

## ACOUSTIC PROPERTIES OF TRIGLYCINE SULPHATE CRYSTALS WITH $\alpha$ -ALANINE IMPURITY

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Received December 12, 2025; accepted January 20, 2026

The frequency- and concentration-dependent absorption coefficients of longitudinal and transverse ultrasonic waves in pure and  $\alpha$ -alanine-doped triglycine sulfate ( $\alpha$ -TGS) crystals ( $\alpha$ -alanine content in crystals is: 5, 10, 15, 20, 25, 30, and 35 mol%) grown from solution at room temperature were studied. It is established that absorption coefficients of ultrasonic waves in  $\alpha$ -TGS crystals with L and S polarization with the increase in concentration of  $\alpha$ -alanine about 1.57 and 1.88 times decrease, and propagation velocity increases by 3.24 and 13.38%, respectively. Decrease of absorption coefficients of transverse ultrasonic waves caused by elastic scattering of phonons on impurity, i.e., decrease of  $\tau$ , at the corresponding dispersive properties of a phonon subsystem.

**Keywords:** Triglycine sulfate; Ultrasonic waves; Velocity of elastic wave; Absorption coefficient; Ferroelectric; Phase transition; Phonon;  $\alpha$ -alanine

**PACS:** 43.35. Ac, 43.35. Bf, 43.35. Cg, 77.84. -s

### INTRODUCTION

Triglycine sulfate (TGS) crystals are model and well-studied ferroelectric crystals [1, 2]. The first works devoted to growing and studying the physical properties of these crystals date back to the middle of the last century [3] and continue to the present day [4-9]. Interest in these crystals is mainly associated with the possibility of creating radiation detectors based on them over a wide spectral range, including the infrared, as substrates for vidicon materials, good prospects for use in various microelectronics devices, and pyroelectric sensors [10, 11], etc.

The dielectric, structural, acoustic and other properties of pure [12] and doped with various impurities [13-17] TGS crystals have been studied quite well. It is known that impurity atoms in crystals affect their acoustic properties, in particular the absorption coefficient and the speed of propagation of ultrasonic waves [18, 19]. Interest in these studies is driven by both the study of the influence of impurities on the ultrasonic properties of TGS crystals and the great potential for their practical use. On the other hand, TGS crystals are a convenient model system for studying the role of structural imperfections in the physical properties of ferroelectrics [14, 15, 17]. The introduction of impurities makes it possible to regulate a number of properties of crystals, and in particular, and most importantly, it allows one to stabilize spontaneous polarization due to the formation of internal fields [17, 20]. In this regard, it is of significant interest to study the acoustic properties of TGS crystals doped with  $\alpha$ -alanine ( $\alpha$ -TGS), namely, the effect of impurities on the propagation and attenuation speeds of ultrasonic waves in them.

This work presents the results of studies of the frequency and concentration dependences of the absorption coefficient of longitudinal and transverse ultrasonic waves in nominally pure and  $\alpha$ -alanine-doped  $\alpha$ -TGS crystals ( $\alpha$ -alanine content in the crystals: 5, 10, 15, 20, 25, 30, and 35 mol% %) grown from solution at room temperature, i.e. in the ferroelectric phase. Indeed, the  $\alpha$ -TGS crystal is one of the currently very well-known model ferroelectrics with a second-order phase transition, in which, due to the strong anisotropy of the relaxation mechanism of absorption and dispersion of ultrasonic waves [21], both relaxation and actual critical (fluctuation) phenomena occur. However, in most studies [22], ultrasonic anomalies in the region of structural phase transition are considered, while the effect of  $\alpha$ -alanine on ultrasonic properties in  $\alpha$ -TGS crystals in the ferroelectric phase has not been studied.

