Coherent Control of Nonlinear Optics

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Texas A&M University, College Station, TX

Abstract

Using quantum coherent effects provides means to control nonlinear optical processes in various media. We predict several new effects: for example, forward Brillouin scattering and enhancement and control of coherent generation in the backward direction by applying only forward propagating fields. The applications range from development of hyper-dispersive materials, improvement of spatial resolution beyond diffraction limit to generation of squeezed and entangled light.

Lebedev Institute, Moscow, Russia; May 19, 2009

Coherent Control of Nonlinear Optics

Outline(*)

Introduction

- Nonlinear Optics
- Phase-matching condition
- Quantum coherence effects
- Controlling phase-matching via coherence effects
- Experimental implementations

Conclusion

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

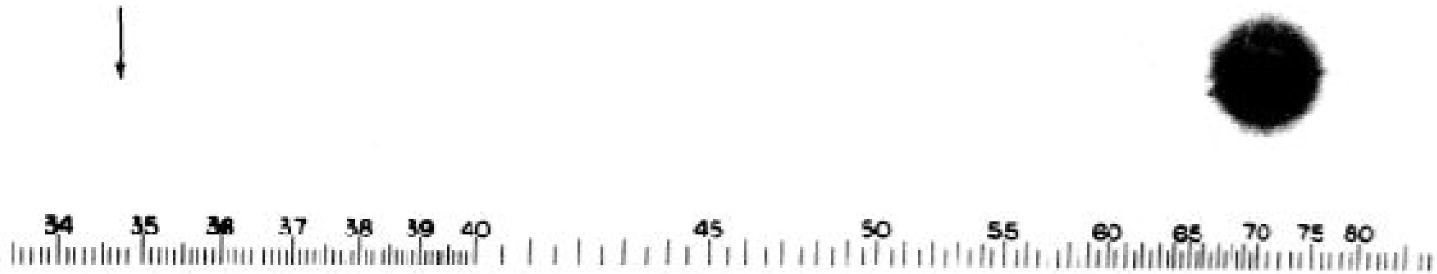
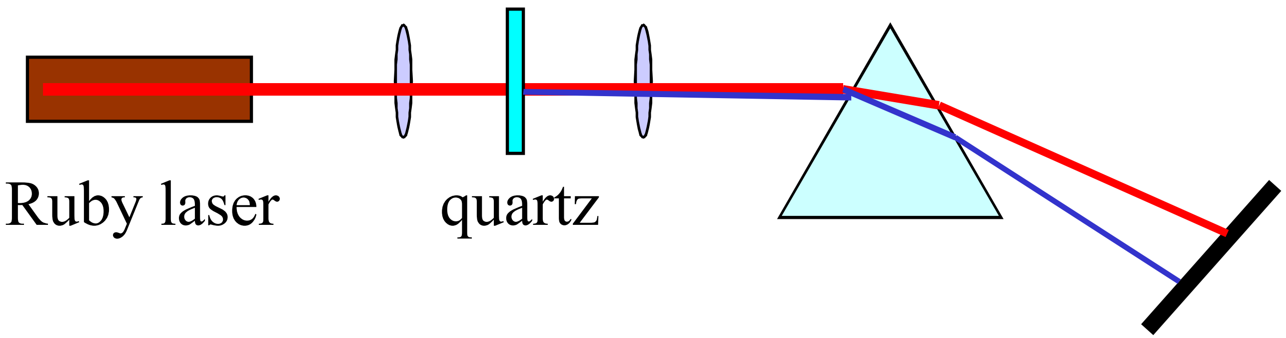


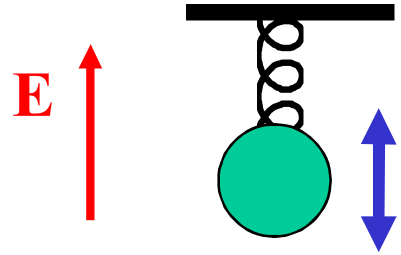
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

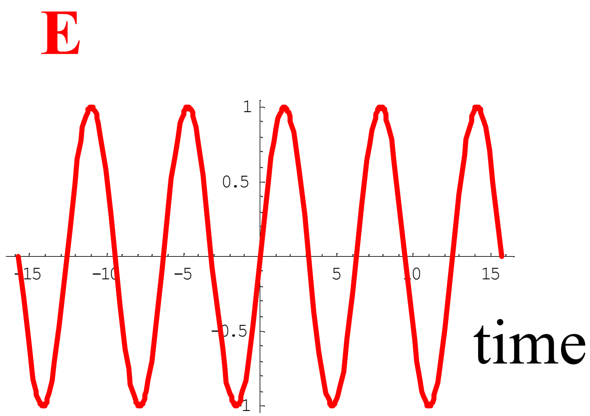


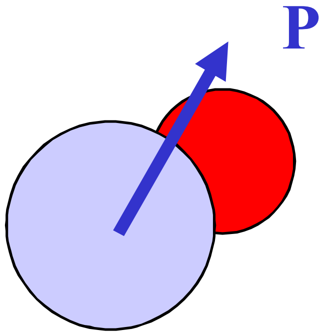
Nonlinear Optics started from here

Linear Optics

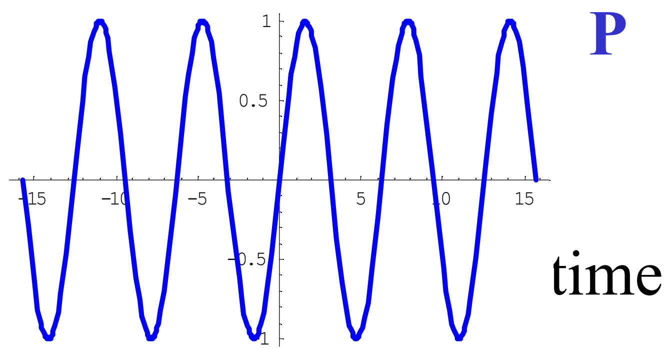
$$P = \chi E$$

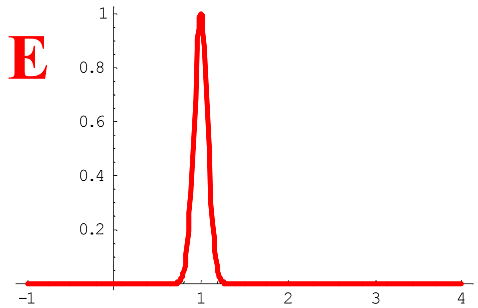




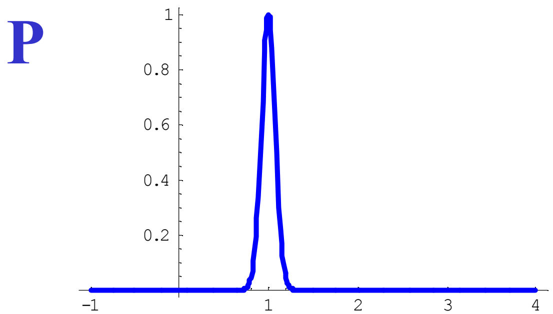


atom or molecule





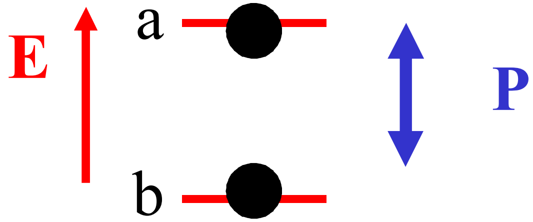
Frequency

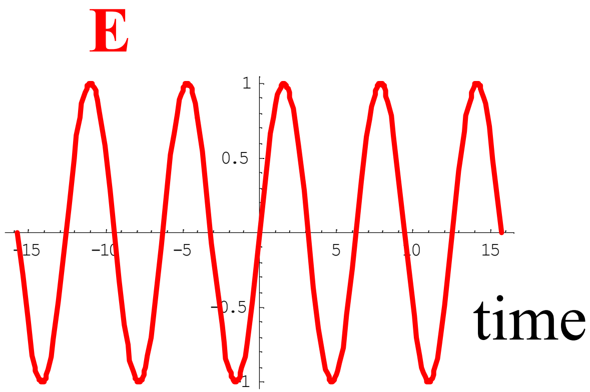


Frequency

Second and third harmonic generation

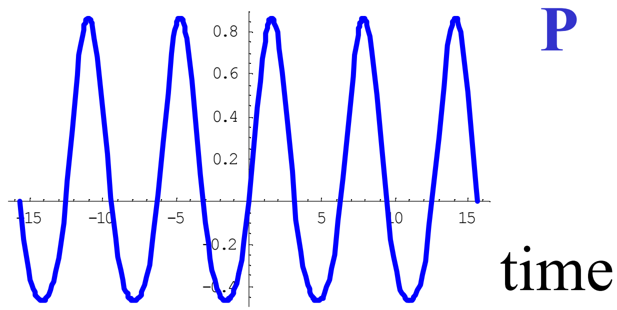
$$P = \chi E + \chi^{(2)} E E + \chi^{(3)} E E E + \dots$$

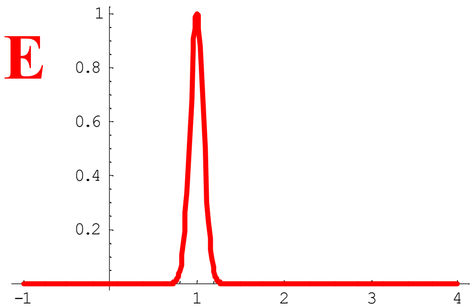




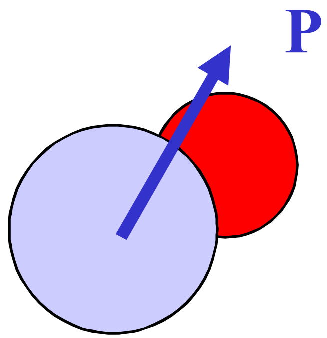
$$a^2 + b^2 = 1$$

$$P \sim a b^* < 1/2$$

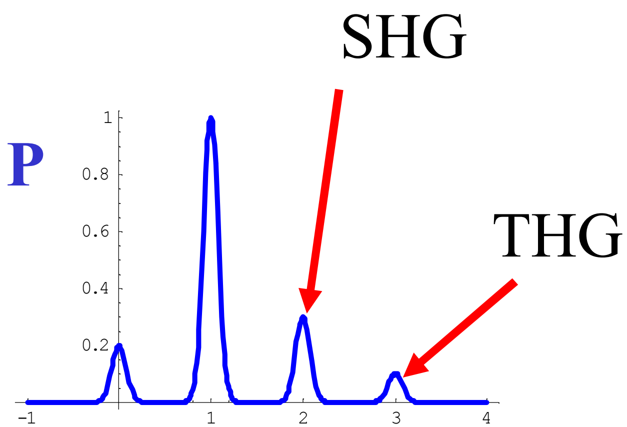




Frequency



atom or molecule



Frequency

Phase-matching

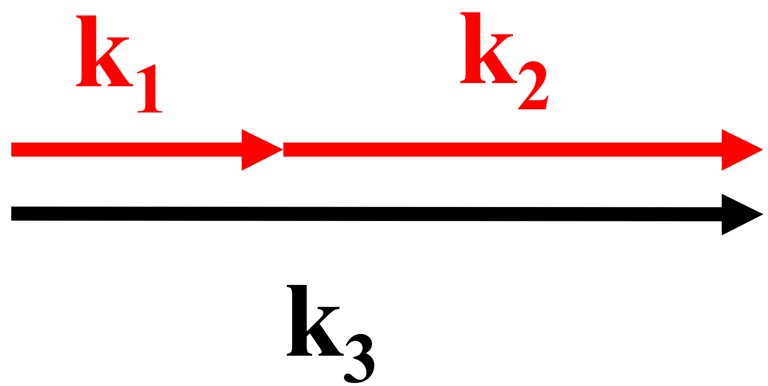
VOLUME 8, NUMBER 1 PHYSICAL REVIEW LETTERS JANUARY 1, 1962

MIXING OF LIGHT BEAMS IN CRYSTALS

J. A. Giordmaine
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received November 29, 1961)

$$\omega_3 = \omega_2 + \omega_1$$

$$k_3 = k_1 + k_2$$



Phase-matching

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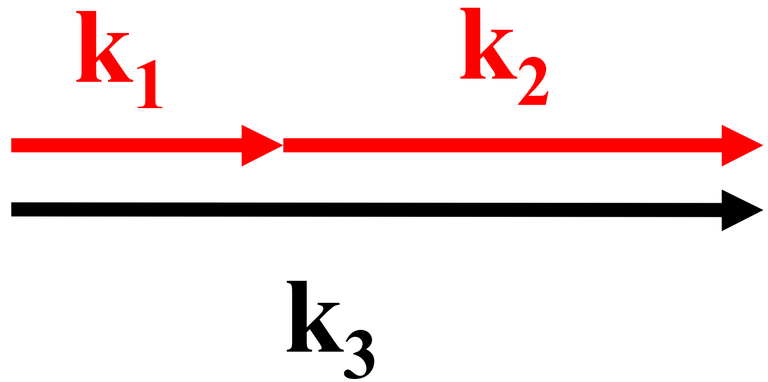
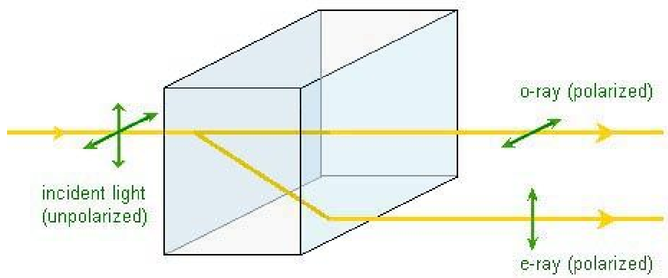
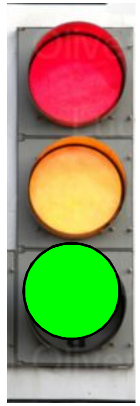




Phase-matching can be done by using crystal birefringence

Control of Coherent Generation



Motivation: controllable phase-matching is desirable to efficiently enhance nonlinear interaction, examples are given for 3- and 4-wave mixing;

Applications: Generation of new frequency bands (X-rays, THz); Generation of quantum states of light (entanglement photons, squeezed light); improvement high resolution spectroscopy; improvement of spatial resolution.

Conclusion: ***YES***

Control of Phase matching for Nonlinear Wave Mixing

How? What is the way to implement such an idea?

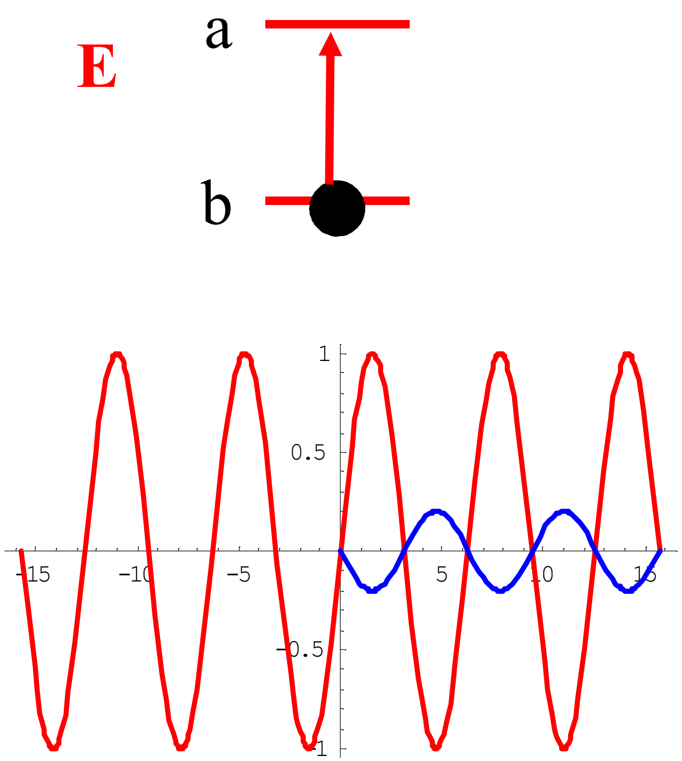
Control of Phase matching for Nonlinear Wave Mixing

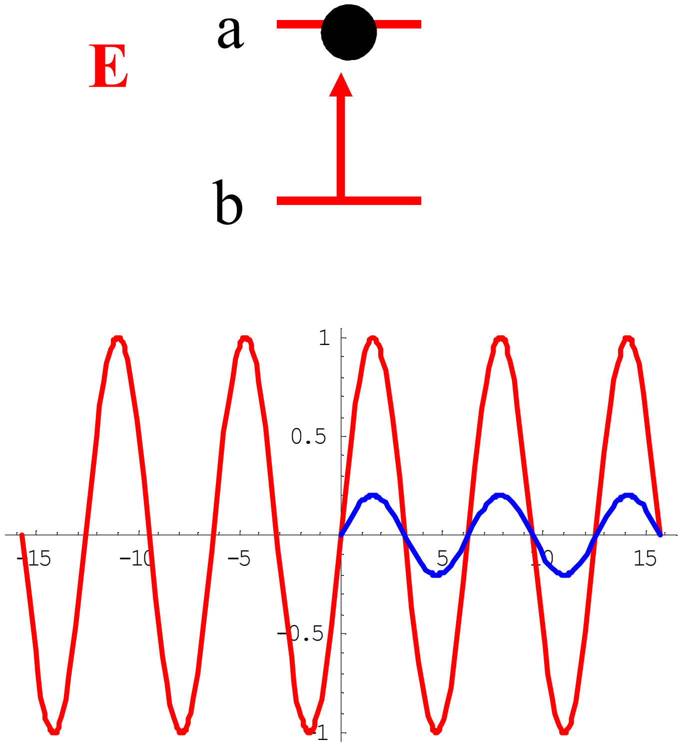
How? What is the way to implement such an idea?

