Ultrafast Meets Ultrasmall: Laser Writing at the Quantum Frontier

Hong-Hua Fang
Tsinghua University
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Our Group @ Tsinghua University

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Research Activity

Light-Matter Interaction @ Nanoscales

Laser Writing

Time-resolved Image & Spectroscopy

Nanophotonics

Optoelectronics

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
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

Intensity

FWHM ~fs

Time (fs)

Laser & Photonics Reviews, 2100029, 2021
I. Introduction

\[ \sigma \propto I^2 \]

Ultrashort Pulse

\[ \tau < 10^{-12} \, s \]

Single-photon absorption

Two-photon absorption

I. Introduction

Ultrafast Laser Ablation


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

Outline

I. Introduction to Ultrafast Laser Writing Techniques

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Laser Writing for Quantum Techniques
Laser Writing for Quantum Techniques

Laser Writing Waveguide


Two-Particle Bosonic-Fermionic Quantum Walk via Integrated Photonics


Macquarie University, Australia

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 centers

1. Quantum Light Source

2. Spin

Quantum Information Process
Nanoscale Sensor
Laser Writing for Quantum Techniques

Color Center as Quantum Nodes

Integrated quantum photonics with color centers

SCIENCE, 2021, 372, 259-264
Laser Writing for Quantum Techniques

Quantum Sensing with color center
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Color Center in AIN

AlN-based integrated photonics

AlN photonic crystal membranes.

Color centers Engineering

High Precision Engineering Color-Center

Engineering NV spatial patterns.

Focused ion beam
◆ High energy, strong scattering
◆ Lattice damage
◆ Degraded optical properties
◆ Low produce yield

ACS Nano 16, 3695−3703 (2020)

Nano Lett. 19, 2121−2127 (2019)
Laser writing color center in AlN Crystal

515 nm Fs laser

Single photon emission

AIN
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

(a) Schematic diagram showing energy levels and transitions.

(b) Graph showing intensity (counts/s) vs. laser power (mW) with different curves for background, corrected, and signal.

(c) Histogram showing counts vs. saturated intensity (10^6 Counts/s).

(d) Polarization diagram showing intensity vs. angle.

(e) Graph showing intensity (kcps/0.2s) vs. time (s) and occurrence.

(f) Graph showing cumulative percentage (%).
Color Center in AlN

[Graphs showing the occurrence and intensity of color centers in AlN]

Point defect $N_{\text{AlO}_N}$

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
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V. Laser Writing Sub-10 nm Structure in Semiconductors
Color centers are atomic defects. How to minimize the lattice damage?

Nano Lett. 2019, 19, 7, 4371–4379
I. Introduction

Optics at critical intensity

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
Room Temperature Single Photon Emission from hBN

Hexagonal Boron Nitride:
✓ van der Waals material
✓ High bandgap semiconductor ($E_g \sim 6$ eV)
✓ Atomically flat
✓ Reduce dielectric disorder
✓ Charge free

If there is a defect, the absorption of the femtosecond laser will be largely increased.
Direct Laser Writing Color Centers Approaching Q-Limit

![Diagram showing pulse energy and breakdown size vs. pulse number.]

- **a** Pulse energy: 4.66 nJ, 4.78 nJ, 4.9 nJ
  - Pulse number: 1
- **b, c** Pulse number: 1, Pulse energy: 4.66 nJ, 4.78 nJ
- **d, e** Pulse number: 2, Pulse energy: 4.66 nJ, 4.78 nJ

Graph: Breakdown size (nm) vs. Pulse number

- Breakdown size: 0, 50, 100, 150, 200, 250 nm
- Pulse number: 1, 2, 3
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
Programmed pattern of color centers in hBN
$g^{(2)}(0) < 0.5$ single photon emitter

The emission from individual sites becomes polychromatic to monochromatic.
Direct Laser Writing Color Centers Approaching Q-Limit

(a) Image showing 2 µm scale.
(b) Graph with data points and linear fit.
(c) Polar plot.
(d) Color map with wavelengths.
(e) Time intensity graph with occurrence data.
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
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)
Now, we can achieve laser writing with feature sizes as small as a few nanometers, i.e., less than 5 nm or $\lambda/100$. 

Can we go further? Can we control the type of laser-induced defect?
Ultrafast Laser-matter Interaction

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:

\[ \Delta E_{kin} = \sqrt{\frac{2 \pi}{3N}} E_{kin} \]

Kinetic energy fluctuations \( \Delta E_{kin} \) of the system comparable with the additional Energy provided by Pondermotive motion \( U \)

\[ r_m \sim \sqrt{\frac{2 \pi}{3N}} E_{kin} \]

~ 3 nm for hBN
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**
- high brightness
- excellent stability
- Long coherence

*Initialization and read-out of spin defects (2020)*

*Quantum sensing imaging with LMs (Several groups, 2022)*

*Boron vacancy*
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Laser Writing Sub-10 nm Structure in Semiconductors

From Dot To Line

Laser Writing Sub-10 nm Structure in Semiconductors

Near Field Optical Effect

Illumination mode

The laser NSOM combination for surface nanostructuring

*Laser & Photon. Rev. 2010, 4, 123-143*
Near Field Optical Effect With AFM tip

Requires precise control
Easy to damage
Difficult to scan large areas

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!