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Terahertz Laser Sources Based on Defference Frequency Generation of Infrared Nonlinear Optical Materials $SnGa_4Q_7$ (Q = S (SGS), Se (SGSe))

Wen-Dan Cheng

Fujian Institute of Research on the Structure of Matter
Chinese Academy of Sciences
Fuzhou, Fujian 350002, China

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Chen-Sheng Lin (Dr. , Associate Prof.)
Zhong-Zhen Luo (Dr.)
Hao Zhang (Assistant Prof.)
Wen-Long Zhang (Dr. Associate Prof.)
Yi-Zhi Huang (Drs. Associate Prof.)
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Power Points at Presentation

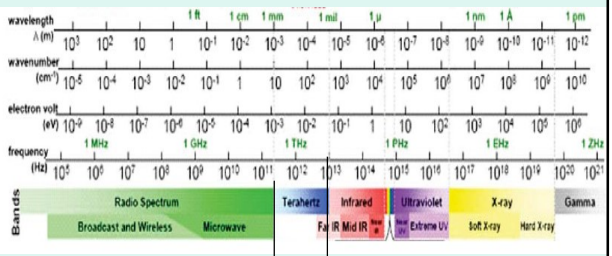
Wen-Dan Cheng, Chinese Academy of Sciences, China

- Simple introductions are about the motivation of this investigation and by the DFG optical process of NLO crystal is the convinient method to generate THz source.
- The evalution performances of THz source, generating from DFG optical process of mid/far Infrared NLO materials $SnGa_4Q_7$ [Q = S (SGS), Se (SGSe)], in view of the theoretical characterizing.
  - The calculated phonon dispersions are employed to determine the infrared transparent cutoff edge. It gives the information of infrared transparance zone.
  - The calculted phonon state densities are used to characterize the contribution of infrared frequency from individual lattice vibration. It gives the clue to inprove or design the wide transparent material in mid/far infrared zone.
  - The calculated electron absorption spectra are employed to determine the UV-visble cutoff edge, and the simmulation lattice vibration spectra are used to calculate the infrared absorption coefficients.

Continue

- The conversion efficiency contributes from the material-self properties, including the cutoff edge of transparent infrared zone, absorption coefficient of THz light, and figure of merit; and from experimental device, including the optical path length, pump light intensity and wavelength of THz wave generation.
- The discussions about findings
  - The the integrate knowledge calculating phonon and photon properties are used to evalute the conversion efficiency of THz source from infrared NLO materials SGS and SGSe.
  - The conversion efficiency of THz source is for SGSe > SGS at the same experimental conditions, which results from a large NLO susceptibility and wide transparent mid/far spectrum of SGSe material.
  - A long length of THz wave will result in a low efficiency for the designed material.
  - A strong intensity of incident light or large size crystal will produce a large THz conversion efficiency of material

☞ Introduction



0.3 THz
1000 μm;10 cm^{-1}

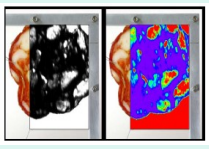
10 THz
30 μm; 800 cm^{-1}

The Chart Copy from C. Corsi, Opto-Electronics Reviews, 2015, 23, 1-23

THz spectral characteristics and Aplications

◆ Transmitted through non conductor and non polar material, such as dry paper, plastic, ceramic, cloth, etc. opaque materials.





◆ Low quantum energy and high ratio between signal and noise
◆It will not have a destructive effect while THz wave incident to the materia, to be beneficial organisms tissue imaging with high fat content

Applications of THz Spectrum





◆to detect and identify the weapons and explosives concealed underneath clothing or packaging

◆to use for biomedical diagnosis in label free genetic analysis and cell imaging

◆Astronomical Observation



As we move from the near-infrared into mid and far-infrared regions of the spectrum, some celestial objects will appear while others will disappear from view

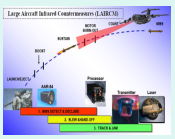
◆Damage Inspections of Industrial Products and Building Structures



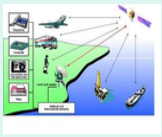
◆Environmental Monitoring and Controls

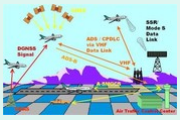


◆Remote and Star-Star Communications, Military Space Engineering of Military, etc.













