
THE PROBLEM OF DISPERSION OF NONLINEAR SUSCEPTIBILITIES IN CRYSTALS

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The structure of the functions $\chi_{ijk}(E)$ of nonlinear crystals is considered. These functions represent an interlink between the microscopic quantum-mechanical description (band spectra) and macroscopic (experimental) parameters of materials. Their study is the central problem of nonlinear optical materials science. In this work, we investigate such nonlinear crystals as KTP (KTiOPO_4), proustite (Ag_3AsS_3), etc. The matrix elements of the corresponding dipole transitions and the parameters of nonlinear optical oscillators are determined. The possibilities of purposeful variations of nonlinear crystal parameters by means of doping, changes of the composition of solid solutions, and radiation processing are analyzed. This opens new prospects for the development of programmable technologies of synthesis of new nonlinear optical materials for laser radiation converters.

1. Introduction

In modern materials science, a new promising area which can be called nonlinear optical materials science has appeared. Its purpose consists in the synthesis and investigation of crystals with improved nonlinear optical characteristics. Till recently, such investigations were generally of empirical character and concerned elementary compounds [1]. At the same time, the basic representatives of such objects have a complicated crystal-chemical structure (for example, KTiOPO_4 , LiB_3O_5 , $\text{BaNaNb}_5\text{O}_{15}$, etc.), which complicates the implementation of the corresponding *a priori* estimates. For the majority of these compounds, there are no calculations of bands. Even if such attempts were sometimes made, their level did not provide the agreement with experimental results, first of all with optical and photoemission spectra. That is why, it was prematurely up to now to discuss the possibility of a theoretical prognosis of nonlinear optical parameters.

The influence of local disturbances of various kinds (impurities, defects, excitons, or surface states) on nonlinear susceptibilities was not investigated as well. At the same time, the doping and the structure ordering along with creation of solid solutions represent the main directions of the purposeful improvement of the characteristics of these materials. All estimations

including the problem of choice of processing modes require the study of the specificities of chemical bonds and the electron density distribution. That is why a reliable search for the corresponding materials calls for the theoretical investigation of electron parameters. Based on the solution of these two interrelated problems, one can solve the central task – the purposeful control over properties and synthesis of nonlinear materials with specified parameters.

In the proposed work, we present a complex approach to the prognosis of nonlinear optical susceptibilities. It includes, on the one hand, the theoretical calculation of the band structure, electron density, and functions $\chi_{ijk}(E)$ and, on the other, the experimental correction of model parameters aimed at their maximal agreement with experiment. First of all, this concerns the nonlinear materials being in good market demand. In our consideration, we fix attention on such nonlinear crystals as KTP and proustite.

2. Optical Spectra, Band Structure, and Electron Density Distribution

A complex crystal-chemical structure of the mentioned materials complicates the problem of first-principles calculations of their band structure from the viewpoint of a reliable agreement of the calculated structure with experimental data. In order to overcome this difficulty, we have proposed an approach whose essence consists in that the model parameters of band calculations are corrected with the use of the data obtained on the basis of the measured structure of polarization spectra in a wide energy range in the depth of the fundamental absorption band and on the basis of their group-theoretic identification. As a rule, we obtained such data with the help of synchrotron radiation sources.

Figure 1 shows the reflection polarization spectra $R(E)$ of a single-crystal KTP sample in the (XZ) cross section, as well as the dielectric functions $\varepsilon_1(E)$ and $\varepsilon_2(E)$ calculated by the Kramers–Kronig relations. The spectra are characterized by a significant anisotropy that

