

15:20~16:00, 10月11日(金)

第6回 レーザー学会「ユビキタス・パワーレーザー」専門委員会

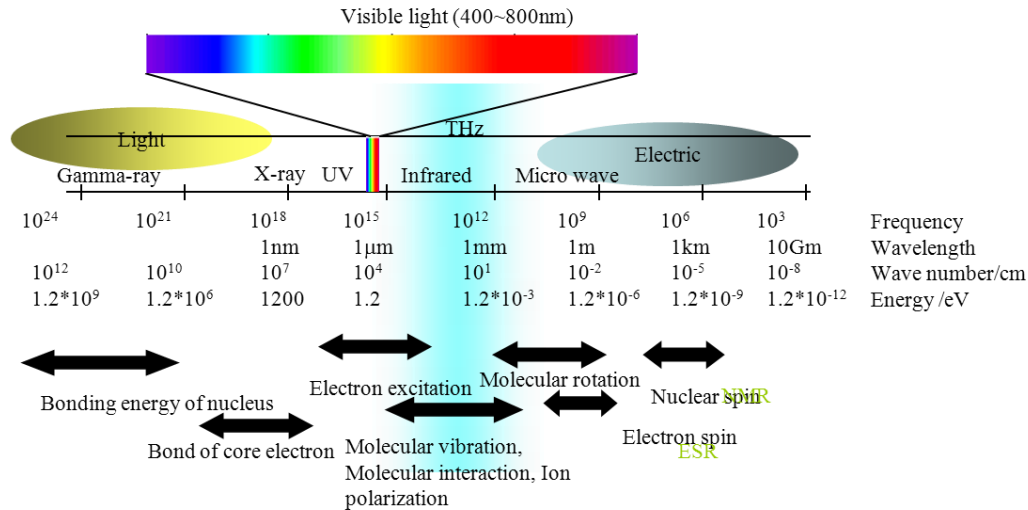
第6回 科学技術交流財団「マイクロ固体フォトニクス」研究会

「小型集積レーザーとTHZ波の最前線」

# チェレンコフ位相整合方式を用いた 非線形光学波長変換法

竹家啓、内田裕久、川瀬晃道

# テラヘルツ波



Terahertz wave lies between visible light and electric wave

## Features

- Moderate transparency
- Easy handling (components)
- Low energy, safety
- Spectroscopy

Terahertz applications

# テラヘルツ応用

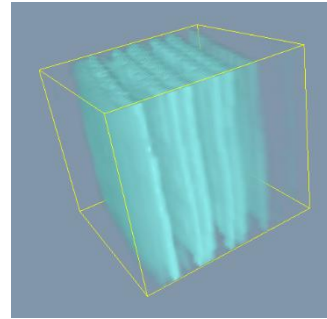
10mm

IF UNDELIVE  
RIKEN  
2-1 Hiroswa,  
BY AIR M

MDMA  
Aspirin  
Methamphetamine

麻薬 アスピリン 覚せい剤  
(試料提供：科学警察研究所)

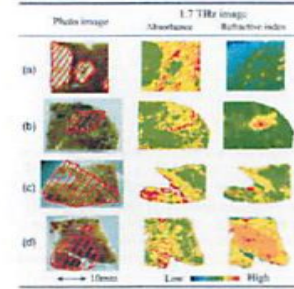
Opt. Exp. 2016  
遮蔽物越しの薬品分析(上:封筒、下、段ボール)



テラヘルツトモグラフィ



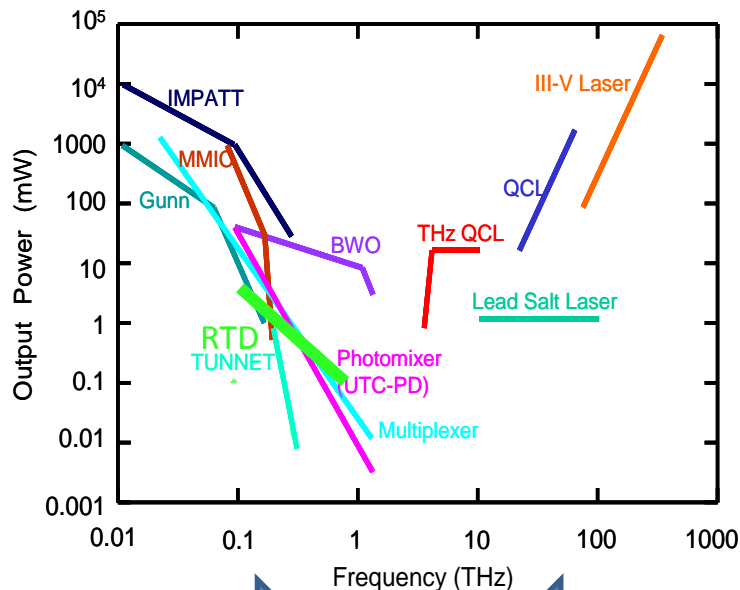
美術品の分析



がん細胞のTHz波測定

セキュリティー  
通信  
分光分析  
etc.

# テラヘルツギャップ



電子回路技術

レーザー、非線形光学技術

Ref: Tonouchi, *Nature Photonics*, 2007.

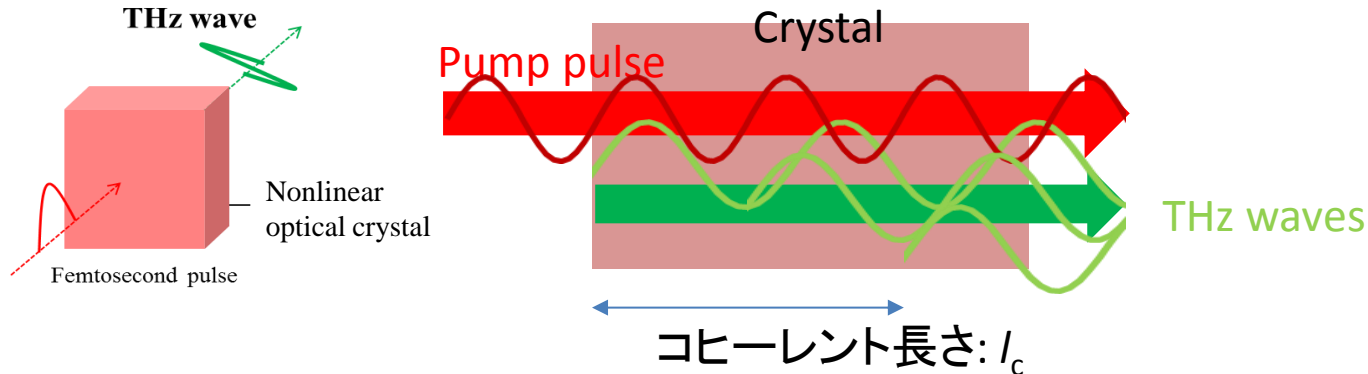
依然として 1-3 THz における光源が不足  
⇒ テラヘルツギャップ

# テラヘルツ波光源

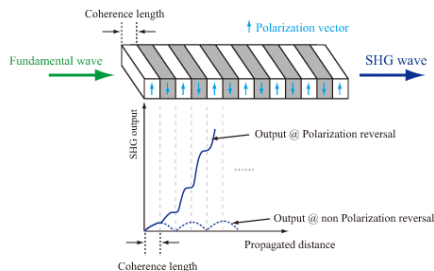
方式	コメント
自由電子レーザー, p型Ge半導体レーザー, CO2ガスレーザー	高出力 装置が大型, 大量の液体Heを消費する, 発生波長域が限定
THz-QCL	225 Kでの動作が実現, CW(Continuous Wave)で140 mW
共鳴トンネルダイオード(Resonant Tunneling Diodes:RTD)	1 THzを超える基本発振も達成
フェムト秒レーザー励起(光伝導アンテナ、 <b>非線形光学結晶</b> など)	THz-TDS、分光計測
<b>非線形光学結晶を用いた波長変換(差周波、パラメトリック発生)</b>	単一周波数を広帯域に発生

結晶を用いたTHz波発生は非線形光学効果を利用するため、デバイスの工夫次第で高出力化、小型化が期待される発生法であり、小型レーザーなどと組み合わせることで、発生方法や応用利用に大きな可能性がある

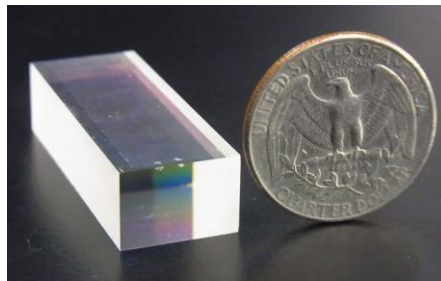
# 非線形光学結晶における位相整合



コリニア(共軸)発生においてはコヒーレント長さを考慮しなければならない。



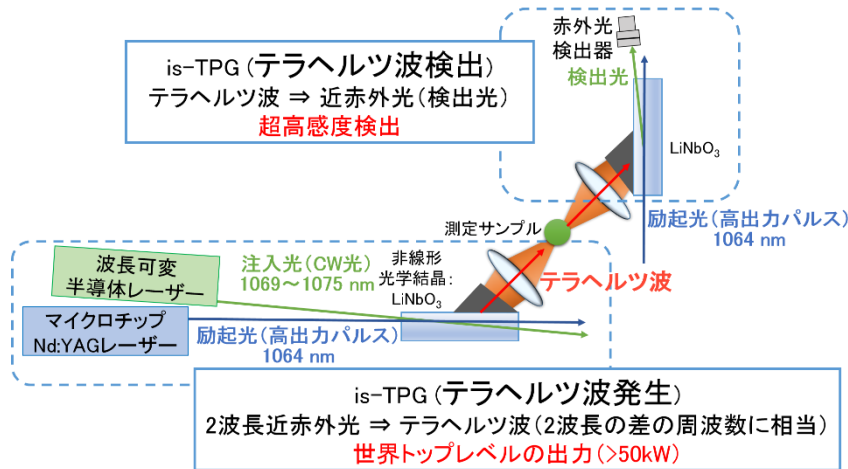
Optipediaより



Ishizuki, Taira et al.,

疑似位相整合などの手法が用いられる

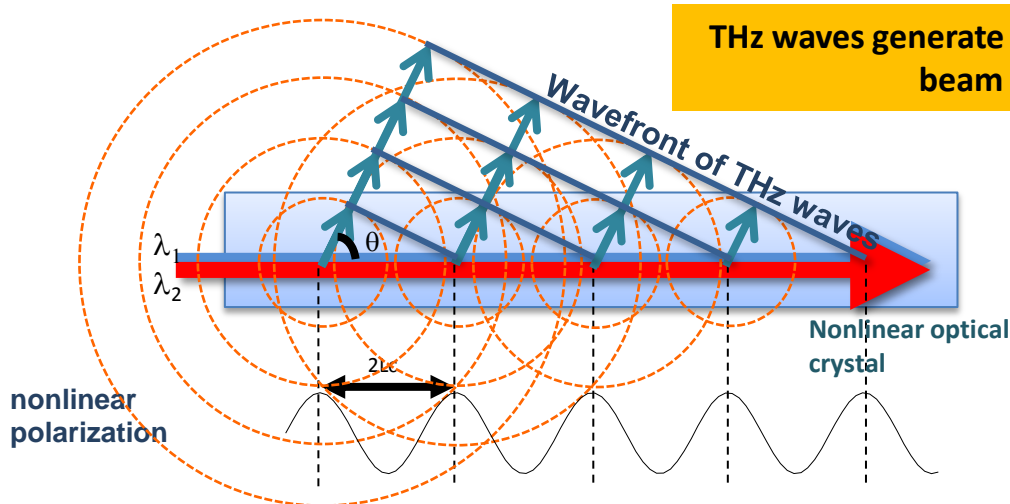
# 非線形光学結晶における位相整合



(パラメトリック発生における位相整合。名古屋大 川瀬研HPより)