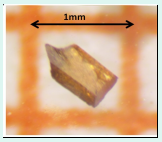
◆The Biggest Bottleneck in THz Developments and Applications

➢for widespread implementation:
The lack THz Source with a high power, low cost, portable, wide wavelength coverage and can work at room temperature;
➢ no device thus far has appeared to have an optimal mix of above properties

◆ Convenient Way:
to generate THz source from the mixing light of two laser frequencies by DFG process using NLO crystal

Requirements:
high conversion efficiency, a large figure of merit (FOM), width infrared transparency, and a large threshold for infrared NLO crystals.

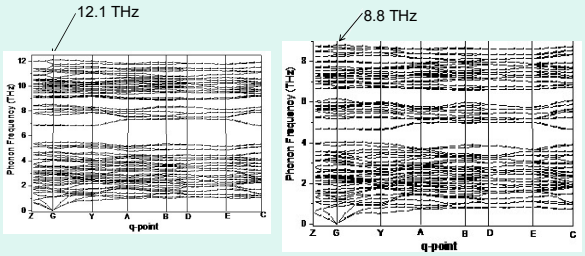
☞ Theoretical characterizing of Optical-Physical Processes





At present, the obtained single crystal size is not suitable for experimental measurements of optical parameters; we can only calculate the physical parameters of various optical processes based on the single crystal structure data.

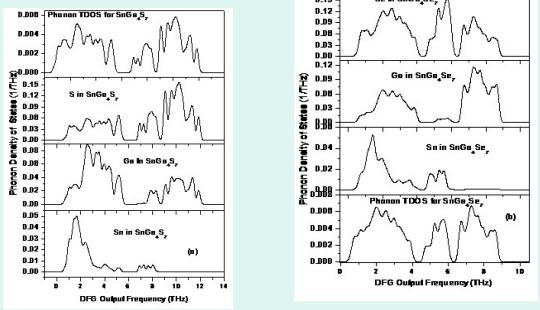
◆Phonon dispersion spectra of SGS and SGSe crystals



➢Phonon dispersion curves give the information of the cutoff edge of infrared transparence

➢The absorption of SGS and SGSe appear at phonon frequency of less than 12.1 THz for SGS and less than 8.8 THz for SGSe.

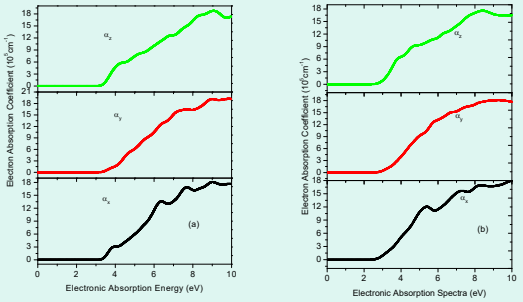
◆Phonon density of states



They give the evidence of lattice vibration contributions at specially designated frequency

The contributions of the lowest frequency band can be assigned as Sn lattice vibrations

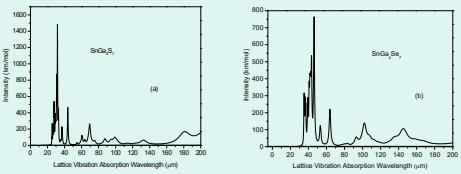
◆ Electronic absorption Spectra of SGS and SGSe



➢ $\alpha=4\pi\chi_2^{(1)}\omega/c$, in which the ω = eV/$\hbar$ and $\chi_2^{(1)}$ is determined by the imaginary part of dielectric function of $\varepsilon(\omega)$.

➢ the cutoff edge of ultraviolet-visible light is at about 3.0 eV and 2.6 eV for the SGS and SGSe crystals, respectively.

