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# Acoustic emission method for investigation of functional materials

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The analysis of the experimental results is carried out at examination of acoustic emission (AE) at dynamic and statical actions. It is shown, that threshold origin AE is accompanied time or stationary values by modifications in dependences of performances and properties of the functional materials (FM) on quantity of the given action. These modifications can be explained by acceleration of processes of relaxation and the degradation, accompanying origin AE at reorganization of structure in local fields FM.

Проведен анализ экспериментальных результатов при исследовании акустической эмиссии (АЭ) при динамическом и статическом воздействиях. Показано, что пороговое возникновение АЭ сопровождается временными или постоянными изменениями в зависимостях характеристик и свойств функциональных материалов (ФМ) от величины данного воздействия. Эти изменения могут быть объяснены ускорением процессов релаксации и деградации, сопровождающих возникновение АЭ при перестройке структуры в локальных областях ФМ.

Many products made of functional materials (FM) exhibit specific individual characteristics and are often in service under various extreme external factors. The testing intended to determine the permissible parameters are based on statistical spot checking models for individual assemblies and components. To describe the macroscale straining of a multicomponent product, it is insufficient to know the properties of individual components and the variation dynamics thereof, but comprehensive studies of the FM product as a whole are required. The external factors resulting in afterward structure changes in FM, including those in local chemical composition and defect concentration, are of critical importance both for the relaxation process dynamics and aging processes during the service. The increasing sophistication of a FM product means obviously a reduced reliability of the obtained permissible parameter values, the determination procedures thereof become more and more complicated [1].

Studies of the aging and relaxation dynamics presents some difficulties; in particular, for threshold processes it is necessary to preform complicated sequential measurements intended to detect the possible changes in the system metastable state. The required frequency of such measurements increases dramatically if it is necessary to determine the time point $\tau$ when the changes occur. That is why for modern FM, other approaches are required that are suitable to study different kinds of external influences, including complex ones.

One of the methods allowing precisely fixing $\tau$ and a threshold of origin of nonreversible changes of properties FM (in a realtime) or an origin of a pre-failure state of a product from FM is the method of an acoustic emission (AE) of materials [2]. The method grounded on physical process of radiation of the pulsing ultrasonic waves, accompanying breakdown (relaxation) of internal stresses during nonuniform evolution of flaws and reorganization of local structure. The AE indicates the starting local structure rearrangement in the material or the product, no matter what is the nature of that rearrangement and what is the kind of external effect (single or complex) causing it [2–4].

To approve the method, it is practical to use it for FM where different AE sources are possible. For example, in alkali halide crystals (AHC), it is the collective motion of dislocations that is essentially the only AE source prior to the failure onset. As for resonators made of $LiNbO_3$ crystals, the block structure rearrangement makes an additional AE source. In $GaP_xAs_{1-x}$ based structures, the 3D defects (the chemical composition inhomogeneities, internal mechanical stresses, etc.) as well as dislocation complexes predominate among the AE sources [4–6].

In this work, presented are the comparative experimental study results for relaxation and degradation processes in FM using AE under the simultaneous control by traditional methods. For piezoelectric $LiNbO_3$ and dielectric LiF, NaCl, and KCl, the external factor was provided by ultrasound mechanical strains (USMS), while for GaP, GaAlAs, and GaPAs light emitting diod structures (LED), by the forward current in the homo- or heterojunction.

The experimental procedures were somewhat different for each of the above-mentioned FM, but a common feature was the AE recording that was performed in the 20 to 200 kHz range (for USMS) or 200 to 500 kHz one (for LED) using a piezoelectric transducer (PET) and the special AE instrument AF-15. When studying USMS, an additional rejection filter was inserted between the PET and AF-15 to filter the US frequencies.

As to the samples to study, $LiNbO_3$ resonators shaped as plates of different thickness and cutting directions having the main resonance frequency along the thickness 1 to 4 MHz provided with electrodes sputtered onto the surfaces; GaP, GaAlAs, and GaPAs structures produced by industry including crystals of (400 to 500)×(400 to 500) $\mu m^2$ size; as well as AHC samples of (30 to 40)×15×10 $mm^3$ size were used.

To generate the ultrasound waves (USW) (including the oppositely directed collinear ones) in AHC and to measure their damping in the 0.7 to 35.0 MHz range, a pair of identical PETs was used. To measure the main frequency $U_f$ and the combination one $U_{2f}$ (deconvolution or the 2nd harmonic) of the generated USW, an additional PET and a S4-74 spectrum analyzer were used. The electric voltage $U_g$ = 10 V at the PET and electrodes of the $LiNbO_3$ plates answers approximately to the strain $\overline{S} = 10^{-4}$ averaged over the sample length.

A direct electric current at the density $J = (0.2 \text{ to } 20)\cdot 10^5$ A/m$^2$ was passed through the LED. The current density $J_i$ through the sample was increased in step-by-step manner as in [4, 7], that is, at each $(i + 1)$-th increase, $J_{i+1} = (2 \text{ to } 1.5 \text{ to } 1.2)\cdot J_i$. At each $J_i$ value, the AE was recorded till all the potentially active AE sources (at the given $J_i$) were actuated reliably (i.e., till the AE was over) to provide that the Kaiser law [2, 3] was met.

