Theoretical Review of a new approach of Lithography at nm Resolution

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Introduction

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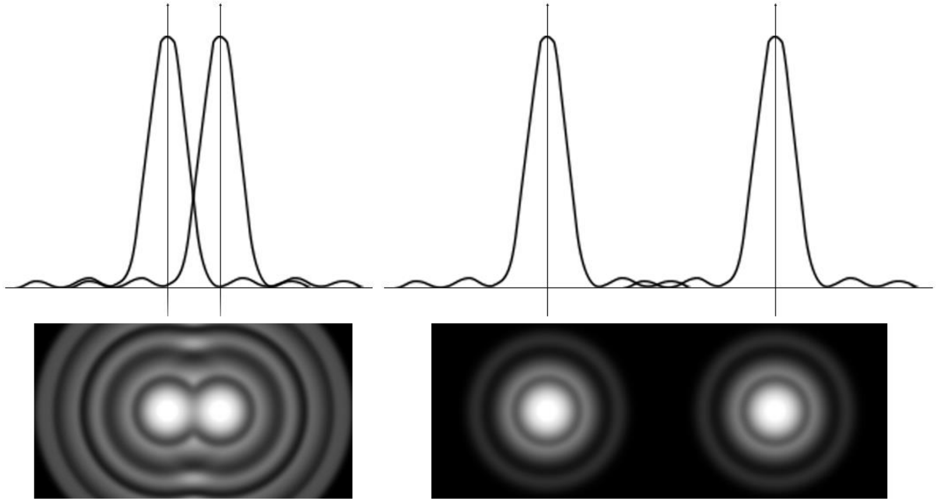
The new potential offered for lithography by the combination of 2 elements:

1. SMLM (Single Molecule Localization Microscopy)
2. EIT (Electromagnetically Induced Transparency)

1 SMLM (Single Molecule Localization Microscopy)



Two light-emitting points can separate in time and space, to break through the Abbe limit.

Abbe limit: optical microscopes using visible light cannot achieve resolutions below 200nm.

SMLM is a "time for space" design that broke through the Abbe limit and greatly improves the resolution of the optical microscope up to 1 nm level. it is the essence of The Nobel Prize in Chemistry in 2014.

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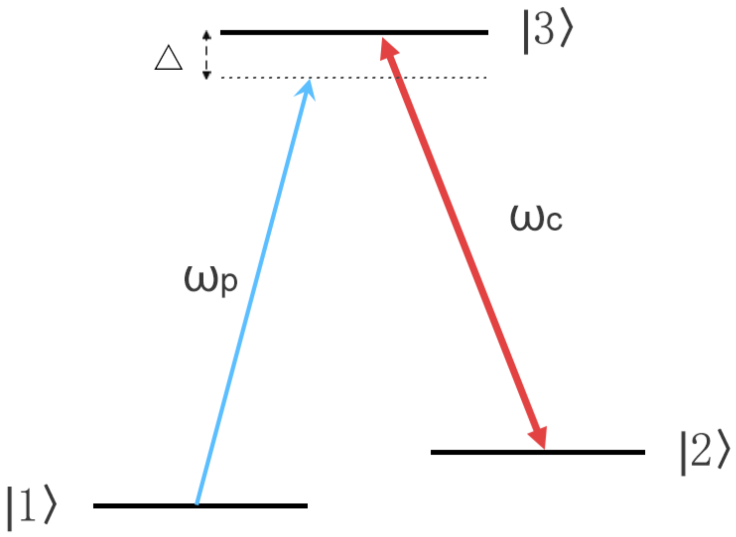
2 EIT (Electromagnetically Induced Transparency)



Energy levels in a Λ-type three-level atomic system, which forms EIT effects.

- Speed of Light in vacuum is 3×10^8m/s.
- EIT can slow down the light, a speed of 17m/s was reported[7].
- EIT is introduced to split the light in time and space.
- The approach is not "time for space" design, because light travel fast, there is no compromise on productivity.

References: [7] Lene Vestergaard Hau, S. E. Harris, Zachary Dutton & Cyrus H. Behroozi, "Light speed reduction to 17 metres per second in an ultracold atomic gas," Nature volume 397, pages594–598 (1999).

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1 **SMLM** + 2 **EIT**

Combination of these 2 elements lead to a new approach of Lithography.