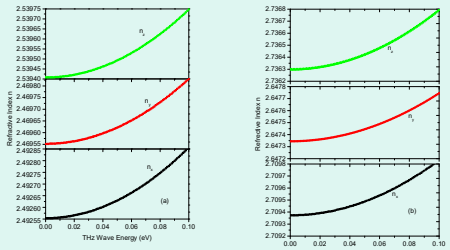
◆Lattice vibration spectra of SGS and SGSe



In terms of the relations between the absorption coefficient α and infrared intensity I, $\alpha(\omega_i) \approx I(\omega_i)C/(10\omega_i)$

the absorption coefficients α of 8.11 cm^{-1} at wavelength 51 μm of Fig (a);
To obtain α of 12.30 cm^{-1} at wavelength of 73 μm of Fig (b)

◆Refractive index and DFG susceptibility in THz zone



➢The refractive index dispersion curves of SGS and SGSe crystals are plotted in these figures localized in the THz wave 0.001 - 0.1 eV (0.24 – 24.2 THz)

➢The average refractive index is larger for SGSe (2.7) than for SGS (2.5)

◆**The second-order susceptibility of DFG process**

$$\chi^{(2)}_{abc}(-\omega_1-\omega_2;\omega_1,\omega_2) = -\sum_{i,j,l}\frac{ie^3}{m^3\omega_1\omega_2(\omega_1+\omega_2)}G^{(2)}_{ijl}(\omega_1,\omega_2)p^a_{ij}p^b_{jl}p^c_{li}$$

$$G^{(2)}_{ij}(\omega_1,\omega_2) = \frac{1}{E_1+E_2-E_{jl}}\left(\frac{f_{il}}{E_2-E_{li}}+\frac{f_{jl}}{E_1-E_{jl}}\right)$$

the contributions of virtual-electron (V-E) to imagine part of susceptibility

$$\chi^{(2,f)}_{abc}(-\omega_3;\omega_1,-\omega_2) = \frac{2e^3\hbar^3}{m^3\Omega}\sum_{i,j,l,K}\frac{\eta\delta(E_3-E_{jl}(K))}{(E_2+E_{li}(K))E_2E_{jl}(K)}\left[\frac{1}{E_2E_{jl}(K)}+\frac{2}{(E_{jl}(K)+E_2)^2}\right]P_{abc}(p^a_{ij}(K)p^b_{jl}(K)p^c_{li}(K))$$

$$P_{abc}(p^a_{ij}(K)p^b_{jl}(K)p^c_{li}(K)) = 0.5P_{bc}(p^a_{ij}(K)p^b_{jl}(K)p^c_{li}(K)+p^a_{jl}(K)p^b_{li}(K)p^c_{ij}(K)+p^a_{li}(K)p^b_{ij}(K)p^c_{jl}(K))$$

the contribution of V-H to imagine part of DFG process

$$\chi^{(2,II)}_{abc}(-\omega_3;\omega_1,-\omega_2)=\frac{2e^3\hbar^3}{m^3\Omega}\sum_{i,j,l,K}\frac{\eta}{|E_{ji}(K)-(E_{ji}(K)-E_2)|}[\frac{\delta(E_3-E_{ji}(K))}{E_2E_{ji}(K)}(\frac{1}{E_2E_{ji}(K)}+\frac{2}{(E_{ji}(K)+E_2)^2})+\frac{\delta(E_3-(E_{ji}(K)-E_2))}{E_2(E_{ji}(K)-E_2)}(\frac{1}{E_2(E_{ji}(K)-E_2)}+\frac{2}{E_{ji}^2(K)})]P_{abc}(p^a_{li}(K)p^b_{ij}(K)p^c_{jl}(K))$$

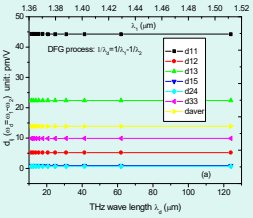
$$P_{abc}(p^a_{li}(K)p^b_{ij}(K)p^c_{jl}(K))=0.5P_{bc}(p^a_{li}(K)p^b_{ij}(K)p^c_{jl}(K)+p^a_{ij}(K)p^b_{jl}(K)p^c_{li}(K)+p^a_{jl}(K)p^b_{li}(K)p^c_{ij}(K))$$

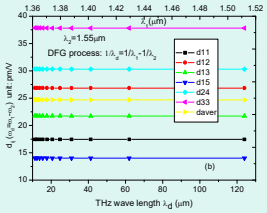
Imagine part $\chi^{(2)} = \chi^{(2,I)} + \chi^{(2,II)}$

The r$\chi^{(2)}$ is obtained by Kramers-Kronig transform from the i$\chi^{(2)}$

Total $\chi^{(2)} = [(r\chi^{(2)})^2+|i\chi^{(2)}|^2]^{1/2}$, and the NLO parameter $2d = \chi^{(2)}$