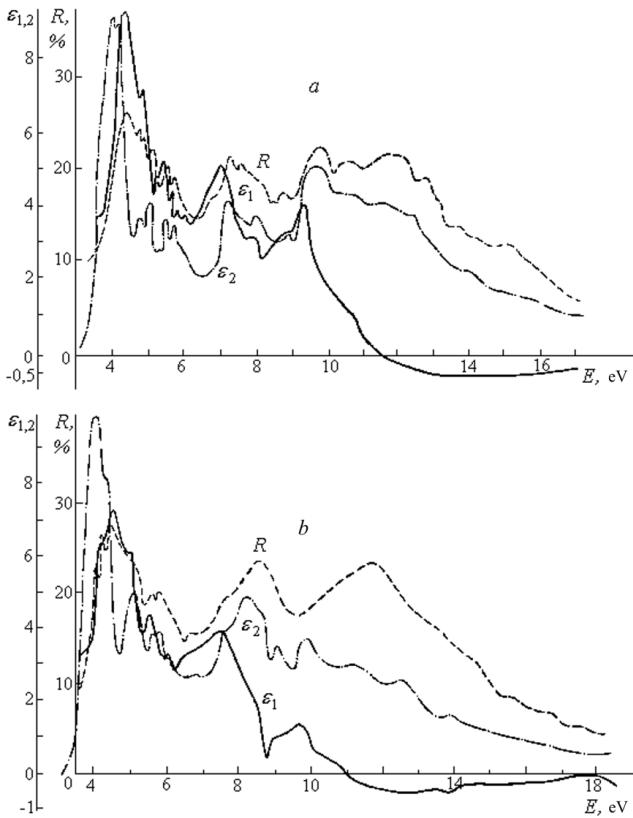


Fig. 1. Spectra of reflection $R(E)$, real $\varepsilon_1(E)$ and imaginary $\varepsilon_2(E)$ parts of the complex permittivity of KTiOPO₄ crystals for the light polarizations $\vec{E} \parallel \vec{X}$ (a) and $\vec{E} \parallel \vec{Z}$ (b)

can be explained in view of the symmetry selection rules (the space group of the KTiOPO₄ crystal is $C_{2v}^9 = Pna2_1$). According to our group-theoretic analysis, the following interband transitions are allowed for the Γ point of the Brillouin zone (BZ):

$\vec{E} \vec{X}$	$\vec{E} \vec{Z}$
$\Gamma_3 \rightarrow \Gamma_1$	$\Gamma_1 \rightarrow \Gamma_1$
$\Gamma_2 \rightarrow \Gamma_4$	$\Gamma_2 \rightarrow \Gamma_2$
	$\Gamma_3 \rightarrow \Gamma_3$
	$\Gamma_4 \rightarrow \Gamma_4$

Using the consistency conditions and the generally accepted notations of high-symmetry points of the Brillouin zone [2], we obtained the system of allowed transitions that explains the anisotropic structure of the KTP spectra:

$$\Gamma_1 \rightarrow \Lambda_1 \rightarrow Z_1 \rightarrow U_1 \rightarrow X_1 \rightarrow D_1 \rightarrow S_1,$$

$$\Gamma_3 \Rightarrow \Lambda_3 \Rightarrow Z_3 \Rightarrow U_1 \Rightarrow X_1 \Rightarrow D_3 \Rightarrow S_{2,4},$$

$$\Gamma_3 \rightarrow \Lambda_3 \rightarrow Z_3 \rightarrow U_1 \rightarrow X_1 \rightarrow D_1 \rightarrow S_1$$

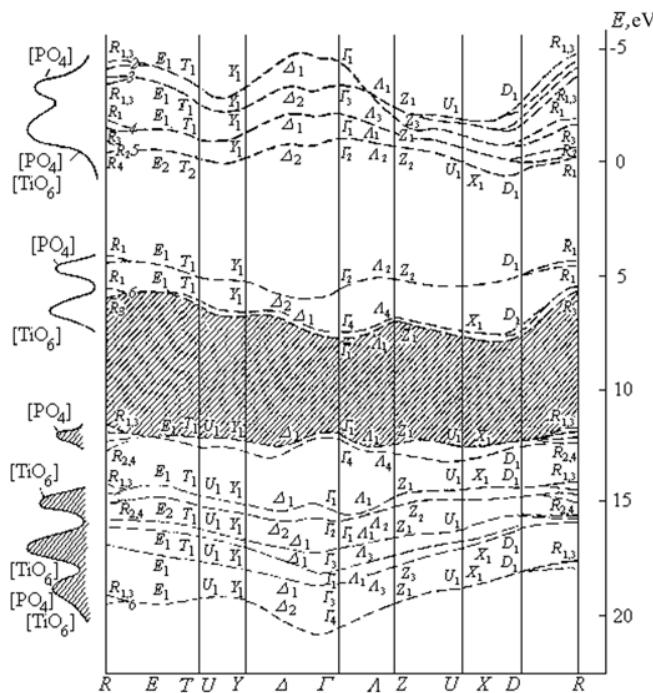


Fig. 2. Band structure of KTiOPO₄ crystal

$$\Gamma_4 \rightarrow \Lambda_4 \rightarrow Z_4 \rightarrow U_1 \rightarrow X_1 \rightarrow D_2 \rightarrow S_{2,4}.$$

When calculating the band structure of KTP, we took the basis functions 4d Ti, 2p O, and 2s,p P into account. With regard for the group-theoretic technique of projection operators, the dimensions of the secular equation of the Hermitian matrix did not exceed 16×16 . The contribution of the 2s O and 4s K terms was calculated in the framework of perturbation theory. As was noted above, the parameters of the secular equations were adjusted by the scale corrections in order to obtain the maximal agreement of the calculated bands with the experimental function $\varepsilon_2(E)$.