Approach:

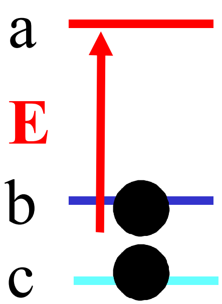
By using Coherence effects

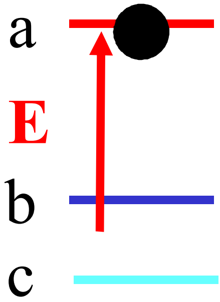
Two-level atom

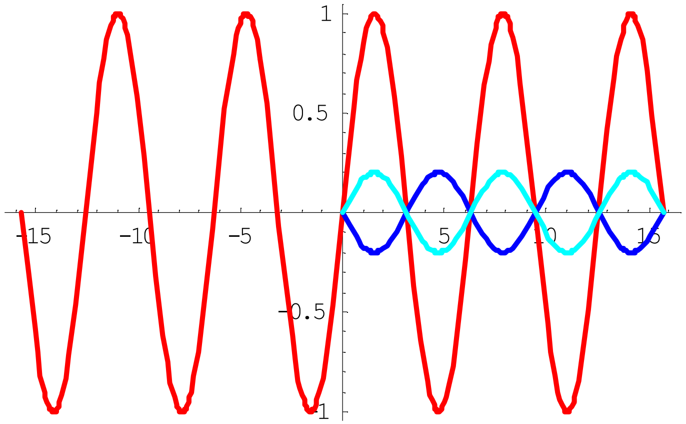


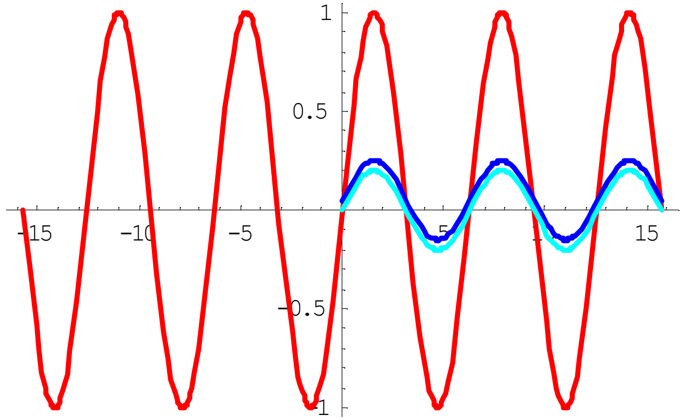


EIT in atomic media
Three-level atom







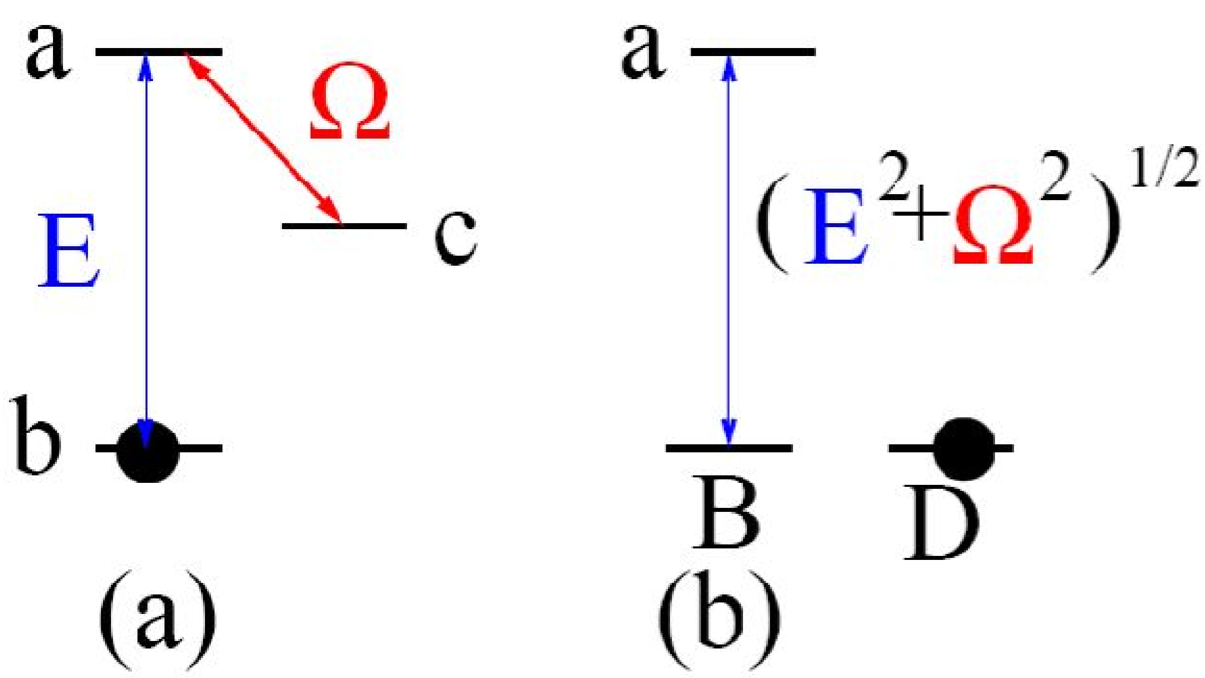


EIT in atomic media: Theoretically predicted: O. A. Kocharovskaya and Ya. I. Khanin, Zh. Eksp. Teor. Fiz., Pis'ma Red 48, 581 (1988). [JEPT Lett. 48, 630 (1988)]; Experimentally observed: K. J. Boller, A. Imamoglu and S. E. Harris, Phys. Rev. Lett. 66, 1360 (1992); V. Sautenkov, et al. PRA 71, 063804 (2005);

Reviews E. Arimondo, in Progrees in Optics, E. Wolf, ed. (Elsevier, Amsterdam, 1996), XXXV, 257-354; Stephen E. Harris, Physics Today, July 1997, 36-42;

M. Fleishhauer, A. Imamoglu, J. Marangos, Rev. Mod. Phys. 77, 633 (2005).

Dark and Bright States



Dark state $$|D\rangle = \frac{\Omega|b\rangle - E|c\rangle}{\sqrt{|\Omega|^2 + |E|^2}}$$

$\langle a|\, H\, |D\rangle = 0$

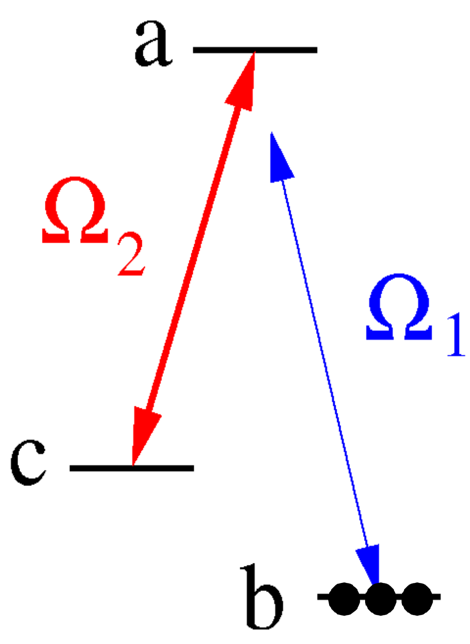
Bright state $$|B\rangle = \frac{\Omega|c\rangle + E|b\rangle}{\sqrt{|\Omega|^2 + |E|^2}}$$

Important remark

Hebin Li, et al., Optical imaging beyond the diffraction limit via dark states, PRA (2008).

Susceptibility of EIT medium

$$\Omega_1 = \frac{\wp_{ab}\mathcal{E}}{\hbar} \qquad \Omega_2 = \frac{\wp_{ca}\mathcal{E}}{\hbar}$$



Hamiltonian

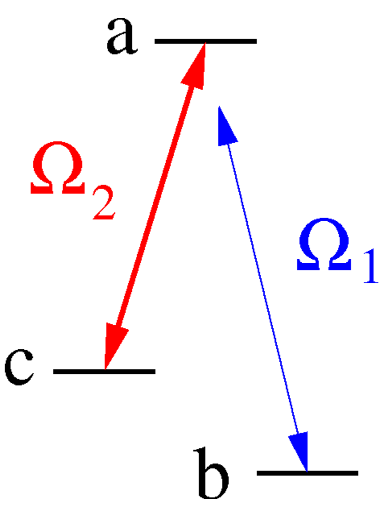
$$V_I = -\hbar[\Omega_1 e^{-i\omega_{ab}t}|a\rangle\langle b| + \Omega_2 e^{-i\omega_{ac}t}|a\rangle\langle c| + h.c.]$$

Density matrix equations:

$$\begin{aligned}
\dot{\rho}_{ab} &= -\Gamma_{ab}\rho_{ab} + i n_{ab}\Omega_1 - i\rho_{cb}\Omega_2 \\
\dot{\rho}_{ca} &= -\Gamma_{ca}\rho_{ca} + i n_{ca}\Omega_2^* + i\rho_{cb}\Omega_1^* \\
\dot{\rho}_{cb} &= -\Gamma_{cb}\rho_{cb} + i\rho_{ca}\Omega_1 - i\rho_{ab}\Omega_2^*
\end{aligned}$$

$$\rho_{ab} = \frac{-i\Omega_1}{\Gamma_{ab} + \frac{|\Omega_2|^2}{\Gamma_{cb}}}$$

$$\Gamma_{ab} = \gamma_{ab} + i(\nu_1 - \omega_{ab}) \qquad \Gamma_{ca} = \gamma_{ca} - i(\nu_2 - \omega_{ac}) \qquad \Gamma_{cb} = \gamma_{cb} + i(\nu_1 - \nu_2 - \omega_{cb})$$



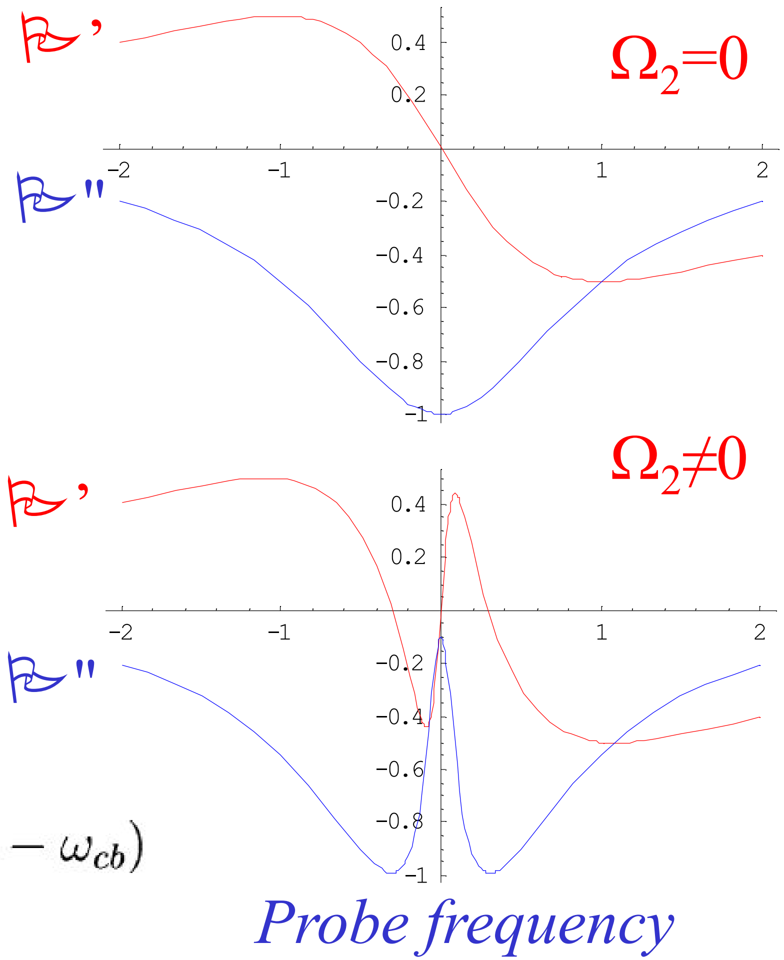
EIT medium

$$\Omega_1 = \frac{\wp_{ab}\mathcal{E}}{\hbar}$$

$$\Omega_2 = \frac{\wp_{ca}\mathcal{E}}{\hbar}$$

$$\chi_{ab} = -\frac{\eta\Gamma_{cb}}{\Gamma_{ab}\Gamma_{cb} + |\Omega_2|^2}$$

$$\Gamma_{ab} = \gamma_{ab} + i(\nu_1 - \omega_{ab}) \qquad \Gamma_{cb} = \gamma_{cb} + i(\nu_1 - \nu_2 - \omega_{cb})$$



Wavevector of the detuned field Ω_1:

$$k_1 = \frac{\nu_1}{c}n = \frac{\nu_1}{c} + \frac{3\lambda^2 N\gamma_r(\nu_1 - \omega_{ab})}{8\pi|\Omega_2|^2} = \frac{\nu_1}{c} + \frac{\nu_1 - \omega_{ab}}{V_g}$$

where $V_g = \dfrac{8\pi|\Omega_2|^2}{3\lambda^2 N\gamma_r}$ is the group velocity.

Please, note here:

EIT can be applied to the search for special relativity and CPT violations that were among the topics of A.D. Sakharov's research

See, for example,
Phys Rev D66 056005 (2002), and
Contemporary Physics 47, 25 (2006).

Also EIT can be applied to clocks, magnetometry, plasma diagnostics including tokamak plasmas

See Anisimov PM, Akhmedzhanov RA,
Zelenskii IV, et al.
JETP 96, 801 (2003)

EIT in various media

From Atoms to Solids

CW and pulsed regimes

S.E. Harris, Phys. Rev. Lett. 70, 552 (1993); ibid. 72, 52 (1994).

V. Sautenkov, Y. Rostovtsev, et al. Phys. Rev. A 71, 063804 (2005);

Atomic and molecular gases, room temperature

A.S. Zibrov, et al., Phys. Rev. Lett. 76, 3925 (1996);

S. Harris, A. Sokolov, Phys. Rev. Lett. 81, 2894(1998)

and BEC

J. Kitching and L.Hollberg, Phys. Rev. A 59, 4685, (1999)

in solids doped by rare-earth ions

B. S. Ham, et al., Opt. Commun. 144, 227 (1997);

B. S. Ham, et al., , Opt. Lett. 22, 1138 (1997)

in semiconductor quantum wells

A. Imamoglu, Opt. Commun. 179, 179 (2000);

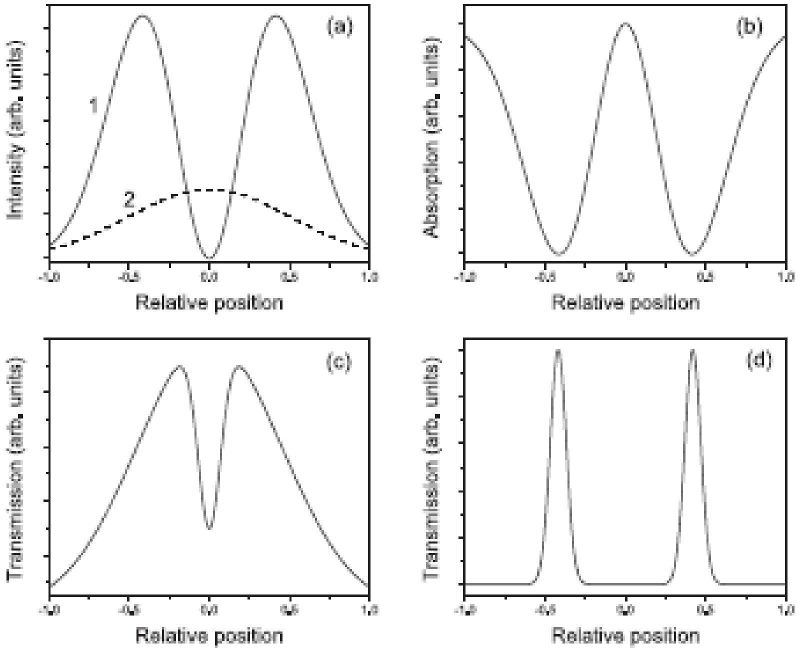
D.E. Nikonov, A. Imamoglu, M.O. Scully, Phys. Rev. B59, 12212 (1999)

different wavelengths: from X-ray to microwaves

C.J. Wei, N.B. Manson, Phys. Rev. A60 2540 (1999);

R. Coussement, Y. Rostovtsev, J. Odeurs, et al.
Controlling absorption of gamma radiation via nuclear level anticrossing,
Phys. Rev. Lett. 89, 107601 (2002).