Although there are great differences, the core essence of lithography is also optical imaging technology, so in principle, microscopy and lithography is same in many aspects.

Theoretical Design Overview

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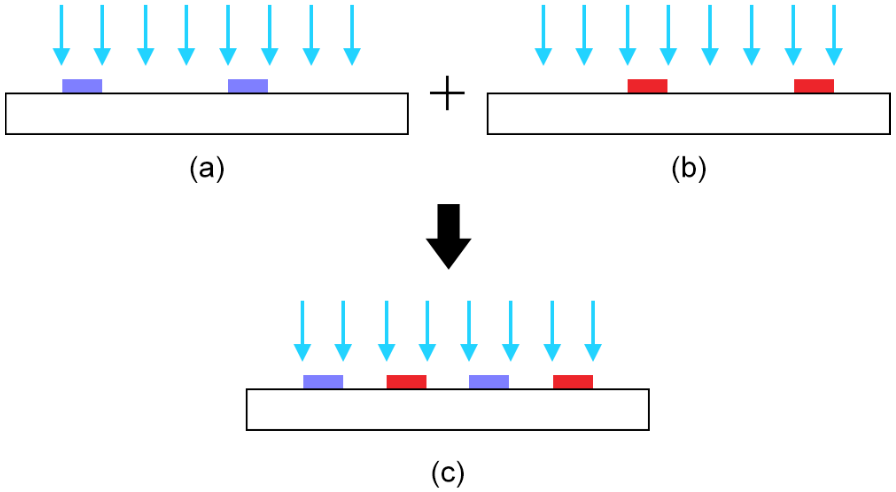
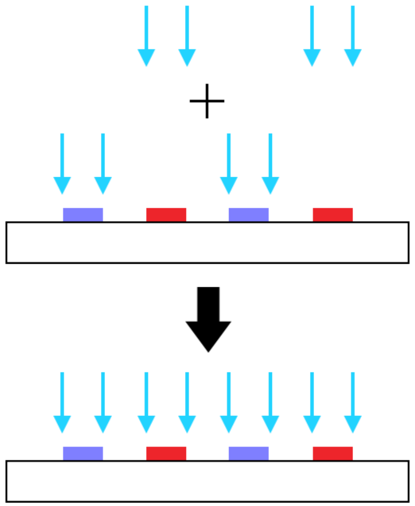
Understand from another perspective:
this approach is a new kind of "Double Patterning"

Split the Mask



Split the Light

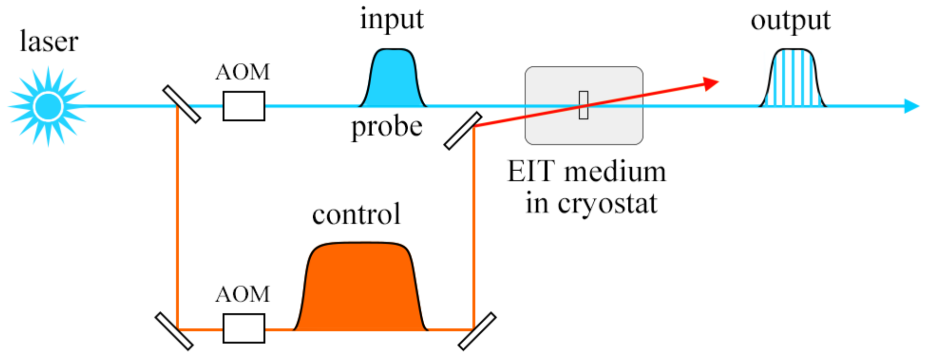


Double Patterning split the mask, high-density mask split into lower-density mask.

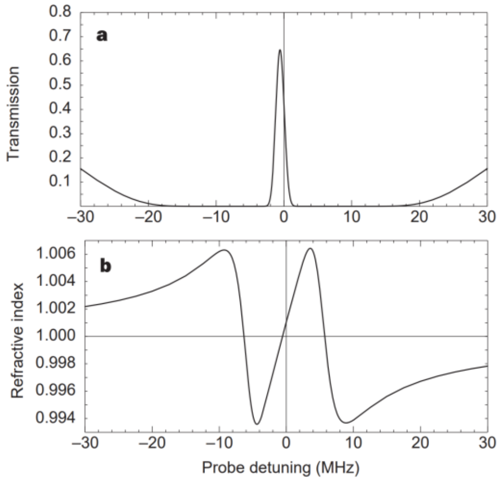
This approach split light beam into separate photons, the advantage is only 1 mask is needed.