Figure 2 presents the band structure of a nonlinear KTiOPO₄ crystal which we have obtained for the first time. The genetic origin of the specific band states and the corresponding electron density distributions are depicted on the left of the figure. These distributions were obtained on the basis of the KTP eigenfunctions. The calculation technique is described in [3] in detail.

The similar investigations were performed for Ag_3AsS_3 , $\beta\text{-BaB}_2\text{O}_4$, LiB_3O_5 , LiNbO_3 , and other nonlinear crystals. For example, Figure 3 demonstrates the $R(E)$, $\varepsilon_1(E)$, and $\varepsilon_2(E)$ spectra of Ag_3AsS_3 crystal (space group $C_{3v}^6 = R3c$). From the symmetry analysis, we obtained the allowed transitions for the $\vec{E} \parallel \vec{C}$ and

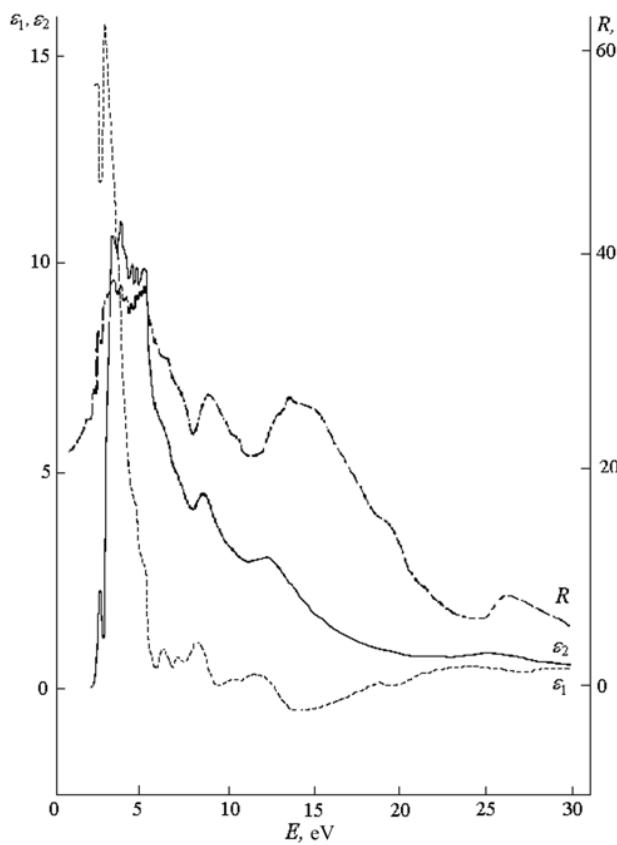


Fig.3. Spectra of reflection $R(E)$, real $\epsilon_1(E)$ and imaginary $\epsilon_2(E)$ parts of the permittivity of Ag_3AsS_3 crystal for the polarization $\vec{E} \perp \vec{C}$

$\vec{E} \perp \vec{C}$ polarizations in the dipole approximation (see Table 1).

The band structure of proustite is shown in Fig.4.