Beating diffraction limit via EIT



$$\chi'' = \frac{\eta \gamma_{cb}}{|\Omega|^2}$$

$$\eta = \frac{3\lambda^2 N}{8\pi} N$$

$$|D\rangle = \frac{\Omega_1 |b\rangle - \Omega_2 |c\rangle}{\sqrt{|\Omega_1|^2 + |\Omega_2|^2}}$$

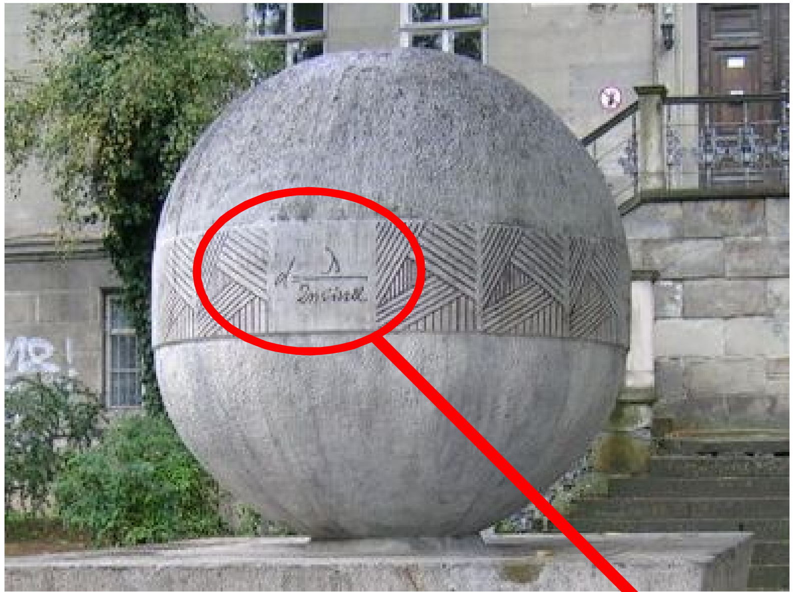
$$\Delta z = L \sqrt{\frac{\gamma_{cb} \Gamma}{|\Omega|^2}}$$

Theory: D.D. Yavuz and N.A. Proite, PRA 76, 041802 (2007), J. Cho, PRL 99, 020502 (2007), A.V. Gorshkov, M.D. Lukin, et. al. PRL 100, 093005 (2008)

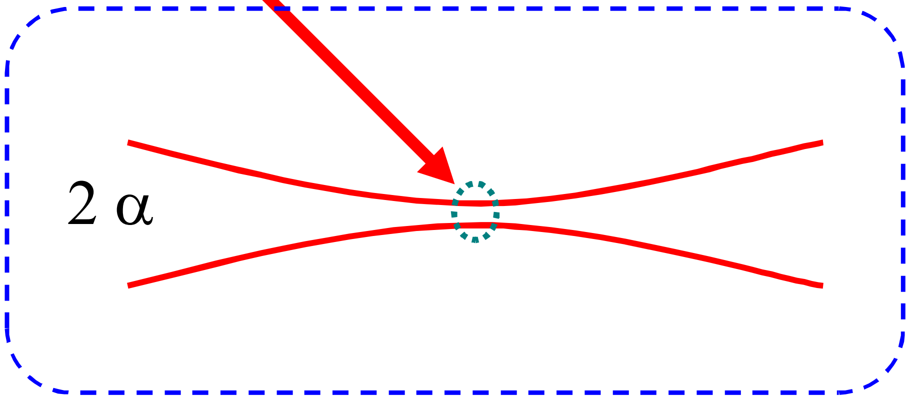
Theory and experiment: H. Li, et. al. PRA (2008).

Beating diffraction limit via EIT

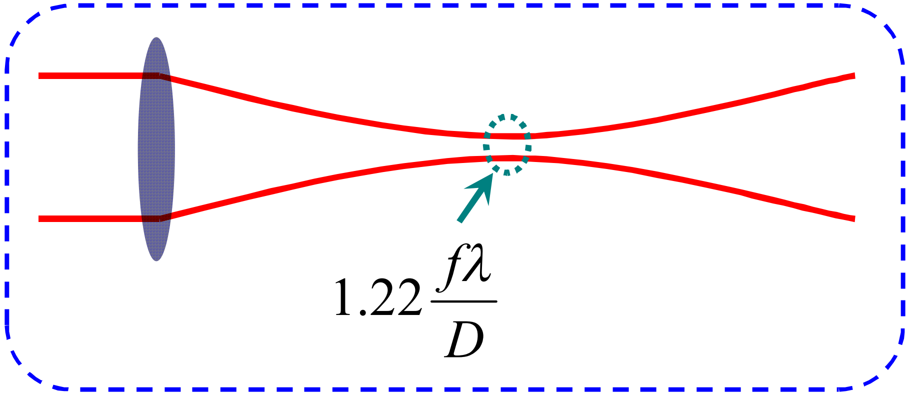
Beating diffraction limit via EIT



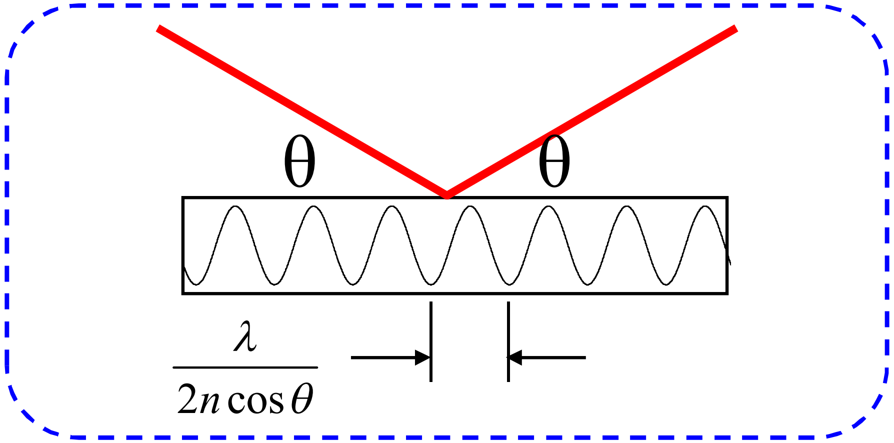
$$d = \frac{\lambda}{2n \sin \alpha}$$



Motivation: go beyond the diffraction limit



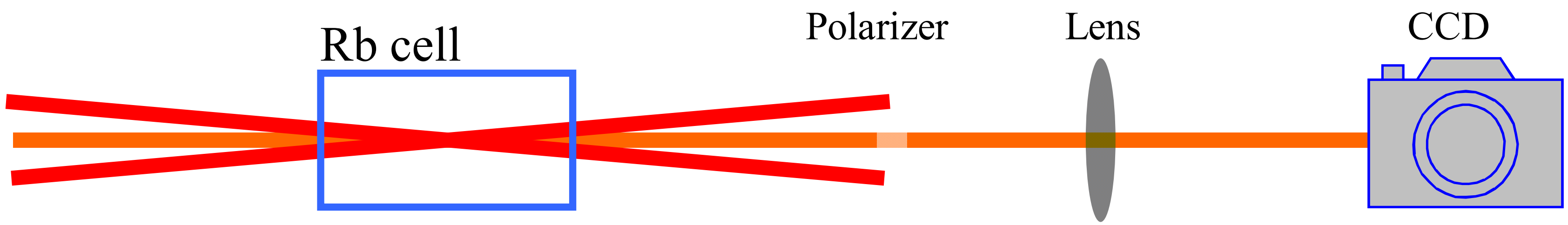
Many applications require a resolution of a small fraction of the wavelength.

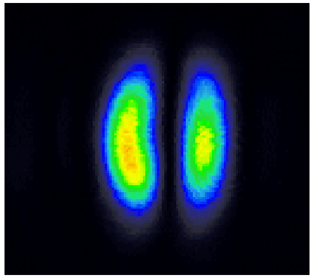


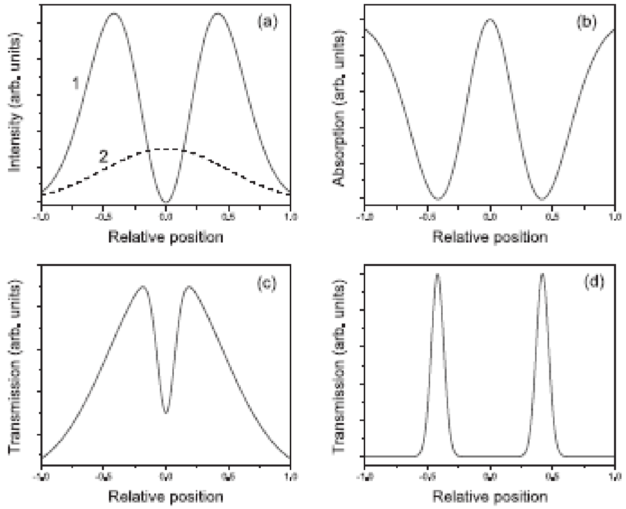
- Localization of atoms
- High resolution microscopy
- Optical lithography

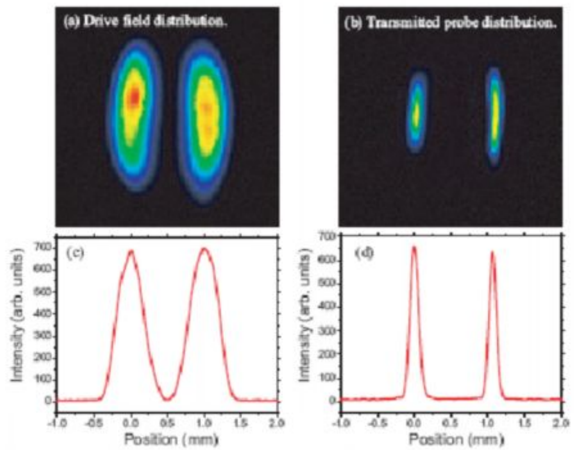
Classically, the best resolution is ~λ/2.

A proof-of-principle experiment







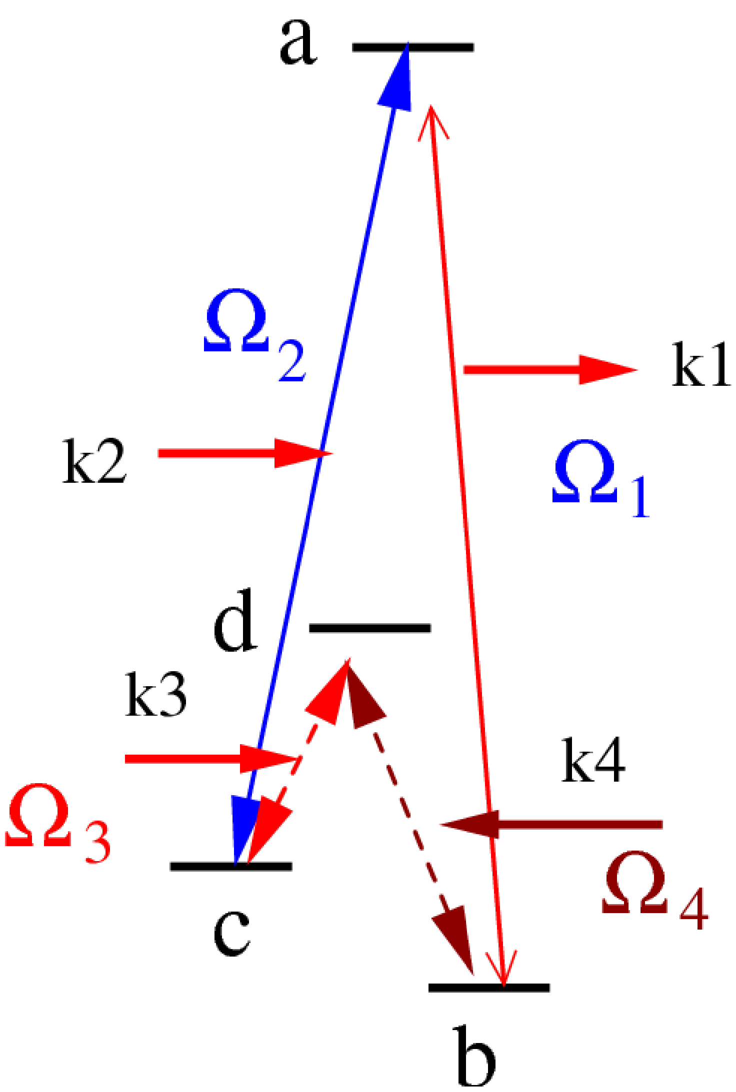


Four-wave mixing

Motivation:

Applications to Quantum Computing
Quantum sensing,
Generation of entangled light,
Generation of NOON states, etc.

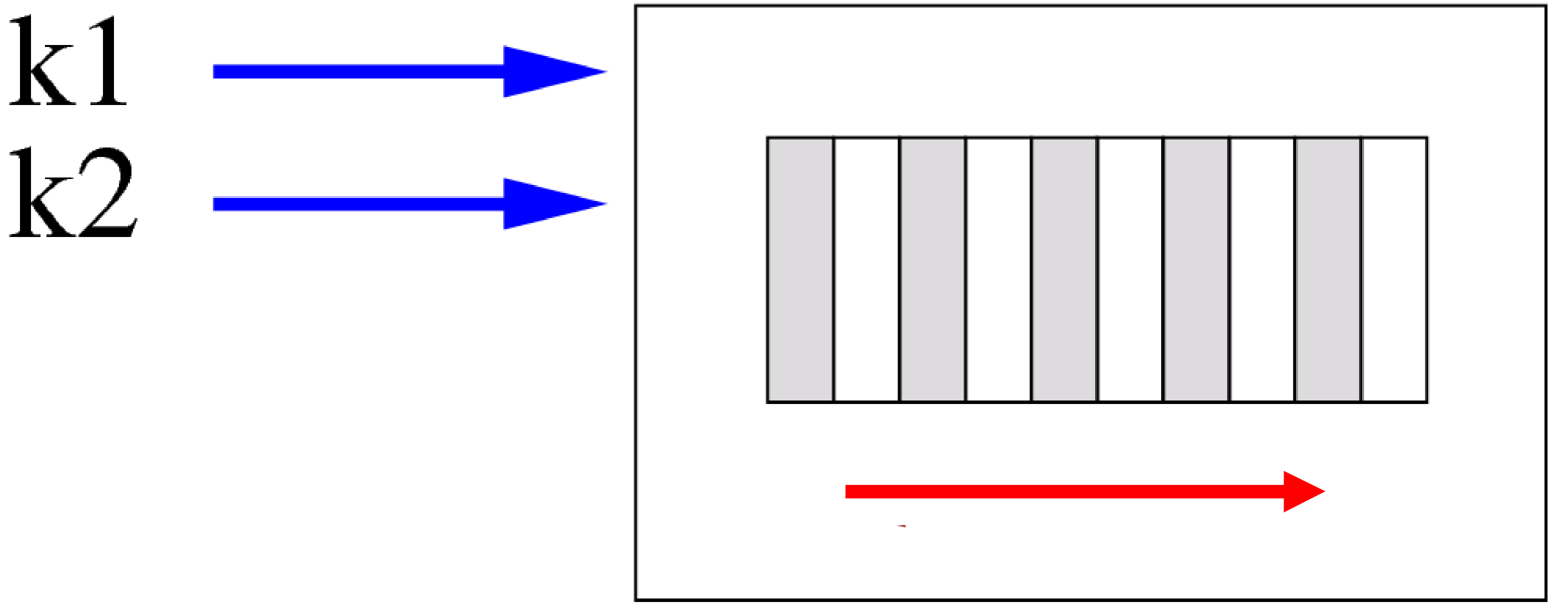
Four-wave mixing



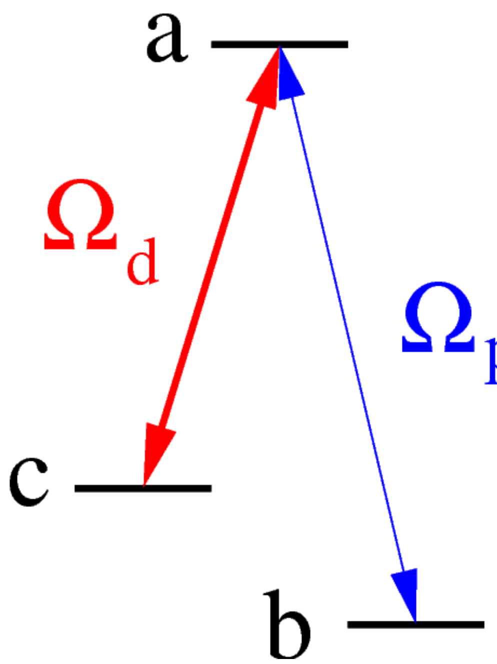
$$\mathbf{k_4 = k_1 - k_2 + k_3}$$

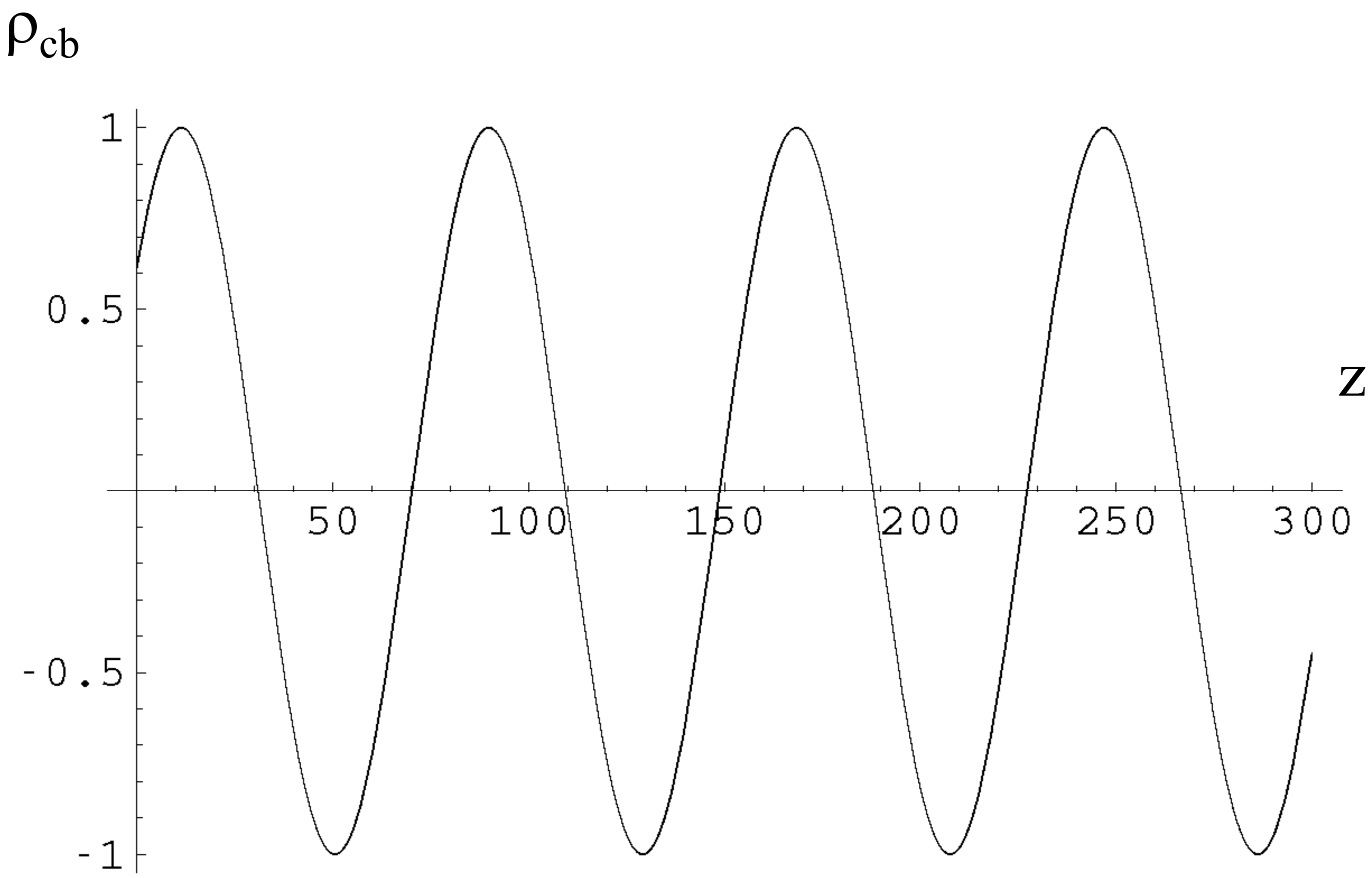
$$\omega_4 = \omega_1 - \omega_2 + \omega_3$$

