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Influence of radiation heat transfer dynamics on crystal growth

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For analysis of the dynamics of forced and spontaneous transitions, a parametric 3D model of the black body radiation spectrum is proposed. Its orthogonal projections are the first and second-order signatures, in the configurations of which dynamic, energy and information aspects of the spectrum of thermal radiation are displayed. Their comparative analysis revealed significantly greater sensitivity of the parameters that reflect the dynamics of radiative transitions to small changes in temperature than the parameters of thermal radiation ($r^{*}(\lambda,T)$, R), which are used in optical pyrometry. It is shown that the influence of external factors on the dynamics of physical processes in crystalline boule is most evident in 3D models of the functional characteristics of the samples. The structure of the functional characteristics of the samples from crystalline boule contains information on the features of defects formation. It is shown that for the effective growth control, interrelated parameters characterizing the features of the dynamics of radiative heat transfer in a growth furnace are needed. The expansion of the number of interrelated parameters that reflect the dynamics of radiative heat transfer makes it possible to carry out the effective parametric control of crystal growth.

Keywords: crystal growth, reproducibility of a crystals properties, individuality of a functional characteristics, radiation heat transfer, geometrization, signature of a functional characteristic, trajectorie of dynamic events, decomposition of a spectrum, parametric control, parametric 3D model.

Для анализа динамики вынужденных и спонтанных переходов предложена параметрическая 3D-модель спектра излучения АЧТ. Ее ортогональные проекции являются сигнатурами 1-го и 2-го порядков, в конфигурациях которых отображаются динамические, энергетические и информационные аспекты спектра теплового излучения. Их сопоставительный анализ выявил существенно большую чувствительность параметров, которые отображают динамику излучательных переходов к малым изменениям температуры, чем параметры теплового излучения ($r^{*}(\lambda,T)$, R), которые используются в оптической пирометрии. Показано, что влияние внешних факторов на динамику физических процессов в кристаллической буле наиболее проявляется в 3D-моделях функциональных характеристик образцов. В структуре функциональных характеристик образцов из кристаллической були содержится информация, связанная с особенностями дефектообразования. Показано, что для эффективного управления ростом необходимы взаимосвязанные параметры, характеризующие особенности динамики радиационного теплообмена в ростовой установке. Расширение количества взаимосвязанных параметров, которые отражают динамику радиационного теплообмена, позволяет осуществить эффективное параметрическое управление ростом кристаллов.

Вплив динаміки радіаційного теплообміну на вирощування кристалів. *В.П.Мигаль, І.О.Клименко, Г.В.Мигаль.*

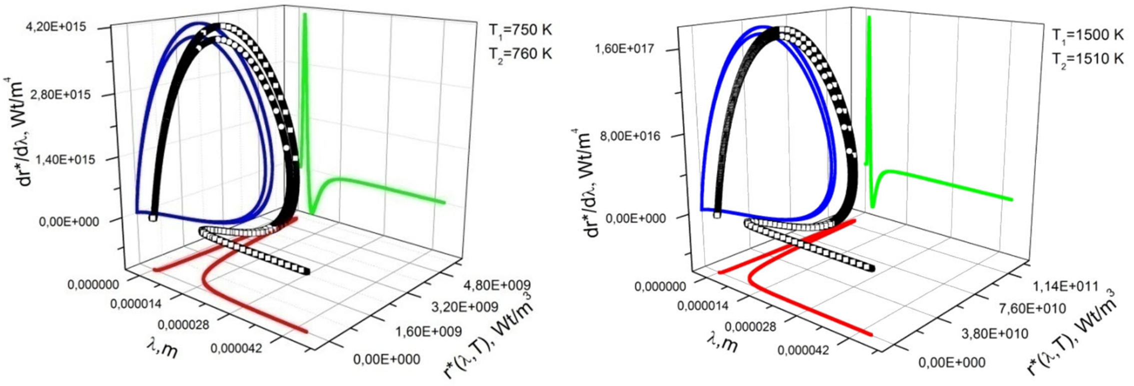
Для аналізу динаміки вимушених і спонтанних переходів запропоновано параметричну 3D-модель спектра випромінювання АЧТ. Її ортогональні проекції є сигнатурами 1-го і 2-го порядків, у конфігураціях яких відображаються динамічні, енергетичні та інформаційні аспекти спектра теплового випромінювання. Їх порівняльний аналіз виявив істотно більшу чутливість параметрів, які відображають динаміку випромінювальних переходів до малих змін температури, ніж параметри теплового випромінювання $(r^*(\lambda,T)$, $R)$, які використовуються у оптичній пірометрії. Показано, що вплив зовнішніх факторів на динаміку фізичних процесів в кристалічній булі найбільш проявляється у 3D-моделях функціональних характеристик зразків. У структурі функціональних характеристик зразків з кристалічної булі міститься інформація, пов'язана з особливостями дефектоутворення. Показано, що для ефективного управління ростом необхідні взаємопов'язані параметри, що характеризують особливості динаміки радіаційного теплообміну у ростовій установці. Розширення кількості взаємопов'язаних параметрів, які відображають динаміку радіаційного теплообміну, дозволяє здійснити ефективне параметричне управління ростом кристалів.