Table 1. Allowed band-band transitions in Ag_3AsS_3

Polarization	Points of the Brillouin zone		
	Γ	Λ, Z, Y	Σ
$\vec{E} \parallel \vec{C}$	$\Gamma_1 \rightarrow \Gamma_1$ $\Gamma_2 \rightarrow \Gamma_2$ $\Gamma_3 \rightarrow \Gamma_3$ $\Gamma_4 \rightarrow \Gamma_4$ $\Gamma_5 \rightarrow \Gamma_5$ $\Gamma_6 \rightarrow \Gamma_6$	$\Lambda_1 \rightarrow \Lambda_1$ $\Lambda_2 \rightarrow \Lambda_2$ $\Lambda_3 \rightarrow \Lambda_3$ $\Lambda_4 \rightarrow \Lambda_4$ $\Lambda_5 \rightarrow \Lambda_5$ $\Lambda_6 \rightarrow \Lambda_6$	$\Sigma_1 \rightarrow \Sigma_1$ $\Sigma_2 \rightarrow \Sigma_2$ $\Sigma_3 \rightarrow \Sigma_3$ $\Sigma_4 \rightarrow \Sigma_4$
$\vec{E} \perp \vec{C}$	$\Gamma_1 \rightarrow \Gamma_3$ $\Gamma_2 \rightarrow \Gamma_3$ $\Gamma_3 \rightarrow \Gamma_1 + \Gamma_2 + \Gamma_3$ $\{\Gamma_4 + \Gamma_5\} \rightarrow \Gamma_6$ $\Gamma_6 \rightarrow \{\Gamma_4 + \Gamma_5 + \Gamma_6\}$	$\Lambda_1 \rightarrow \Lambda_3$ $\Lambda_2 \rightarrow \Lambda_3$ $\Lambda_3 \rightarrow \Lambda_1 + \Lambda_2 + \Lambda_3$ $\Lambda_4 \rightarrow \Lambda_6$ $\Lambda_5 \rightarrow \Lambda_6$ $\Lambda_6 \rightarrow \Lambda_4 + \Lambda_5 + \Lambda_6$	$\Sigma_1 \rightarrow \Sigma_1$ $\Sigma_2 \rightarrow \Sigma_1 + \Sigma_2$ $\Sigma_3 \rightarrow \Sigma_3 + \Sigma_4$ $\Sigma_4 \rightarrow \Sigma_3 + \Sigma_4$

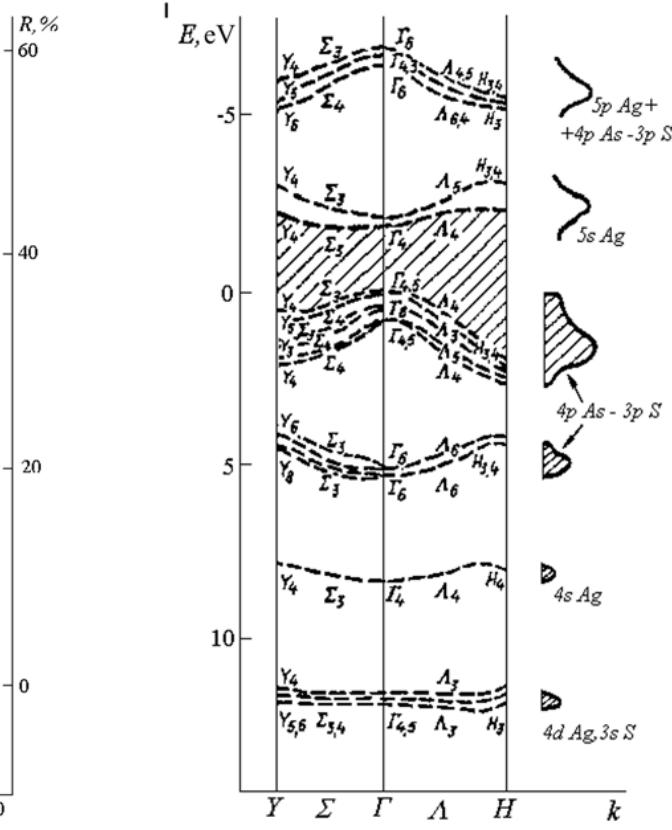
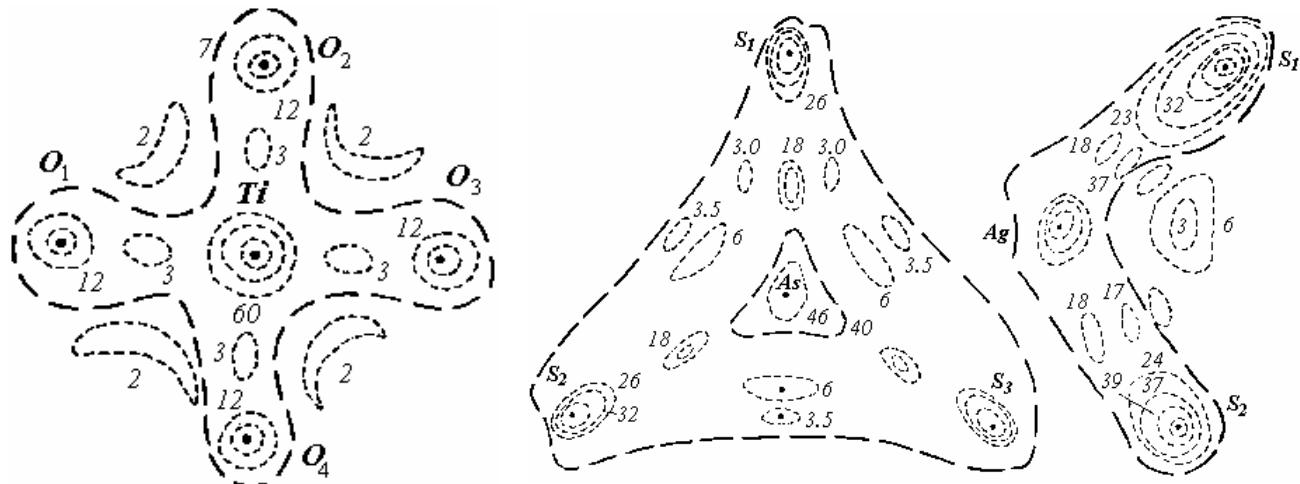