Using two forward propagating beams, we prepare coherent grading in the medium



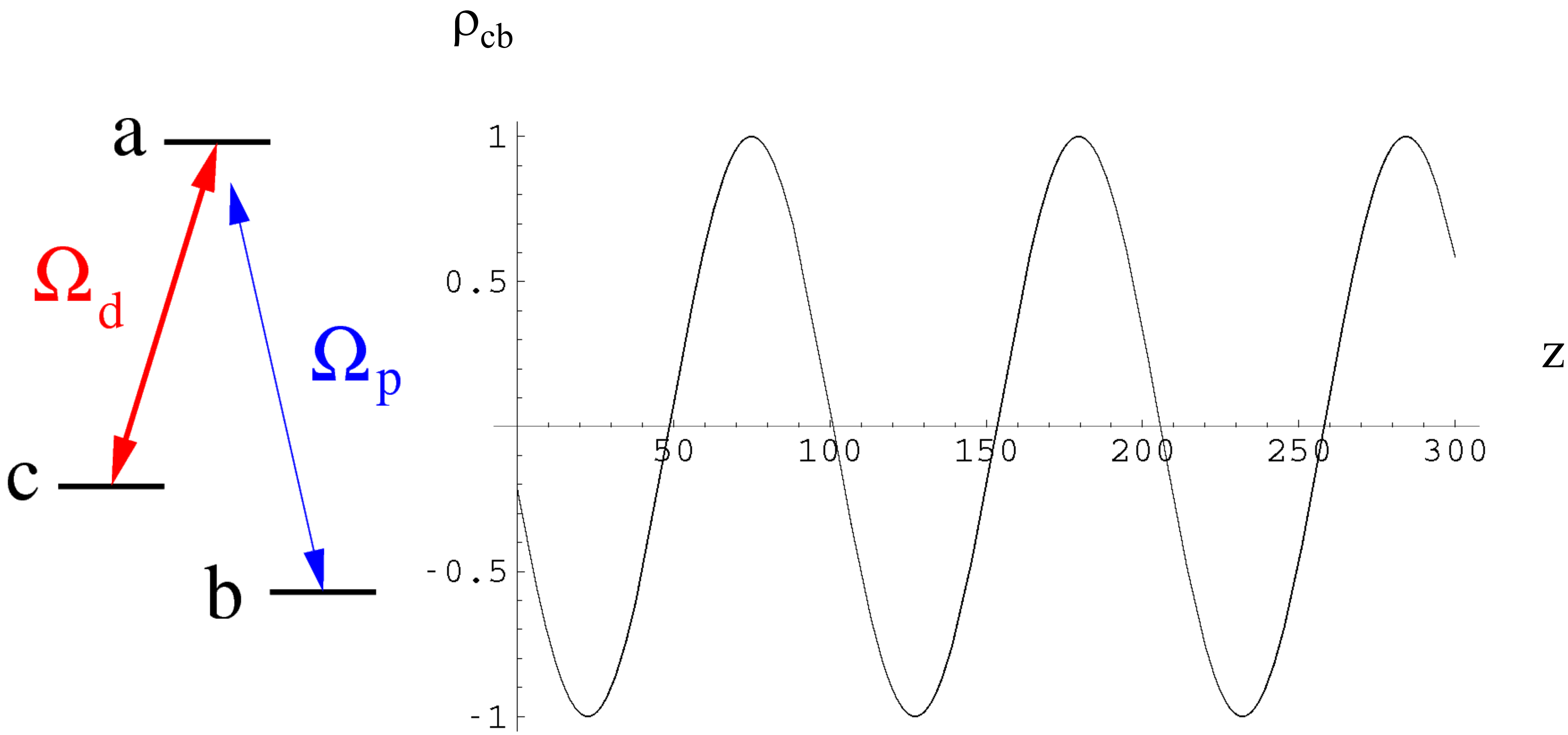
Coherence at b-c transition at the two-photon resonance





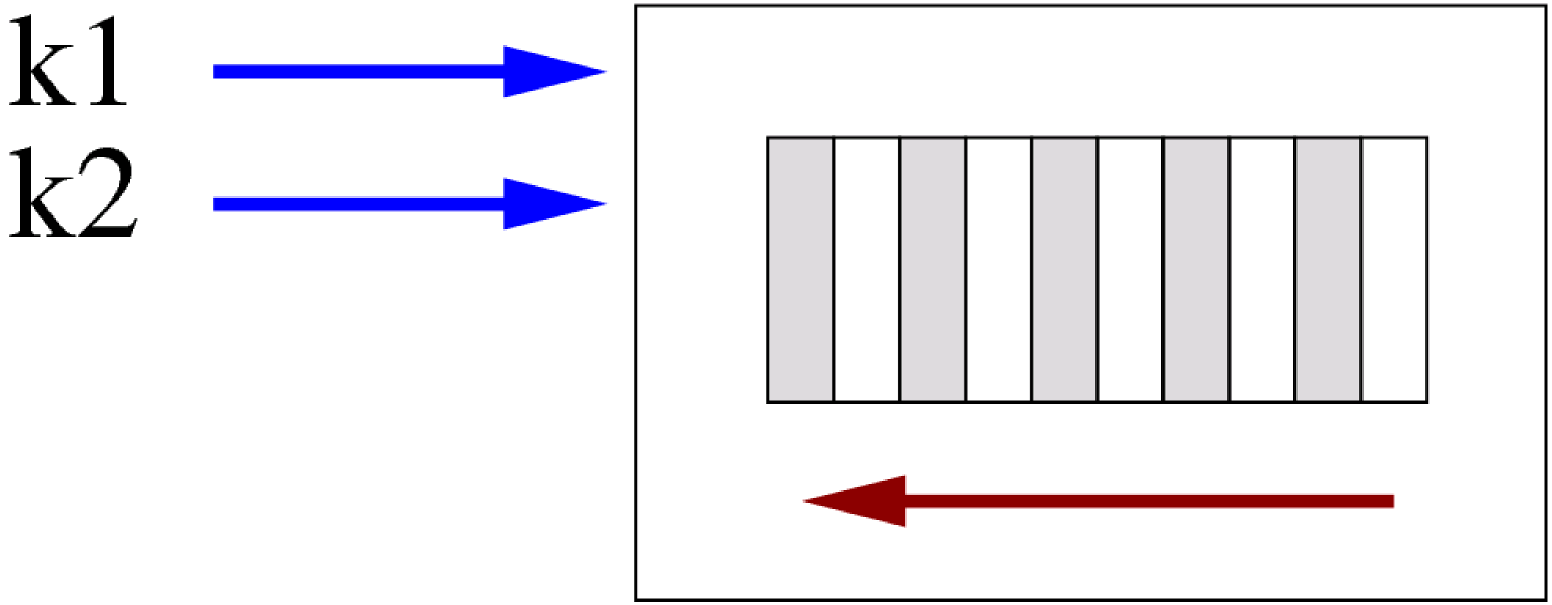
$$\delta = \nu_1 - \omega_{ab} = 0$$

Coherence of b-c transition off the two-photon resonance

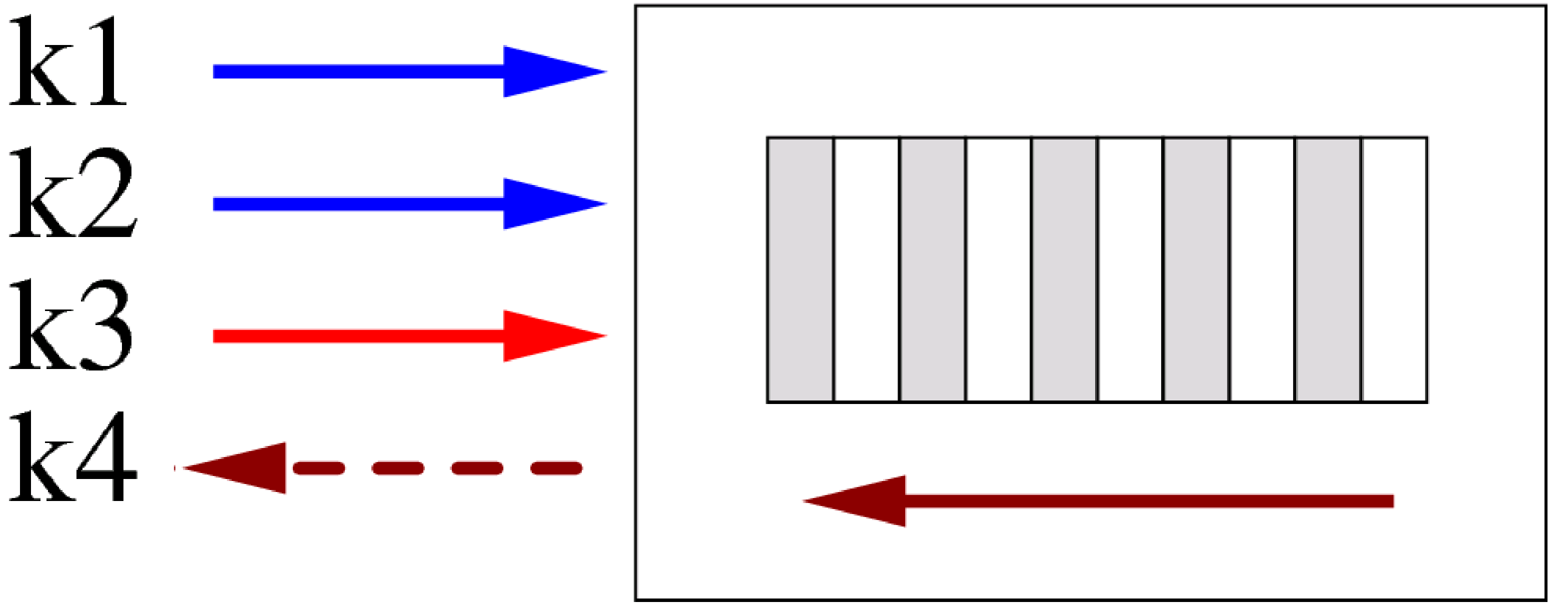


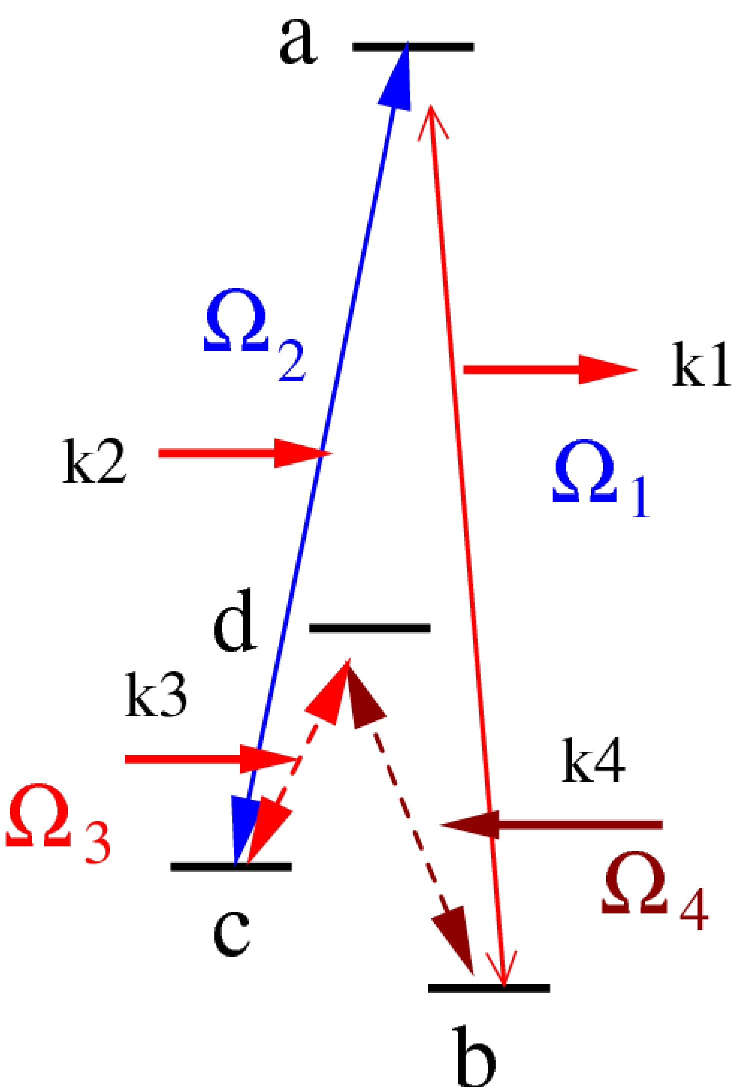
$$\delta = \nu_1 - \omega_{ab} < 0$$

Using two forward propagating beams, we prepare coherent grading in the medium



Then, the third propagating beam is scattered in the backward direction





$$\rho_{cb} \sim -\Omega_1 \Omega_2^*$$

$$\rho_{cb} \sim \exp[i(k_1 - k_2)z]$$

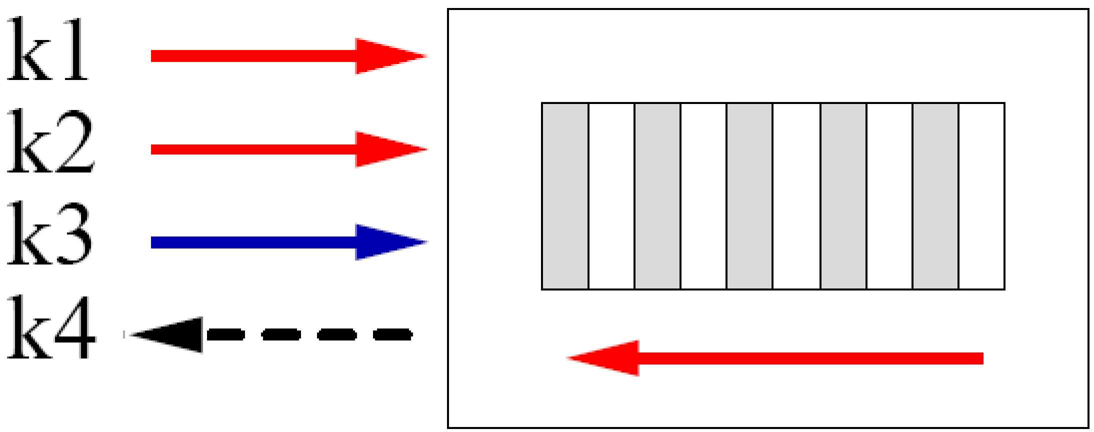
$$\frac{\partial}{\partial z}\Omega_4 \sim \rho_{cb}\Omega_3$$