1. Introduction

Inefficiency of controlling the crystallization from a melt, variety of defects and multifactor dependence of crystal parameters on controlled and uncontrolled influences affect their stability and reproducibility of the crystals properties [1]. In particular, there are problems with growing from the melt of structurally perfect large crystals A^2B^6, as well as with formation in them unique sensory properties. Difficulties in obtaining information *in situ* and the variety of growth factors, the hidden relations between which determine the specific features of the defect formation process, lead to ambiguous parameters for the growth of A^2B^6 crystals (the rate of temperature change, the phase transition temperature, etc.). A result of this is the individuality of the functional characteristics (FC) of samples from different parts of the crystalline boule. It complicates analysis of results of their complex research and makes the identification difficult, as well as the selection of crystals for further use as functional elements of devices [2]. All this is naturally associated with: a) difficulties in monitoring and growth control, b) lack of reliable information about the cause-effect relationships between the combination of growth factors and properties of each part of the crystal boule. Obviously, there are hidden or unaccounted factors that influence the crystal growth process. It can be changed by the external environment parameters, the human factor and the features of the automatic temperature control system in a crystal growth furnace, etc. The modern growth furnace is a complex dynamic system, for the effective control of which it is necessary to get information on the causal relationships between the growth conditions and the FC of obtained crystals.

It is very important to control not only technological growth indicators (growth rate, temperature gradients, etc.), but also difficultly determined physical parameters of the melt and the growing crystal when automating the process of crystal growth. In most methods of A^2B^6 crystals growth from the melt, the temperature and the specified temperature gradients are monitored based on the calculated model of radiative heat transfer [3]. The rate of heating and cooling of the crucible is, as a rule, linear. It is obvious that any perturbation (mechanical, electrical, etc.) causes spatio-temporal inhomogeneities of physical processes. Therefore, the long process of a large-sized crystalline boule growing and its cooling is accompanied by the formation of a variety of structural defects. Their spatially inhomogeneous distribution in the sections of the crystalline boule contains information about local perturbations and changes in the parameters of the external environment, which is necessary to optimize the control of the growth furnace.

The founder of cybernetics N.Viner showed that the principles of control and communication are similar. He also introduced so notion as signal organization (its structure) its space-time ordering and drew attention to the fact, that the time series of parameters contains a minimum related information about its structure and is not effective for control [4]. Therefore, the interaction of elements of the complex dynamic system in control process is considered exclusively as the informational one. At the same time, this interaction determines



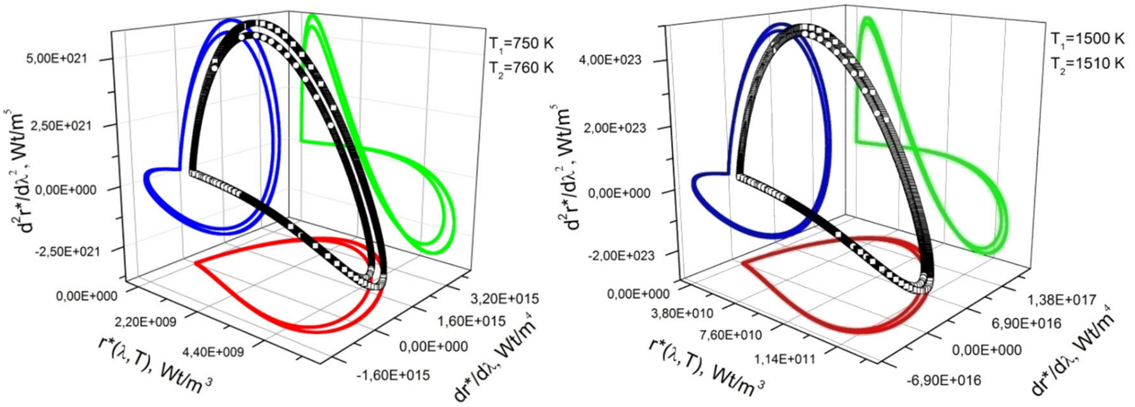
a) b)

