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Power scaling of widely-tunable monochromatic terahertz radiation by stacking high-resistivity GaP plates

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A high-resistivity GaP crystal was used to generate monochromatic THz pulses with peak output powers reaching 722 W at 108.1 μm by mixing two coherent beams at about 1 μm based on phase-matched difference-frequency generation. By stacking two and three GaP plates with their second-order nonlinear coefficients being switched between the adjacent ones, we have increased the peak power at 120.3 μm from 433.4 W to 1.36 and 2.36 kW, respectively. 2.36 kW corresponds to the photon conversion efficiency of 25%, which is two orders of magnitude higher than our previous result. In contrast, if they are stacked for having the same sign of the nonlinear coefficients, the wavelength corresponding to the highest peak power is red-shifted to 204.8 and 303.9 μm, respectively. Such a result indicates that there is an optimal interaction length for each specific output wavelength. © *2010 American Institute of Physics*. [doi:10.1063/1.3292585]

Zinc-blende semiconductors can be used to generate terahertz (THz) waves through either optical rectification using an ultrafast laser with a sufficiently-wide bandwidth or difference-frequency generation (DFG). Indeed, a ZnTe crystal was used to generate an average power of 150 μW.[1] On the other hand, resonantly-enhanced second-order nonlinearities significantly increased the output power from InAs to 57 μW.[2,3] ZnTe can be broadband-phase-matched at the pump wavelength of 800 nm.[1,4] However, at such a wavelength, two-photon absorption (TPA) (Ref. 5) may limit the output power generated by ZnTe. Although GaAs can be phase-matched at 1.4 μm, its TPA coefficient is within the same order of magnitude as ZnTe. Phase-matching was achieved in GaP crystals for both noncollinear[6] and collinear[7] configurations at the mixing wavelengths of around 1 μm. GaP is more advantageous than ZnTe and GaAs for scaling up the output powers since its TPA coefficient is two orders of magnitude lower.[8] THz waves hold promise for imaging[9] and chemical identification.[10]

In this letter, we present our results on power scaling of THz radiation based on DFG in high-resistivity and low-dislocation-density GaP plates. We have stacked GaP plates such that three nonzero elements of second-order nonlinear susceptibility have the same sign or opposite signs from one plate to the next. These two stacking configurations have yielded completely different results in terms of the highest output peaks and corresponding wavelengths. The highest output peak power measured by us is about 2.36 kW, which is two orders of magnitude higher than our previous result.

Each of the semi-insulating undoped (110) GaP plate used in our experiment has a resistivity of >1.0 MΩ-cm and a dislocation density of 2.25×10^5 cm^{-2}. It has a diameter of 48.5 mm and a thickness of 663 μm. Both of the (110) facets are polished. In order to generate monochromatic THz pulses, two coherent radiation beams were a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser at 1.064 μm with pulse duration of 10 ns and pulse energy of 20.7 mJ (the highest used in our experiment) and the idler output from a β-BaB_2O_4-based optical parametric oscillator (OPO) pumped by the third-harmonic output of the Nd:YAG laser. The highest pulse energy for the idler beam is 18.7 mJ whereas its pulse duration is 5 ns. These two beams were collimated, combined, and then illuminated the GaP plates. The beam radius for both of the mixing beams at the GaP plates was measured to be 1.0 mm. Therefore, the highest peak intensity for the idler beam was 120 MW/cm^2, which is a factor of 5.4 below the highest intensity of 650 MW/cm^2 used without causing any damage.[11] They collinearly propagated in a direction perpendicular to the two large facets of the GaP plates with their polarizations parallel to the $[1,\bar{1},0]$ and $[0,0,1]$ directions, respectively. Under such a configuration, the effective second-order nonlinear coefficient is much higher than that from a (100) GaP crystal.[7,12] Germanium and polyethylene windows were used to block the two mixing beams. The THz radiation generated by DFG was attenuated, collimated by a parabolic mirror, and then focused onto a power meter by using another parabolic mirror. The THz output wavelengths were calibrated by an etalon.