非線形光学結晶を用いる際には、励起光とTHz光の位相整合を考慮しなければならない。さらに、結晶自身によるTHz波の吸収も無視できない。

# チェレンコフ位相整合



THz waves generate each point, when pump beam propagate a NLO crystal

$$\cos \theta = \frac{\lambda_{THz} / n_{THz}}{2Lc} = \frac{\lambda_{THz} / n_{THz}}{\lambda_1 \lambda_2 / (n_1 \lambda_2 - n_2 \lambda_1)} \doteq \frac{n_{opt}}{n_{THz}}$$

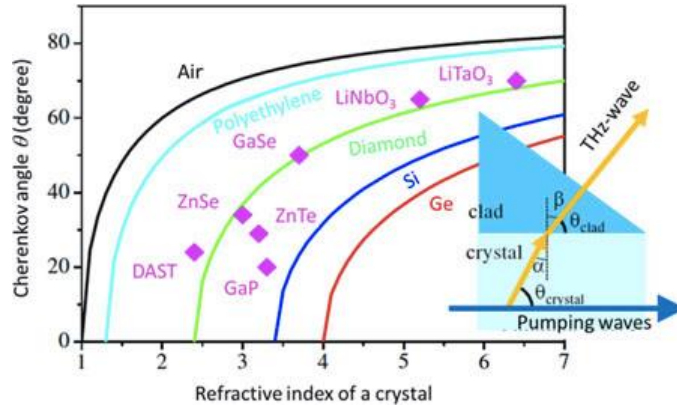
結晶内を伝搬する励起光の位相速度が、発生するテラヘルツ波より早いとき ( $n_{THz} > n_{opt}$ )、チェレンコフ位相整合条件が満たされ、特定の方向に波が強め合う。

位相整合条件を考慮しないでいい。  
表面発生を利用しているため結晶による吸収も軽減できる。

This is an appropriate method to generate THz waves, taking advantage of NLO crystal.



# プリズム結合チェレンコフ位相整合方式 Prism-Coupled Cherenkov Phase-Matched (PCCPM) THz wave generation

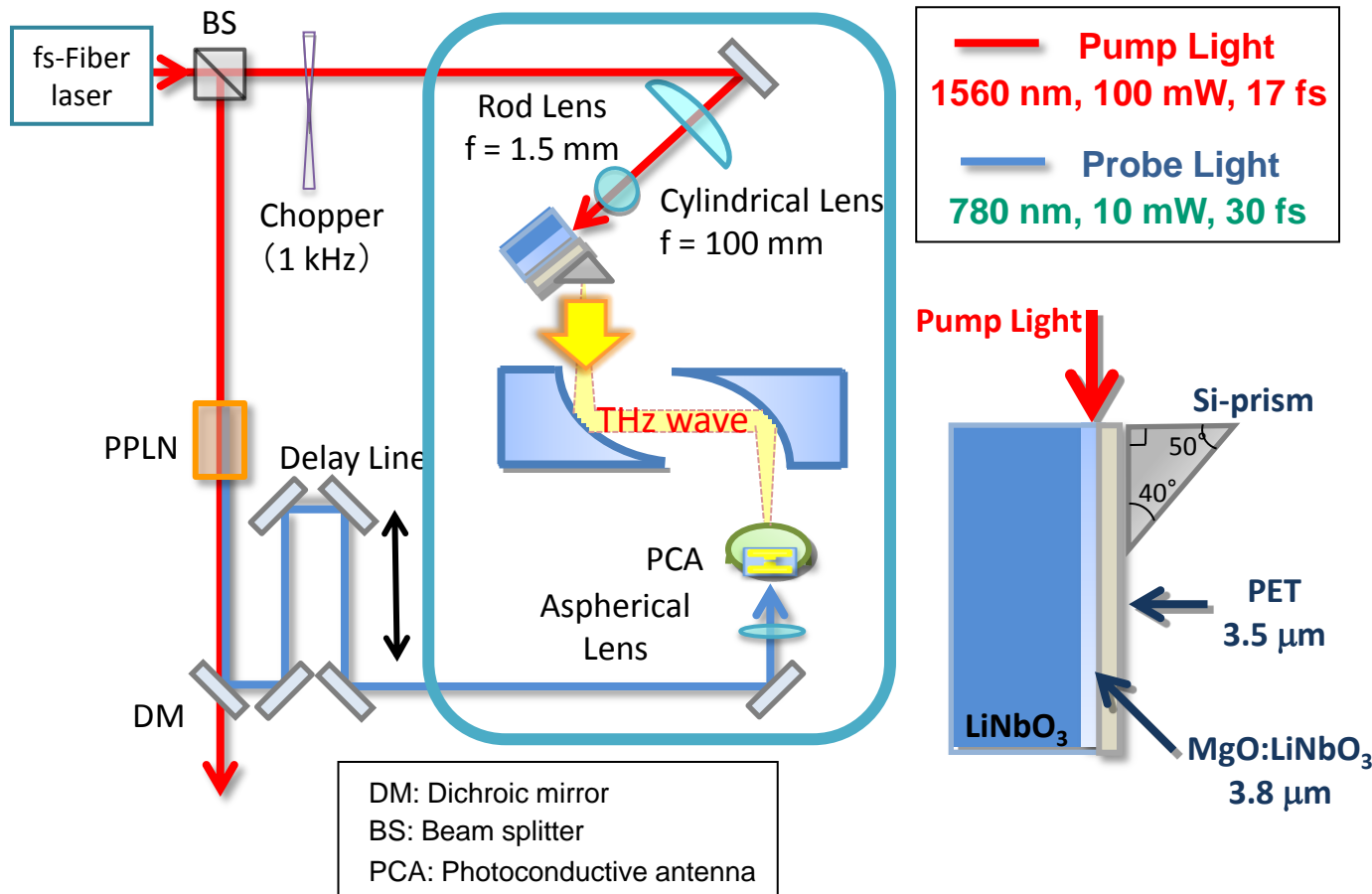


$$\begin{aligned} \theta_{clad} &= \frac{\pi}{2} - \arcsin\left(\frac{n_{THz}}{n_{clad}} \sin\left(\frac{\pi}{2} - \theta_{crystal}\right)\right) \\ &= \frac{\pi}{2} - \arcsin\left(\frac{n_{THz}}{n_{clad}} \sin\left(\frac{\pi}{2} - \arccos\left(\frac{n_{opt}}{n_{THz}}\right)\right)\right) \\ &= \arccos\left(\frac{n_{opt}}{n_{clad}}\right) \end{aligned}$$

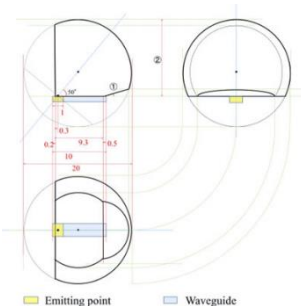
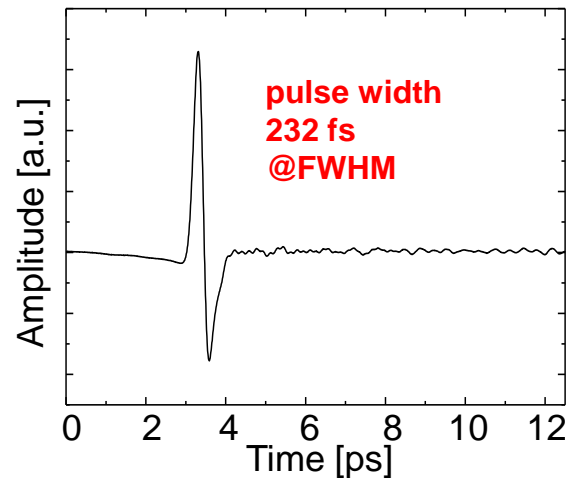
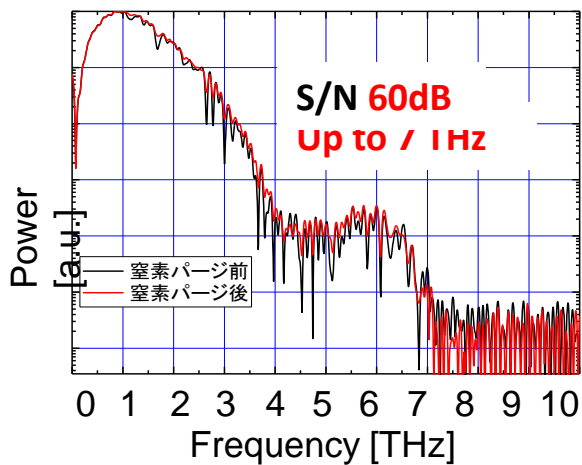
- $\cos \theta_{clad}$  ..... clad内のテラヘルツ波の放射角
- $n_{crystal}$  ..... テラヘルツ波に対する結晶の屈折率
- $n_{clad}$  ..... テラヘルツ波に対するcladの屈折率
- $\cos \theta_{crystal}$  ..... 結晶内のテラヘルツ波の放射角
- $n_{opt}$  ..... 励起光に対する結晶の屈折率

適切なClad材をプリズムとして選択することで、結晶のTHz帯における屈折率を無視してテラヘルツ波を取り出せる。→非線形光学結晶はTHz帯に複雑な分散を持つ結晶が多いが、それらを考慮せず幅広く結晶選択の自由がある