Theoretical Design Overview

Schematic diagram of one implementation example



Schematic diagram of the approach



Effect of probe light detuning. a, Transmission profile. b, Refractive index profile. Referenced from [7]

- AOM(Acousto-Optic Modulator) is used to shift the light frequency to required.
- If probe light pass through alone, EIT medium is opaque.
- Control light is turned on first, to ensure EIT is formed.
- Quantum coherent interference happen, a sharp transparent windows for probe light is created.
- Steep dispersion occur, the group velocity of probe light is slowed down.
- Control light is adiabatically shut down, the probe light will be "stopped" and stored in the medium as atomic spin wave, a dark-state polaritons.
- Later Control light is turned on, atomic spin wave will be recovered with original frequency and propagation direction.

References: [7] Lene Vestergaard Hau, S. E. Harris, Zachary Dutton & Cyrus H. Behroozi, "Light speed reduction to 17 metres per second in an ultracold atomic gas," Nature volume 397, pages594–598 (1999).

Theoretical Design Overview

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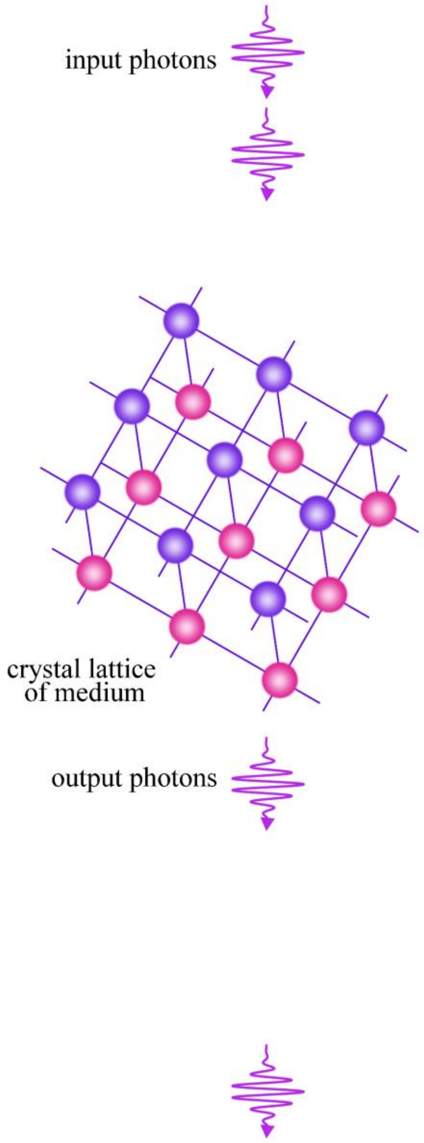
Prediction of the potential of new approach

	Double Patterning	Potential of new approach
Added cost	Related cost of splitting Patterns Added masks fabrication cost Added etch process Repositioning loss	1 exposure, No added cost
Limit	Number of splitting normally < 5	Number of Light splitting can $> 10^8$
Resolution	decided by light wavelength	can reach atom level, $\leq$1nm

Theoretical Design Overview



The approach is not "time for space" design

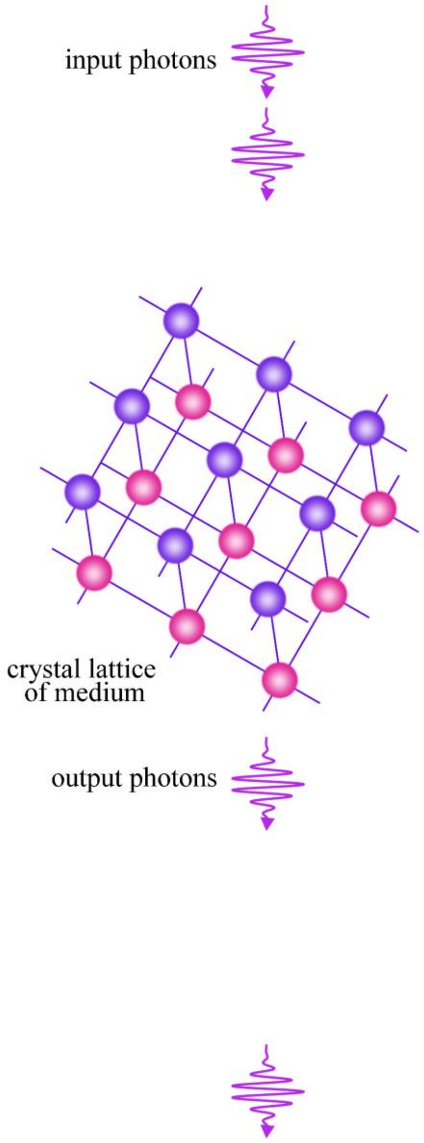
Estimate the order of magnitude of space traveled by light:

