Applied Physics Letters

A monochromatic and high-power terahertz source tunable in the ranges of 2.7–38.4 and 58.2–3540 μm for variety of potential applications

Wei Shi and Yujie J. Ding

Citation: Appl. Phys. Lett. **84**, 1635 (2004); doi: 10.1063/1.1649802
View online: http://dx.doi.org/10.1063/1.1649802
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v84/i10
Published by the American Institute of Physics.

APPLIED PHYSICS LETTERS VOLUME 84, NUMBER 10 8 MARCH 2004

A monochromatic and high-power terahertz source tunable in the ranges of 2.7–38.4 and 58.2–3540 μm for variety of potential applications

Wei Shi and Yujie J. Ding[a)]
Department of Electrical and Computer Engineering, Lehigh University, Bethlehem, Pennsylvania 18015

(Received 16 September 2003; accepted 22 December 2003)

Based on phase-matched collinear difference-frequency generation in a single GaSe crystal, continuously tunable and coherent radiation in the extremely wide ranges of 2.7–38.4 and 58.2–3540 μm has been achieved. This unique source has the additional advantages of high coherence (narrow linewidth) and simple alignment. The peak output power for the terahertz radiation reaches 209 W at the wavelength of 196 μm (1.53 THz), which corresponds to a power conversion efficiency of 0.055%. Moreover, the terahertz transmission spectra on DNA macromolecules and protein were directly measured, demonstrating some potential and important applications of this terahertz source. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1649802]

A tunable monochromatic terahertz (THz) source is the key to biomedical diagnostics, THz spectroscopy, and chemical identification.[1,2] It can be used to directly probe a DNA binding state[2] and to detect skin cancer. Furthermore, it can be used to study gaseous molecules and biological macromolecules.[3] Label-free genetic diagnostics[1] and detection of explosives also require a THz source to be tuned in a large range. In all these examples a wide tuning range has an advantage of directly probing characteristic resonances in the THz domain. Besides broad tunability a THz source should have advantages of narrow linewidth, simple alignment, stable THz output, and compactness.

Besides free-electron lasers, high-power THz waves were generated based on synchrotron radiation of relativistically accelerated cyclotron electrons.[4] Another attractive approach is based on interminiband transitions of a semiconductor heterostructure.[5(a)] Indeed, at 90 K the output peak power can reach 2.6 mW at 87 μm (3.5 THz).[5(b)] However, tuning in a broad range remains to be greatly challenging for such a scheme. Optical-heterodyne mixing can only produce low output powers.[6] Other promising mechanisms proposed recently[7] have not been implemented yet. Among all the schemes, parametric processes such as difference-frequency generation (DFG) in nonlinear optical (NLO) crystals such as $LiNbO_3$ and DAST are quite promising.[8,9] However, large absorption coefficients of $LiNbO_3$ and DAST in the THz domain result in low efficiencies and limited tunability.[9,10] Among all the NLO crystals GaSe is the most superior for the THz generation. First, it has the lowest absorption coefficients (α) in the THz wavelength region.[11–13] Consequently, GaSe has the largest figure of merit for the THz generation ($d_{\mathrm{eff}}^2/n^3\alpha^2$), which is several orders of magnitude larger than that for bulk $LiNbO_3$ at 300 μm.[11–13] Furthermore, since GaSe has anomalously large birefringence phase matching (PM) can be achieved in an ultrabroad wavelength range. Even though GaSe has potential to reach mirrorless parametric oscillation by just using a single pump beam,[12] DFG offers the advantages of relative compactness, simplicity for tuning, straightforward alignment, much lower pump intensities, and stable THz output powers and wavelengths. Indeed, unlike the parametric oscillation, DFG does not require a complicated alignment procedure even if wavelength tuning is required. Recently, we implemented a THz source tunable in the range of 56.8–1618 μm (0.18–5.27 THz) based on DFG in GaSe.[14] The peak output power reaches 69.3 W at 196 μm, which corresponds to the power conversion efficiency of 0.018%. In our experiment, we have obtained a linewidth of 0.77 μm (6000 MHz). In principle, this linewidth can be reduced further by using the pump beams with much narrower linewidths.

In this letter, we demonstrate that GaSe can be used to produce much wider tuning ranges and higher peak powers. The peak intensity for the Nd:YAG pump beam focused onto the GaSe crystal is about 17 MW/cm^2, which is much lower than that used for achieving the parametric oscillation in $LiNbO_3$.[10] The second pump beam is an idler output beam from an optical-parametric oscillator with the energy per pulse 3–5 mJ. To improve the tuning ranges and peak powers, we used a single GaSe crystal with the length of 20 mm along the optic axis, which is much longer than the previous GaSe crystals. In addition, this new crystal has much better optical and surface quality. Therefore, the linear absorption coefficients in the near-IR region are lower. Furthermore, the damage threshold is higher.