# LiNbO<sub>3</sub>結晶+チェレンコフ発生方式を用いたTHz-TDS光学系



# LiNbO<sub>3</sub>スラブ導波路からのテラヘルツパルス



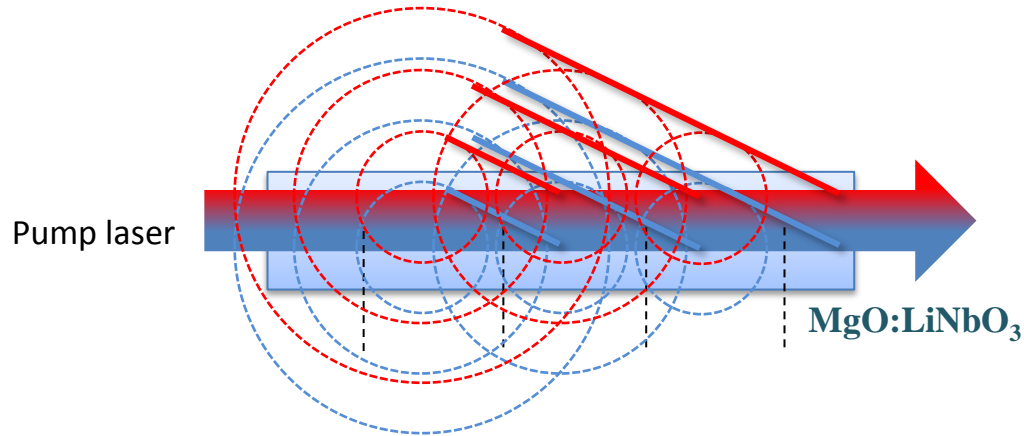
# ビーム径による位相不整合

However, there are still some problems on Cherenkov phase matching

## Phase mismatch

A finite beam width of pump beam makes many waves at the same time

These waves create interference and cancel each other



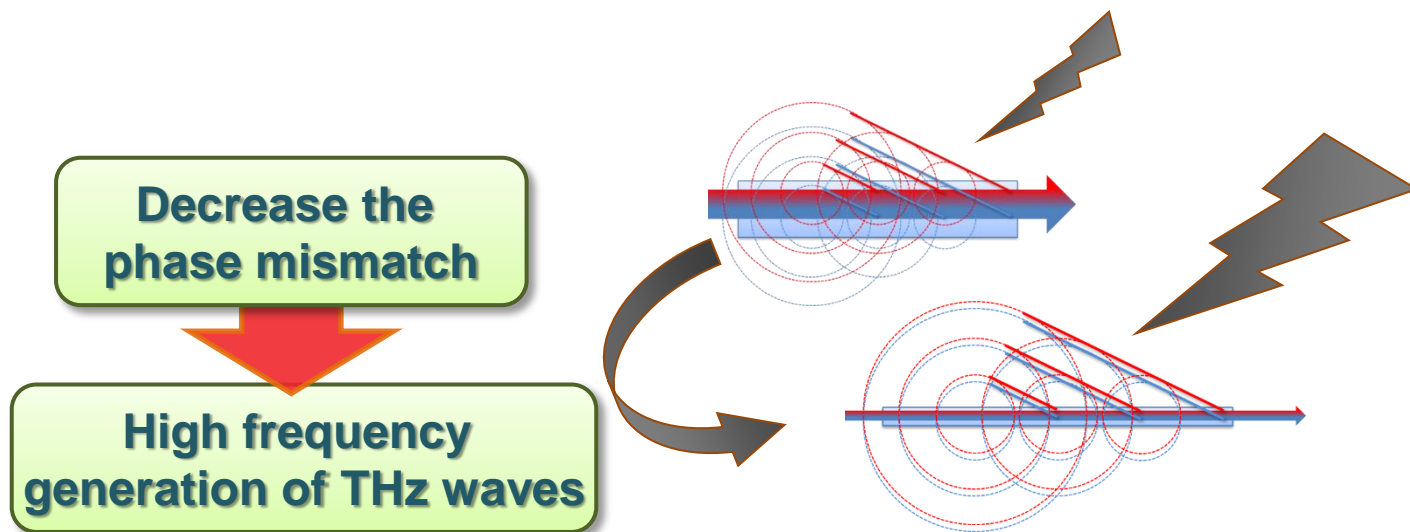
Phase mismatch prevent effective THz wave generation in *high frequency area*

# 位相不整合の解消

There is a limit to reduce the diameter of pump beam

薄い結晶を用いることで解消を狙う

Using waveguide which has thin thickness,  
It is possible to reduce the beam width further



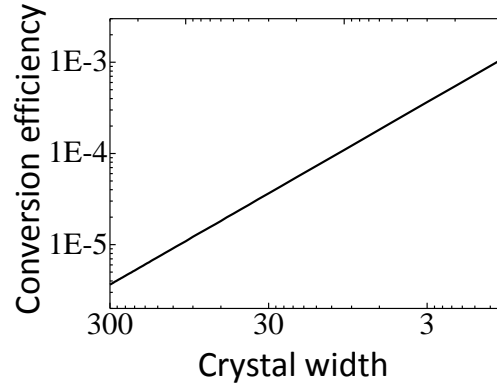
# ビーム径 ( $\approx$ crystal width) と変換効率

Equation of surface generation

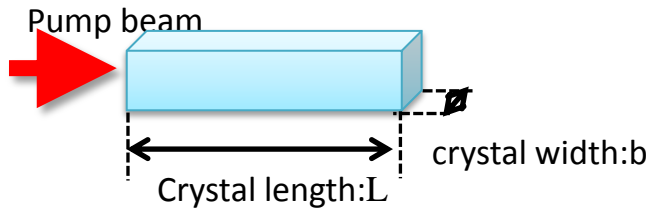
$$P_{3\omega} = \frac{32W_0 d_{33}^2 L P_1 P_2}{n_\Omega N_1 N_2 \lambda^2 b}$$

$W_0$  : Wave impedance  
 $d_{33}$  : Nonlinear optical coefficient  
 $P_1, P_2$  : Pump power  
 $n_\Omega$  : Refractive index at THz region  
 $N_1, N_2$  : Refractive index at near-IR  
 $\lambda$  : Wavelength of THz waves  
 $L$  : Crystal length  
 $b$  : Crystal width

Y.AVETISYAN, et.al, Appl. Phys. B 73, 511-514(2001)



Conversion efficiency and crystal width

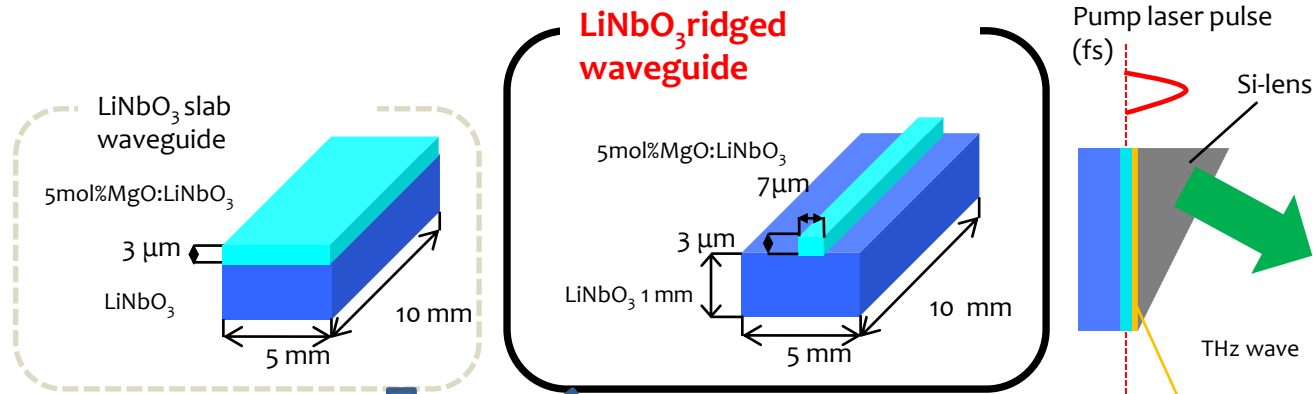


**Using narrow crystal,**

**THz waves generate effectively**

**Ridge crystal**

# リッジ導波路

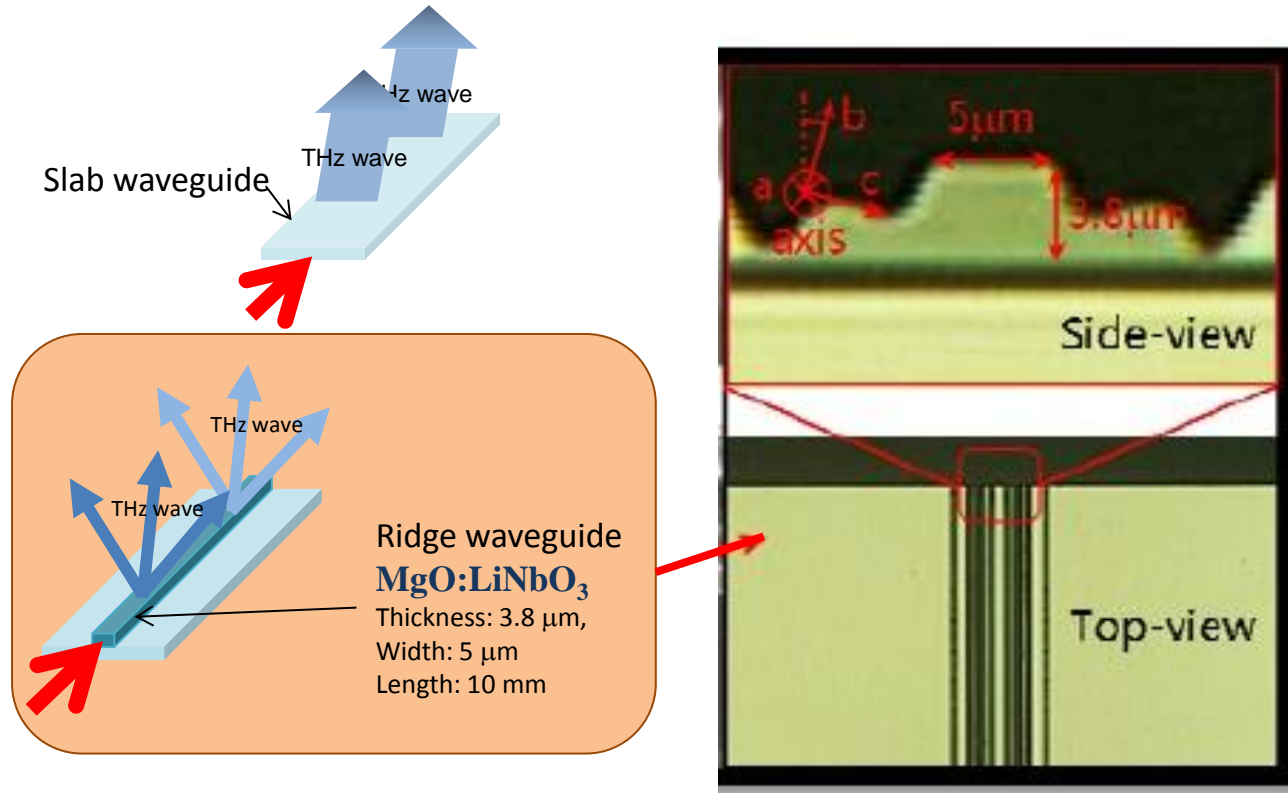


Advantages of waveguide

- Suppress of phase mismatch
- Long interaction length
- Decrease an absorption by crystal
- High power density of pump
- High efficiency