Fig.4. Band structure of proustite

The interpretation of band structures and the calculation of electron densities allowed us to simulate the space distribution of electron charges for the most principal clusters of the crystals under study. These patterns give a possibility to estimate the nature of chemical bonds and, therefore, their influence on the functions of nonlinear susceptibilities.

Fig. 5. Contours of the electron density in a $[TiO_6]$ cluster of $KTiOPO_4$ single crystalsFig. 6. Electron density distribution in the $[AsS_3]$ and $[S\text{-}Ag\text{-}S]$ clusters of Ag_3AsS_3 single crystals

3. Nonlinear Susceptibilities and Nature of Chemical Bonds

Figures 5 and 6 demonstrate the diagrams of the electron density distributions for $[TiO_6]$ of KTP crystal and $[AsS_3]$ and $[S\text{-}Ag\text{-}S]$ clusters of Ag_3AsS_3 crystal. Based on these data, we determined the covalence parameters f_h as the ratio of the electron charge at the middle of a chosen chemical bond (ρ_c) to the averaged charge near atoms (ρ_a): $f_h = \rho_c/\rho_a$. For $[TiO_6]$, we obtained $f_h = 0.89$; for the $[AsS_3]$ triads, $f_h = 0.85$; and for the $[S\text{-}Ag\text{-}S]$ chains oriented along the Z -axis of proustite, $f_h = 0.48$. The corresponding bond ionicities are as follows: $f_i^{[TiO_6]} = 0.11$; $f_i^{[AsS_3]} = 0.15$; and $f_i^{[S\text{-}Ag\text{-}S]} = 0.52$.

By the example of three chosen clusters, we will clarify the factors that determine the corresponding charge distributions. As one can see from Fig. 5, the local electron charge distribution in a $[TiO_6]$ cluster is caused by the elongation of the oxygen p -orbitals in the direction of the cation, whereas the $p\text{-}p$ anion bonds are weakly hybridized. In the case of an $[AsS_3]$ cluster, it is clear from Fig. 6 that the contribution of the $S\text{-}S$ component is essentially lower than that of the $Ag\text{-}S$ component of the chemical bond, which is explained by peculiarities of the geometric (pyramidal) structure of the cluster. Finally, a completely different pattern of the charge distribution is characteristic of the $[S\text{-}Ag\text{-}S]$ chain cluster. The covalence degree for these bonds is the lowest, because the charge localization near the silver ion is rather high. At the same time, the presence of two elongated charge couplings testifies

to the defining contribution of the hybridized $5sAg\text{-}3pS$ bonds.

In order to clarify the effect of the chemical bond nature on nonlinear susceptibilities, we traced the variation of the $\chi_{ijk}(E)$ function under conditions of the substitution of potassium ions in KTP. Changing the composition of a substitutional solid solution and thus varying the quantity f_h , we established that, at $f_h \cong 0.17$, the quantity χ_{ijk} in the neighborhood of the basic maximum essentially increased. This fact forces to perform a more detailed investigation of the dependence of $\chi_{ijk}(E)$ on the chemical bond parameters.

Our specially performed investigations of the system of solid solutions $Ag_3As_xSb_{1-x}S_3$ demonstrated [4] that, at $x \cong 0.1$, χ_{ijk} reaches its maximum at the oscillation frequency of a neodymium laser.