Figure 1(a) shows the external PM angular tuning curves for the type-oee and type-eoo collinear DFG THz radiation. The pump and output wavelengths were verified by an infrared spectrometer and a home-made scanning THz etalon. One can see that the output wavelengths are consistent with those calculated from DFG. For the type-oee PM, the monochromatic output has an extremely wide tuning range of 58.2–3540 μm (0.0848–5.15 THz) has been achieved. This is the widest tuning range ever achieved besides the free-electron lasers. The short-wavelength cutoff for the THz output is due to the presence of a narrow lattice absorption band for GaSe, which peaks at 40 μm.[16] On the other hand, the long-wavelength end is limited by the measurable THz signal since it decreases as the output wavelength increases.[17] By using the type-eoo configuration on the same crystal, the

[a)]Electronic mail: yud2@lehigh.edu

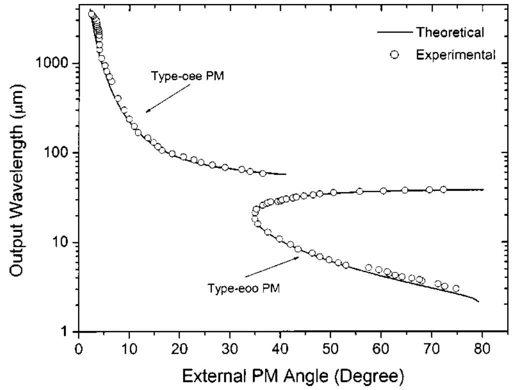

FIG. 1. Type-oe-e and type-eo-o phase-matched output wavelengths vs external phase-matching angle. Dots—data; solid curves—calculation by using phase-matched conditions and refractive index dispersions (Ref. 15).

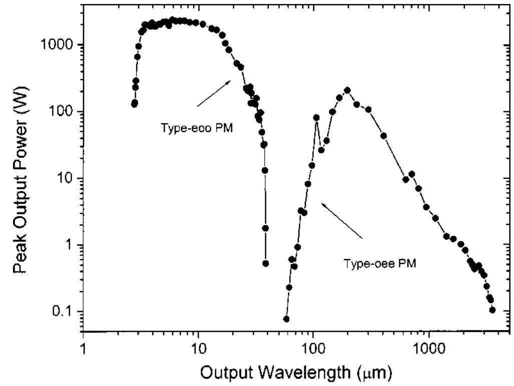

FIG. 2. Output peak power vs output wavelength for two types of phase-matched DFG.