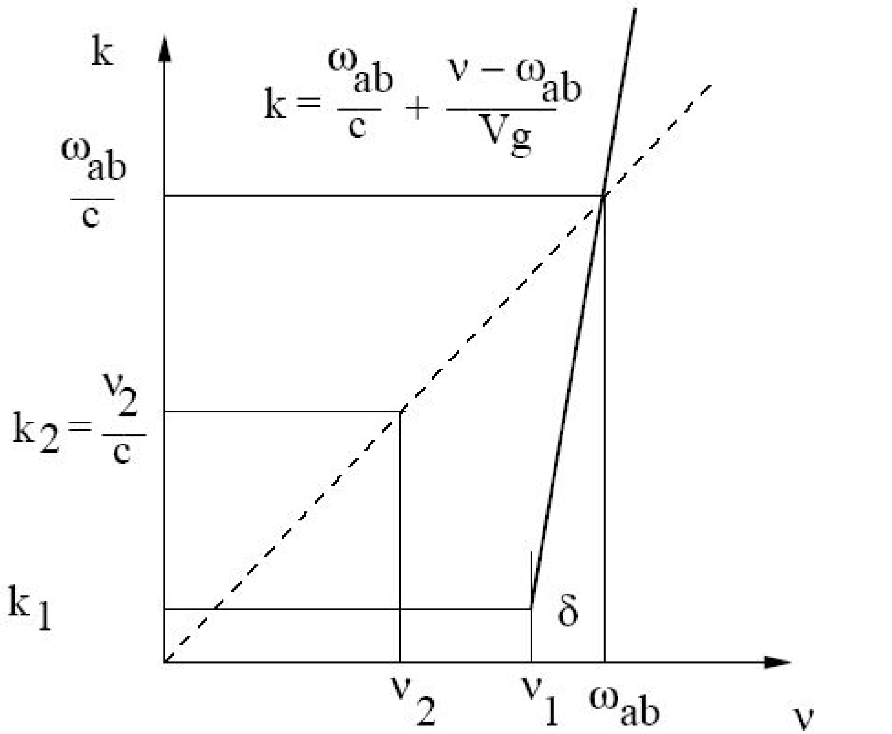
$$\sim \Omega_1 \Omega_2^* \Omega_3$$

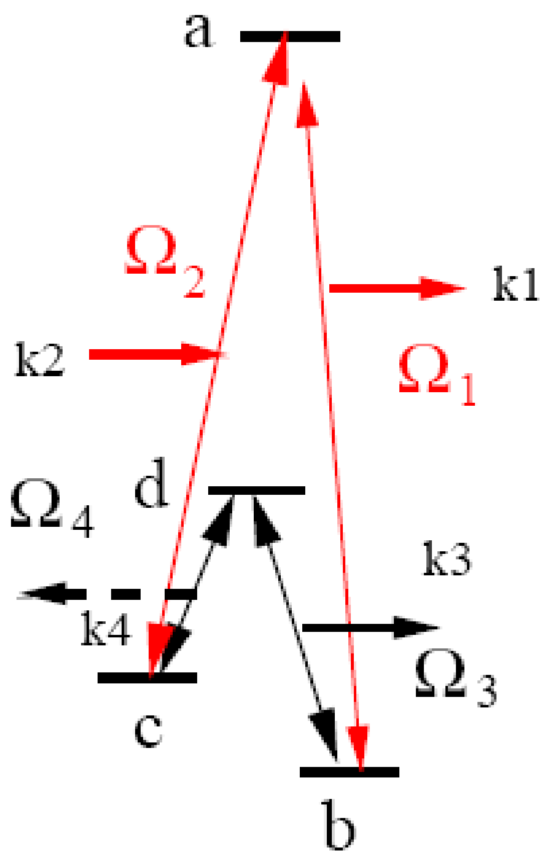
$$\sim e^{i(k_1 - k_2 + k_3 - k_4)z}$$

$$\nu_4 = \nu_1 - \nu_2 + \nu_3$$

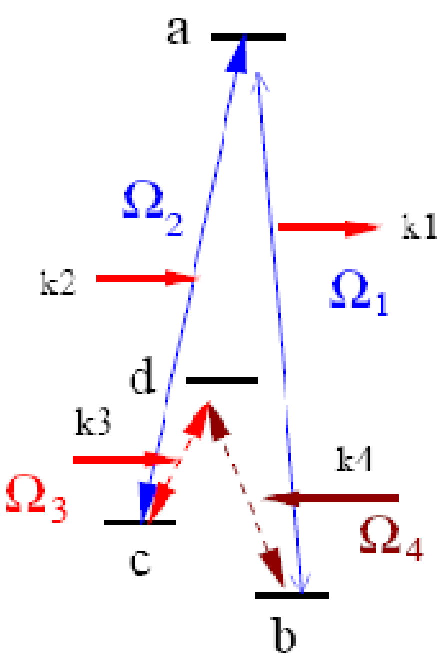
$$k_4 = k_1 - k_2 + k_3$$







$$\Delta k = k_1 - k_2 = \frac{\omega_{cb}}{c} + \frac{\nu_1 - \omega_{ab}}{V_g}$$



Signal field is given by $\Omega_4 \sim \rho_{cb}\Omega_3 e^{i(\Delta k + k_3)z}$

Phase-matching condition $k_4 = k_1 - k_2 + k_3$

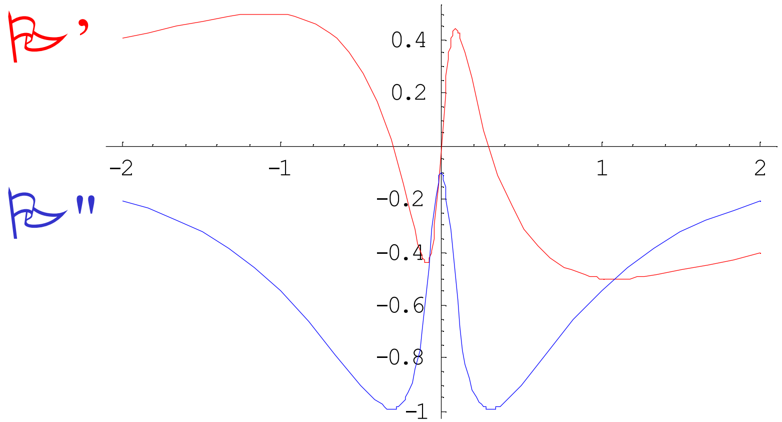
Detuning should satisfy $|\delta\nu| = 2|k_4|V_g = 2|k_4|\dfrac{8\pi|\Omega_2|^2}{3\lambda^2 N\gamma_r}$

In terms of susceptibility, since:

$$k_1 = \frac{\nu_1}{c}n_1 \simeq \frac{\nu_1}{c}\left(1 + c\frac{\nu_1 - \omega_{ab}}{\nu_1 V_g}\right) = \frac{\nu_1}{c}\left(1 - c\frac{2|k_4|}{\nu_1}\right)$$

$$\chi_{ab} = 2(n_1 - 1) = -4\frac{\lambda_{ab}}{\lambda_{db}}$$

The effect can be implemented for IR and FIR

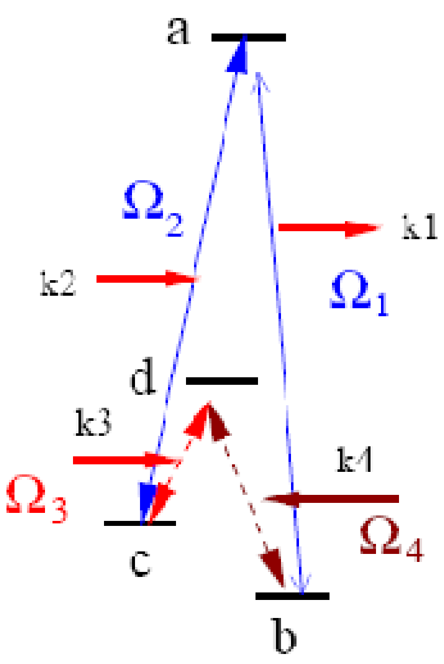


Probe frequency

and the condition for EIT is

$$\frac{|\Omega_2|^2}{\gamma_r|\delta\nu|} \geq 1$$

Thus, density is given by $N = \dfrac{16k_4}{3\lambda^2}\left(\dfrac{|\Omega_2|^2}{\gamma_r|\delta\nu|}\right) \simeq \dfrac{16k_4}{3\lambda^2}$



Estimation

Density is given by

$$N = \frac{16k_4}{3\lambda^2}\left(\frac{|\Omega_2|^2}{\gamma_r|\delta\nu|}\right) \simeq \frac{16k_4}{3\lambda^2}$$

NO (a resonant transition at 236 nm, $A^2\Sigma^+ - X^2\Pi$), vibration frequency of 1900 cm^{-1}, 5.26 μm

$N_{NO} = 8 \cdot 10^{15}\ \mathrm{cm}^{-3}$

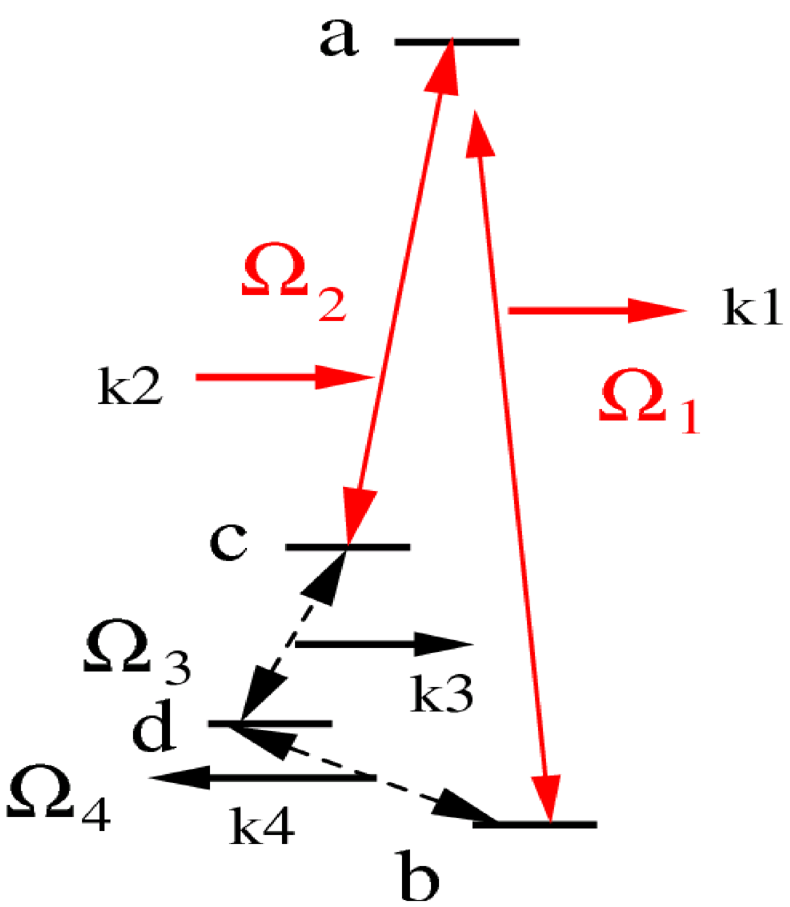
NO_2 (a resonant transition at wavelength 337 nm, vibrational frequency of 750 cm^{-1}, 13.3 μm)

$N_{NO2} = 1.4 \cdot 10^{15}\ \mathrm{cm}^{-3}$

For transition between rotational levels $\simeq 10\ \mathrm{cm}^{-1}$,
the required molecular density of NO and NO_2 molecules is $N \simeq 1.2 \cdot 10^{13}\ \mathrm{cm}^{-3}$

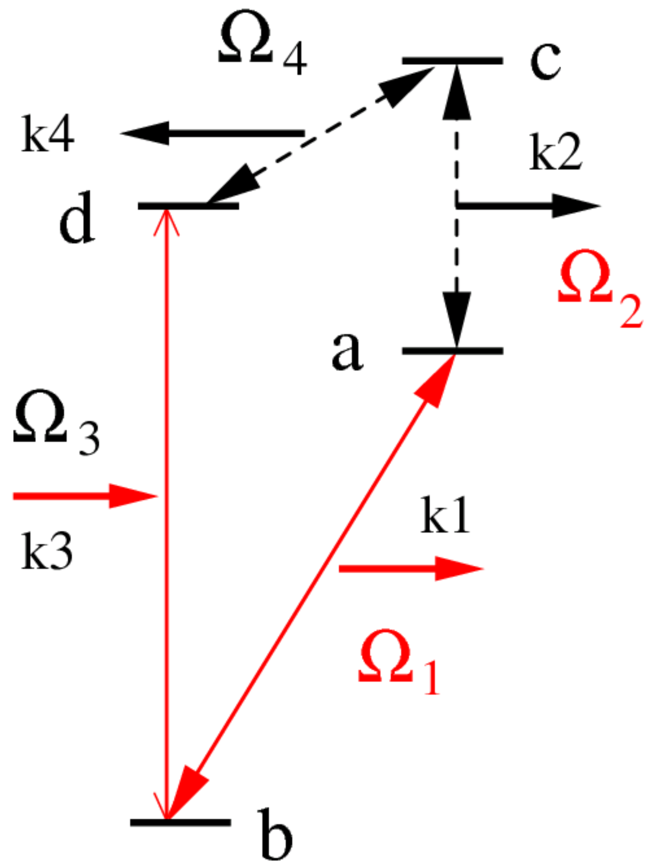
Alternative Schemes

Ladder- Λ

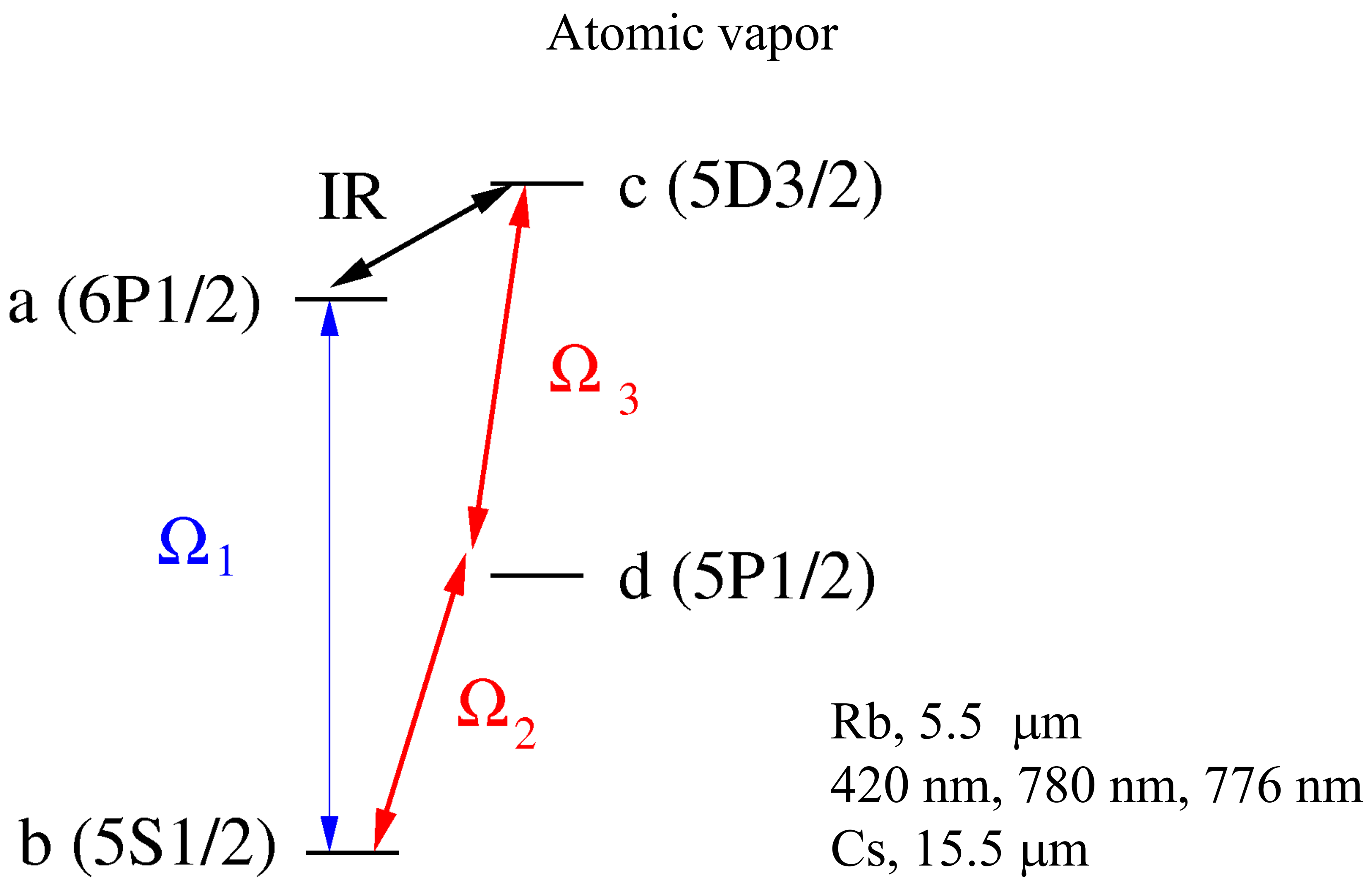


$$k_4 = k_1 - k_2 - k_3$$

Λ-V



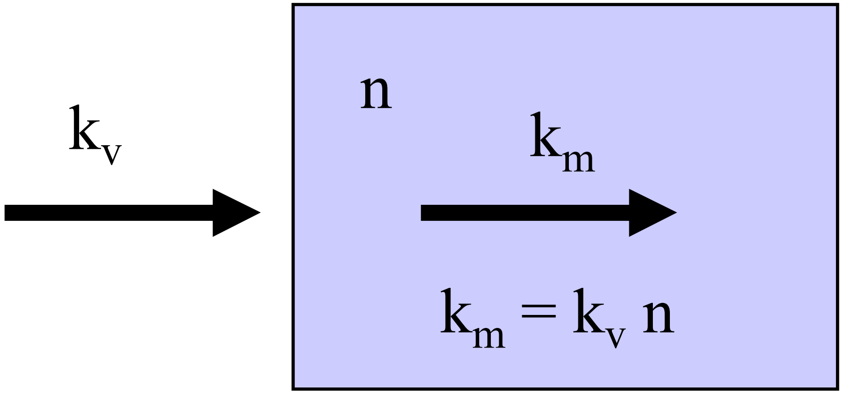
$$k_4 = k_1 + k_2 - k_3$$

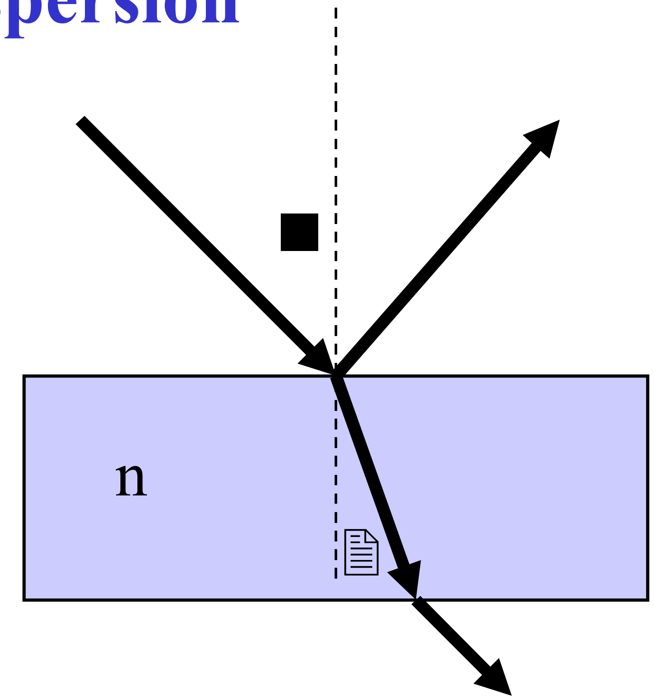

Atomic vapor
IR
c (5D3/2)
a (6P1/2)
Ω 3
Ω1
d (5P1/2)
Ω2
b (5S1/2)
Rb, 5.5 μm
420 nm, 780 nm, 776 nm
Cs, 15.5 μm
852 nm, 920 nm, 456 nm

The presented results based on resonant steep dispersion of the medium

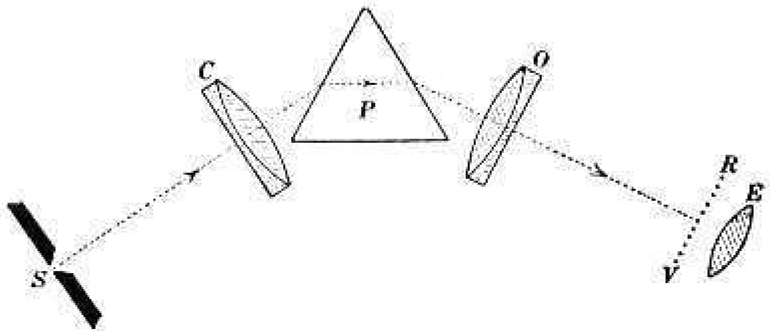
But could it be observed experimentally?

