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EFFICIENT AND TUNABLE THz RADIATION BASED ON SELENIDE AND SULFIDE CRYSTALS

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Abstract: It has been studied terahertz spectra by time-domain spectroscopy, the far infrared properties of different selenite, sulfide and telluride compounds and crystals. Propose a new method to extract, from the experimental data, the refractive index and absorption of such materials that exhibit strong absorption lines. Has been illustrated this method by measuring the terahertz response of doped $TlInS_2$ crystals. Has been reported the observation of a strong absorption peak at 1.242 THz in InSe, which could be attributed to the excitation of TO phonon (half layer shear mode).

Keywords*:* Terahertz, time-domain spectroscopy, selenide, sulfide.

1. Introduction

Telluride and selenide materials exhibit amazing properties in view of various promising applications like storage of electricity, nonlinear optics, etc. Some of them show specific spectral features in the terahertz (THz) range, while some others can be used to generate THz waves by optical rectification. Therefore, a precise characterization of the THz response of such materials is necessary for developing optimized THz devices, and also to have a better knowledge of the physical phenomena occurring at THz frequencies in the materials. Due to its large achievable bandwidth and its great dynamics, THz time-domain spectroscopy (TDS) is a valuable tool to perform this characterization. Nevertheless, has been obliged to modify the classical code of extraction of parameters (refractive index n and coefficient of absorption α of the materials) as these materials present spectral bands of high absorption, in which almost no THz signal is transmitted through the samples. Has been applied this modified procedure to study low dimensional $A^{III}B^{III}C_2^{VI}$ compounds [1], such as $TlInS_2$, GaSe and InSe.

2. Principle of the extraction THz-TDS technique

The classical THz-TDS method, has been recorded the temporal waveforms impinging onto and transmitted by a sample. Then a numerical fast Fourier transform (FFT) of both signals is performed. The ratio of the transmitted and incident FFT spectra gives the transmission coefficient of the sample. As the origin of time is preserved between the two requested measurements, both modulus and relative phase of the transmission coefficient are obtained. If the sample is a slab with parallel sides, the index of refraction and the coefficient of absorption could be accurately determined using inverse electromagnetic code [2]. This code, as well as derived ones, requires inputting the absolute transmission phase, which is obtained by a linearization of the measured phase at low frequencies. In materials exhibiting a high absorption band, the transmitted signal within this band could be weaker than noise. In this case, the transmission coefficient is almost zero in modulus, and its phase is unknown, which makes extraction codes inoperative. The usual solution to this problem is to perform THz-TDS in reflection. However, the disadvantage of the reflection technique is its weak precision due to

difficulty to get a good reference signal. This latter is supplied by a metallic mirror located at the position of the sample, whose coefficient of reflection is supposed to be 100 %. A small error of the sample position, for example a few μm shift from the position of the mirror, leads to dramatic errors, mostly regarding the absorption of the sample.

Has been developed a combined technique [3], which takes benefit of both transmission and reflection spectra by THz-TDS' spectroscopy. The basic idea is to roughly estimate *n* from reflection data over the whole THz spectrum. In the first spectral region of transparency, *n* is more precisely obtained from transmission data. Then we correct the effect of any error in position of the reflection experiment by equalizing the *n* values obtained in reflection and in transmission. In the second band of transparency, and similarly in the other transparency regions, the missing phase in transmission is retrieved by comparing the *n* values extracted from corrected-reflection and transmission. Thus *n* is nicely determined from transmission data in the regions of transparency, while we save the reflection values in the regions of absorption. As well, absorption id determined from transmission in the transparency bands, while we keep the corrected reflection values in the absorption peaks. However, in the present study, materials are not transparent at lower frequencies and the extraction method [3] must be modified. The refractive index is estimated over the whole spectrum by performing a Kramers-Kronig transformation of the absorption obtained in transmission. The error due to the saturated transmission in the absorption bands or to missing absorption peaks located outside the experimental spectral window, is spread over the whole spectrum. Then the phase correction in reflection and the phase retrieval in transmission are performed as explained previously. Of course, the method can be applied only if the error in the Kramers-Kronig refractive index, due to the missing absorption data, is smaller than the error induced by a 2π phase error (Fig.1, 2).