As for other nonlinear crystals, in particular borate oxides, the dominant contribution to the nonlinear susceptibility in these materials is made by the $sB\text{-}pO$ bonds. Varying the geometry of chemical bonds or substituting components, one can influence the susceptibility χ_{ijk} in the necessary spectral region. As we have demonstrated by the example of these materials, a change of the relative position of clusters essentially influences the nonlinear coefficients. In particular, the nonlinear susceptibilities in LiB_3O_5 and $\beta\text{-}BaB_2O_4$ are larger by orders of magnitude than those in lithium tetraborate ($Li_2B_4O_7$) single crystals due to the reorientation of boric clusters.

Quantum-chemical estimates made in [5] yield the optimal value of the covalence parameter $f_h \approx 0.17$ –

0.20 in the case of localization of the nonlinear susceptibilities.

4. Dispersion of Nonlinear Susceptibilities

The problem of the dispersion of $\chi_{ijk}(E)$ is the least investigated one in the physics of nonlinear crystals. It is due to the fact that, in the same way as in the calculations of electron density distributions, a preliminary information on the band structure of crystals should be available.

The electron component of the second-rank tensor of nonlinear susceptibility was calculated according to the standard technique of perturbation theory:

$$\begin{aligned} \chi_{ijk}(E) = & \frac{e^3 N}{2!} \sum_{\mu} G_{ijk}^{(\mu)} \sum_k \sum_{\alpha, \beta} \hat{P} \left[\frac{\langle 0 | i | \alpha \rangle \langle \alpha | j | \beta \rangle \langle \beta | k | 0 \rangle}{(2E + E_{\alpha})(E + E_{\beta})} + \right. \\ & \left. + \frac{\langle 0 | j | \alpha \rangle \langle \alpha | i | \beta \rangle \langle \beta | k | 0 \rangle}{(E_{\alpha} - 2E)(E_{\beta} - E)} + \frac{\langle 0 | j | \alpha \rangle \langle \alpha | k | \beta \rangle \langle \beta | i | 0 \rangle}{(E + E_{\beta})(E_{\alpha} - E)} \right], \end{aligned} \quad (1)$$

where \hat{P} denotes the commutator, i.e. the operator of permutations for all band states, $G_{ijk}^{(\mu)}$ is the geometric factor, E and $2E$ are the interband and double resonances with participation of virtual states, 0 is the index of the actual lower (filled) band states (V -zones), α and β are the indices of the upper (empty) band states (C -zones), $\langle \alpha | i | \beta \rangle$ is the dipole matrix element of the transition between the α -th and β -th band states:

$$\begin{aligned} \langle \alpha | i | \beta \rangle = & \int U_{\alpha, \vec{k}} \exp[i\vec{k}(\vec{r}_i - \vec{r})] \vec{e}_{\alpha} A_i \vec{e}_{\beta} U_{\beta, \vec{k}}^* \times \\ & \times \exp[-i\vec{k}(\vec{r}_i - \vec{r})] d^3 \vec{r} = |U_{\alpha, \vec{k}} U_{\beta, \vec{k}}^*| \vec{k} \vec{e}_{\alpha} \vec{e}_{\beta} A_i, \end{aligned} \quad (2)$$

$\vec{e}_{\alpha}, \vec{e}_{\beta}$ stand for the vectors of the corresponding orbitals, and A_i is the vector-potential of the incident radiation.

As one can see from (1), $\chi_{ijk}(E)$ is determined by three dominant factors: the matrix elements of dipole transitions, collection of virtual states, and geometric factor of basic clusters. It is easy to see that the contributions of the imaginary part of the permittivity $\varepsilon_2(E)$ in the region of the interband and double resonances, i.e. $\varepsilon_2(E)$ and $\varepsilon_2(2E)$, will be defining for the structure of the $\chi_{ijk}(E)$ function.

The real part of the nonlinear susceptibility is derived with the help of the Kramers-Kronig relations.

To calculate (1), we used the tetrahedron technique, i.e. the integration was performed by means of the

partition of segments of the irreducible BZ into parallelepipeds which were divided into still smaller tetrahedral cells with energy derivatives obtained from the band dispersion $E(\vec{k})$. In this case, it was assumed that the matrix elements inside each cell are constant. The accuracy of calculations increased with decrease in the partition step. The summation was performed over many (above 90) points of the BZ. All computer calculations of the matrix elements were carried out with the use of the Gaussian form of wave functions. According to the analysis performed on the basis of quantitative estimates of the data, the most important as for the contribution to the nonlinear susceptibilities are two conduction bands (C , C') and two valence bands (V , V'). The primed notations correspond to virtual band states.