Dispersion of the index of refraction
Leads to angular dispersion





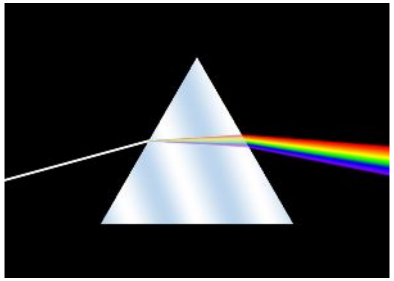
Sin■/Sin▯= n



$$\frac{d\theta}{d\lambda} = -\frac{\sin\varphi}{n^2}\frac{dn}{d\lambda}$$

Angular dispersion

Prisms:



$$d\theta/d\lambda = 10^{-4}\ \mathrm{nm}^{-1}$$

Diffraction gratings

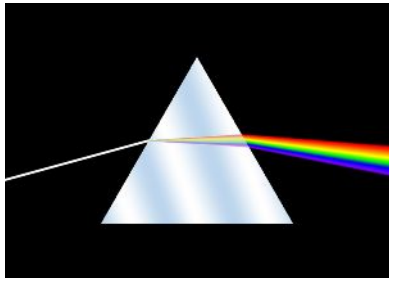
Interferometers

$$d\theta/d\lambda = 10^{-3}\ \mathrm{nm}^{-1}$$

What is angular dispersion of media with excited quantum coherence?

Angular dispersion

Prisms:

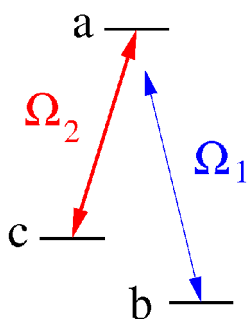


$$d\theta/d\lambda = 10^{-4}\ \mathrm{nm}^{-1}$$

Diffraction gratings
Interferometers

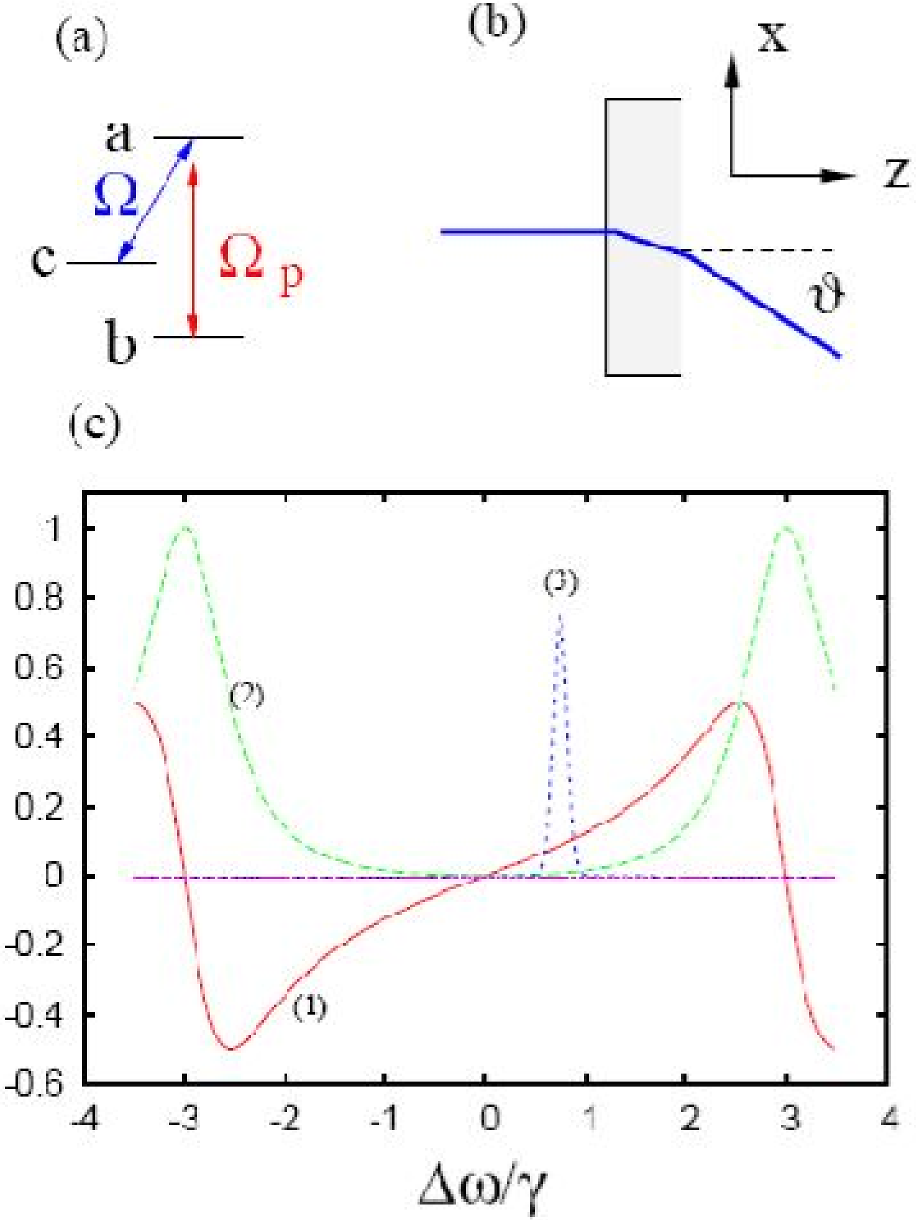
$$d\theta/d\lambda = 10^{-3}\ \mathrm{nm}^{-1}$$

The medium with excited quantum coherence



$$d\theta/d\lambda = 10^{3}\ \mathrm{nm}^{-1}$$

An ultra-dispersive medium controlled by coherent field



Propagation equation

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2}$$

Geometrical optics equaiton

$$\frac{d}{ds}\left(n\frac{d\vec{R}}{ds}\right) = \nabla n$$

Index of refraction is given by

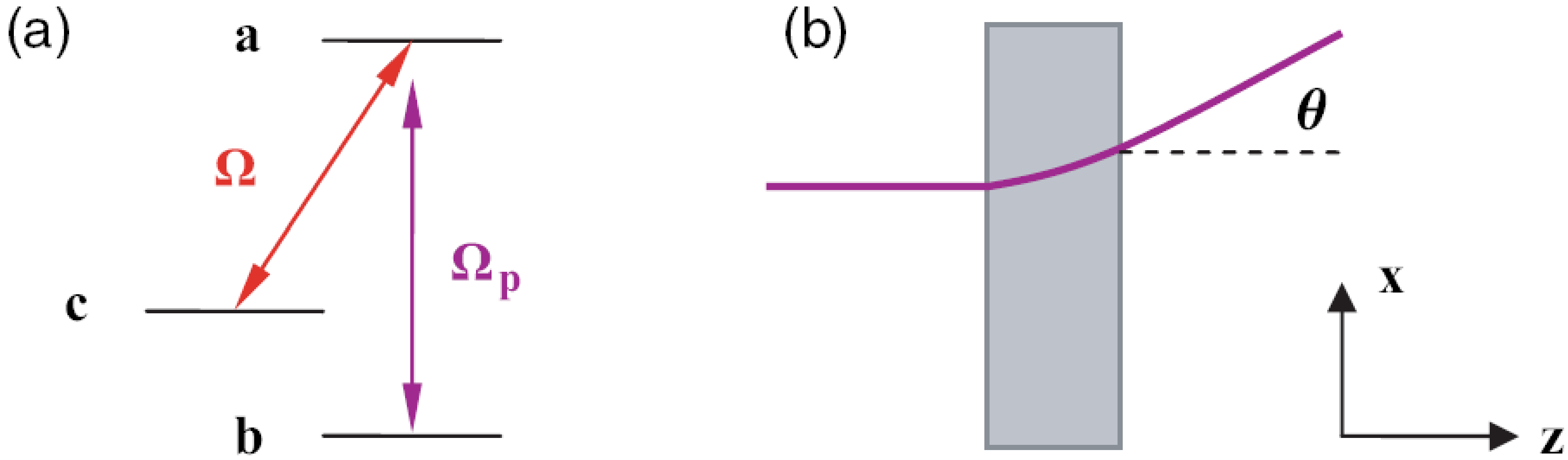
$$n = 1 + \frac{\omega - \omega_{ab}}{kV_g} \qquad V_g = \frac{|\Omega|^2}{\eta\gamma_r}$$

Estimate for the bending angle is

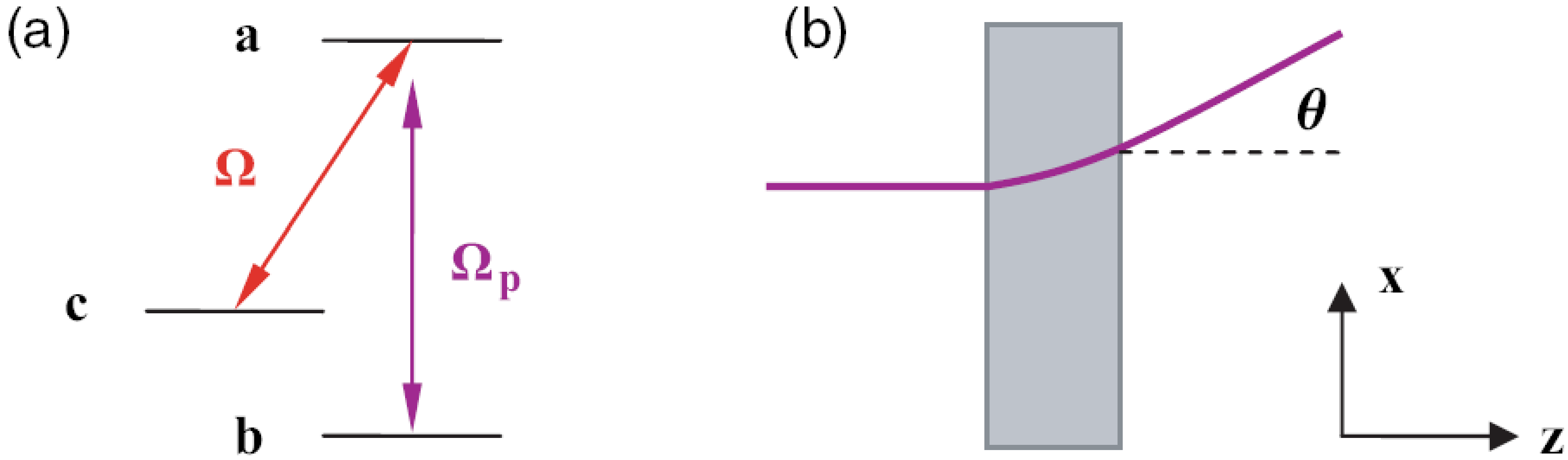
$$\theta = \alpha L = \frac{\Delta\omega}{kV_g}\frac{\Delta V_g}{V_g}\frac{L}{D} = \frac{\Delta\omega}{\gamma_{cb}kD}\frac{\Delta V_g}{V_g}\frac{\gamma_{cb}L}{V_g}$$

$$\theta \simeq 0.1 - 1,$$

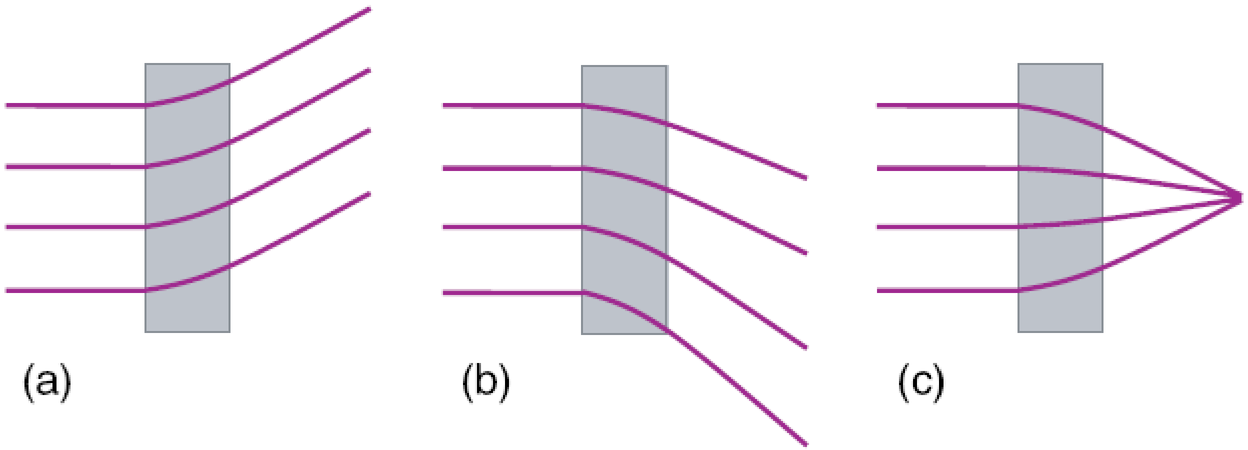
Propagation of the probe beam



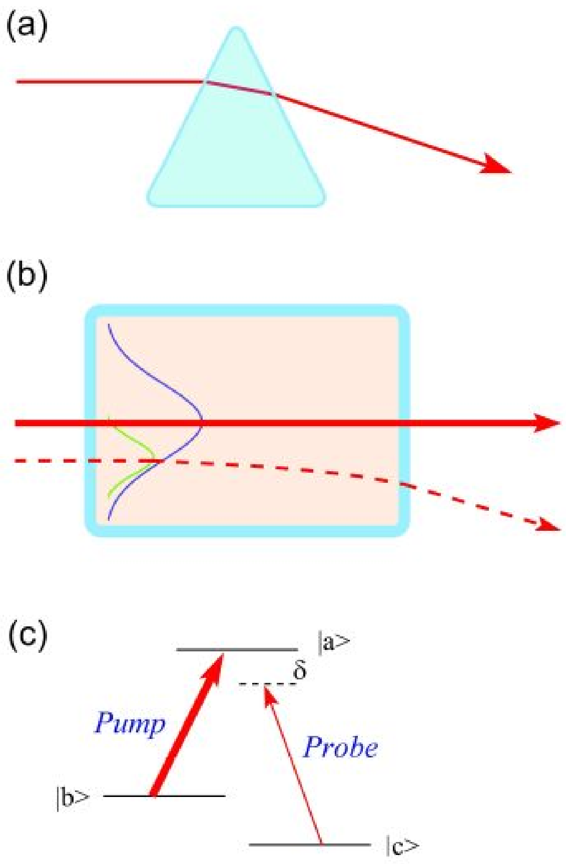
Propagation of the probe beam



Depending on the profile of the pump beam:



An ultra-dispersive prism build from EIT medium



Eikonal equation:

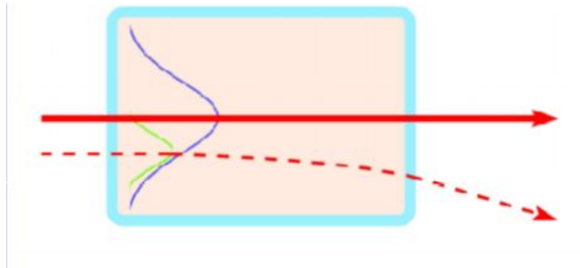
$$(\nabla\Psi)^2 = k^2 = \frac{\omega^2}{c^2} n^2$$

Bending angle is given by

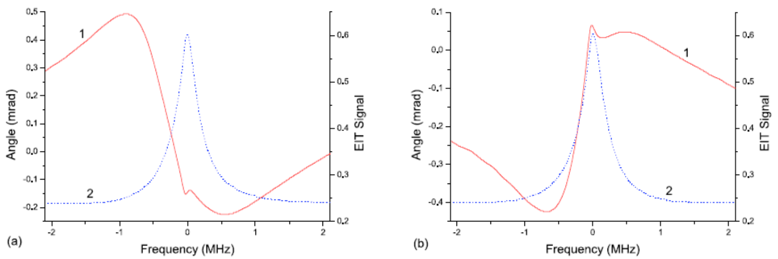
$$\theta \simeq L \nabla n$$

(b) Configuration of probe and control fields inside Rb cell.
(c) Simplified levels diagram of Rb atom.