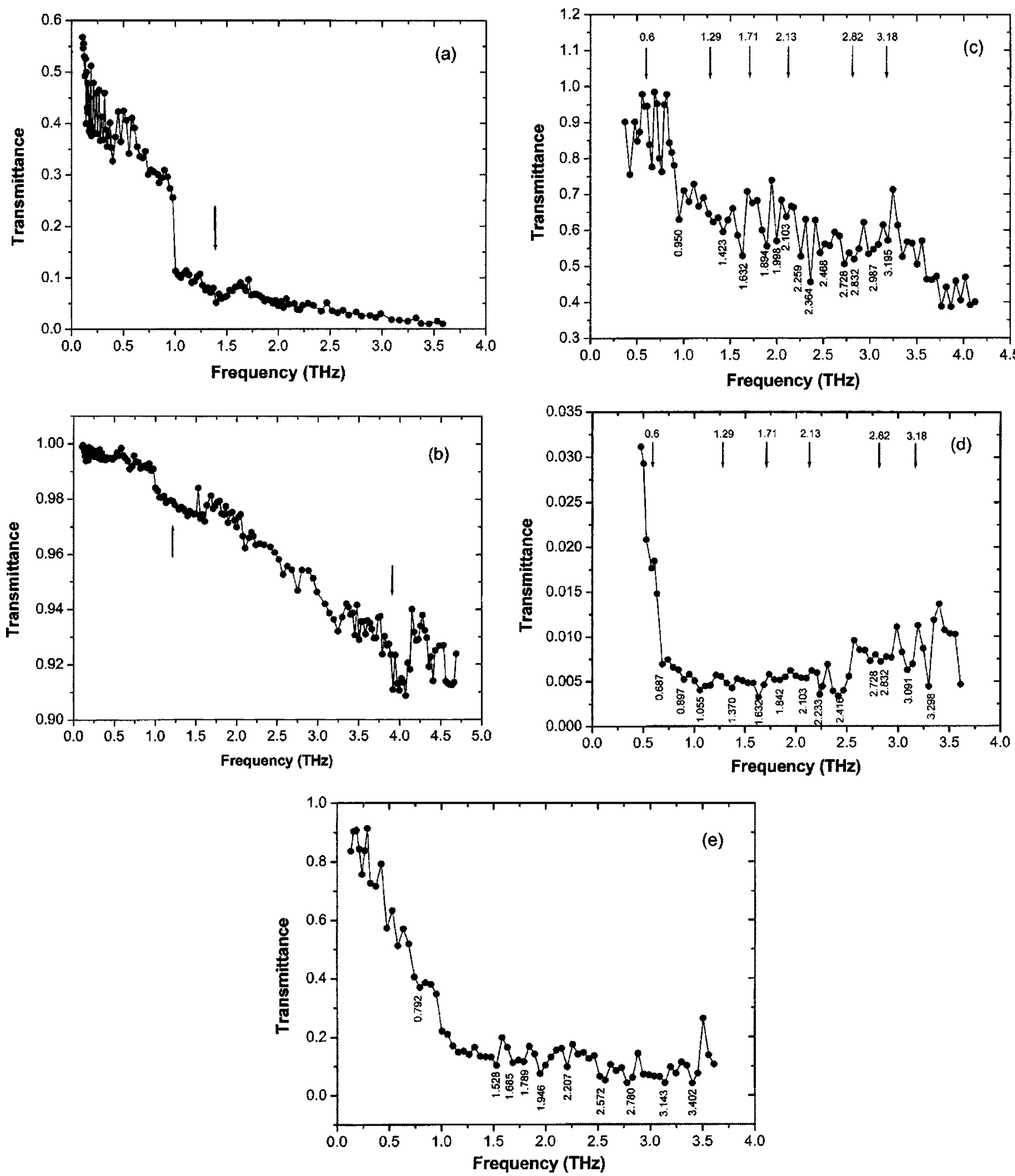

FIG. 3. Directly-measured transmission spectra, i.e., transmittance vs frequency: (a) water. Arrow marks location of the *B* band. (b) Polystyrene foam. Arrows mark locations of *B* band and vibrational band. (c) Herring DNA. (d) Salmon DNA. For (c) and (d), arrows mark locations of predicted resonance peaks based on Ref. 21. (e) Salmon protein.

coherent radiation from 2.7 to 38.4 μm (111–7.81 THz) has also been achieved (Fig. 1). This tuning range already covers high-frequency end of the THz regime defined in the range of 0.1–10 THz. It represents significant improvement over the tuning range of 2.7–28.7 μm achieved previously.[18] One can see that the obtained output wavelengths almost cover the entire range of the THz band defined as 0.1–10 THz except for a narrow gap of 5.15–7.81 THz (58.2–38.4 μm) based on the combination of two types of the PM.

The generated output beams in the two PM configurations had pulse duration of 5 ns and a repetition rate of 10 Hz. Their pulse energies were measured by a pyroelectric detector and calibrated bolometer (see Fig. 2). The maximum output peak powers for the two PM configurations are measured to be 2364 W at 5.87 μm (51 THz) and 209 W at 196 μm (1.53 THz) corresponding to the power conversion efficiencies of 0.75% and 0.055%, respectively. These output powers and conversion efficiencies are also improved significantly compared with the previous results.[14,18] When the THz wave is tightly focused, the corresponding intensity reaches the level of MW/cm^2 at 196 μm, which can be used to explore a number of new fundamental, nonlinear coherent effects in the THz domain. According to Fig. 1, the PM angles are small for type-oe-e interaction, which is important since the effective NLO coefficient is correspondingly high while the conversion efficiency is also high. Moreover, in the range of 70–2000 μm (0.15–4.29 THz) the THz peak power can be kept to be higher than 1 W. Such an extremely wide tuning range with a high level of the output peak powers can be quite valuable to a number of potential applications.[1,2,3,19] Indeed, we have directly measured transmission spectra of water, polystyrene foam, herring and salmon DNA, and salmon protein with our preliminary results shown in Fig. 3. For water there is a broad absorption peak in the range of 1.09–1.5 THz (200–275 μm), see Fig. 3(a). This is due to bending of intermolecular hydrogen bonds (i.e., *B* band).[20] On the other hand, for polystyrene foam there is a relatively sharp peak near 4.07 THz (73.7 μm) [Fig. 3(b)]. We believe this is due to vibrational band of the one-dimensional polymer chains. Moreover, polystyrene foam has extremely low absorption coefficients and refractive indices of about unity in the THz range. Besides the polystyrene foam, we have also measured herring and salmon DNA's [Figs. 3(c) and 3(d)]. In our experiments, nonoriented films of DNA's or proteins were used, which were obtained by dissolving DNA or protein powders in distilled water to form gels and then drying the gels in air. According to Figs. 3(c) and 3(d), there are a number of distinct modes in addition to the common phonon modes (i.e., at 1.632, 2.728, and 2.832 THz). These resonance frequencies are consistent with the predictions[21] and the measurements made via FTIR.[22] On the other hand, the spectrum for the salmon protein is quite different from those of the DNA's [Fig. 3(e)]. We believe we will significantly increase signal-to-noise ratios if the average over several scans is taken.