SGS and SGSe crystals belong to class m, and have ten nonvanishing tensors (d_{11}, d_{12}, d_{13}, d_{15}, d_{24}, d_{26}, d_{31},d_{32}, d_{33}, and d_{35}), only six independent tensors (d_{11}, d_{12}, d_{13}, d_{15}, d_{24}, and d_{33})





The figures give the plots of these six components and their average value within the λ_3 wavelengths from 10 to 120 μm for the SGS and SGSe crystals

◆Conversion efficiency of THz source

DFG

$$\eta_d=\eta_d^0\exp[-(\alpha_{p1}+\alpha_{p2}+\alpha_d)L/2]\frac{\sin^2(\Delta kL/2)+\sinh^2[(\alpha_{p1}+\alpha_{p2}-\alpha_d)L/4]}{(\Delta kL/2)^2+[(\alpha_{p1}+\alpha_{p2}-\alpha_d)L/4]^2}$$

$$\eta_d^0=\frac{8\pi^2d_{eff}^2L^2I_{p1}}{\varepsilon_0 n_{p1}n_{p2}n_d c\lambda_d^2}\quad(SI)\qquad \eta_d^0=\frac{512\pi^5d_{eff}^2L^2I_{p1}}{n_{p1}n_{p2}n_d c\lambda_d^2}\quad(cgs)$$

R. L. Rutherland, *Handbook of Nonlinear Optics* Second Edit.,, 2003, *Chapter* **2**, pp 33-100.

The conversion efficiency η_d is divided into two contributions:

- one depends on the properties of material-self, $\eta_d \propto d_{eff}^2 e^{\alpha}\sin(\Delta k)/n^3$
- The other one depends on the conditions of experiment, $\eta_d \propto L^2 I_p/(c\lambda_d^2)$

Table 1. The calculated parameters and conversion efficiency for studied crystals

Output	WL	Abs.	Int.	size	R(n$_x$n$_y$n$_z$)	d_{eff}	Conv.
$SnGa_4S_7$							
	25	trans.	40	0.5	15.6327	13.9018	5.891
	34	5.157	40	0.5	15.6320	13.9012	0.937
	80	4.391	40	0.5	15.6314	13.9007	0.212
	80	4.391	160	0.5	15.6314	13.9007	0.848
$SnGa_4Se_7$							
	25	trans.	40	0.5	19.6289	24.7002	14.813
	34	trans.	40	0.5	19.6277	24.6974	8.007
	34	trans.	40	0.8	19.6277	24.6974	20.501
	80	3.617	40	0.5	19.6267	24.6952	0.6265

Units: WL(μm); Abs (cm^{-1}); Int (MW/cm^2); size (cm); d_{eff} (pm/V); Conv(x 10^{-5})

$d_{eff} = d_{aver}$

It can conclude several rules:

1)a large NLO coefficient will obtain a large efficiency for different materials under the same experimental conditions;

2) wide infrared transparency will result in large conversion efficiency and linear absorption will reduces the THz conversion efficiency for the different materials;

3) a long wavelength of THz generation will result in a low efficiency for the designed material;

4) a strong intensity of incident light (limited in damage threshold of material) or large optical path (limited in crystal size) will produce a large THz conversion efficiency of material

5) At the same experimental conditions, THz conversion efficiency SGSe > SGS

☞ Findings and Conclusions

- a new way to calculate NLO coefficients based on DFG process
- the relation between the infrared absorption coefficient and infrared spectrum intensity $\alpha(\omega_i) = I_i \cdot C/(10\omega_i)$
- The cutoff edge of infrared transparent zone is determined by the calculations of phonon frequency dispersion
- The calculated phonon density of states gives the contributions of the individual phonon frequency
- The linear refractive indices and NLO parameters are used to determine the figure of merit
- The conversion efficiency of THz light is relative with the figure of merit, cut-off edge of transparent infrared spectrum, absorption coefficient of THz light
- SGSe crystal appears with a large THz light conversion efficiency, which results from a large NLO susceptibility and wide transparent mid/far spectrum
- The evaluation of integrate knowledge from characterizing the phonon, photon and electron properties, to design THz source we should choose more wide transparent material at mid/far infrared zone or avoid the terahertz absorption band of material, and choose a material with a large FOM value.