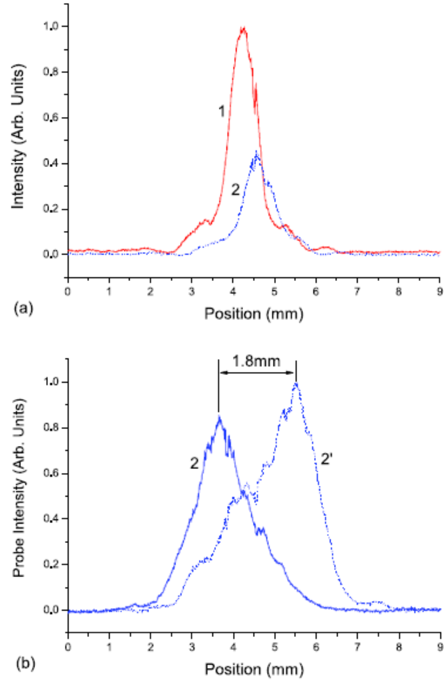
An ultra-dispersive prism: experimental results



Experimental setup



Angular dependence on detuning



Distribution of field vs position
(a) At the cell, (b) at 2 meter distance

Conclusion

We show that angular dispersion of the prism is the six orders of magnitude is higher than glass prisms and gratings.

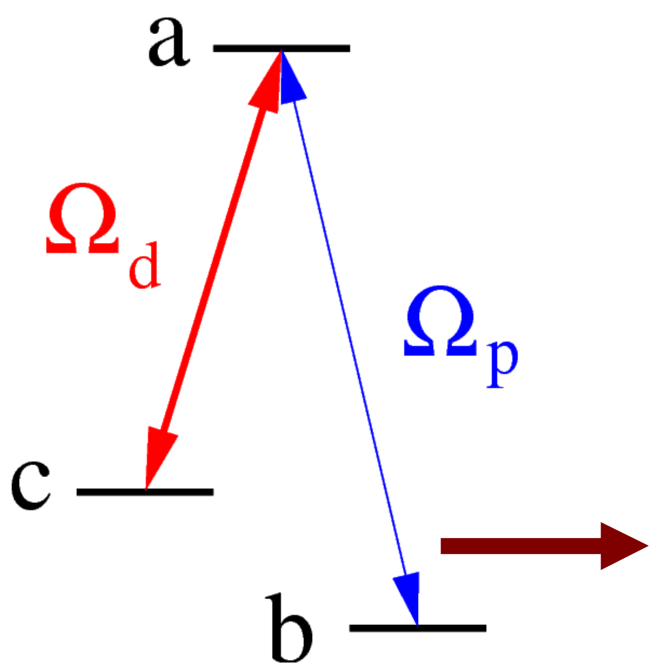
Application to solids is very promising

V.A. Sautenkov, *et al.*, Ultra-dispersive adaptive prism, quant-ph/0701229

It might be nice…

But, is there any experimental implementation?

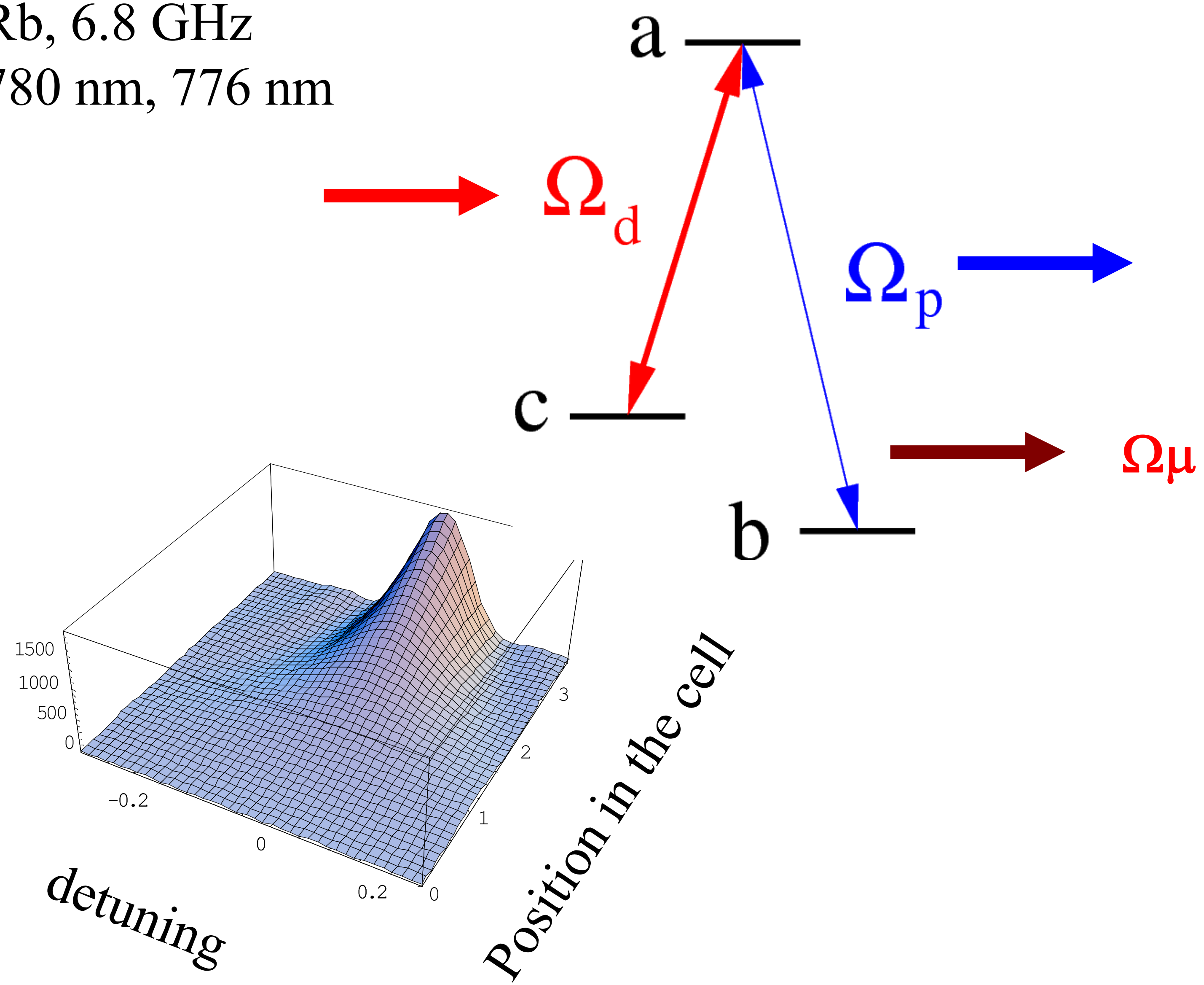
Proof-of-principle experiment in Rb atomic vapor
(work in progress at TAMU)



Rb, 6.8 GHz
780 nm, 776 nm

Proof-of-principle experiment in Rb atomic vapor
(work in progress)

Rb, 6.8 GHz
780 nm, 776 nm



Proof-of-principle experiment in Rb atomic vapor
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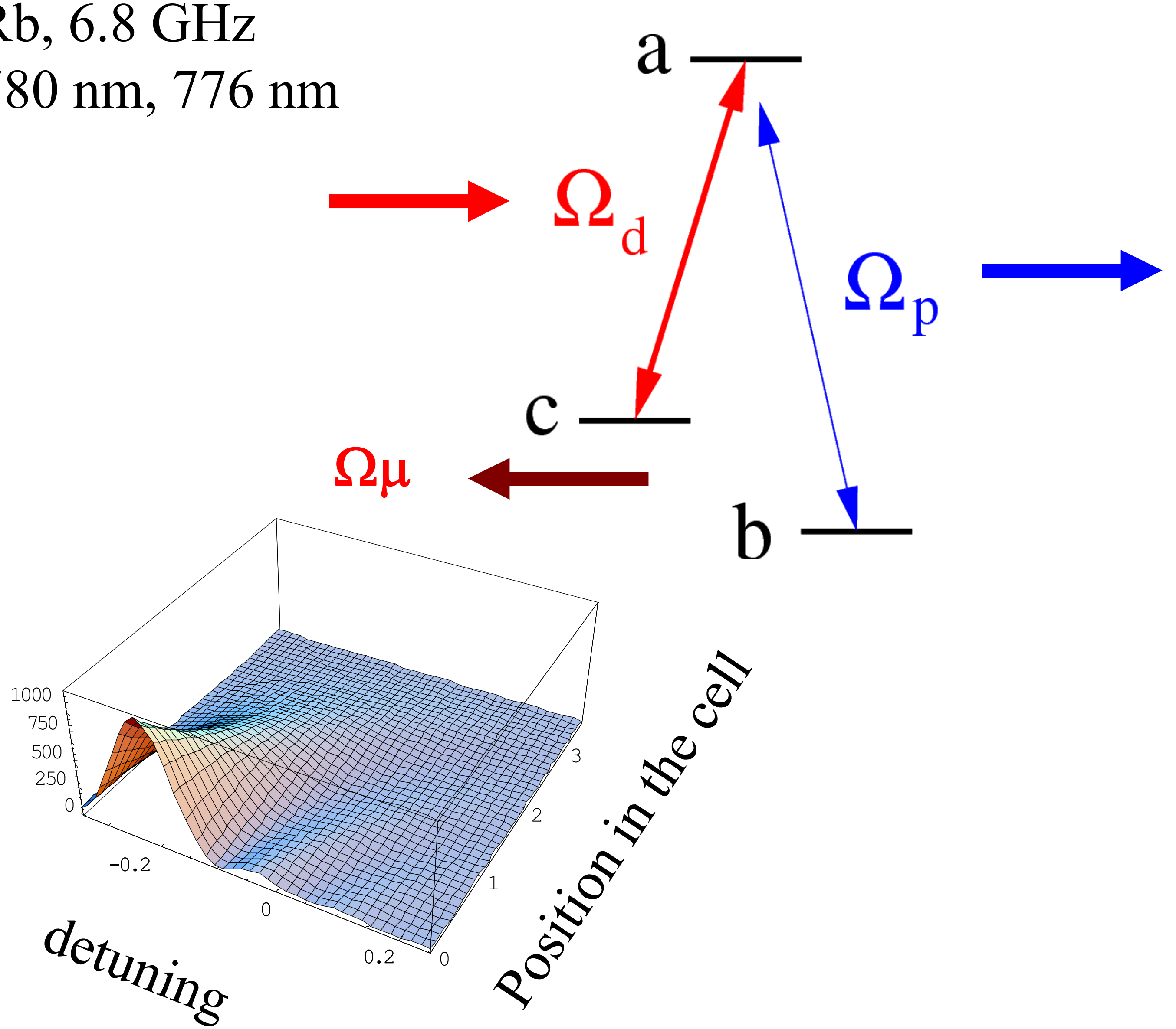
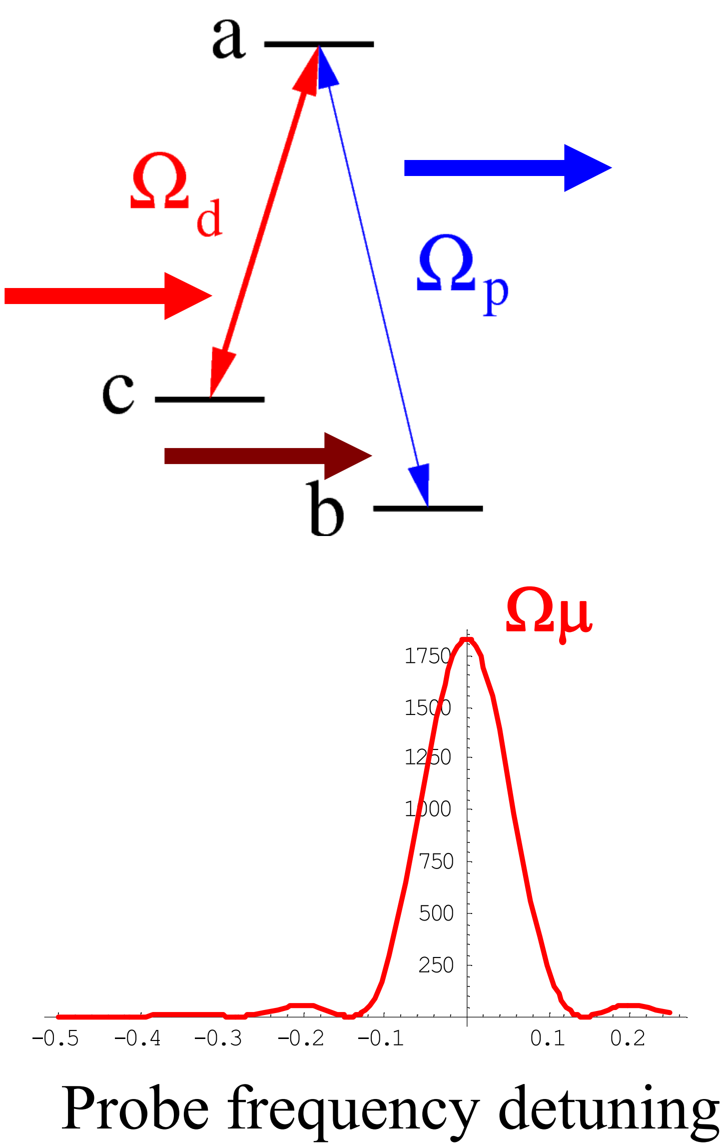
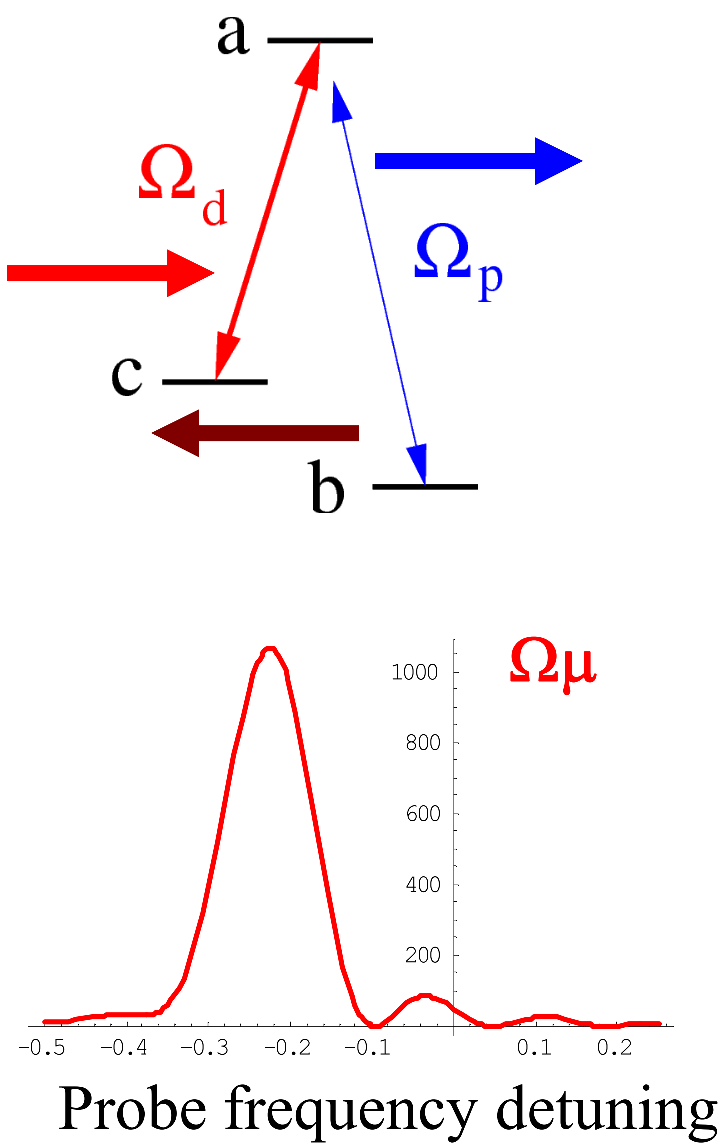
Rb, 6.8 GHz
780 nm, 776 nm



Proof-of-principle experiment in Rb atomic vapor
(work in progress at TAMU)