Fig. 1. Black body radiation spectrum, their derivatives and signatures: (a) at $T = 750$ K and 760 K, (b) at $T = 1500$ K and 1510 K.

dynamic, energy and thermodynamic features of the system functioning, in the analysis of which it is possible to use the fundamental laws, principles and criteria of physics. It is obvious that improvement of the crystals growth technology requires the use of interdisciplinary cyber-physical approach, based on adaptation of the known cybernetic laws and principles (the decomposition principle, the feedback law, etc.) to the analysis of different nature physical processes [5].Therefore, further improvement of the crystal growth technology requires the identification of causally related factors, their formalization and accounting in the control model. Naturally, this requires the interdisciplinary approach to identifying the related information that is hidden in the structure of different physical and technological characteristics, as well as the structure of the samples FC from each crystalline boule. Therefore, the main purpose of the work was the development of tools for interdisciplinary approach to identify the features of the dynamics of radiative heat transfer, which mostly affect the structure of crystals FC and their individuality.

2. Geometrization of physical processes and their 3D-models

Information, related to growth conditions of A^2B^6 crystalline boule (controlled and uncontrolled effects, etc.), contain different nature FC of each sample. For example, in the spectral and temporal photoresponse of ZnSe crystals, this information can be detected if the photoresponse on crystal FC is numerically differentiated and represented in the generalized phase space (variable-state-velocity), where the variable is a parameter that is the argument of the functional dependence [6, 7]. Such processing allowed to increase sensitivity of the sensor based on ZnSe by an order of magnitude. By analogy, black body radiation spectrum can be represented in some generalized space. To do this, we use the Planck's law, which is a mathematical model of the equilibrium black body radiation spectrum. For its analysis we also use the idea of A.Einstein on stimulated emission. He applied the statistical principle of detailed balance, according to which the speed of direct and inverse processes is the same, for the physically demonstrative derivation of the Planck's law. Let us continue the analysis of the dynamics of radiative transitions, whose intensity dependence on wavelength determines the black body radiation spectrum. To do this, we numerically differentiate the Planck's law and obtained results represent in the space with coordinates (λ, $r^*(\lambda, T)$, $dr^*(\lambda, T)/d\lambda$) as a trajectory of dynamic microstates. Its orthogonal projections are: the well-known black body radiation spectrum $r^*(\lambda, T)$; dependence of the derivative $dr^*(\lambda, T)/d\lambda$ on the wavelength, as well as the first order signature, represented in coordinates $r^*(\lambda, T) - dr^*(\lambda, T)/d\lambda$. Its configuration displays the parameters relationship. The black body radiation spectrum at $T = 750$ K and 760K, $T = 1500$ K and 1510 K, their derivatives and signatures are shown in Fig. 1 (a) and (b), respectively. Temperature values were chosen from the crystallization and polymorphic transformation regions of A^2B^6 crystals, and the temperature difference $\Delta T = 10$ K is



a) b)

Fig. 2. Parametric 3-D models of black body radiation spectrum at temperatures T = 750 K and 760 K (a) and at temperatures T = 1500 K and 1510 K (b).

close to the average daily temperature changes of the heater and the external environment in summer. When use the wavelength derivative, the radiation spectrum is naturally divided into two spectral regions, differing in sign of the derivative and in the steepness $dr^*(\lambda, T)/d\lambda$. This simplifies the precise determination of the function maximum position $r^*(\lambda, T)_{max}$. It can be seen from the figure that the parameter $dr^*(\lambda, T)/d\lambda$ is more sensitive to temperature change than $r^*(\lambda, T)$.