### MATERIAL AND METHODS

To carry out ultrasonic studies, an acousto-optical installation was used [23], operating in a pulsed mode and consisting of a system for excitation of ultrasonic waves and a system for recording continuous He-Ne laser radiation with a wavelength  $\lambda_0 = 632.8 \text{ nm}$  diffracted by ultrasonic waves. High-frequency longitudinal and transverse ultrasonic waves with frequencies in the range of 0.2-1.2 GHz were excited using quartz piezo transducers  $x$  or  $y$  slice, 70-100  $\mu\text{m}$

thick. The studied samples were parallelepipeds with an average size of  $8 \times 9 \times 10$  mm, oriented along the main crystallographic axis [001]. The attenuation coefficient of ultrasonic waves was determined by the Bragg diffraction of light on ultrasonic waves [23] based on the dependence of the intensity of diffracted light  $I$  and on the distance to the transducer  $x_1$  and  $x_2$  using the following formula:

$$\alpha, \text{ cm}^{-1} = \frac{1}{x_1 - x_2} 10 \lg \frac{I(x_1)}{I(x_2)}. \quad (1)$$

The accuracy in determining the attenuation coefficient was  $\sim 5\%$ . At the same time, the speed of propagation of ultrasonic waves was determined from measurements of the angle of Bragg diffraction of light by the following formula:

$$v = \frac{\lambda_0 \nu}{2 \sin \theta}, \quad (2)$$

where  $\lambda_0$  – is the wavelength of laser radiation,  $\nu$  – is the frequency of ultrasonic waves, and  $\theta$  – is the Bragg diffraction angle.

## RESULTS AND DISCUSSIONS

The experimentally obtained results of the attenuation coefficient and speed of ultrasonic waves propagating along the [001] crystallographic direction at a frequency of 500 MHz in doped and nominally pure  $\alpha$ -TGS crystals at room temperature are given in Table 1.

**Table 1.** Concentration dependence of the attenuation coefficient and propagation velocity of ultrasonic waves to TGS crystals at temperature  $T = 300$  K.

<i><math>\alpha</math>-alanine content in TGS, %</i>	<i><math>q \parallel z</math></i>		<i><math>v \times 10^5</math> cm/s</i>	
	<i><math>\alpha_l</math></i>	<i><math>\alpha_s</math></i>	<i><math>v_l</math></i>	<i><math>v_s</math></i>
<i>TGS pure</i>	22.2	35	3.858	1.985
<i><math>\alpha</math>-TGS, 5 %</i>	20.7	32.1	3.876	2.032
<i><math>\alpha</math>-TGS, 10 %</i>	19.1	29	3.898	2.078
<i><math>\alpha</math>-TGS, 15 %</i>	18.1	26.1	3.925	2.110
<i><math>\alpha</math>-TGS, 20 %</i>	17.2	23.1	3.951	2.140
<i><math>\alpha</math>-TGS, 25 %</i>	16.2	21.5	3.962	2.171
<i><math>\alpha</math>-TGS, 30 %</i>	15.4	20.1	3.973	2.203
<i><math>\alpha</math>-TGS, 35 %</i>	14.3	18.5	3.983	2.250

As can be seen from Table 1, the attenuation coefficients of ultrasonic waves with L and S polarization decrease by approximately 1.57 and 1.88 times, respectively, with increasing  $\alpha$ -alanine concentration, and the propagation speed increases by 3.24 and 13.38 %, respectively. The concentration dependence curves of the absorption coefficient and the propagation speed of ultrasonic waves are described by curves of the following form:

$$\alpha_l = -60.61346 + \frac{82.8359}{(1 + (c/533.7909) \cdot 0.830231)};$$

$$\alpha_s = 9.232921 + \frac{25.733609}{(1 + (c/23.32901) \cdot 1.382119)};$$

$$v_l = 4.019122 - \frac{0.160101}{(1 + (c/17.87642) \cdot 1.819346)};$$

$$v_s = 4.022538 - \frac{2.037637}{(1 + (c/265.7397) \cdot 0.9357489)};$$

where  $c$  the  $\alpha$ -alanine content is in %.

