Dispersion equations for the entire transparency range of GaSe

J.F. Molloy[a,b], M. Naftaly[b], Yu.M. Andreev[a,c], K. Kokh[d], G.V. Lanskii[a,c], and V.A. Svetlichnyi[c]

[a]Siberian Physical-Technical Institute of Tomsk State University, Tomsk, Russia
[b]National Physical Laboratory, Teddington TW11 0LW, UK
[c]Institute of Monitoring of Climatic and Ecological Systems SB RAS, Tomsk, Russia
[d]Institute of Mineralogy and Geology SB RAS, Novosibirsk, Russia

***Abstract*—High optical quality GaSe samples with faces parallel and orthogonal to the optical axis are manufactured. o- and e-wave dispersions are studied by THz time domain spectroscopy. New dispersion equations are designed that are suitable for application in the entire transparency range from the near IR through the mid-IR and further into the THz.**

I. Introduction

The layered ε-GaSe has been known from 1934 and promises efficient optical frequency conversion into mid-IR and THz range due to its outstanding physical properties [1]. However, in spite of these promising properties and long-term success of in-lab applications, GaSe is still difficult to grow as large high-optical-quality crystals, due to technological reasons. It is also challenging to process in arbitrary orientations due to its layered structure with very weak inter-layer electrostatic bonding [2]. In particular, the above-mentioned difficulties have resulted in the publication of only a few papers for both *o*- and *e*-wave dispersion of refractive index across the entire transparency range from the near and mid-IR, and further into the THz regime [3–5]. These equations exhibit broadly varying birefringence from 0.49 [4] to 0.79 [3]. Surprisingly, the birefringence estimated by using dispersion equations cited in the world-famous and widely used Handbook [1] after Ref. [4] differ drastically from the much larger birefringence and smaller o-wave refractive index recorded by the few known THz-TDS measurements [3, 6, 7].

The GaSe crystals studied were grown by modified vertical Bridgman method with a heat field rotation and possess both high optical quality (absorption coefficient $\alpha<0.03$ cm^{-1}) and uniformity [8-10]. Recently developed, original cutting and polishing technologies [9, 10] have enabled us to fabricate samples with faces parallel and orthogonal to the optical axis. THz-TDS was used to obtain the *o*- and *e*-wave refractive indices between 0.2–4 THz with spectral resolution from 3.75 to 30 GHz, to identify the most adequate dispersion equations among known examples, and to design modified dispersion equations for the entire transparency range of GaSe showing birefringence identical to the available experimental data.

II. Results

First, the criterion was designed to select adequate spectra of refractive indices recorded by THz-TDS. Namely, only spectra that demonstrated good point-to-point reproducibility of the data and a highly regular etalon oscillation pattern (noise free, identical dispersion gradients, peak positions and magnitudes and periods) were considered. Such spectra confirm that the sample under study is free of defects and deformation, and not wedged. Then the selected results, with the addition of widely used data for IR dispersions in [1], were approximated in the form of new Sellmeier equations:

$$n_o^2 = 10.6409 + \frac{0.3788}{\lambda^2 - 0.1232} + \frac{6963.32}{\lambda^2 - 2198.85} + \frac{0.017\lambda^2}{\lambda^2 - 262177.5577} \quad (1)$$

$$n_e^2 = 5.76 + \frac{0.3879}{\lambda^2} - \frac{0.2288}{\lambda^4} + \frac{0.1223}{\lambda^6} + \frac{0.3706\lambda^2}{\lambda^2 - 1780.3} \quad (2)$$

Fig. 1 presents estimated dispersions by using known and designed dispersion equations.

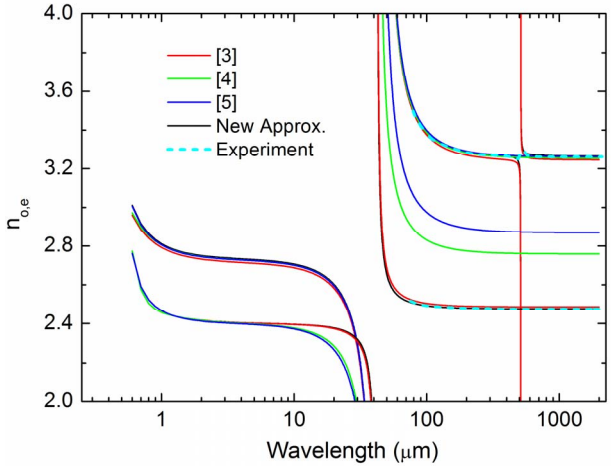

Fig.1. Plots of o- and e-wave dispersions for GaSe.