The results of calculations of the most important matrix elements at point Γ of the BZ for the nonlinear crystals under study are presented in Table 2. These results indicate the contribution of the specific subbands to the nonlinear susceptibilities.

5. Parameters and Nature of Nonlinear Optical Oscillators

The dispersions of the second-order nonlinear susceptibilities for KTiOPO₄, Ag₃AsS₃, and LiNbO₃ crystals are presented in Fig. 7. The function $\chi_{ijk}(E)$ for KTP that covers the region below 13 eV has three dominant peaks at 4.19, 5.06, and 5.45 eV caused by transitions between the p -orbitals of oxygen, which form the top of the valence band, and the C -zone. The intermediate states 3pP and 2pO serve as virtual ones. The other maxima are less intense.

T a b l e 2. Matrix elements for the nonlinear crystals

Crystal	Transition type	Matrix element, rel.un.
KTiOPO ₄	$V - C'$	0.287
	$V - C$	0.376
	$V' - C'$	0.289
	$C - C'$	0.065
Ag ₃ AsS ₃	$V - C'$	0.205
	$V - C$	0.312
	$V' - C'$	0.223
	$C - C'$	0.056
LiNbO ₃	$V - C'$	0.427
	$V - C$	0.539
	$V' - C'$	0.323
	$C - C'$	0.101

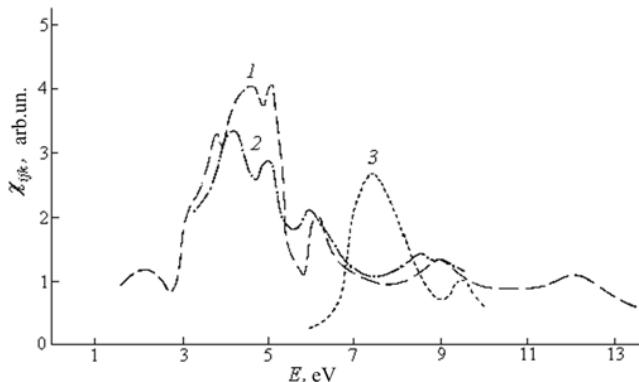


Fig. 7. Dispersions of the nonlinear susceptibilities $\chi_{ijk}(E)$ of Ag_3AsS_3 (1), KTiOPO_4 (2), and LiNbO_3 (3) single crystals

The typical structures of the functions $\chi_{ijk}(E)$ are also observed for other crystals. In this connection, they were resolved into partial components. In the framework of the model of nonlinear oscillators, we determined the oscillator strengths by the relation [6]

$$F = \frac{\pi V \mu c^2}{e^2 N_{ef} N} E_{0i} H_i \chi_{ijk}^{\max}, \quad (3)$$

where V is the volume of a unit cell, H_i is the half-width of the i -th component of the function $\chi_{ijk}(E)$, and $\frac{\pi V \mu}{e^2 N} \cong \text{const}$. All calculations were carried out in relative units. The obtained results are given in Tables 3 and 4, where η_i indicates the partial contribution of the corresponding nonlinear oscillators to the nonlinear susceptibility.

As one can see from Table 3, the dominant contribution to the nonlinear susceptibility of Ag_3AsS_3 crystals is made by the first long-wave oscillator ($E = 4.56$ eV). It is related to transitions from the hybridized V -zone (4pAs – 3pS) to the C -zone 5s Ag, as well as the 2*E*-contribution due to the resonance 4p As levels. The other transitions are characterized by small matrix elements. A certain increase of $\chi_{ijk}(E)$ in the region close to 12.1 eV results from the transitions with participation of the 4dAg, 3sS, 5pAg, 4pAs, and 3pS band states.

T a b l e 3. Parameters of nonlinear optical oscillators for Ag_3AsS_3

E_{0i} , eV	H_i , eV	$\chi_{ijk}^{(\max)}$	F_i , rel.un.	η_i , %
4.56	2.30	4.06	42.58	46.8
6.16	0.52	2.02	6.47	7.1
7.06	0.62	0.93	4.08	4.5
8.07	0.58	0.76	3.56	3.9
8.96	1.11	0.84	8.35	9.2
11.01	0.56	0.63	3.88	4.3
12.12	1.05	0.91	11.58	12.7

A characteristic feature of KTiOPO_4 single crystals consists in that their nonlinear susceptibility is determined by two oscillators – the first and third ones (Table 4). The first oscillator (4.19 eV) represents the superposition of the E -transition from the top of the $[\text{PO}_4]$ valence band to the 3d band of titanium that forms the bottom of the conduction band, as well as transitions from the lower $[\text{TiO}_6]$ branch of the valence band. However, the most intense oscillator is formed due to the superposition of two lower subbands genetically formed by the hybridized $[\text{PO}_4]$ and $[\text{TiO}_6]$ subbands contributing to $2E$ -terms. The contribution of the upper C band is less by an order of magnitude. The largest matrix elements appear along the $Y-\Gamma$ and $\Gamma-Z$ directions of the Brillouin zone.