Speed of Light in vacuum is $3 \times 10^8 \mathrm{m/s}$, **Maximum Space traveled per second**.

Take Number of Light splitting	$\mathrm{Num} = 10^8$
Take the No-interfering distance	$\mathrm{D} = 10^{-6} m = 1\, micrometre$
Space traveled by light	$\mathrm{S} = \mathrm{Num} \times \mathrm{D} = 10^2 m$

Estimation suggests there is big margin from Maximum limit, because light travel fast, there is no compromise on productivity.

Theoretical Design Overview



The essence of the new approach

EIT medium rearrange the photons of probe light in time and space.

When propagating in medium, group velocity of photons is slow inside atom range.

So photons will concentrate in the atomic range, even without storage process, photons will be rearranged. After storage, photons are orderly as atomic lattice.

Theoretical Design Details Analysis

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Choose EIT medium

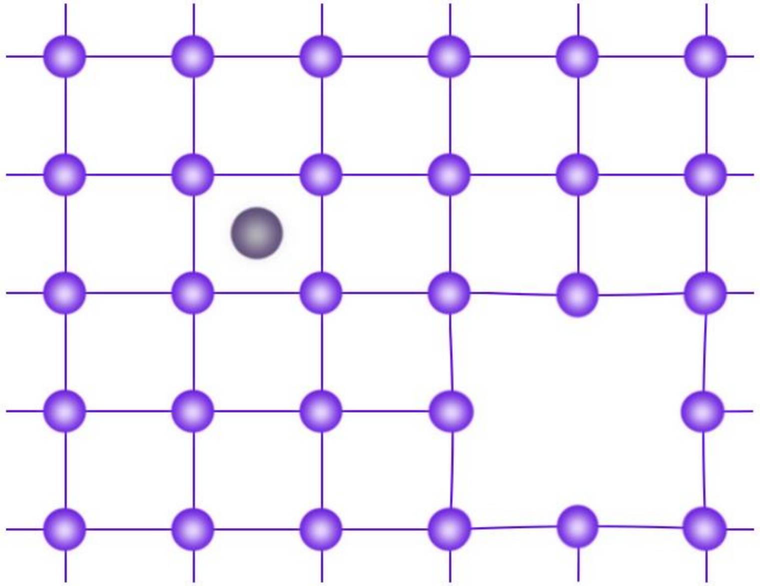
	Gas	Solid	
		optical crystals doped with rare earth elements	pure rare earth compound crystal
Common examples	Cs(Cesium) Rb(Rubidium)	Pr^{3+}:Y_2SiO_5 (Praseodymium doped yttrium silicate crystal)	EuCl3·6H2O (Europium chloride hexahydrate)
Shortcomings	low atom density, apt to atomic diffusion motion	lattice defect, large inhomogeneous broadening	

Pure rare earth compound crystal is recommended, Perfect crystal can be accurate to atom level.

Ce (Cerium) is a potential choice of rare earth element

The transition frequency of medium is better to be close to the frequency of available photo resist(436nm, 365nm, 248nm and 193nm), the smaller the better. EuCl3·6H2O has a transition frequency with 579.7033nm of wavelength, so we need to look for other frequency, another rare-earth element Ce (Cerium) is a potential choice, it has absorption peaks located at 205nm, 243nm, and 293nm, all of which are in the ultraviolet band.