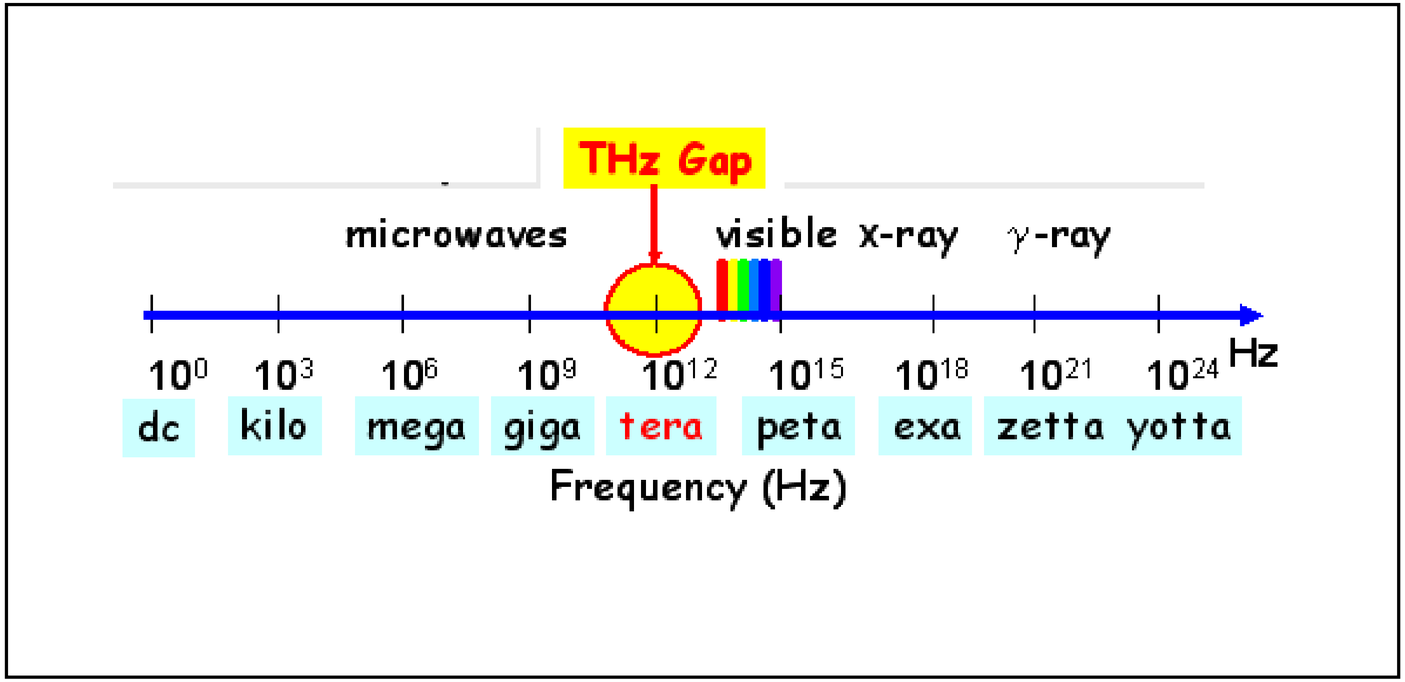




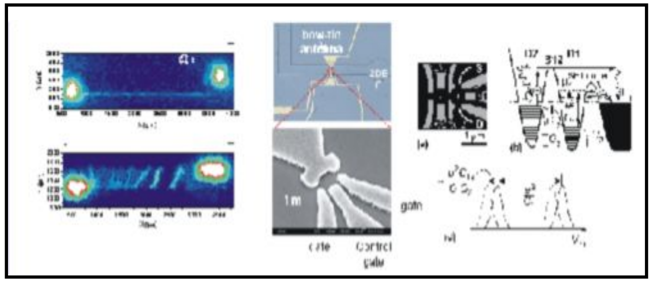
Application to Ruby Crystal and Generation of THz

(work in progress at the Institute of Applied Physics RAS by R. Akhmedzhanov's group)

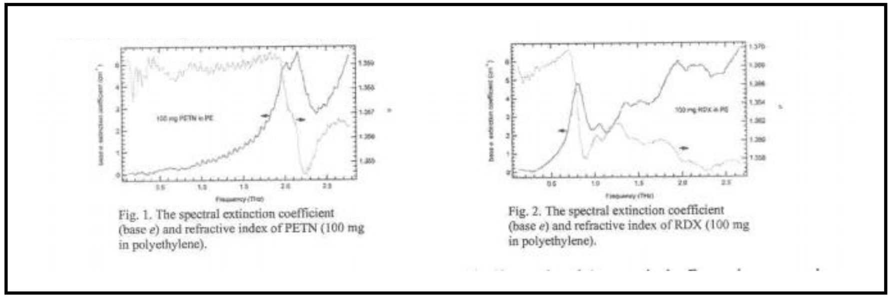
What is THZ?



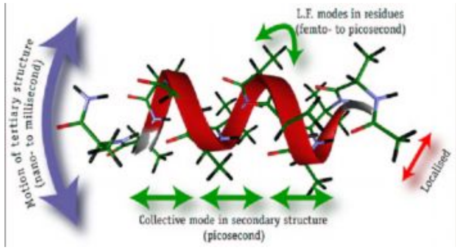
Possible applications of ultrashort pulses of THz radiation



physics and engineering



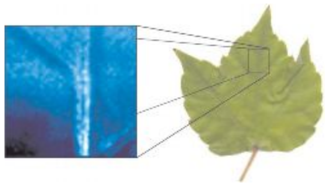
defense and security

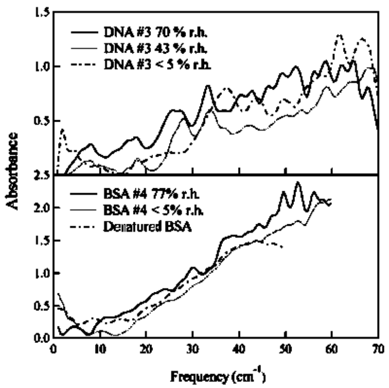


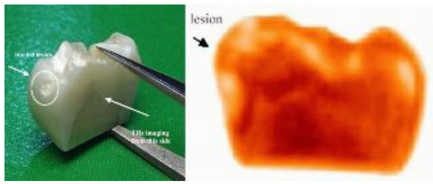
chemistry

biology

medicine







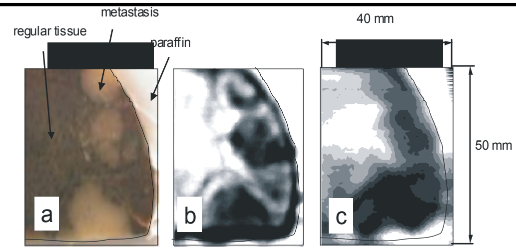
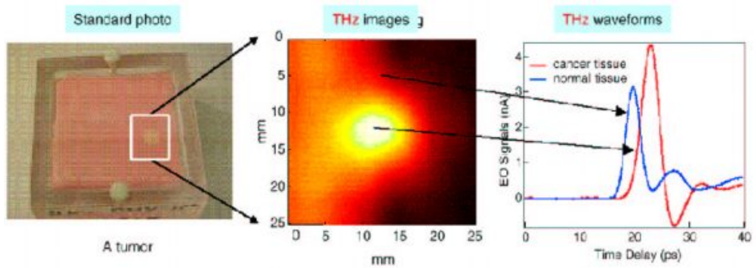
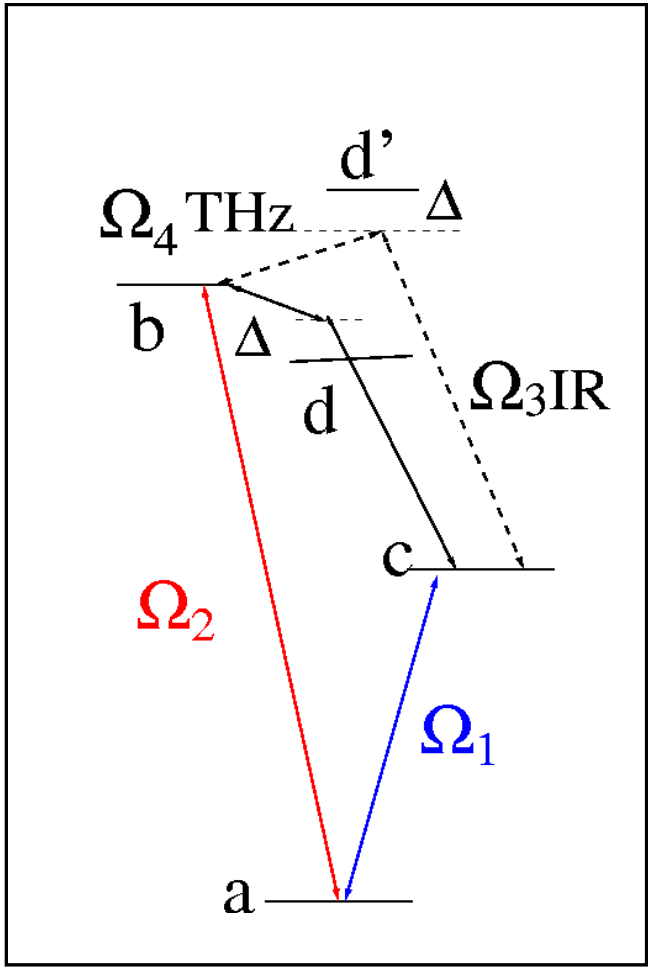


Figure 1. Histo-pathological sample of a human liver which contains tumours. (a) Photography. (b) Image obtained with pulsed THz spectroscopy and an integration window from 0.2 to 0.5 THz. (c) CW THz image obtained at 230 GHz.



Coherent generation of short THz pulses: from atomic and molecular gases to solids



Ba, Ca, Rb_2, K_2, …

Ω_1 and Ω_2 are optical fields in two-photon resonance to excite coherence between levels b and c. Ω_3 is the IR field, and Ω_4 is generated THz radiation (V-Λ shown by dashed lines, Ladder-V by solid lines). Atomic medium: For example, Rb levels are a = $5S_{1/2}$, b = $10P_{1/2,3/2}$, c = $6P_{1/2,3/2}$, d(d') = $8D_{3/2,5/2}$($9D_{3/2,5/2}$).

M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Efficient nonlinear frequency conversion with maximal atomic coherence, Phys. Rev. Lett. **77**, 4326-4329 (1996); M. D. Lukin, P. R. Hemmer, M. Löffler, and M. O. Scully, Resonant enhancement of parametric processes via radiative interference and induced coherence, Phys. Rev. Lett. **81**, 2675-2678 (1998)

Efficiency of the method

Hamiltonian

$$V_I = -\hbar[\Omega_2 e^{-i\omega_{ac}t}|a\rangle\langle c| + \Omega_1 e^{-i\omega_{ab}t}|a\rangle\langle b| + h.c.]$$
$$-\hbar[\Omega_3 e^{-i\omega_{dc}t}|d\rangle\langle c| + \Omega_4 e^{-i\omega_{db}t}|d\rangle\langle b| + h.c.]$$

Equations:

$$\frac{\partial\rho}{\partial\tau} = -\frac{i}{\hbar}[H,\rho] - \frac{1}{2}(\Gamma\rho + \rho\Gamma) \qquad \frac{\partial\Omega_\alpha}{\partial z} = -\kappa_\alpha\Omega_\alpha - i\xi\eta_\alpha\rho_\alpha$$

$$\Omega_i = \wp_i\mathcal{E}_i/\hbar \quad \xi = \int F_{nm}(x,y)dxdy/S \qquad \eta_\alpha = \nu_\alpha N\wp_\alpha^2/(2\hbar\epsilon_0 c) \qquad \kappa_4 = \lambda/D^2$$

Generated field is given by

$$\Omega_4 = \xi\int_0^L dz e^{i\delta kz}\eta\tau\rho_{cb}\Omega_3^* = \xi\frac{\sin(\delta kL)}{(\delta kL)}\eta\tau\rho_{cb}L\Omega_3^*$$

$$(\delta k = k_4 - k_3 - k_1 + k_2),\ \xi \simeq 1$$

Efficiency:

$$\epsilon = \frac{I_4^L}{I_3^0} = \xi^2\mathrm{sinc}^2(\delta kL)\left(\frac{4\pi^2\wp_v\wp_j N\rho_{bc}\tau L}{\lambda\hbar}\right)^2$$

Note: depletion of Ω_3 is not taken into account

Coherent generation of THz radiation in Ruby at room temperature

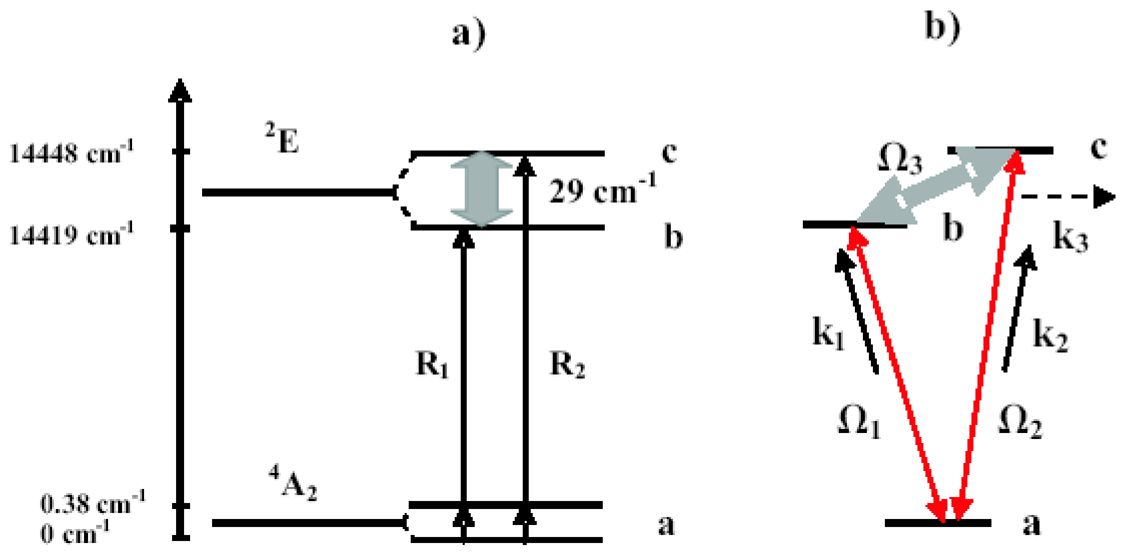
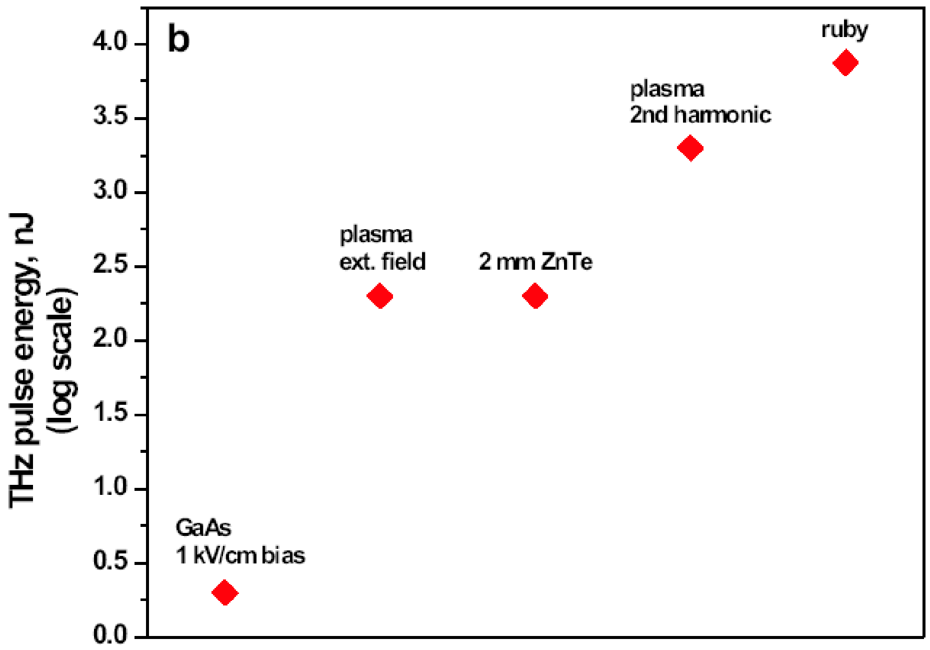
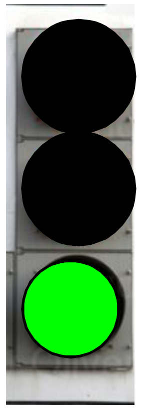


FIG. 1: a) Three-level V energy system in ruby proposed for generation of 29 cm^{-1} THz pulses; b) Model V system of energy levels with two co-propagating fields 1 and 2 inducing coherence between levels b and c.


b
THz pulse energy, nJ
(log scale)
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
GaAs
1 kV/cm bias
plasma
ext. field
2 mm ZnTe
plasma
2nd harmonic
ruby

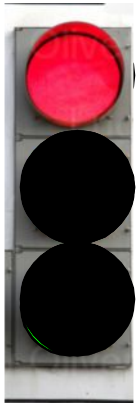


Conclusion:

We show that in EIT atomic and molecular systems coherent backscattering is possible. It allows one

- to generate spatially entanglement photon states,
- to control phase-matching,
- to perform nonlinear light steering,
- to improve spatial resolution

We have shown the experiments in support of theoretical results, although some experiments are in progress.



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Thank you for your attention ☺

Thanks to collaborators:

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J. Dowling, Louisiana State University
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M. Gubin, Lebedev Institute, RAS, Russia
I. Novikova, E. Mikhailov, College of William&Mary
N. Kalugin, NMT
G. Kurizki, Weizmann Institute of Science, Israel
S. Suckewer, R. Miles, Princeton University
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TAMU
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EE **R. Navel, P.R. Hemmer**
Chemistry **J. Bevan, J. Laane**
Math **G. Gordon**

$$$ Funding: ONR, CRDF