Terahertz Laser Sources Based on Difference Frequency Generation of Infrared Nonlinear Optical Materials SnGa_4Q (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.)
Yi Yang (Dr.)

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Introduction
THz spectral characteristics and Applications

- 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
- 0.3 THz
- 1000μm; 10 cm⁻¹
- 10 THz
- 30μm; 800 cm⁻¹

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 integrated knowledge calculating phonon and photon properties are used to evaluate 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.
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

- Industrial Maintenance

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 of 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 transparency
- 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.

Electronic absorption Spectra of SGS and SGSe

- $\alpha = \log_{10}I/I_0$, in which the $I$ = eV/h and $\log_{10}$ is determined by the imaginary part of dielectric function of $\epsilon(\omega)$.
- The absorption of SGS and SGSe appear at phonon frequency of less than 12.1 THz and infrared of about 3.0 eV and 2.6 eV for SGS and SGSe, respectively.

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

$\chi^{(2)}(\omega_1 - \omega_2, \omega_1, \omega_2) = \sum_{\nu} \frac{\alpha^2}{\epsilon} \frac{\rho_2^2}{\epsilon_2} G^{(2)}(\omega_1, \omega_2)$

$G^{(2)}(\omega_1, \omega_2) = \frac{1}{\epsilon_2 - \epsilon_1} \frac{1}{\epsilon_2 - \epsilon_1}$

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

$P_{\mu}(\rho_1(\mathbf{K})\rho_\nu(\mathbf{K})\rho(\mathbf{K})) = 0.1 \rho_1(\mathbf{K})\rho_\nu(\mathbf{K})\rho(\mathbf{K})$
the contribution of V-H to imagine part of DFG process

\[ n_{\text{eff}} = \frac{\gamma_2}{\gamma_1} \sum_{\text{V-H}} \frac{1}{E_{\text{p}}(K) - E_{\text{i}}(K) - E_{\text{f}}(K)} \frac{1}{E_{\text{p}}(K) - E_{\text{i}}(K) + E_{\text{f}}(K)} \]

\[ \psi_{\text{imag}}(K) = \psi_{\text{imag}}(K) \psi_{\text{imag}}(K) + \psi_{\text{imag}}(K) \psi_{\text{imag}}(K) \]

Imaginary part \( \chi^{(2)} = \chi^{(2)} + \chi^{(2)} \)

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

Total \( \chi^{(2)} = \chi^{(2)} + \chi^{(2)} \), and the NLO parameter \( 2d = \chi^{(2)} \)

Conversion efficiency of THz source

\[ \eta_{\text{eff}} = \frac{\gamma_2}{\gamma_1} \sum_{\text{V-H}} \frac{1}{E_{\text{p}}(K) - E_{\text{i}}(K) - E_{\text{f}}(K)} \frac{1}{E_{\text{p}}(K) - E_{\text{i}}(K) + E_{\text{f}}(K)} \]


The conversion efficiency \( \eta_{\text{eff}} \) is divided into two contributions:

- one depends on the properties of material-self, \( \eta_{\text{eff}} = \frac{d_{\text{eff}}}{d_{\text{eff}}} \varphi(\Delta k) \rho^2 \)
- the other one depends on the conditions of experiment, \( \eta_{\text{eff}} = L^2 \rho / (\Delta k) \rho^2 \)

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 SGS > 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 \( \alpha(\lambda) = |\lambda/(100\lambda)| \)
- 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 to the figure of merit, cutoff edge of transparent zone is determined by the calculations of material.
- SGS 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/infrared zone avoid the terahertz absorption band of material, and choose a material with a large FOM value.