Therefore, increase in the black body temperature of 10 K is mostly appeared in signatures whose configurations reflect the nature of the relation between $r^*(\lambda, T)$ and $dr^*(\lambda, T)/d\lambda$. The components of configurations of the first-order signatures are similar, and the increment of the area with increasing temperature is naturally associated with increase in the intensity of the radiative transitions in a certain wavelength interval. We note that as the temperature is increased twice, the maximum of the radiation spectrum is shifted, the scale $dr^*(\lambda, T)/d\lambda$ changes significantly, and the relative change in the signature area decreases.

It is possible to find related information on the growth conditions by transforming the FC of samples from the crystal boule slices into parametric 3D models. For analyzing the signatures configurations we can use the tools of interdisciplinary approach to identifying and investigating the individuality of the animate and inanimate nature objects functioning [7]. The approach is based on parametric geometrization of the dynamics of the objects functioning in space (state-slope-curvature), as a result of which a one-dimensional series of parameters is transformed into its 3D model. Considering the physical meaning of the derivatives, this model can be analyzed in the space of dynamic events (state-velocity-acceleration). Thus, analysis of 3D models of time and spectral photoreponse of the crystals showed that their configurations geometrically represent closed trajectories in the space of dynamic events [8]. The orthogonal projections of the trajectory are the 1st and 2nd order photoreponse signatures, with the help of which the natural decomposition into the components of the configurations of the corresponding cycles is carried out [9]. So, configuration of the signature in the plane (state-velocity) can be analyzed as a dynamic cycle, and in the plane (state-acceleration) as an energy cycle. In this case, the structure of the hidden relationships reflects the signature configuration on the plane (velocity-acceleration), and the powers of the interconnections are information characterizing the ratio of the quadrants areas. Related information on the growth conditions is mainly contained in the space-time distribution of the components of the signature configuration. It appears itself in the orderliness of the photoreponse structure components, and also statistically in the distribution of the powers of the information arrays at the quadrants. Reconfiguration of the signature during the processing (laser, acoustic, etc.) indicates that related information about the influence of the defect structure appears itself in the configuration of its second-order signature of photoreponse. Therefore, the powers of information arrays [5] are so important that they allow us to formalize the structure of interrelations in the form of a matrix of balance indicators. It is a cybernetic model of control of generation-recombination processes in the crystal.

Similar to the study of the kinetics structure and spectral photoreponse by means of their parametric 3D models, we construct such models of the black body radiation spectrum for temperatures $T = 750$ K and 760 K (Fig. 2a), and also for temperatures $T = 1500$ K and 1510 K (Fig. 2b).The most related information contains a parametric 3D model. In this model, the black body radiation spectrum transforms into closed trajectory of the dynamic events, the probability of each of them is determined by the multiplication $r^*(\lambda, T) \times dr^*(\lambda, T)/d\lambda \times d^2r^*(\lambda, T)/d\lambda^2$ and is represented by a point (see Fig. 2).

Given above the first-order signature and two new second-order signatures are the orthogonal projections of the 3D model. Configuration of the second-order signature $dr^*(\lambda,T)/d\lambda - d^2r^*(\lambda, T)/d\lambda^2$ is located in 4 quadrants, which reflects the natural decomposition of the equilibrium radiation spectrum on information arrays, the powers of which are proportional to the covered areas. These interrelationships determine the natural structure of the balancing of the rates of direct and inverse processes, which, in accordance with the principle of detailed balance, form the back body radiation spectrum. Thus, when the temperature was doubled, the signature configurations remained almost unchanged, which statistically confirmed the same number of forward and reverse absorption transitions and radiation transitions. Attention should be paid to the dependence of the linear density of probable events on the trajectory on the wavelength, and also to the asymmetry of the configuration of the second-order signature $dr^*(\lambda,T)/d\lambda - d^2r^*(\lambda, T)/d\lambda^2$. Thus, the steepness and curvature of the signature configurations of the black body radiation spectrum are more sensitive than the parameters of thermal radiation $r^*(\lambda, T)$ and the integrative parameter $R(T)$, which are used in optical pyrometry.