Crystal defects & Impurities

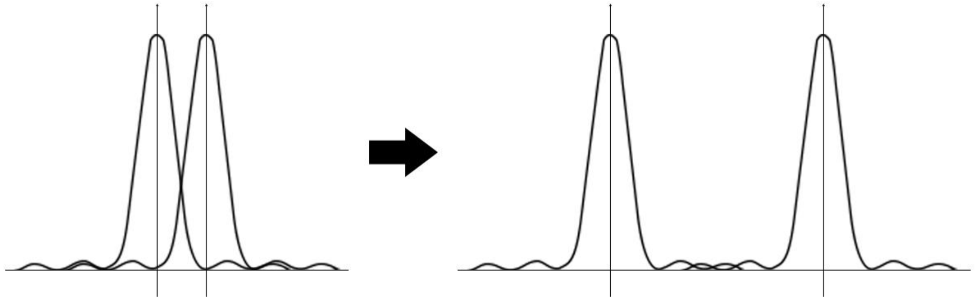


Impurities or defects have no big impact on the No-interfering distance.

Processing accuracy depend on the crystal lattice structure. There is big fault tolerance, crystal lattice is required to provide enough No-interfering distance.

Impurities can have some impact on exposure uniformity and are prone to scattering noise, but have no big impact on the reliability of lithography.

The group velocity (v_g) limitation



If the distance between each photon is large enough, there is no interference.

In EIT medium, Probe light is compressed by a Ratio = c/v_g (c is speed of light in vacuum).

Estimate the order of magnitude:

Crystal lattice constant of the medium: $d \approx 1$ nm
No-interfering distance: $D \approx 193-1000nm$

if possible, a bigger D is better.

There is a maximum v_g limit to ensure: $\text{Ratio} > \frac{D}{d}$

Theoretical Design Details Analysis

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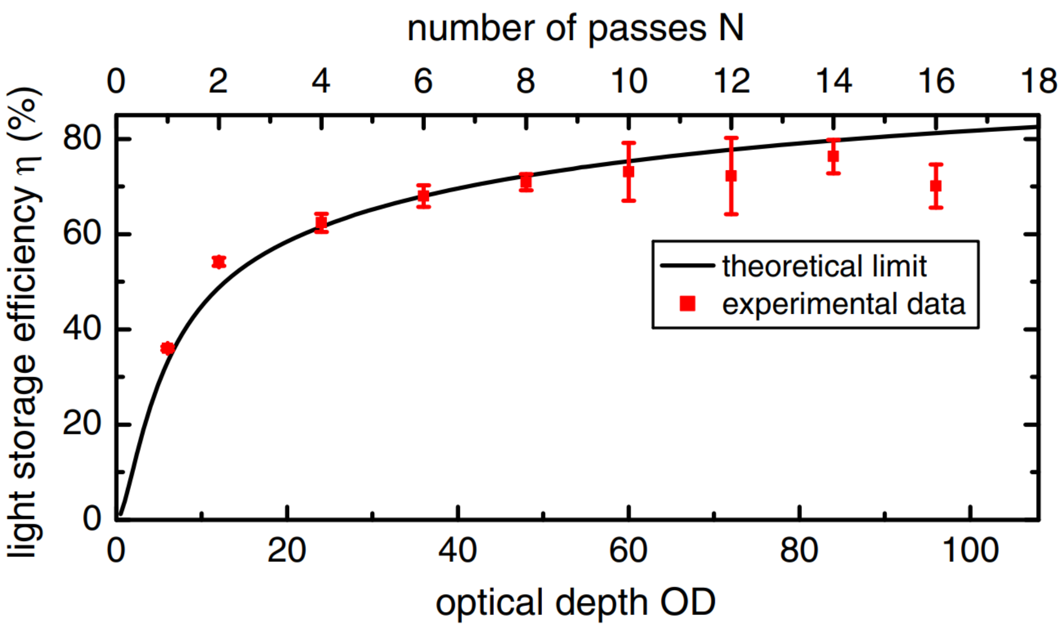
The efficiency of quantum storage



Maximal light storage efficiency η, experimental data(red solid squares) and theory (black solid line), versus number of passes N (i.e., optical depth OD). Reference from [13].

Storage efficiency is the ratio of output photons to input photons.