By measuring the transmission and reflection spectra of each GaP plate using Fourier transform infrared spectroscopy, we have deduced the absorption spectrum. According to Fig. 1, the absorption coefficient is 2.2 cm^{-1} at 120 μm, which is seven times lower than that from our previous sample. This implies that we could stack up to seven plates to effectively increase the output power. Based on the dispersion relation,[13] the coherence length for the THz generation is linearly increased with the wavelength from 581 μm at 76.5 μm. At 304 μm, e.g., the coherence length becomes 7.37 mm. We measured the THz output powers at different output wavelengths for a single GaP plate. According to Fig. 2, the THz output radiation was tuned continuously from 3.91 THz (76.7 μm) to 454.9 GHz (659.5 μm). Such a wide tuning range was achieved by slightly adjusting the frequency (wavelength) of the idler beam generated by OPO.

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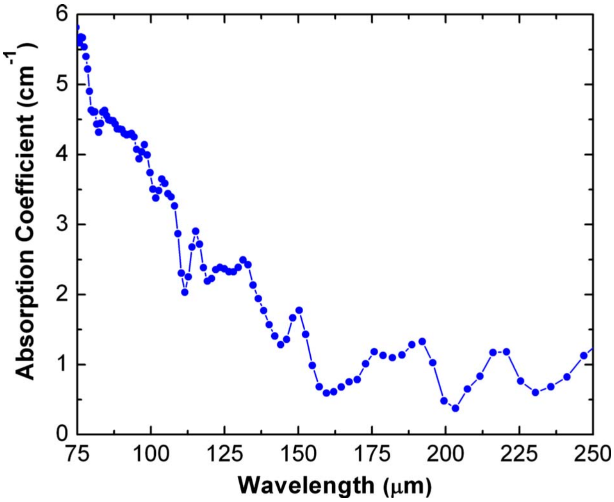

FIG. 1. (Color online) Absorption coefficient is plotted vs wavelength following the measurement of transmission and reflection spectra on a single GaP plate.

Within such a range, the plate thickness is either comparable to or much shorter than the coherence length calculated by us. Therefore, the THz generation from the single plate is phase-matched. At 2.775 THz (108.1 μm), the output power reached the highest value, i.e., 722 W. Using the pulse width of ~5 ns, the highest average power of the generated THz radiation is 36.1 μW at the average input power of 207 mW at 1.0642 μm and 187 mW at 1.0748 μm, respectively. Such an output power is a factor of 46 larger than the highest power achieved on GaP so far.[7]

By using a (110) GaP wafer, we can access a much higher value of the effective second-order nonlinear coefficient.[12] Second, the two parallel facets of the GaP crystal can induce Fabry–Perot effect, which can also contribute to the enhancement of the THz output power. In fact, from Fig. 2 one may notice that the THz output power changes periodically. On the low-frequency side, i.e., for the frequencies below 2.5 THz, the frequencies at most of the peaks and valleys can match those calculated based on Fabry–Perot effect. If we only assume such an effect for the THz wave, the ratio of a local maximum power and its adjacent minimum value should be about 3.3. For the frequencies higher than 2.5 THz, the frequencies at the peaks and valleys are no longer consistent with those predicted by Fabry–Perot effect. In addition, the modulation depths are much larger. We believe these modulations are primarily caused by the water-vapor absorption. Third, the absorption of the THz pulses generated by DFG is much less than that for the previous crystal.[7] As a result, the frequency at the highest output power from our crystal (2.775 THz) is much higher than that measured previously, i.e., 1.73 THz.[7] There was the subsequent absorption of the THz pulses by a rather thick crystal at relatively high frequencies.[7] Such an assumption is further supported by the fact that the high-frequency limit measured on our crystal is significantly higher than the previous value. Fourth, since the dislocation density of our GaP crystal is remarkably low, the damage threshold is significantly increased. As a result, we were able to use much higher peak powers for the two mixing beams.