6. Conclusions

Investigations of the dispersion of nonlinear optical susceptibilities give a possibility to estimate the contribution of various clusters to their formation. Thus, technologists possess a reliable instrument for the purposeful control over nonlinear optical parameters.

Based on the proposed approach, one can estimate the partial contribution of a certain structural fragment to a specific component of the tensor of corresponding rank, determine the contributions of the electron and lattice subsystems, plan possible variations of the composition of the corresponding clusters and their geometric arrangement aimed at the optimization of nonlinear optical susceptibilities.

Of basic importance is the choice of the basic structural fragment, since variations of its composition can influence $\chi^{(NL)}$. In particular, for Ag_3AsS_3 , such fragments are $[\text{AsS}_3]$ triads and $[\text{S}-\text{Ag}-\text{S}]$ chains. Substituting cations or anions, one can change parameters of the chemical bond and, therefore, the susceptibility. A change of the structural ordering of clusters is especially efficient, which is confirmed by the example of borate oxides.

T a b l e 4. Parameters of nonlinear optical oscillators in KTiOPO_4

E_{0i} , eV	H_i , eV	$\chi_{ijk}^{(\max)}$	F_i , rel.un.	η_i , %
4.19	1.84	5.94	45.80	23.7
4.98	0.58	4.20	12.13	6.0
5.99	2.21	4.81	63.67	32.9
7.10	0.83	3.02	17.79	9.2
7.62	0.12	2.03	1.86	0.96
8.12	0.71	3.12	17.98	9.3
8.51	0.44	3.15	11.79	6.1
8.94	0.54	2.98	14.38	7.5

It is worth noting that not every crystal has a reserve to increase the corresponding susceptibility. For example, BeO single crystals are characterized by much lower susceptibilities than those of isostructural crystals CdS, CdSe, and SiC. The doping or the creation of radiation defects will result in the disorientation of the geometric structure, which makes such a procedure unpromising for the improvement of nonlinear properties.

The knowledge of the parameters of nonlinear oscillators gives a possibility to foresee a change of the susceptibility in the case of the formation of a solid solution by means of the substitution of an anion. The point is that the cation substitution results mainly in a change of the electron s-states. An increase of the corresponding susceptibility requires at least one additional oscillator, moreover an anisotropic one. For example, it can be an oscillator that genetically originates from p- or d-orbitals of the corresponding chemical element. For example, the dominant role for binary compounds belongs to an oscillator genetically related to the p-anion bonds.

I feel my pleasant duty to express the respect and the sincere gratitude to unforgettable Antonina Fedorivna Prykhot'ko. I am also grateful to Mykhailo Semenovych Brodyn and many friends from the Institute of Physics of the NAS of Ukraine.

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ПРОБЛЕМА ДИСПЕРСІЇ НЕЛІНІЙНИХ СПРИЙНЯТЛИВОСТЕЙ КРИСТАЛІВ

Я.О. Довгий

Р е з ю м е

Розглянуто структуру функції $\chi_{ijk}(E)$ нелінійних кристалів. Ці функції є зв'язуючою ланкою між мікроскопічним квантовомеханічним описом (зонні спектри) та макроскопічними (експериментальними) параметрами матеріалів. Їх вивчення є центральною проблемою нелінійнооптичного матеріалознавства. Досліджено такі нелінійні кристали, як КТР (КТіОРО₄), прустит (Ag₃AsS₃) та ін. Визначено величини матричних елементів відповідних дипольних переходів та параметри нелінійних оптических осциляторів. Проаналізовано можливість цілеспрямованих змін параметрів нелінійних кристалів методами легування, змін складу твердих розчинів, радіаційної обробки. Це відкриває перспективи розроблення програмованих технологій синтезу нових нелінійнооптических матеріалів для перетворювачів лазерного випромінювання.