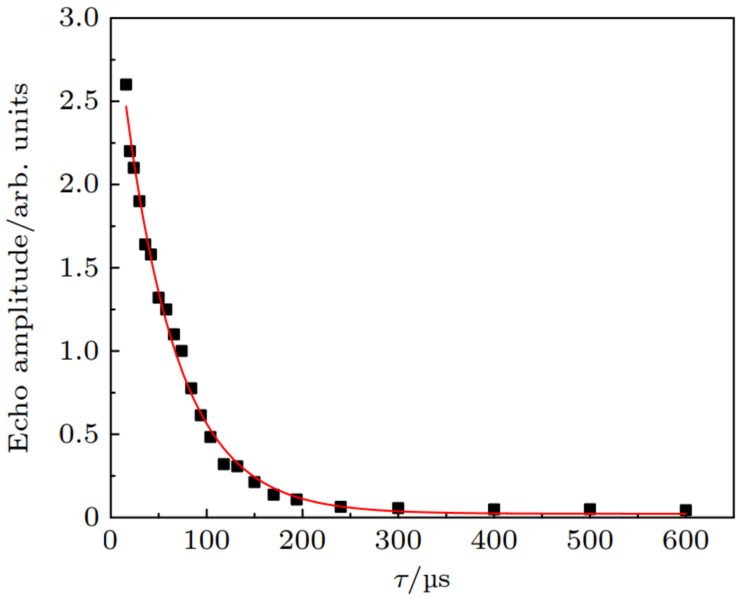
Quantum storage is one mechanism: rearrange the photons, efficiency of 100% is ideal.

Estimate the order of magnitude of quantum storage efficiency:

It was reported that a light storage efficiency of (76.3±3.5)% can be achieved[13]. It suggests that efficiency can be improved.

References: [13] Daniel Schraft, Marcel Hain, Nikolaus Lorenz, and Thomas Halfmann, "Stopped Light at High Storage Efficiency in a Pr3+:Y2SiO5 Crystal," Phys. Rev. Lett. 116, 073602 – 17 February 2016.

The storage time



The decay curve of two-pulse photon echo amplitude with time in EuCl3·6H2O. Referenced from [5].

The storage time need to be long enough, so that the rearrangement is completed.

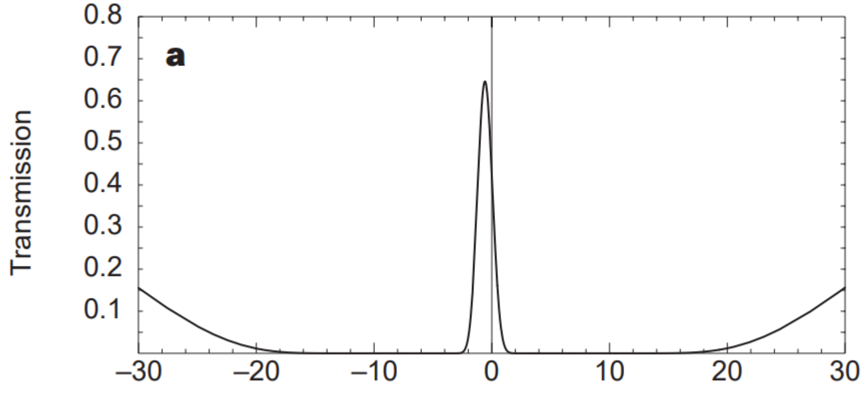
Since in this implementation example, photon is temporarily saved, we don't need storage time of very long.

It was reported the transition coherence time of pure rare earth compound crystal EuCl3·6H2O (Europium chloride hexahydrate) is 55.7 μs[5], since the order of magnitude level of transfer time at atomic-scale for photon is below femtosecond scale, this order of magnitude level is enough for temporary storage purpose.

References: [5] Li Zong-Feng, Liu Duan-Cheng, Zhou Zong-Quan, Li Chuan-Feng, "Atomic frequency comb optical memory in EuCl3·6H2O crystal," Acta Phys. Sin., 2021, 70(16): 160302.

Theoretical Design Details Analysis

The issue of noise



Effect of probe light detuning. a, Transmission profile, when EIT is formed, a sharp transparent windows for probe light is created. Referenced from [7]

Low-noise is quite important for lithography.

Control light is stronger in intensity and turned on first, thus to ensure EIT is formed, the particles are imprisoned on ground state, become dark-state, so essentially it is a dissipation-free mechanism, it is low-noise.