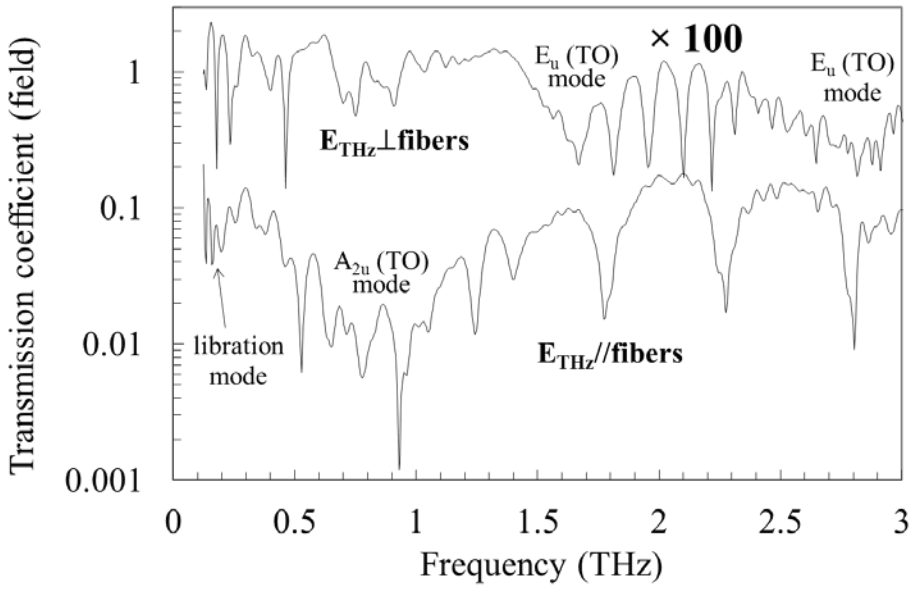

Fig. 1. Transmission coefficient of $TlGaTe_2$ crystal samples versus frequency for THz E-field polarizations.

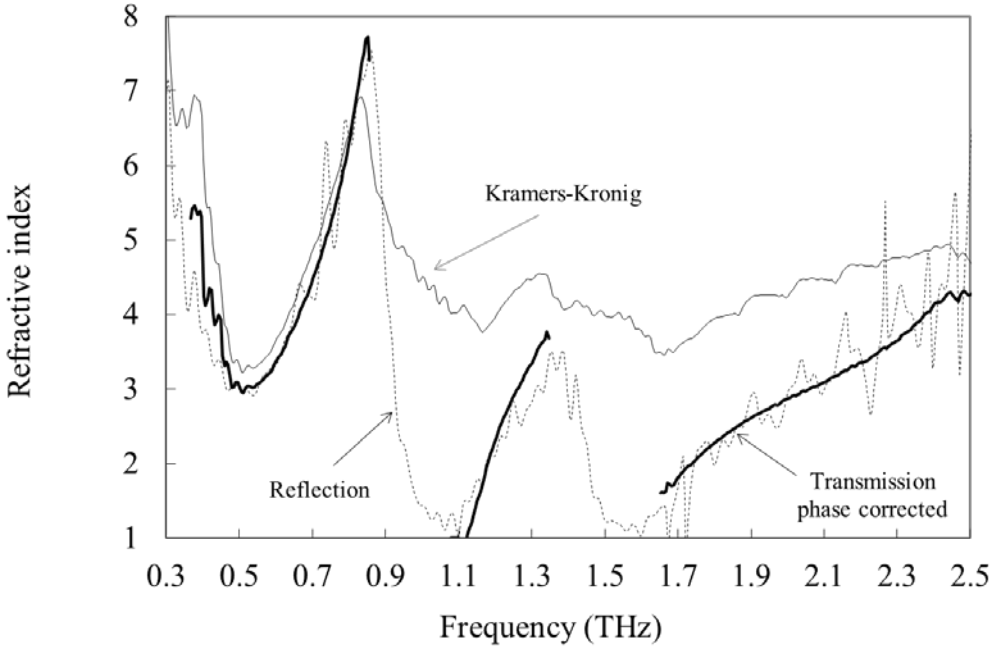

Fig. 2. Refractive index of $TlInS_2$: Co versus frequency obtained with the different THz-TDS methods explained in the text.

3. Results and discussion

It has been applied this method to determine refractive index (*n)* and absorption coefficient (*α)* of low dimensional $A^{III}B^{III}C_2^{VI}$ compounds, like doped $TlInS_2$, $TlGaTe_2$, etc. These materials grow as films of nano-fibers. The fibers exhibit a hexagonal structure made of layers of A and C atoms, which form $A^{III,3+}C_2^{VI,2-}$ nanochains, separated by a layer of B atoms. Electrical hoping-type conductivity occurs in the metallic B layer, and thus this conductivity shows a strong anisotropy. Below a threshold temperature, the experimental DC-conductivity of the samples varies linearly as 1/*T*, which is typical of ionic-type conductivity [1]. Above this temperature, the conductivity increases dramatically: this phenomenon is called super-ionic conductivity. In the case for example of $TlGaTe_2$, this sharp increase of conductivity happens due to the phase transition accompanying the disordering of the Tl^+ sublattice. Fig. 4 shows the coefficient of transmission of $TlInS_2$:Co versus frequency. Several absorption peaks in the spectrum could be attributed to the excitation of phonons (A_{2u} and B_u). The absorption line at approximately 0.2 THz, seen only when the THz field is aligned along the nanofibers, occurs at a frequency lower than the lowest phonon peak (A_{2u}). Therefore, this resonance is probably related to the libration oscillation of the $Ga^{3+}Te^{2-}$ nanofibers [4].