In Fig.1 it is seen that available and designed dispersion equations give similar result in the mid-IR where they differ only in the data close to transparency edges (validity ranges). It should be mentioned that in fact mid-IR dispersions for GaSe considered in [4, 5] originate from the data in [12].

In Fig.1 it is also seen that o-wave dispersions estimated for the THz range are also almost identical; data in [3] accounts for the abnormal dispersion caused by absorption peak of the rigid phonon mode $E'^{(2)}$ centered at 0.59 THz. It is evident that abnormal dispersions caused by other phonon absorption peaks should be considered in the design of dispersion equations. Besides, data for e-wave dispersions determined by

semi-empirical method, using measured o-wave dispersions and experimental data on phase matching show large differences. The validity ranges of the designed equations are estimated as 0.65–18 μm and 0.2–4 THz.

Fig.2 shows for comparison the birefringence spectra that are calculated using the dispersion equations from this work and from the literature.

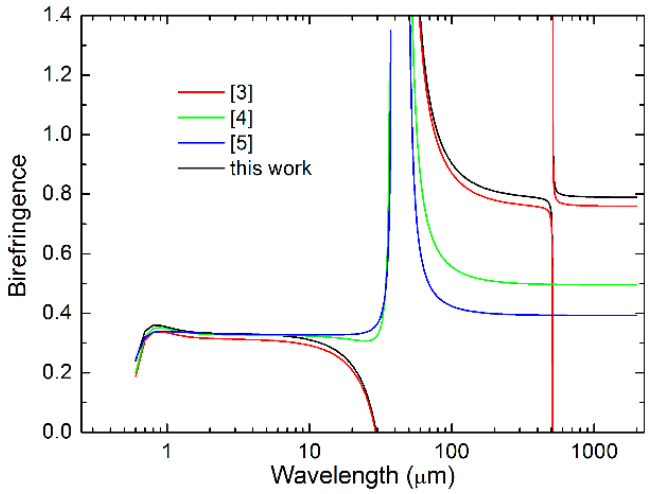

Fig. 2. Plots of calculated birefringence for GaSe.

In Fig.2 it is seen that birefringence estimated for the mid-IR range by using the designed equations (Eqs. 1 & 2) is in good agreement with the birefringence estimated by using equations from [1, 4], whereas values from the data in Ref. [3] are somewhat underestimated. The similarity of the calculated dispersion curves for ordinary refractive index, depicted in Fig.2, reflects the simplicity of direct measurements of cleaved samples. Indirect (semi-empirical) estimation of e-wave refractive index for the processed crystals in [4, 5], from experimental phase matching data, leads to noticeable differences between the available data, as well to differences in the calculated birefringence. This is due to the difficulties in processing GaSe to manufacture samples with facets parallel to optical axis. The designed dispersion equations (Eqs. 1 & 2) agree well with the experimental THz-TDS data for birefringence B=0.79, which are also close to the data cited and somewhat underestimated data in [3].

Eqs. 1 & 2 do not confirm the estimations obtained by using the dispersion equations recommended by Handbook [1] after Ref. [4]. It is interesting to emphasize that in spite of the wrong data for e-wave dispersion these dispersion equations are still widely used and show reasonable difference in estimated and external experimental phase matching angles of less than 10 degrees that is often acceptable in practice. It can be proposed that the dispersion equations in [4] are still useful for PM estimation due to the weak input of $\boldsymbol{k}_3$ vector into $\boldsymbol{k}$-vector sum (PM conditions) [4]. The correct data on THz dispersion properties can be much more important for other applications of GaSe crystals such as for electro-optics or polarization optics, or estimation of dispersion properties for solid solution GaSe crystals.

Anyway, further work should be directed to improve the dispersion equations for GaSe at local spectral ranges close to transparency edges and accounting for the input of phonon absorption peak, or free charge carries in GaSe crystals doped with elements that are leading to strong changes in free charge carriers concentration such as Al [13].

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