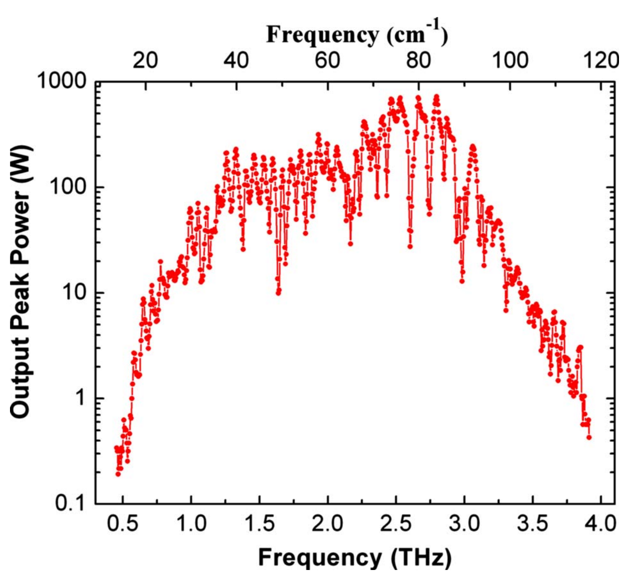

FIG. 2. (Color online) THz output power was measured vs output wavelength for a single GaP plate. As discussed in the text, the modulations were caused by convolution between Fabry–Perot effect for THz wave and water-vapor absorption.

We measured the dependence of the THz output power on the input power for one of the mixing beams. Based on the linear-least-square fit to the power dependences, we have determined the internal conversion efficiency to be 0.067% (the photon conversion efficiency of 6.8%). Without taking into consideration absorption losses, considering the Fabry–Perot effect for the THz wave, and assuming that the three nonzero elements of the second-order nonlinear optical susceptibility for GaP are identical, the maximum conversion efficiency for the THz generation in terms of peak powers is given by:[14]

$$\eta_{\max} = \frac{P_{\mathrm{THz}} T_{\mathrm{THz}}^{-1}}{P_1 T_1} = \frac{16 d_{\mathrm{eff}}^2 L^2}{\varepsilon_0 c n_1 n_2 n_{\mathrm{THz}} \lambda_{\mathrm{THz}}^2} \frac{P_2}{\pi w_0^2} \frac{T_2}{T_{\mathrm{THz}}^2}, \qquad (1)$$

where max is used to designate the local maximum values due to Fabry–Perot effect, d_{eff} is the effective nonlinear coefficient, P_{THz} is the THz output peak power, P_1 and P_2 are the input peak powers at 1.064 μm and at the idler wavelength, respectively, λ_{THz} is the output wavelength, L is the thickness of the GaP wafer, w_0 is the beam radius for the pump beams, n_i are the refractive indices of the GaP crystal at the respective wavelengths of the three interacting waves, and T_i is the Fresnel transmission coefficients at single surface. Assuming $d_{\mathrm{eff}} \approx 47$ pm/V, we have obtained $\eta_{\max} \approx 0.015\%$ from Eq. (1). This value is about a factor of 4.5 lower than our experimental value. We believe the difference between two may be attributed to the uncertainty for the value of d_{eff} used in Eq. (1).

In addition to one GaP plate, we have attempted to stack two and three GaP plates. We have stacked them according to two different configurations, i.e., three elements of second-order nonlinear susceptibility for GaP have (A) same sign and (B) opposite signs between the adjacent GaP plates. Figure 3 illustrates our result obtained for the stacking configuration A. One can see that the wavelength corresponding to the highest output power has been significantly red-shifted from 108.1 to 204.8 to 303.9 μm. The corresponding power is reduced from 722 to 698 W to 611 W. 611 W corresponds to a photon conversion efficiency of 16% inside the stacked plates. These results indicate that there is an optimal interaction length at each specific output wavelength. We believe

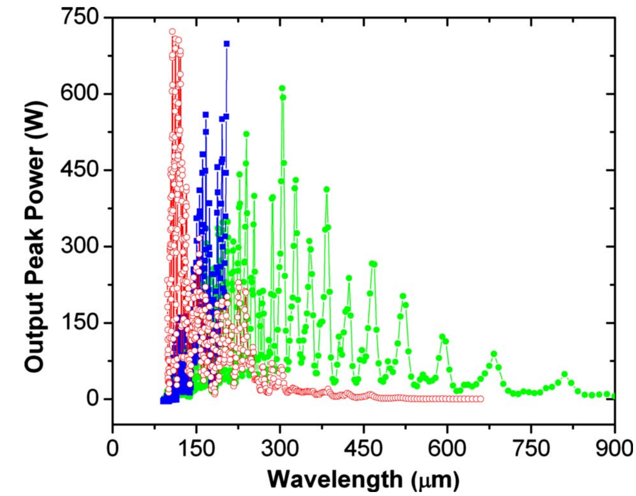

FIG. 3. (Color online) Output peak power vs wavelength: open circles—single GaP plate; squares—stacked two GaP plates; dots—stacked three GaP plates. For both of the stacked two and three plates, second-order nonlinear coefficients have same sign between adjacent plates.