LiNbO<sub>3</sub> ridge waveguide as THz emitter

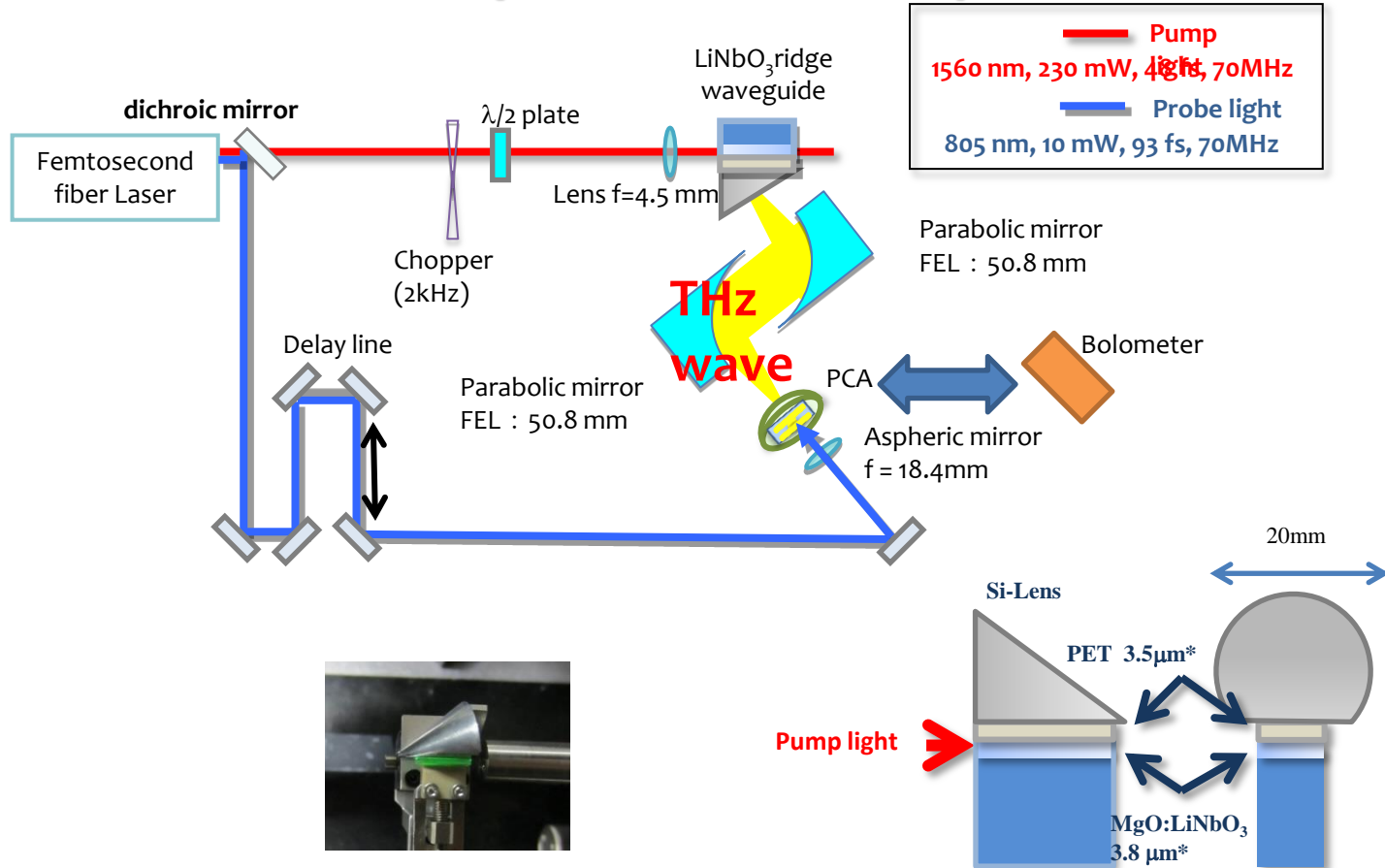
# リッジ導波路



Ridge waveguide has higher efficiency, lower absorption

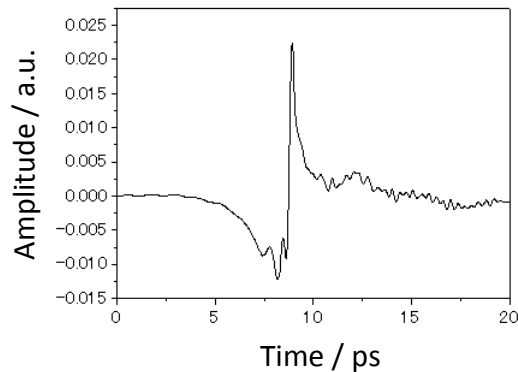


# Experimental setup

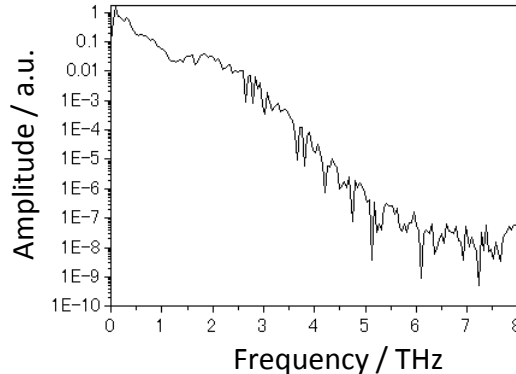


# リッジ導波路からのテラヘルツ波出力

THz output from ridge LiNbO<sub>3</sub> waveguide via time domain measurement



Time domain  
spectrum



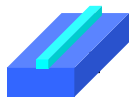
Frequency spectrum

## Condition

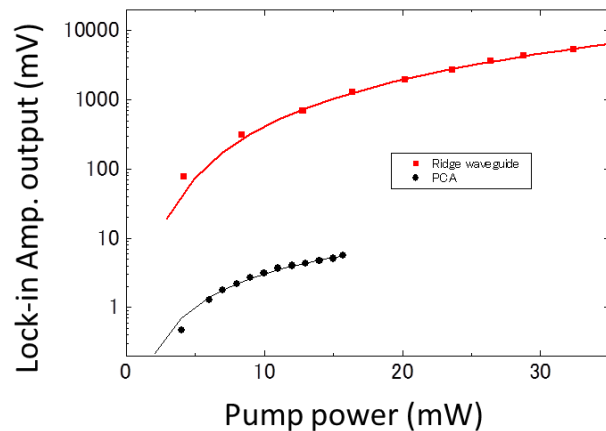
- time constant : 3 s
- step : 3  $\mu\text{m}$
- data points : 4096 points
- average : 1 times

**Dynamic range is over 80 dB,  
frequency spectrum is up to 7 THz**

# テラヘルツ波出力

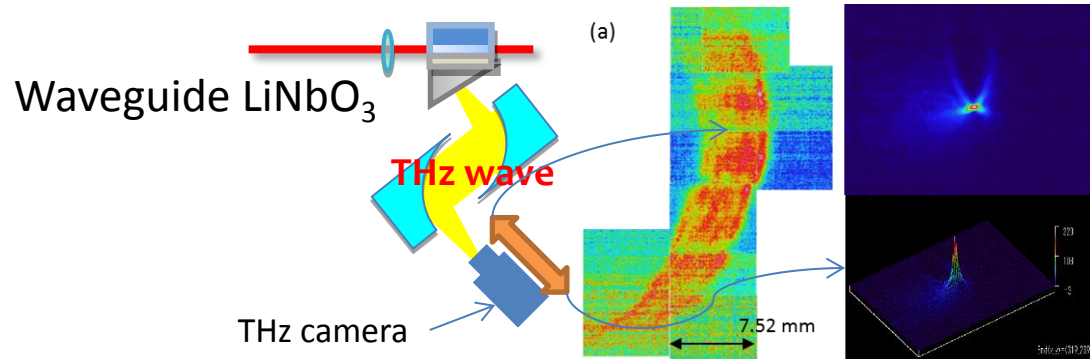
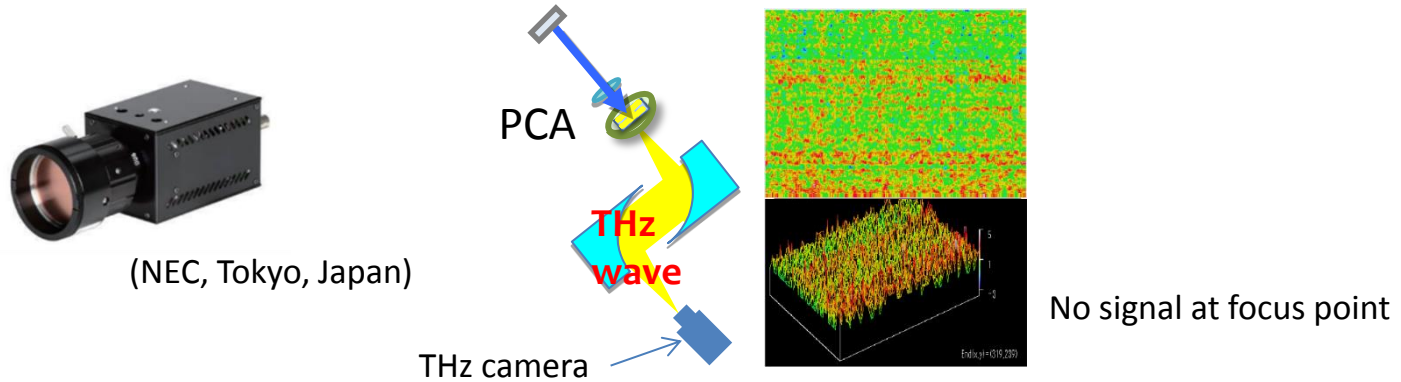


LiNbO <sub>3</sub> waveguide		PCA		
Power	Output Power by Si bolometer	Power	Output Power by Si bolometer	
32.4 mW	5400 mV	15.7 mW	5.59 mV	966 times



THz output power from waveguide (red) and PCA (black) against pump power. The power was measured by a Si bolometer.