3. Results and discussion

For dynamic systems the parametric control is the most effective, for implementation of which interrelated parameters are needed. Therefore, it is critically important to select the interrelated parameters for controlling the radiative heat transfer during crystallization and cooling. It can be seen from the above that the most sensitive parameters are the parameters $r^*(\lambda, T)$, $dr^*(\lambda,T)/d\lambda$ and $d^2r^*(\lambda, T)/d\lambda^2$. The correlations of these parameters determine the configuration of the 3D model of the black body radiation spectrum, in which the thermodynamics of thermal radiation is most appropriately displayed. Orthogonal projections of the 3D model are the first and second order signatures, in the configurations of which the interrelations between naturally separated portions of the spectrum are displayed. Therefore, they are more informative and sensitive to small temperature changes than the thermal radiation spectrum $r^*(\lambda, T)$. In particular, the configuration of the first-order signature can be analyzed as a certain cycle of realized microstates, which reflects the dynamics of forced and spontaneous transitions in two spectral regions, differing in the sign of the derivative of the black body radiation spectrum. The increase in temperature twice does not change the configuration of the signature, however its area increased significantly. As shown in [7, 8], the signature area, which covers possible microstates, can be represented as the power of their subset W. Since its natural logarithm is the Boltzmann entropy $H = \ln W$, increase in the signature area with temperature indicates the increase in entropy H. From comparison of the signatures in Fig. 1, it can be seen that when the temperature is doubled, the area and, accordingly, the entropy increase by four orders of magnitude. At the same time, the area of the black body radiation spectrum, which reflects the energy luminosity $R(T)$, increased by a factor of 16 only in accordance with the Stefan-Boltzmann law. In this case, the known Wine laws are reflected in the nature of changes in the configuration of the signature. We note that when the temperature is doubled, the configuration of the second-order signature $r^*(\lambda, T) - d^2r^*(\lambda, T)/d\lambda^2$ does not change, which reflects the equilibrium of the thermal radiation. As can be seen from the figures, the parametric representation of the thermal radiation spectrum turned out to be more informative when change in the radiation temperature than the usual spectrum. Thus, the change in temperature even by 10 K (within 0.5...1.0 %) is barely noticeable in the usual black body radiation spectrum. However, they are clearly visible in its signatures (see Fig. 1 and 2), namely in the displacement of individual components and in increasing the entropy ratio H_B/H_H. In this case, both the well-known laws (the Stefan-Boltzmann, Wine) and the hidden statistical patterns of changes in the structure of the black body radiation spectrum are reflected in the nature of signature changes. They are of interest for optical pyrometry.

The second order-signatures are of special interest. Thus, as the temperature is doubled, the configuration of the second-order signature $r^*(\lambda, T) - d^2r^*(\lambda, T)/d\lambda^2$, the ratio of the areas of its components does not changed, that indicates the equilibrium of thermal radiation. The information aspects of the black body radiation spectrum are most apparent in the signature $dr^*(\lambda,T)/d\lambda - d^2r^*(\lambda, T)/d\lambda^2$, in the configuration of which the structuring information appear naturally. Indeed, the signature covers in 4 quadrants the areas Φ_1, Φ_2, Φ_3, Φ_4, which reflect the powers of information arrays. Their ratios are the balance indicators of the powers of natural relationships. Therefore, the hidden structure of interrelations in the black body radiation spectrum can be formalized with help of the corresponding matrix of the powers balance indicators B_{ij}, which are equal to the ratio of areas of all quadrants to each other ($B_{12} = \Phi_1/\Phi_2$, etc.) [5, 6]. Therefore, the configuration of the signature $dr^*(\lambda,T)/d\lambda - d^2r^*(\lambda, T)/d\lambda^2$ can serve as a natural model of structure of the black body radiation spectrum.

