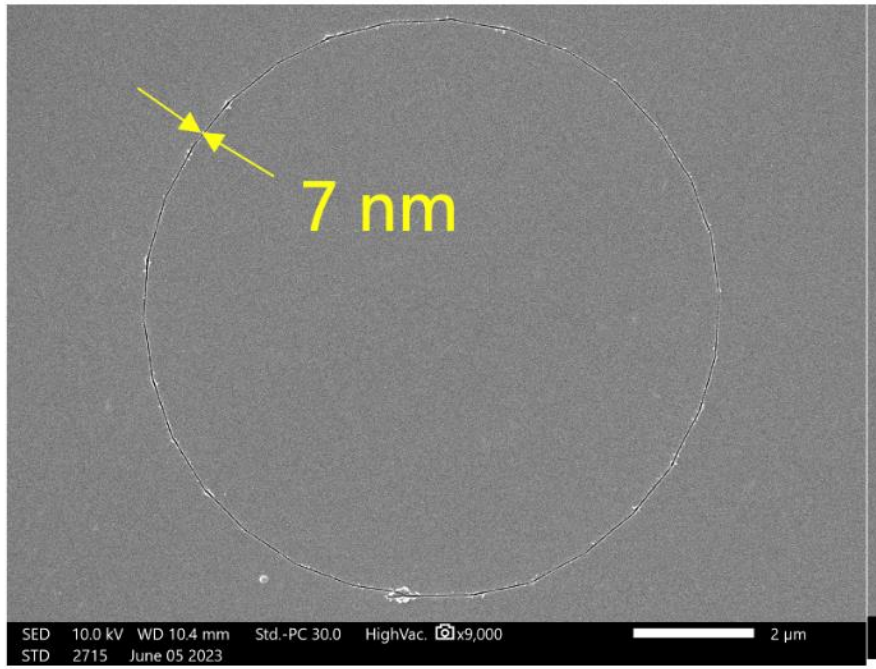
Light: Science & Applications volume 9, Article number: 41 (2020)

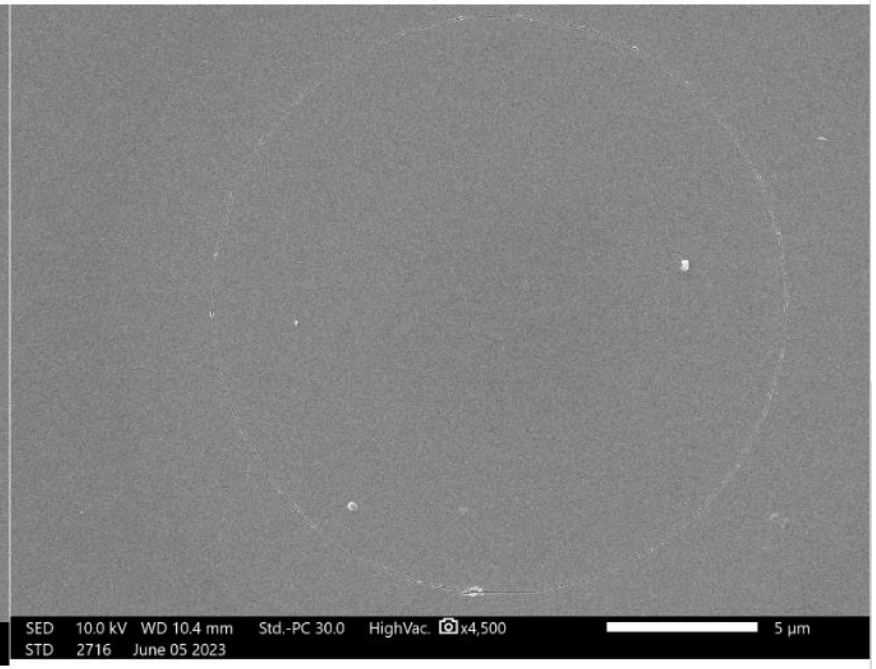
Laser Writing Sub-10 nm Structure in Semiconductors

From Dot To Line









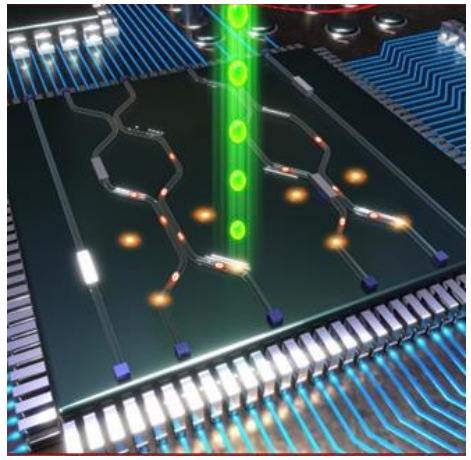
Tip-free
Mask-free
Resist-free
Technique

Summary

1) **Ultrafast laser writing is a highly effective method for developing quantum technologies with color centers by enabling precise engineering at the nanoscale.**

2) **. The color centers produced through laser writing exhibit exceptional precision, stability, and efficiency.**

3) **Laser writing is a very promising technique for quantum devices fabrication**



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Thank you for your attention!