# テラヘルツカメラを使用した空間分布計測

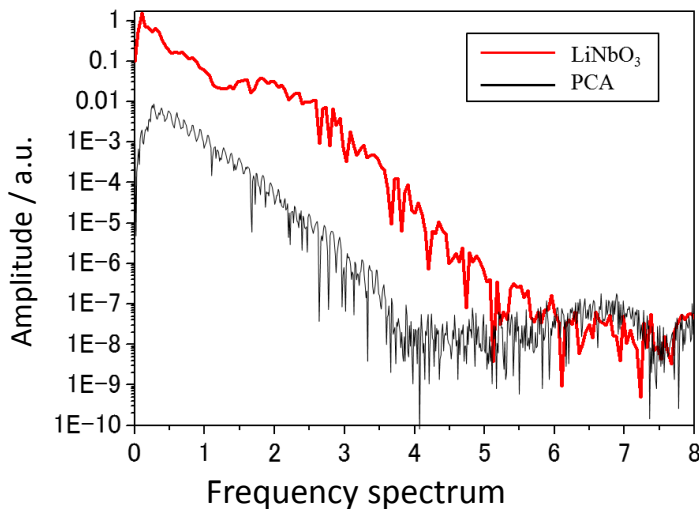


Signal is observable even at not focus point

# 出力比較



LiNbO <sub>3</sub> waveguide		PCA		
Power	Output Power by Si bolometer	Power	Output Power by Si bolometer	
32.4 mW	5400 mV	15.7 mW	5.59 mV	966 times



**Output from LiNbO<sub>3</sub> waveguide is 1000 times larger than that from PCA Further, DR is over 80 dB, frequency spectrum to 7 THz**

# 無機結晶ではなく、有機結晶を用いた場合は？

ここまでの結果は無機結晶 $\text{LiNbO}_3$ を用いた結果。  
さらに高性能な有機結晶を用いるとどうなるか。

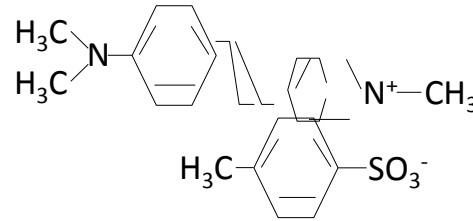
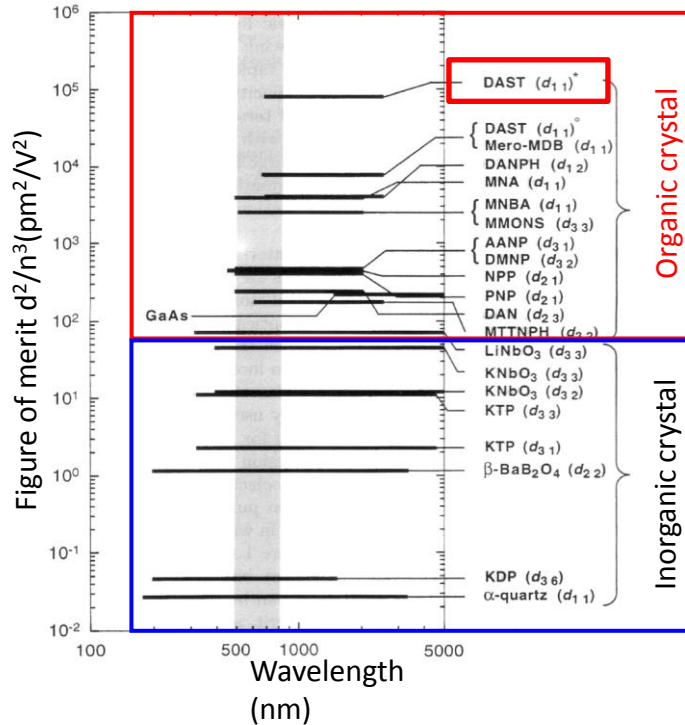
一般的に有機結晶は非線形光学性能 $d$ が無機結晶より100倍ほど大きい。ここで波長変換効率は $d$ の二乗に比例

チェレンコフ位相整合方式は結晶の選択性に制限が小さい

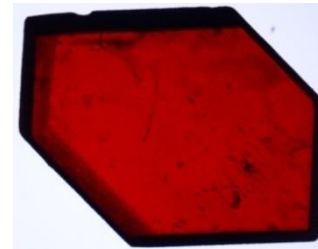
**有機非線形光学結晶 (bulk DAST and OH1, OH1 thin film)を用いてテラヘルツ波発生を行った。**

# 有機非線形光学結晶: DAST

Figure of merit of NLO crystals



Structure formula of DAST(4-dimethylamino-N-methyl-4-stilbazolium tosylate)

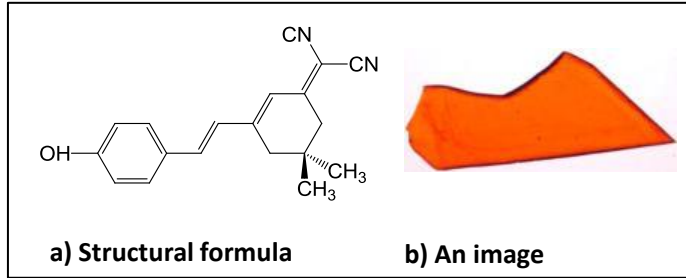


DAST crystal

DAST crystal has highest D constant  
(However DAST crystal has strong absorption)

# 有機非線形光学結晶: OH1

OH1:2-(3-(4-Hydroxystyryl)-5,5-dimethylcyclohex-2-enylidene)malononitrile

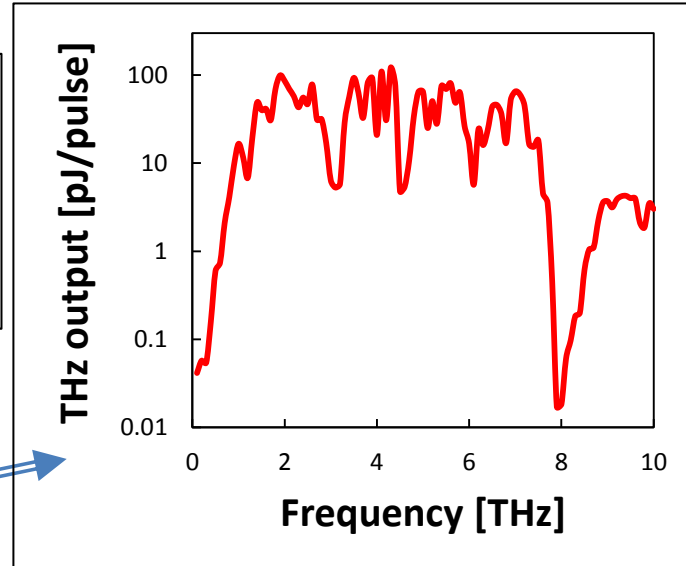


OH1 crystal

Features of OH1 crystal

- High figure of merit (c axis)  
 $d_{33}=120\text{pm/V}@1.9\mu\text{m}$
- 昇華性 (easily change to air)

A possibility to make thin film OH1 crystal using physical vapor deposition .



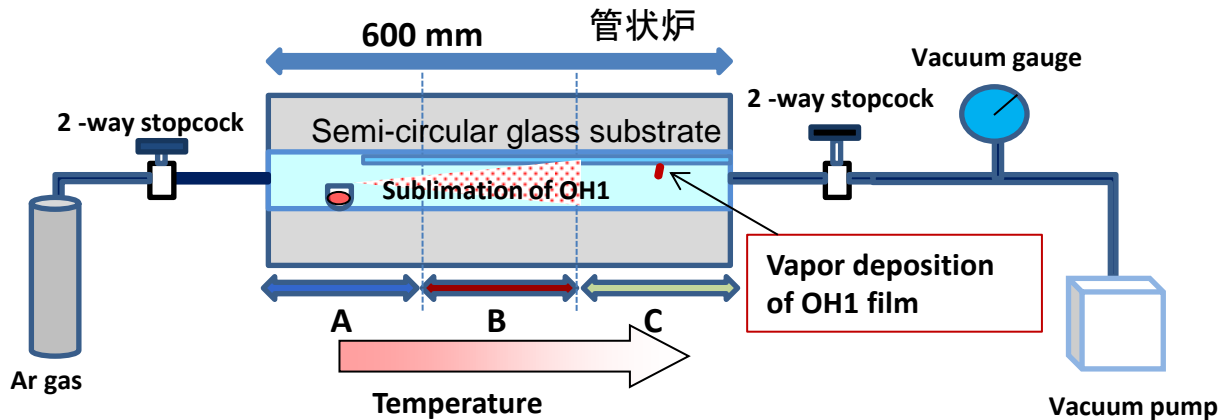
THz generation from OH1 crystal using difference frequency mixing. H. Uchida, et al. *Applied Physics B* 111. 3 (2013): 489-493

OH1 crystal can generate broadband THz waves.

OH1 crystal has a potential of THz emitter as waveguide structure



# 気相成長によるOH1薄膜結晶作成



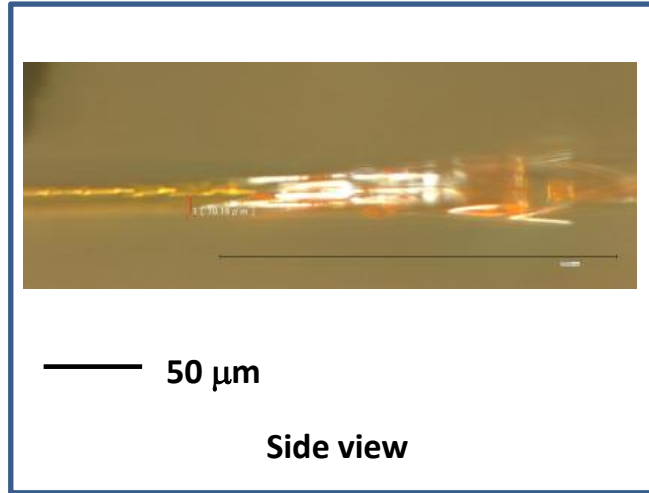
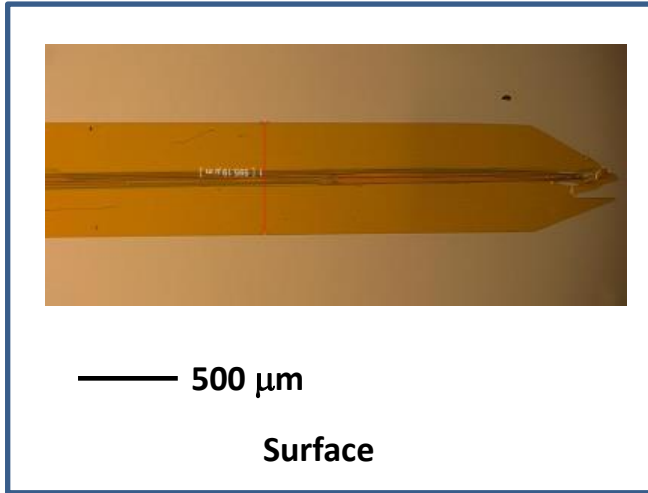
OH1 thin film manufacturing apparatus using sublimation method with a tubular furnace

**Making condition**  
Temperature A : 180 °C  
                  B : 120 °C  
                  C : 90 °C  
Pressure       0.07 MPa (Negative pressure)  
Holding time  24 hours