In conclusion, our result presented here has set a world record for a widely tunable, monochromatic (narrow-linewidth), and tabletop THz source. Indeed, an efficient and coherent THz wave tunable in the two extremely wide ranges of 2.7–38.4 and 58.2–3540 μm, with typical linewidths of 6000 MHz, has been achieved by us. These tuning ranges are much wider than those achieved so far based on all the schemes besides the free-electron lasers. Furthermore, the wide spectral bands achieved by us can be easily accessible by rotating the GaSe crystal to change the PM angle and/or to select the configuration. The maximum peak output power has reached 209 W at 196 μm with a pulse width of about 5 ns. GaSe is the best nonlinear optical material ever used for the THz generation in terms of the absorption coefficient in the THz domain and nonlinear coefficient. The transmission spectra of DNA's directly measured by us can eventually become valuable for characterizing and perhaps identifying biological macromolecules.

This work has been supported by AFOSR and NSF.

[1] P. G. O'Shea and H. P. Freund, Science **292**, 1853 (2001).
[2] M. Nagel, P. H. Bolivar, M. Brucherseifer, H. Kurz, A. Bosserhoff, and R. Büttner, Appl. Phys. Lett. **80**, 154 (2002).
[3] D. L. Woolard, T. R. Globus, B. L. Gelmont, M. Bykhovskaia, A. C. Samuels, D. Cookmeyer, J. L. Hesler, T. W. Crowe, J. O. Jensen, J. L. Jensen, and W. R. Loerop, Phys. Rev. E **65**, 051903 (2002).
[4] G. L. Carr, M. C. Martin, W. R. McKinney, K. Jordan, G. R. Neil, and G. P. Williams, Nature (London) **420**, 153 (2002).
[5] (a) R. Köhler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, G. Davies, D. A. Ritchie, R. C. Lotti, and F. Rossi, Nature (London) **417**, 156 (2002); (b) G. Scalari, L. Ajili, J. Faist, H. Beere, E. Linfield, D. Ritchie, and G. Davies, Appl. Phys. Lett. **82**, 3165 (2003).
[6] E. R. Brown, F. W. Smish, and K. A. McIntosh, J. Appl. Phys. **73**, 1480 (1993).
[7] I. Vurgaftman and J. R. Meyer, Appl. Phys. Lett. **75**, 899 (1999); L. Friedman, G. Sun, and R. A. Soref, *ibid.* **78**, 401 (2001).
[8] F. Zernike Jr. and P. R. Berman, Phys. Rev. Lett. **15**, 999 (1965).
[9] K. Kawase, T. Hatanaka, H. Takahashi, K. Nakamura, T. Taniuchi, and H. Ito, Opt. Lett. **25**, 1714 (2000).
[10] K. Imai, K. Kawase, J.-I. Shikata, H. Minamide, and H. Ito, Appl. Phys. Lett. **78**, 1026 (2001).
[11] V. G. Dmitriev, G. G. Gurzadyan, and D. N. Nikogosyan, *Handbook of Nonlinear Optical Crystals* (Springer, Berlin, 1999), pp. 119–125, 166–169.
[12] Y. J. Ding and I. B. Zotova, Opt. Quantum Electron. **32**, 531 (2000).
[13] Y. J. Ding and J. B. Khurgin, Opt. Commun. **148**, 105 (1998).
[14] W. Shi, Y. J. Ding, N. Fernelius, and K. L. Vodopyanov, Opt. Lett. **27**, 1454 (2002).
[15] K. L. Vodopyanov and L. A. Kulevskii, Opt. Commun. **118**, 375 (1995).
[16] E. D. Palik, *Handbook of Optical Constants of Solid* (Academic, New York, 1998), Vol. III.
[17] A. Yariv, *Quantum Eletronics* (Wiley, New York, 1988), p. 401.
[18] W. Shi, Y. J. Ding, X. Mu, and N. Fernelius, Appl. Phys. Lett. **80**, 3889 (2002).
[19] W. S. Holland, J. S. Greaves, B. Zuckerman, R. A. Webb, C. McCarthy, I. M. Coulson, D. M. Wather, W. R. F. Dent, W. K. Gear, and I. Robson, Nature (London) **392**, 788 (1998).
[20] H. R. Zelsmann, J. Mol. Struct. **350**, 95 (1995).
[21] M. Bykhovskaia, B. Gelmont, T. Globus, D. L. Woolard, A. C. Samuels, T. Ha-Duang, and K. Zakrzewska, Theor. Chem. Acc. **106**, 22 (2001).
[22] J. W. Powell, G. S. Edwards, L. Genzel, F. Kremer, A. Wittlin, W. Kubasek, and W. Peticolas, Phys. Rev. A **35**, 3929 (1987).