Fig. 2 and 3 present respectively the *n* and *α* spectra of $TlInS_2$:Co. In the transparency regions, *n* is precisely obtained from transmission TDS while, in the absorption peaks, its value deduced from reflection is noisier. Absorption obtained from transmission TDS exhibits a strong saturation at the resonances, while values derived from reflection data are affected by a strong error due to a mispositionning of the reference mirror, which is corrected using our method (thick curve).

n and *α* spectra of InSe and GaSe are depicted in Fig. 4. The rigid layer phonon mode $E'^{(2)}$ of GaSe at 0.596 THz is clearly observed. In InSe, we can see a similar peak at 1.242 THz, which can be attributed to a TO half-layer shear phonon-mode, as already reported from Raman spectroscopy [5]. To the best of our knowledge, our observation is the first one in THz

spectroscopy, as published far infrared studies of InSe [6-7] were performed with FTIR instruments that do not reach the 1-THz range. Fiq.5 presented Terahertz generation and detection in GaSe and InSe crystals.

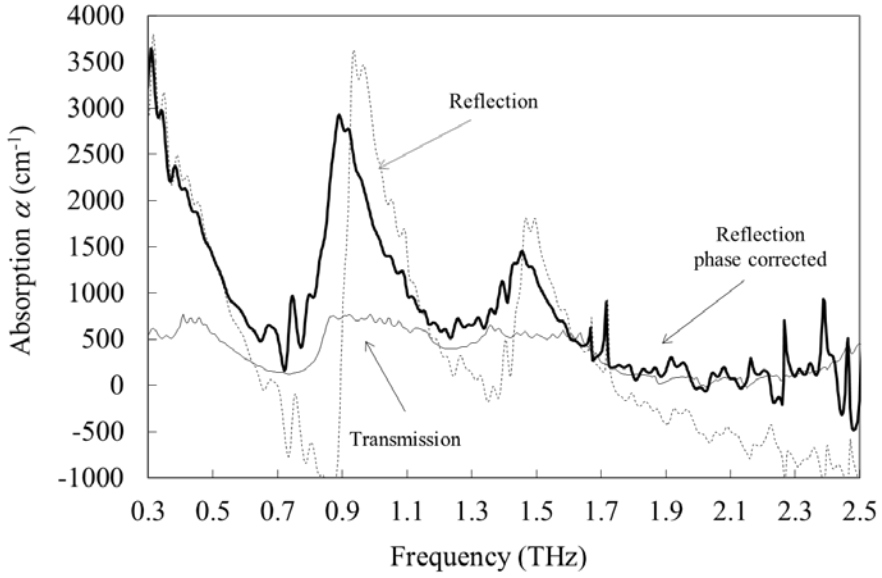

Fig.3. Absorption coefficient of $TlInS_2$:Co versus frequency obtained with the different THz-TDS methods explained in the text.