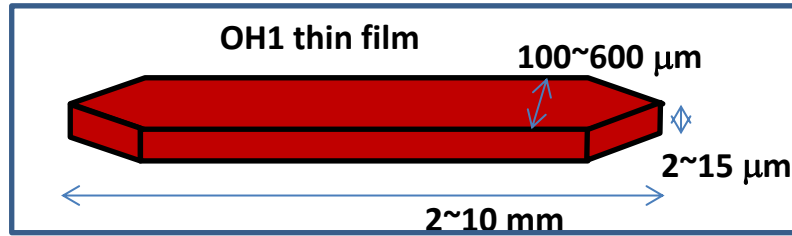


Manufactured OH1 film

# OH1 薄膜



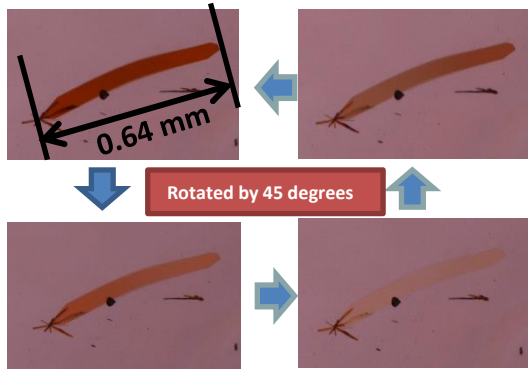
Optical microscope image of OH1 thin film



schematic diagram of manufactured OH1

**An OH1 thin film is made by a simple PVD.**

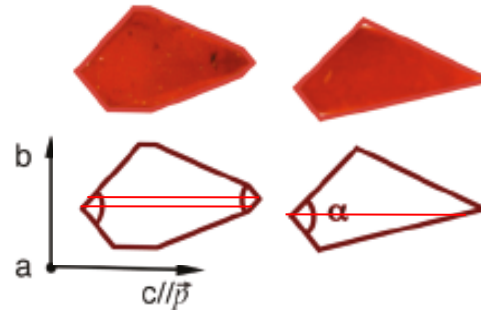
# OH 1 薄膜結晶の評価



An extinction ratio observation using polarized light microscopy

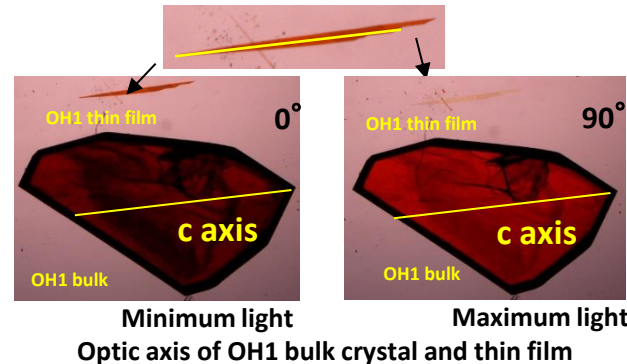
An extinction ratio was regularly changed with a rotation of polarizing plate

A single crystal thin film was grown



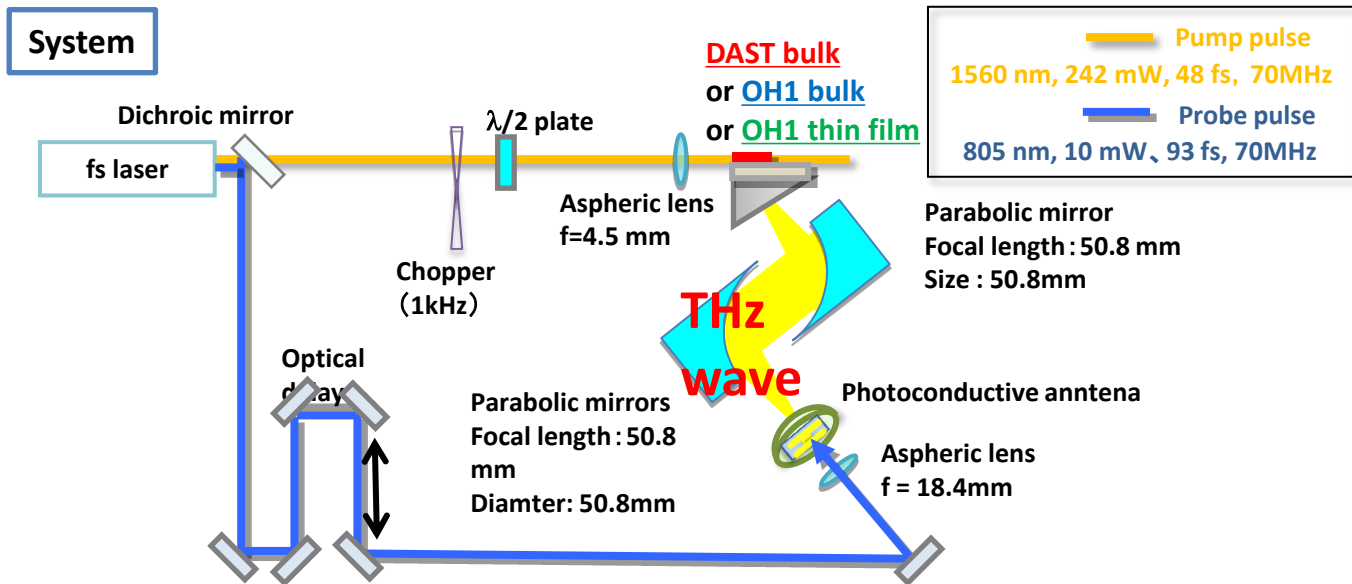
Optic axis and OH1 crystal habit

S. J. Kwon, et al. *Cryst. Growth Des.* 10 (2013): 1552-1558

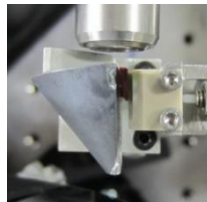


Same extinction change was observed as bulk crystal  
→Crystal habit is same as of bulk

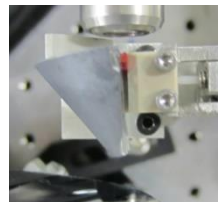
# DAST, OH1結晶からのテラヘルツ波発生



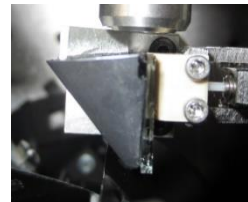
A coupling of crystals to Si lens  
※OH1 is connected to slide glass plate



**DAST bulk**

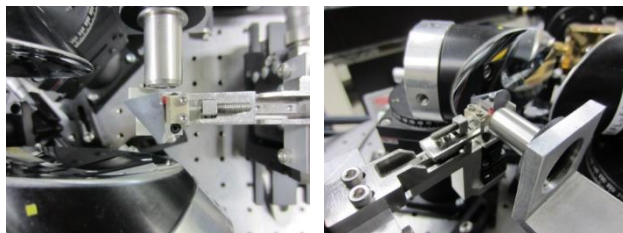


**OH1 bulk**



**OH1 thin film**

# OH1バルク結晶からのテラヘルツ波

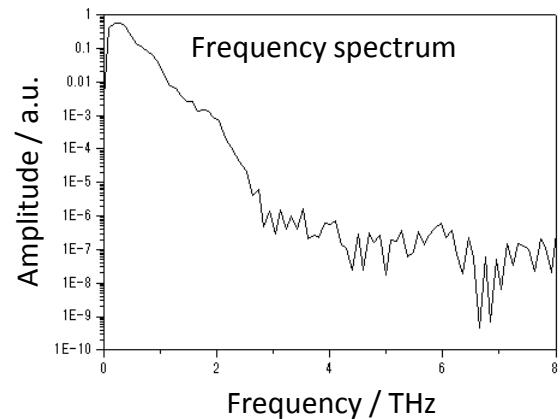
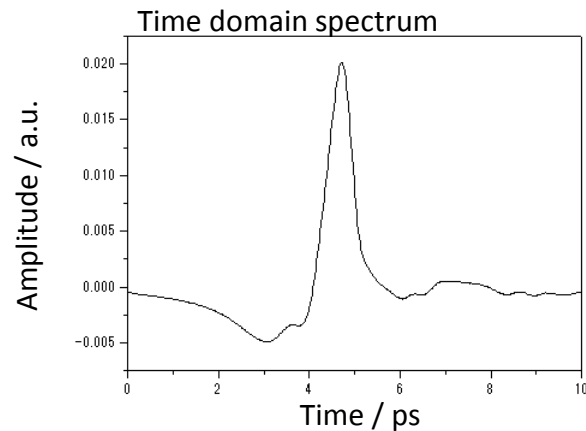


mount of OH1 crystal

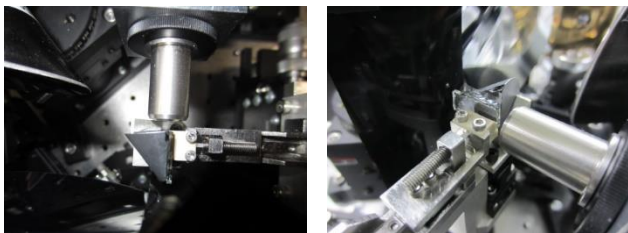


1 mm

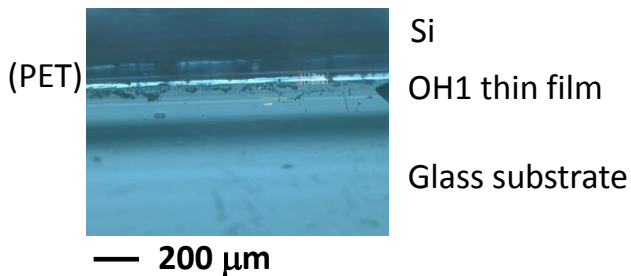
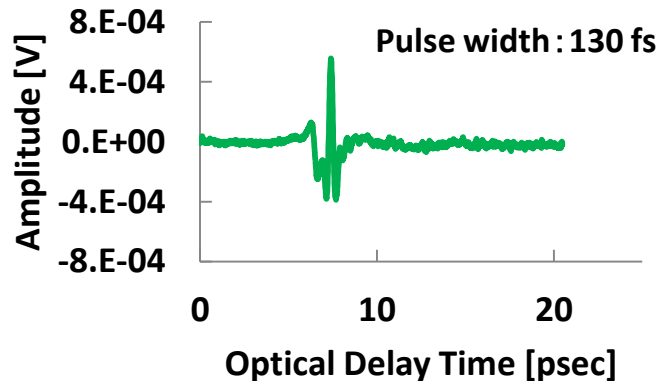
Si lens and OH1 crystal



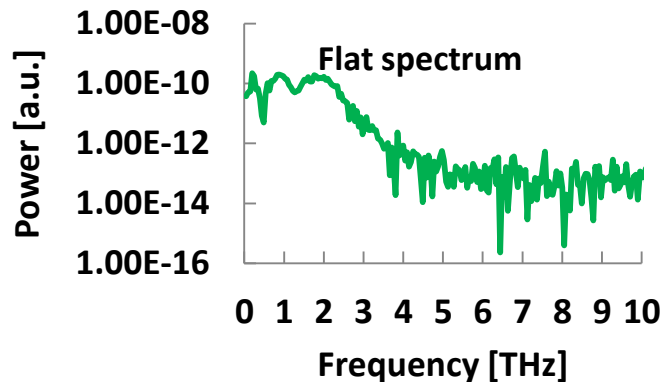
# OH1薄膜結晶からのテラヘルツ波発生



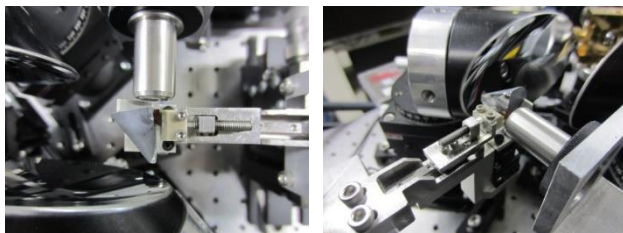
A mount of OH1 thin film



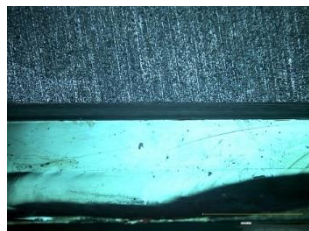
Si lens and OH1



# DAST結晶からのテラヘルツ波発生



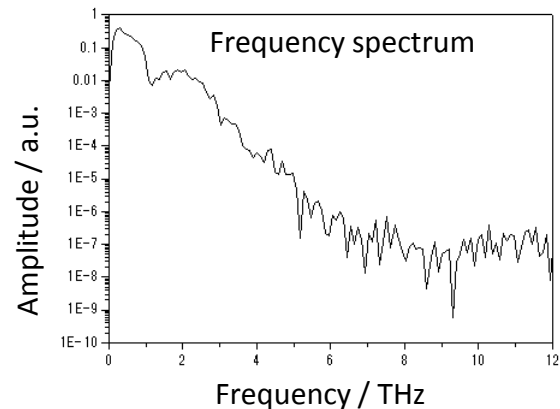
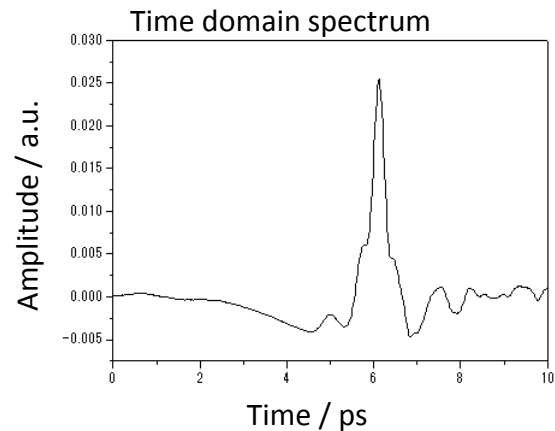
DAST crystal