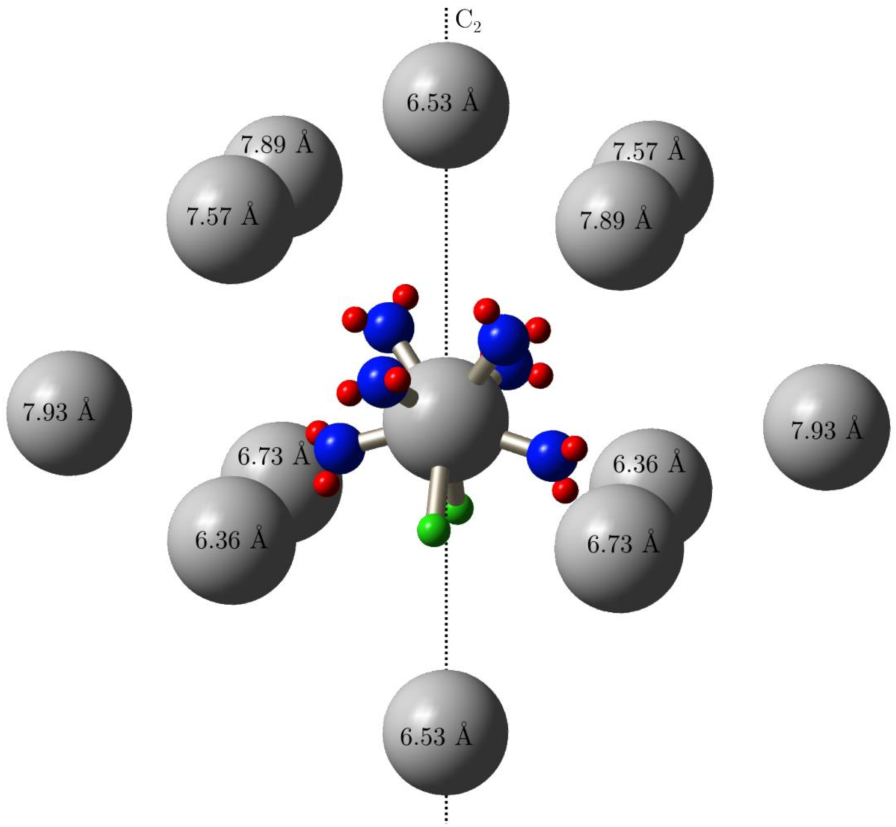
In principle, scatter noise is the main noise sources, it is mainly due to impurity and lattice defect of crystal growth, this can be improved.

SHB(Spectral hole burning) is suitable for noise filtering using EIT medium as **atomic pool**, after SHB preparation, saturated absorption happen, the medium will become transparent, while opaque to other light. It has a similar effect as EIT.

Noise is an issue that can be improved.

References: [7] Lene Vestergaard Hau, S. E. Harris, Zachary Dutton & Cyrus H. Behroozi, "Light speed reduction to 17 metres per second in an ultracold atomic gas," Nature volume 397, pages594–598 (1999).

The inhomogeneous broadening



The environment of an Eu^{3+} ion in $EuCl_3.6H_2O$, Referenced from [3]

- **Rare earth doped crystal** like $Pr^{3+}:Y_2SiO_5$ (Praseodymium doped yttrium silicate crystal) has large inhomogeneous broadening of several GHZ, this means that only a small part of the atoms can participate in the EIT process, so the efficiency of quantum storage is low.

- **Pure rare earth compound crystal** $EuCl_3{\cdot}6H_2O$ (Europium chloride hexahydrate) is better, with its inhomogeneous broadening around 100 MHz, but still need improvement.

- Pure rare compound crystal $EuCl_3{\cdot}6H_2O$ was reported that by isotopically purifying the ^{37}Cl in ^{35}Cl, a narrow optical inhomogeneous linewidth of 25 MHz is obtained, further improvement is also possible[3].

- A narrow optical inhomogeneous linewidth means all atoms can participate in the EIT process, so the efficiency of quantum storage is high.

References: [3] R. L. Ahlefeldt, M. R. Hush, and M. J. Sellars, "Ultranarrow Optical Inhomogeneous Linewidth in a Stoichiometric Rare-Earth Crystal," Phys. Rev. Lett. 117, 250504 –15 December 2016.

Conclusions

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SMLM has verified that optical imaging technology can reach nm resolution.

EIT has verified that slow light and light storage is a robust technique.

Though further investigation is required, from available verification and theoretical inference, this approach show the **potential prospects** in lithography:

Light storage provide us a tool to manipulate photons at atomic level, the diameter of an atom is with an order of magnitude of 0.1 nanometer, we estimated it could lead to outstanding **resolution level**.

The dark-state polaritons of EIT is dissipation-free, which means **low-noise**, critical for its potential volume application in lithography.

Acknowledgment

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Thank you！ 谢谢