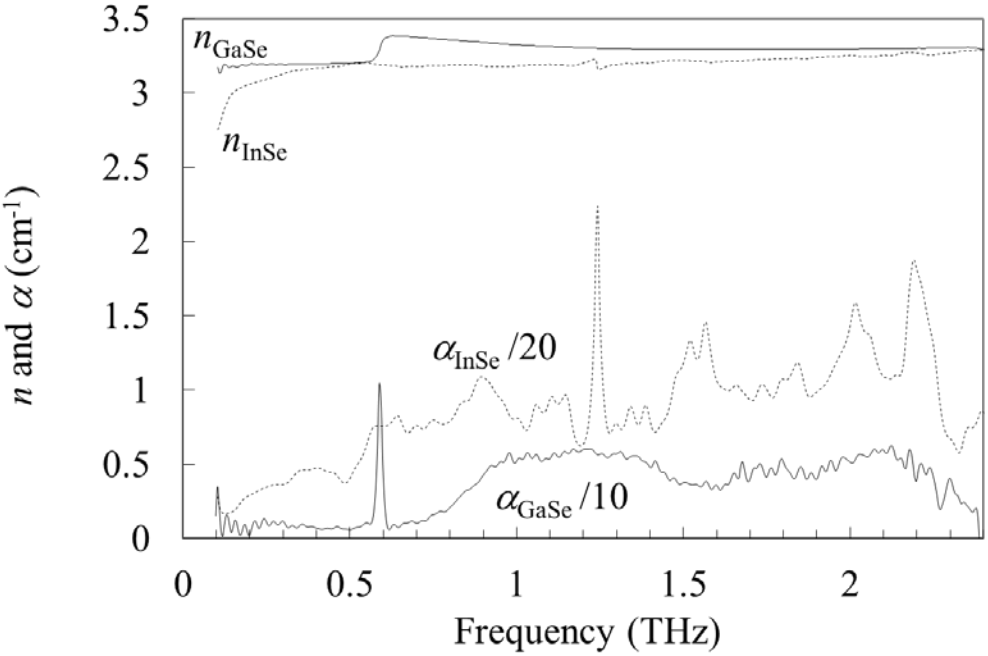

Fig.4. n and α versus frequency of InSe and GaSe.

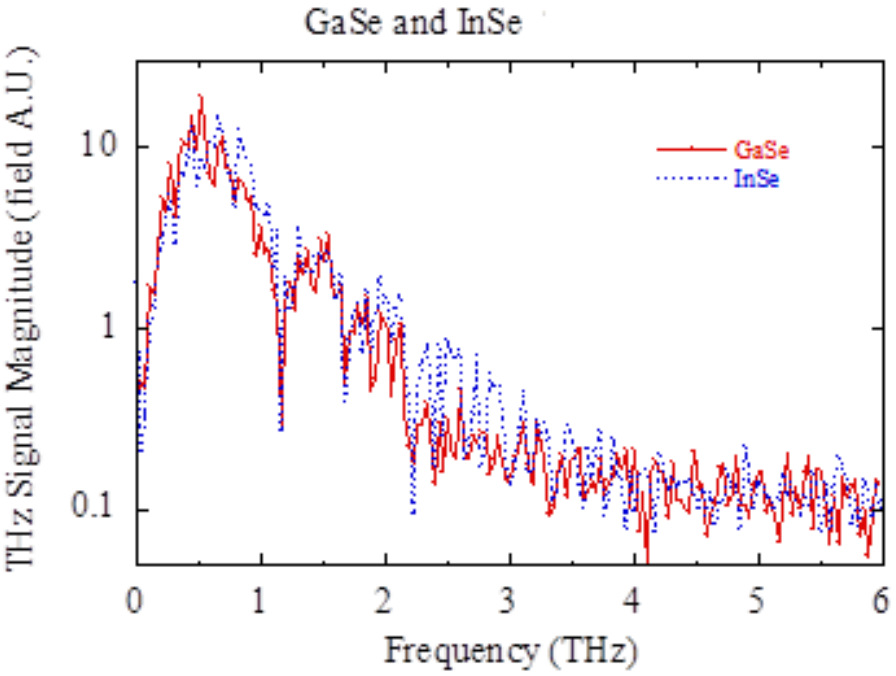

Fig. 5. Terahertz generation and detection in GaSe and InSe crystals.

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ЭФФЕКТИВНОЕ И ПЕРЕСТРАИВАЕМОЕ ТЕРАГЕРЦОВОЕ ИЗЛУЧЕНИЕ НА СЕЛЕНИДНЫХ И СУЛЬФИДНЫХ КРИСТАЛЛАХ

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Резюме: Методами спектроскопии времени пролета изучены терагерцовые спектры кристаллов $TlInS_2$, $TlGaTe_2$, InSe и GaSe. Методом получения оптических констант, в материалах имеющих сильные линии поглощения в терагерцовой области спектра, получены оптические константы допированных кристаллов $TlInS_2$. Показано наличие терагерцового излучения в кристаллах InSe и GaSe.

Ключевые слова: Терагерцовая спектроскопия, временная область, селенид, сульфид.

SELENİD VƏ SULFİD KRİSTALLARINDA EFFEKTİV VƏ TEZLİYİ DƏYİŞƏN TERAHERS ŞUALANMANIN TƏDQİQİ

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Xülasə: Terahers spectroscopiyası metodu ilə $TlInS_2$, $TlGaTe_2$, InSe və GaSe kristallarının terahers spektrləri tədqiq edilib. Spektrin terahers oblastında güclü udulma spektral xətləri olan kristallarda optik əmsalların alınması metodu ilə, aşqarlanmış $TlInS_2$ kristallarının optik əmsalları alınmışdır. InSe və GaSe kristallarında terahers şualanması müşaidə olunub.

Açar sözlər: Terahers spektroskopiyası, uçuş zamanı , selenid, sulfid.