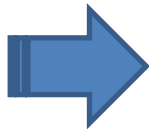
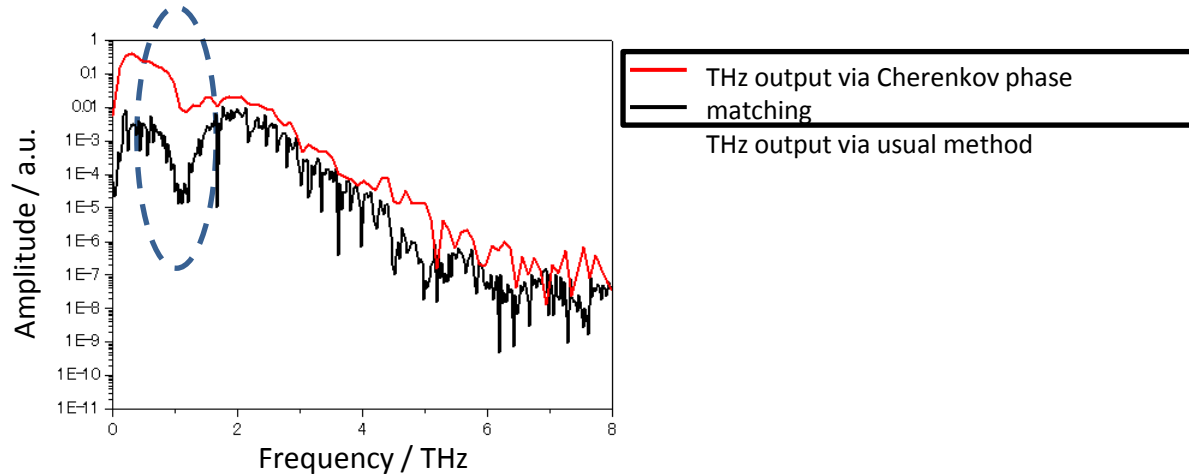
Si  
(PET)  
DAST

1 mm

Side view of DAST crystal and Si lens



# 結晶による吸収の軽減

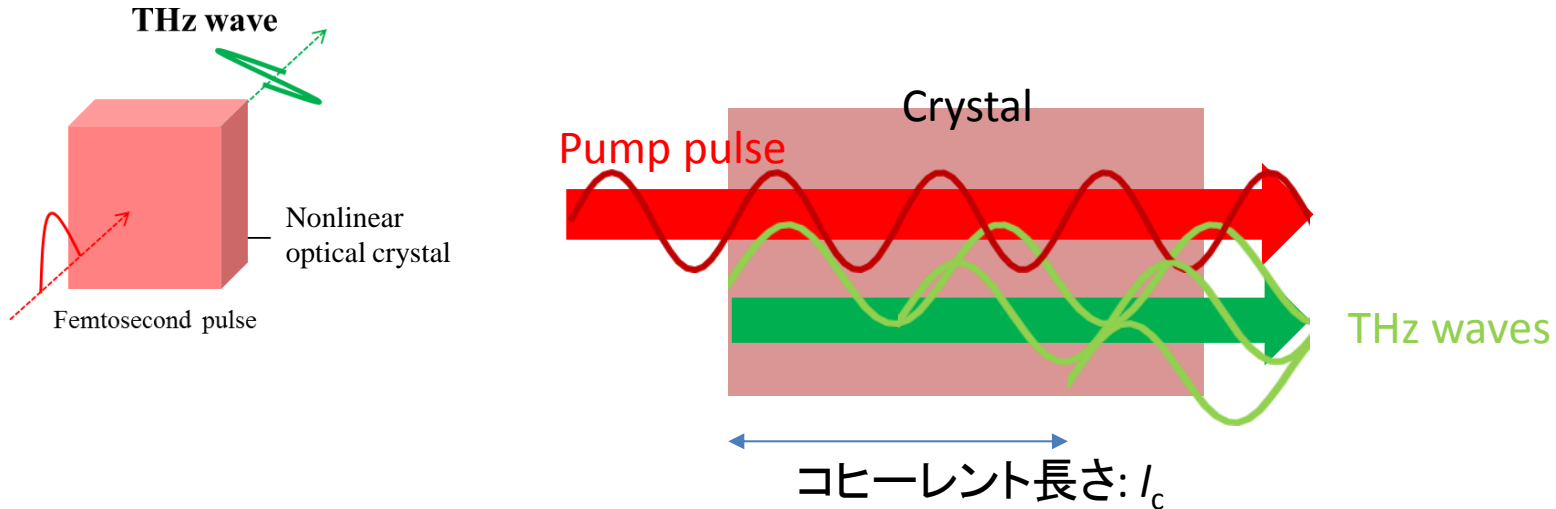


Using Cherenkov phase matching, we achieve cancellation of the absorption around 1.1 THz of DAST crystal



# 励起波長に依存しない テラヘルツ波発生 of 検討

# 共軸発生での位相整合



励起光とTHz光の位相整合により、コヒーレント長さ(使用できる結晶の厚み)が制限される

もし長いコヒーレント長さ(結晶厚み)を利用できれば効率よくTHz波を発生できる

# LiNbO<sub>3</sub> のコヒーレント長さ@共軸発生

Coherent length is determined by

$$l_c = \frac{c}{2 \times \nu \times |n_{THZ}(\nu) - n_g(\lambda)|}$$

$l_c$  : coherent length

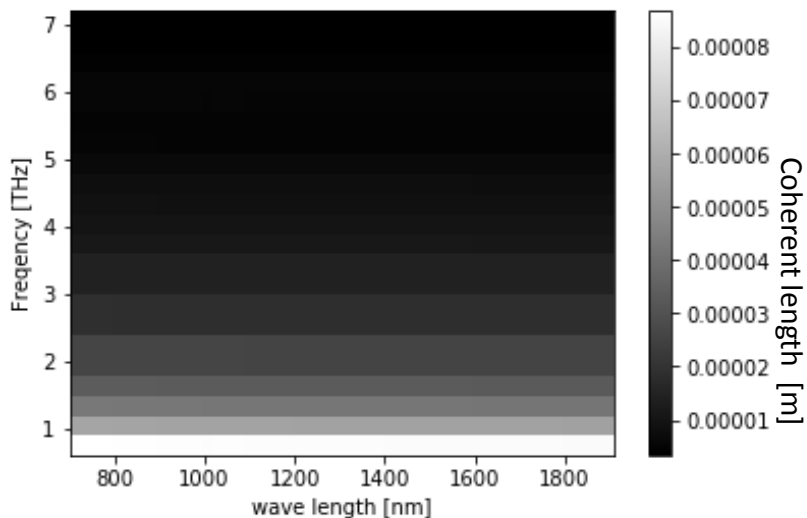
$c$  : light speed

$\nu$  : THz frequency

$n_{THZ}$  : Refractive index of crystal at THz

$n_g$  : Group refractive index of light

$\lambda$  : wavelength



(K. Takeya, *Opt. Lett.*, 2018)

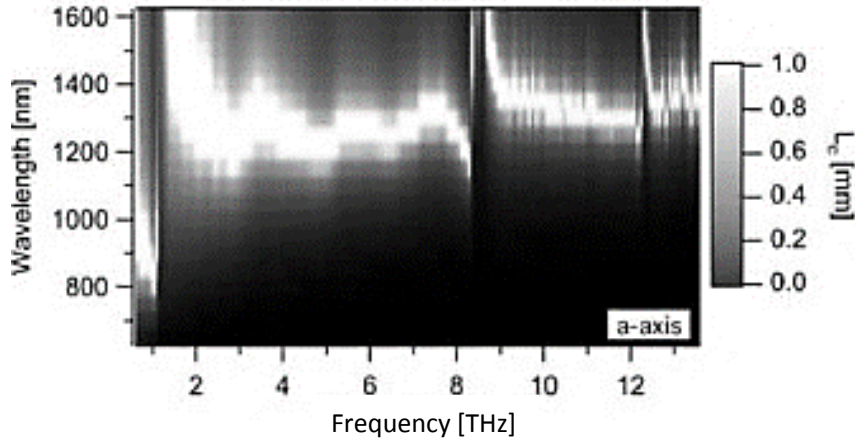
コヒーレント長さ：  
数十  $\mu\text{m}$  @ broad wave length



Collinear phase matching on  
LiNbO<sub>3</sub> is difficult

# DAST のコヒーレント長さ@共軸発生

Coherent length of DAST ( $a$ -axis)



*Paul D. Cunningham, et. al., Optics Express, (2010)*

DAST crystal

- Coherent length :
    - over 1 mm @ 1200~1400 nm
    - Under 0.1 mm @ other wavelength
- to select appropriate wavelength is important.