Proceeding from the foregoing, it is expected that the configuration of the signature, which reflects the structure of the black body radiation spectrum, may be inherent in signatures of other crystals FC. Indeed, similar configurations of the signatures of the photoreponse kinetics spectra and luminescence spectra were observed in the study of ZnSe crystals [9] and the temperature dependences of the photodielectric response. Consequently, in the structure of FC of a group of the samples from the single crystalline A^2B^6 boule, there is related information about the growth conditions. It can be assumed that there is a relationship between the space-time distribution of locally concentrated features of the FC signatures and the type of structure defects in the samples. This is evidenced by the individuality of kinetics of photoreponse and luminescence in the samples from the single crystalline boule and the similarity of their structures, as well as the results of their wavelet analysis [12]. The similarity of the signature configurations is inherent in the FC of other crystals. Configurations are also found in human electrophysiological signals (electrocardiogram, etc.) [7]. Note that the development of the idea of N.Wiener on the similarity of the control principles are the laws of reciprocal adaptation proposed by the ergonomist V.Venda [10]. They are consistent with the basic principles and laws of the general theory of systems and complement them. In particular, the necessary and sufficient condition for the emergence and development of any system is the presence of internal processes (between the system components) and external (the systems with external environment) reciprocal adaptation, which is based on the relationship between structure and function. In the signature configurations, the reciprocal adaptation of physical and technological processes is displayed. Therefore, the similarity of the signatures configurations of different crystals FC can be development of technological heredity [11, 12]. It is important that in the system analysis of the 1st and 2nd order signatures of the black body radiation spectrum and different FC of crystals, universal means can be used, as well as fundamental laws, principles and criteria.

Thus, the universality of the approach developed made it possible to compare the dynamics of equilibrium thermal radiation with the dynamics of photoinduced non-equilibrium processes (photoresponse, photoluminescence) and obtain complementary information [6, 7]. The similarity of the dynamics structures of these processes makes it necessary to take a fresh look at the requirements for the automation of technological regimes for obtaining the crystalline boule. In particular, it is necessary to pay special attention to: a) the need to increase the sensitivity of temperature sensors and optical pyrometry, b) the fluctuations of the controlled parameters and the response time to them, c) the need to harmonize the dynamics of different processes. That is why it is expedient to provide parametric regulation of the crystal growth process by means of interrelated and highly sensitive parameters that reflect the dynamic features of radiative heat transfer. They, as shown by the analysis of the signatures of the black body radiation spectrum, are most sensitive to external and internal factors. We also note that it is possible to determine the interconnected parameters and statistical indicators from the 3D models of FC, the application of which will simplify the identification and selection of the samples with the same FC, and also optimize the thermal and acoustic processing of the crystals.

4. Conclusions

In the monograph [13], B.Kadomtsev showed that in evolution of the complex dynamical systems, the appearance of bifurcation points is possible, where the dynamic behavior of an object can strongly depend on small perturbations, i.e. from signals passing either from the environment or from other dynamic systems. Therefore, attention is paid to the dynamics of forced and spontaneous transitions, for analysis of which the parametric 3D model of the black body radiation spectrum is proposed. Its analysis revealed a significantly greater sensitivity of the dynamic parameters of thermal radiation to small changes in temperature than the parameters of thermal radiation $(r)^*(\lambda,T)$, R), which are used in optical pyrometry. In this case, the scale changes of its parameters ($dr^*(\lambda,T)/d\lambda$, $d^2r^*(\lambda, T)/d\lambda^2$, H, B_{ij}) indicate the need to take into account the dynamics of physical processes occurring during cooling in different sections of boules, the temperature of which differs only by a few kelvins. This makes it possible to reveal the influence of the radiative heat transfer dynamics on the formation of technologically inherited properties of the crystals. Indeed, as it turned out, even small changes in temperature can determine the specific features of the dynamics of radiative heat transfer. If we take into account the fact that the growing boule usually lasts for several days, even the average daily temperature fluctuations in the laboratory can influence the growth processes dynamics. In addition, different stages of post-crystallization cooling (adjacent parts of the boule, whose temperature differs by several kelvins) are characterized by different dynamic behavior, which is accompanied by defect formation and formation of a residual stress field. They in A^2B^6 crystals cause a variety 3 of photoelectric and other phenomena. Therefore, the dynamics of the radiative heat transfer can be either a system-forming factor of the ordering of the defective structure or system-destructive. Obviously, this explains the influence of the cooling rate on the structure of the spectral photoresponse of ZnSe crystals, revealed in [5].

Thus, the conceptual foundations of the developed approach allow us to expand the number of interrelated parameters by differentiating and integrating various characteristics that reflect the dynamics of physical processes in growing large-sized crystals. Analysis of the crystals FC and 3D models built on their basis allows to obtain related information on the influence of external factors on the crystal growth. The use of universal means for revealing the structure of the both crystals FC and technological and thermal processes is promising for improving the growth technology, modifying the crystals physical properties [14], as well as thermal, acoustic and other crystal treatments.

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