The study showed that in pure TGS crystals the attenuation of ultrasonic waves is proportional to the square of the frequency, and in  $\alpha$ -TGS the attenuation of longitudinal ultrasonic waves decreases by approximately 20 %, and the nature of the frequency dependence remains quadratic. The attenuation coefficient of transverse ultrasonic waves is very sensitive to the admixture of  $\alpha$ -alanine and in some samples the attenuation coefficient is almost halved compared to pure TGS crystals. As for the frequency dependence of the attenuation of these waves, it is observed that it is proportional to the square of their frequency. It was also shown that the attenuation of ultrasonic waves in TGS and  $\alpha$ -TGS crystals is practically independent of temperature, with the exception of the region of the ferroelectric phase transition [30] observed in them at a temperature of  $T = 54^\circ\text{C}$ . In Figure 1 shows the concentration dependence of the attenuation of ultrasonic waves in  $\alpha$ -TGS crystals.

As can be seen from Fig. 1, with an increase nonlinear in the concentration of  $\alpha$ -alanine in TGS crystals, the attenuation coefficient of ultrasonic waves of both polarizations decreases (Fig. 2).

It should be assumed that the decrease in the attenuation coefficient of ultrasonic waves in nominally pure and doped  $\alpha$ -TGS ferroelectric crystals is associated with a decrease in the phonon relaxation time [20].

A theoretical consideration of the attenuation of longitudinal and transverse elastic waves in crystals with impurities was carried out in [24], where, under the condition  $\Omega\tau \ll 1$ , for crystals with impurities, the relaxation time is taken as:

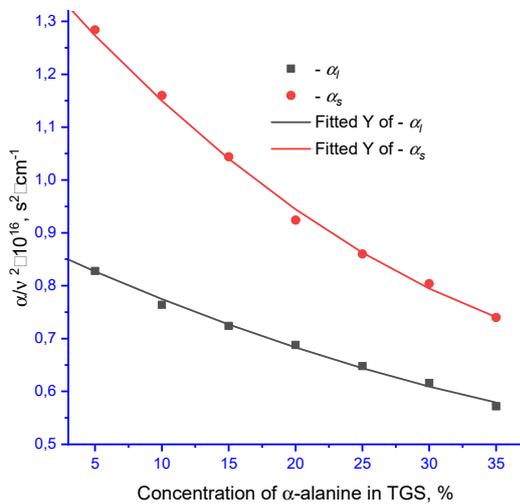
$$\tau = \left( \tau_i^{-1}(k_T) + \tau_l^{-1}(k_T) \right)^{-1}, \quad (3)$$

where,  $k_T$  – is the wave vector of thermal phonons;  $\tau_i$  – is the relaxation time associated with inelastic phonon scattering;  $\tau_l$  – is the relaxation time associated with elastic scattering by impurities.

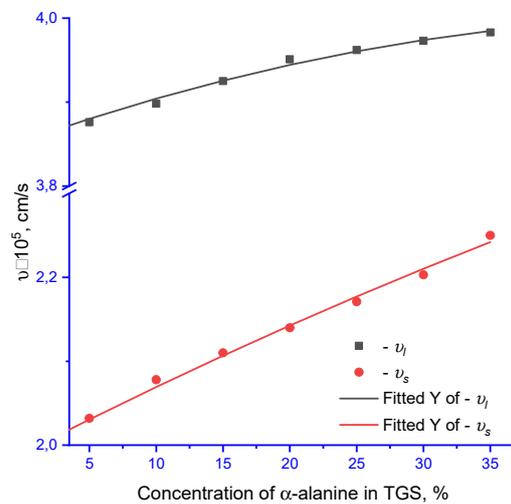
Taking into account the relationship between the indicated relaxation times, an expression for determining the attenuation coefficient of transverse elastic waves is obtained in the following form:

$$\alpha = \beta(\Omega^2 T) \rho v^2 \sum_{\vec{k}, j} C(\vec{k}, j) \tau(\vec{k}, j) \gamma^2(\vec{k}, j), \quad (4)$$

where,  $\beta$  – is a numerical factor of the order of unity, the value of which depends on the type of local equilibrium distribution of thermal phonons;  $\Omega$  and  $v$  – frequency and speed of ultrasonic waves;  $C(\vec{k}, j)$  – heat capacity of the  $(\vec{k}, j)$  phonon branch;  $\gamma$  – effective anharmonicity constant, depending on the degree of order of the crystal.