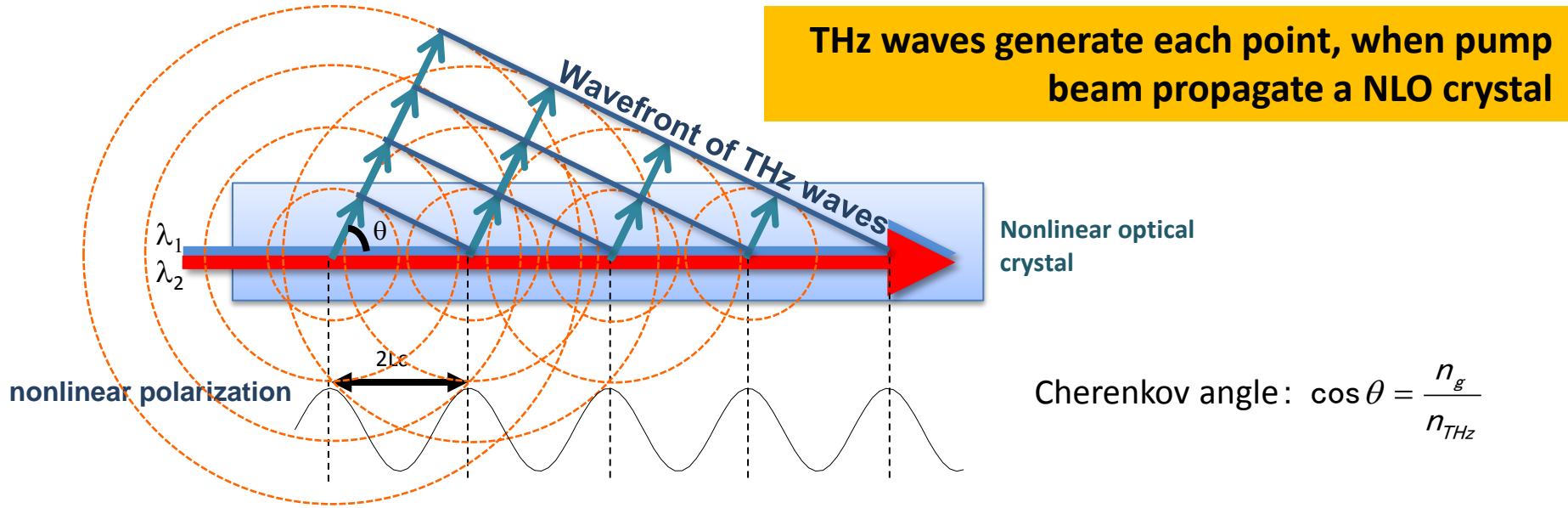
Coherent lengths are generally short

→The selection of the wavelength is problem

→Effective THz generation is not easy in many crystals

If possible, wavelength free THz generation is good

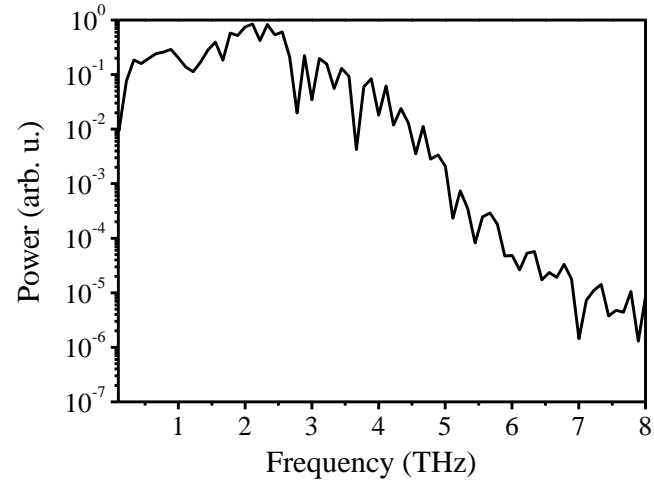
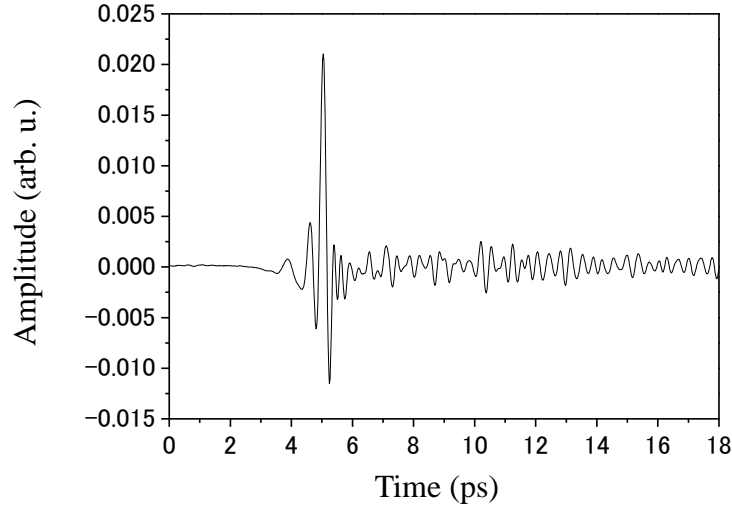
# Cherenkov phase matching



$V_{\text{pump}} > V_{\text{THz}} \Rightarrow$  the Cherenkov phase-matching condition is satisfied.

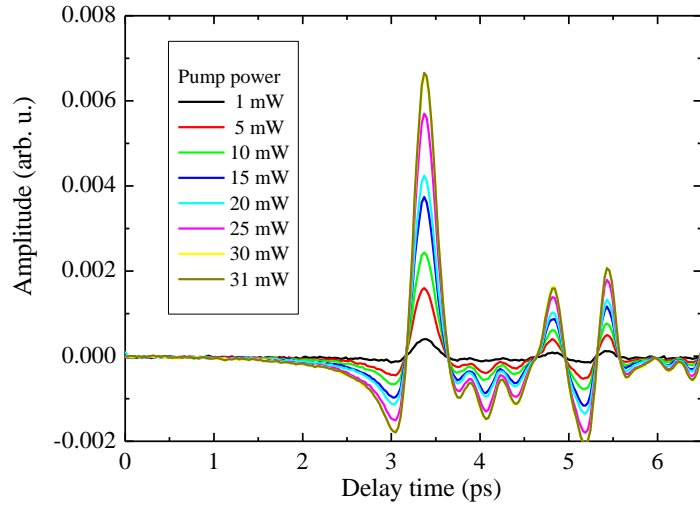
The directions of pump beam and THz waves are different, so that the phase matching is probably determined by only Cherenkov angle.

# Results: pumped by 1560 nm

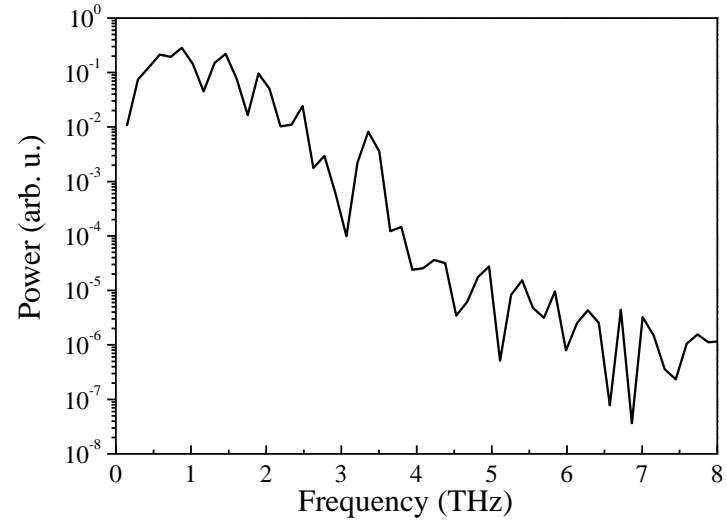


THz output from LiNbO3 crystal with pump wavelength of 1560 nm

# Results: pumped by 800 nm



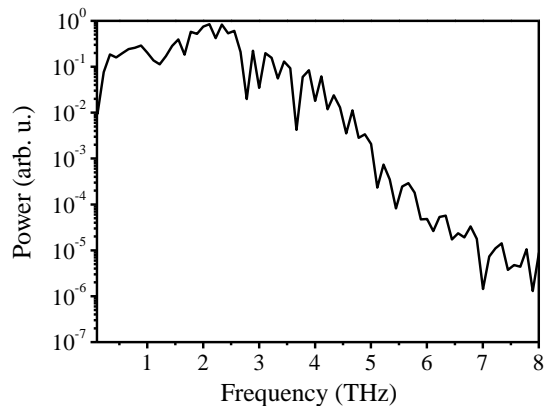
Time domain spectrum



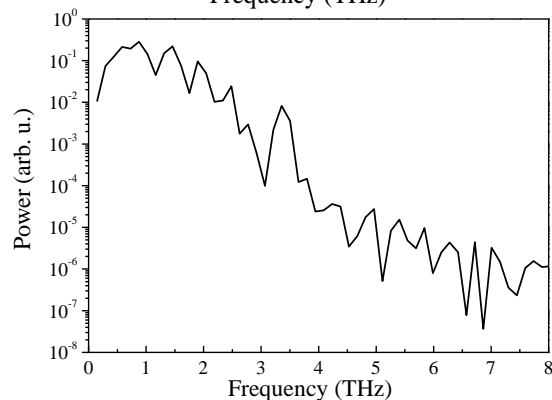
Frequency spectrum

THz output from LiNbO3 crystal with pump wavelength of 800 nm

# 異なる励起波長によるTHz発生と比較



Pumped by 1560 nm (pulse width 65 fs)

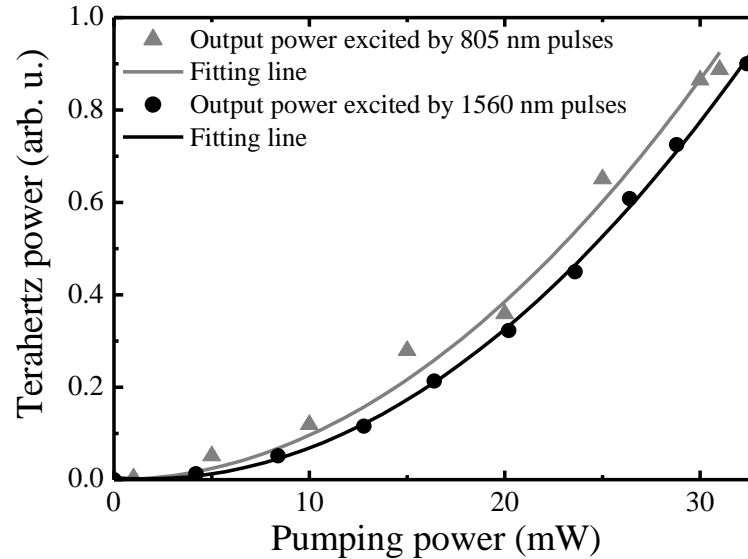


Pumped by 805 nm (pulse width 130 fs)

Broadband THz output were observed at both wavelength.



# Pump power dependence of THz power



(K. Takeya, *Opt. Lett.*, 2018)

Incident light intensity dependence of generated THz waves. The results show pumping at wavelengths of 1560 (gray) and 805 nm (black). Both show that they are proportional to the square of the pump light intensity.

# Conclusion

- チェレンコフ位相整合方式を用いることで、 $\text{LiNbO}_3$  結晶から広帯域、高DRのテラヘルツ波を発生
- 有機結晶DAST、OH1を用いたテラヘルツ波発生が観測できた。
  - DAST結晶からは結晶自体の吸収を減少した広帯域発生を観測。
  - OH1結晶の気相成長による作成
  - OH1結晶からもテラヘルツ波発生
- 励起波長に依存しない広帯域テラヘルツ波発生を観測

# Acknowledgement

- This work was supported by JSPS KAKENHI Grant Number 00296013 and the Japan Science and Technology Agency

**Thank you for your attention**