that the red-shift of the optimal output wavelength is primarily caused by the combination of the linear wavelength dependence of the coherence length and wavelength-dependent output power reflected by Eq. (1). Therefore, at the wavelengths of 108.1, 204.8, and 303.9 μm, the coherence lengths should be roughly close to 663 μm, 1.326, and 1.989 mm, respectively.

Under the stacking configuration B our result is completely different. One can see from Fig. 4 that the wavelength corresponding to the highest output power is kept at 120.3 μm for the two and three plates. Therefore, quasi-phase matching is achieved at this wavelength. This implies that the coherence length at 120.3 μm is about 663 μm. On the other hand, using the dispersion relation,[13] we have calculated coherence length to be 1.65 mm. We believe that the factor of 2.5 between the two is probably caused by the inaccuracy in the THz index of refraction calculated by using the dispersion relation.[13] Using 663 μm as the coherence length, we have determined the index of refraction at 120.3 μm to be 3.217. This value is just 3.8% lower than that calculated by using Ref. 13. Such an estimate illustrates the fact that the coherence length is quite sensitive to the index of refraction at the output wavelength. At such a wavelength, the output peak power was significantly increased from 433 W to 1.36 kW and 2.36 kW, corresponding to two and three plates, respectively. The peak power of 2.36 kW corresponds to an average power of 118 μW. Compared with our previous result,[7] we have increased the peak power by a factor of 151, i.e., two orders of magnitude. The internal power conversion efficiency deduced from our experiment is 0.22%, corresponding to the photon conversion efficiency of 25%.

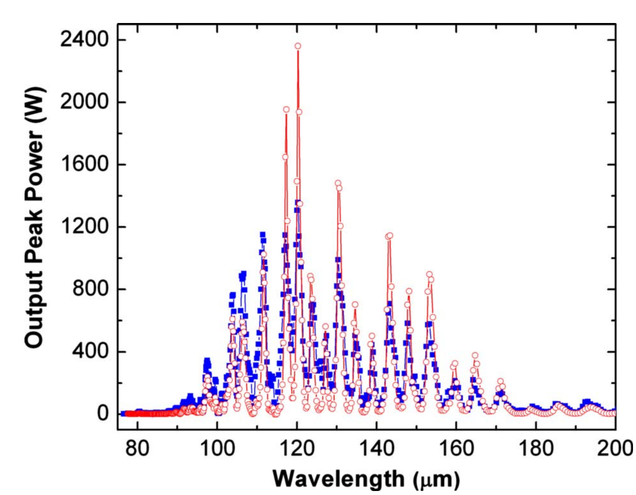

FIG. 4. (Color online) Output peak power vs wavelength: squares—stacked two GaP plates; open circles—stacked three GaP plates. For both of the stacked two and three plates, second-order nonlinear coefficients have the opposite signs between adjacent plates.

In conclusion, we have significantly increased the peak and average output powers generated from a single GaP plate based on phase-matched difference-frequency generation. The highest output power achieved by us 722 W at 2.78 THz. By adjusting the frequency for one of incoming radiation beams, we have achieved the tuning range of 455 GHz–3.91 THz. We have investigated THz generation from stacked two and three GaP plates under two configurations. When different plates have the same sign of the second-order nonlinear coefficients, the wavelength corresponding to the highest output power is red-shifted to 204.8 and 303.9 μm. These results indicate that there is an optimal interaction length at each specific output wavelength. When adjacent plates have the opposite signs of the second-order nonlinear coefficients, the output peak power has been significantly increased from 433 W to 1.36 kW and 2.36 kW. 2.36 kW is amounted to an improvement over previous result by two orders of magnitude. Therefore, just three stacked GaP plates with the total thickness of less than 2 mm are capable of reaching the significant spatial depletion of the pump photons.

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