**Figure 1.** Concentration dependence of  $\alpha/v^2$  values in  $\alpha$ -TGS for longitudinal and shear vibrations



**Figure 2.** Concentration dependence of the propagation speed of longitudinal and shear ultrasonic waves in  $\alpha$ -TGS

A theoretical consideration of the attenuation of longitudinal and transverse elastic waves in crystals with impurities was carried out in [24], where, under the condition  $\Omega\tau \ll 1$ , for crystals with impurities, the relaxation time is taken as:

$$\tau = \left( \tau_i^{-1}(k_T) + \tau_l^{-1}(k_T) \right)^{-1}, \quad (3)$$

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### CONCLUSIONS

It has been established that with an increase in the concentration of  $\alpha$ -alanine in TGS crystals, the attenuation coefficient of ultrasonic waves of both polarizations decreases nonlinearly, while the speed of propagation increases nonlinearly.

The concentration dependences of the attenuation coefficient and propagation velocity of ultrasonic waves of both polarizations in TGS crystals with an admixture of  $\alpha$ -alanine can be approximated by the following expressions:

$$\alpha_l = -60.61346 + \frac{82.8359}{(1 + (c/533.7909) \cdot 0.830231)};$$

$$\alpha_s = 9.232921 + \frac{25,733609}{(1 + (c/23.32901) \cdot 1.382119)};$$

$$v_l = 4.019122 - \frac{0.160101}{(1 + (c/17.87642) \cdot 1.819346)};$$

$$v_s = 4.022538 - \frac{2.037637}{(1 + (c/265.7397) \cdot 0.9357489)},$$

where  $c$  the  $\alpha$ -alanine content is in %.

It has been established that the attenuation coefficients of ultrasonic waves with L and S polarization decrease by approximately 1.57 and 1.88 times, respectively, with increasing  $\alpha$ -alanine concentration, and the propagation speed increases by 3.24 and 13.38 %, respectively.

According to (4), it follows that a decrease in the attenuation coefficient of transverse ultrasonic waves can be due to elastic scattering of phonons on impurities, i.e., a decrease in, with the corresponding dispersion properties of the phonon subsystem. Thus, according to equation (4), a decrease in  $\alpha_l$ , and  $\alpha_s$  may be due to a decrease in the effective anharmonicity constant  $\gamma$  of crystal lattice vibrations under the influence of defects.

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**АКУСТИЧНІ ВЛАСТИВОСТІ КРИСТАЛІВ ТРИГЛІЦИН СУЛЬФАТУ З ДОМІШКОЮ  $\alpha$  АЛАНІНУ**  
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Досліджено частотну та концентраційну залежність коефіцієнта поглинання поздовжніх та поперечних ультразвукових хвиль у чистих та легуваних  $\alpha$ -аланін-тригліцинсульфаті ( $\alpha$ -TGS) кристалах (вміст  $\alpha$ -аланіну в кристалах: 5, 10, 15, 20, 25, 30 та 35 мол.%), вирощених з розчину за кімнатної температури. Встановлено, що коефіцієнти поглинання ультразвукових хвиль у кристалах  $\alpha$ -TGS з L та S поляризацією зі збільшенням концентрації  $\alpha$ -аланіну приблизно в 1,57 та 1,88 раза зменшуються, а швидкість поширення збільшується на 3,24 та 13,38% відповідно. Зменшення коефіцієнтів поглинання поперечних ультразвукових хвиль, спричинене пружним розсіюванням фононів на домішці, тобто зменшення  $\tau$ , при відповідних дисперсійних властивостях фононної підсистеми.

**Ключові слова:** тригліцинсульфат; ультразвукові хвилі; швидкість пружної хвилі; коефіцієнт поглинання; сегнетоелектрик; фазовий перехід; фонон,  $\alpha$ -аланін