

Comment on “GaSe_{1-x}S_x and GaSe_{1-x}Te_x thick crystals for broadband terahertz pulses generation” [Appl. Phys. Lett. 99, 081105 (2011)]

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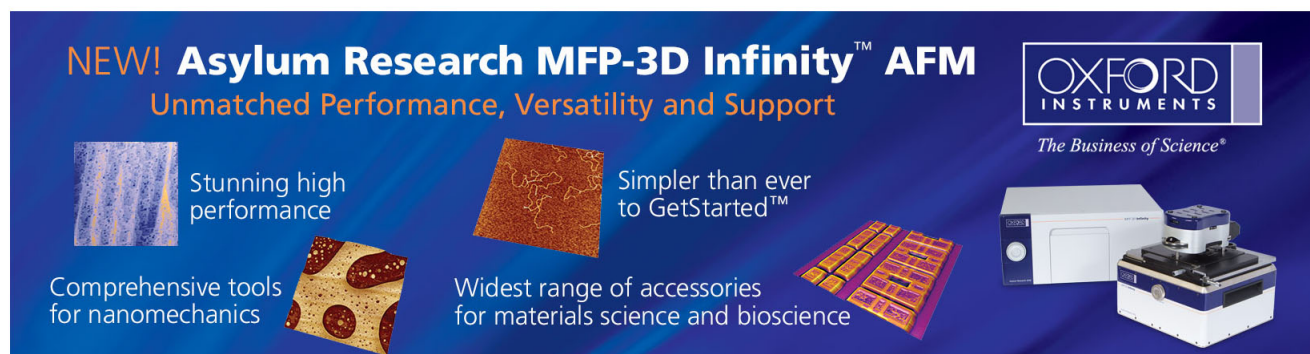
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Comment on “GaSe_{1-x}S_x and GaSe_{1-x}Te_x thick crystals for broadband terahertz pulses generation” [Appl. Phys. Lett. 99, 081105 (2011)]

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In a recent letter, Nazarov *et al.*¹ claimed that the broadband terahertz pulse generation can be achieved in thick GaSe, GaSe_{1-x}S_x, and GaSe_{1-x}Te_x crystals. By doping Te and S in GaSe crystals, the absorption peak at 0.59 THz which affects the spectra of THz generation can be canceled out. In this comment, we point out that Nazarov *et al.* did not consider the absorption peak at higher frequency of 1.77 THz while the Te and S were doped into GaSe crystals as shown in Fig. 1.

By a homemade THz time-domain spectroscopy (TDS) system with a high signal-to-noise ratio and within a low humidity (<3%) environment, the rigid layer mode² $E'(2)$ was clearly observed at ~ 0.59 THz for GaSe as shown in Fig. 1, especially for the thick crystals. However, this $E'(2)$ mode will be gradually suppressed as increasing the concentration of Te, which is consistent with the results in Fig. 3(b) of Ref. 1. Meanwhile, the other peak, $E''(2)$ mode, at high frequency of ~ 1.77 THz grows up with more Te-doping,³ which was not presented in Fig. 3(b) of Ref. 1. Similar phenomenon was also observed in S-doped GaSe crystals as shown in Fig. 1. As the explanation in Ref. 1, the intercalation of S and Te atoms to the interlayer space would increase the interlayer bonding and affect the rigid layer mode $E'(2)$. That is why the interlayer vibration mode $E''(2)$ inside one layer (included two Ga atoms and two Se atoms) could be created due to the frailer bonding between two Ga atoms inside one layer as illustrated in the inset of Fig. 1.

The influence of THz-generation spectra at low-frequency side is reasonably reduced by shrinking the absorption peak at ~ 0.59 THz. However, the appearance of absorption peak at ~ 1.77 THz would certainly degrade the THz generation efficiency at high-frequency side. Actually, this absorption effect was already revealed by a significant deep at around 1.77 THz in Fig. 4(a) of Ref. 1, which was essentially disregarded by authors. Therefore, the broadband

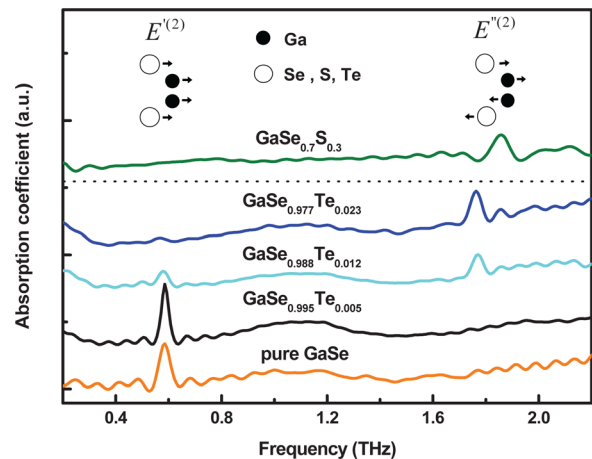


FIG. 1. THz absorption spectra in GaSe, GaSe_{0.995}Te_{0.005}, GaSe_{0.988}Te_{0.012}, GaSe_{0.977}Te_{0.023}, and GaSe_{0.7}S_{0.3} single crystals obtained by the THz TDS measurements. Inset illustrates the various atomic vibration modes in GaSe_{1-x}A_x (A = Te, S) crystals.

THz generation with flat at high-frequency side cannot be really achieved by the strategy of S- and Te-doping in thick GaSe crystals.

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