In $LiNbO_3$ piezoelectric, the AE has been observed in the frequency range of the electromechanical resonance for the resonator. The AE onset is associated with the exceeding of the threshold vibration amplitude value, the vibration spectrum of the resonator being changed at the same amplitude, too. Time correlations have been found for the change in nonlinear properties of $LiNbO_3$ resonators (3rd and 2nd harmonic amplitudes) and the AE parameters under variations of the strain amplitude. On Fig.1a dependence of a ratio of voltages of 2-nd harmonics and a base frequency on the measuring transformer $U_{2f}/U_f$ from a voltage on electrodes of plate $U$ (upper diagram) as well as amplitude of AE-signals (medium diagram) and AE count rate (lower diagram) from $U$ are given.

A threshold increase and hysteresis is observed in $U_{2f}/U_f$ and AE count rate dependences on $U$; the curves are not reiterated at repeated increase/decrease of $U$. In Fig. 1a, the index (1) means measurements at increasing $U$ while (2), those at decreasing one. Since the AE phenomenon is associated with irreversibility, and thus, with non-reiterative character of processes occurring in the material [2, 3], the fact observed agrees well with the behavior of AE signal amplitudes having maxima shifted mostly towards larger strains as compared to the $U_{2f}/U_f$ maxima.

It has been shown that in LiF, NaCl, and KCl dielectrics, the threshold onset of the intense AE may answer to a change in the 3rd order elastic moduli and the absorption coefficient of elastic waves. There is a set of frequencies (1 to 35 MHz) related together by the relationship $f_n = f_0/n$ for which the external influence threshold required to onset AE is lowered significantly.

Fig. 1b shows the $U_{2f}/U_f(U_g)$ (1) as well as $U'_{2f}/U'_f(U_g)$ (2) dependences, the latter being the total contribution from nonlinearity of the generator and emitting USW transducers. The dependence $U_{AE}$ (3) is the voltage of continuous AE that arises as sev-

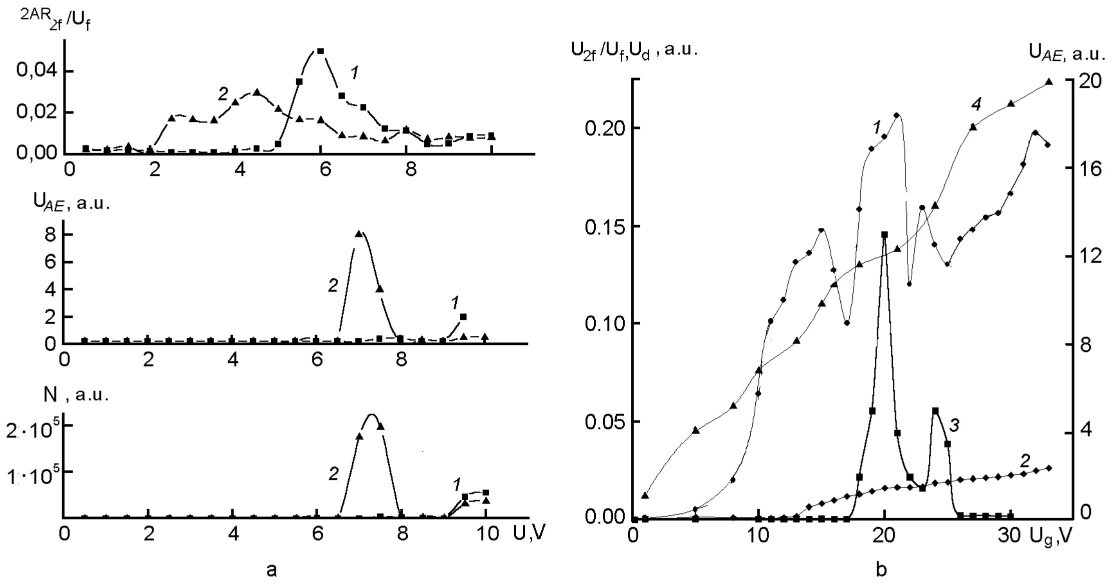

Fig. 1. Time correspondences in the AE onset and changes in FM nonlinear properties under USMS for $LiNbO_3$ resonators (a): the index (*1*) means measurements at increasing *U;* (*2*), those at decreasing one. b — those for LiF single crystals.

eral groups of noise pulses followed by development into pseudo-ordered emission, as in [8]. The curves (4) present the dependence of the ultrasound $U_d$ (that has passed the sample) from $U_g$, respectively.

Fig. 1b shows the "final" (after a significant drop of the AE count rate) dependences for $U_{2f}/U_f$ that were fast (up to several times per second) changed by 20 to 50 % in the middle region during AE.

For GaP, GaAlAs, and GaPAs structures, it has been found that, due to natural aging ($5{\cdot}10^8$ s), the threshold (for the AE onset) $J_i$ value is shifted out of the nominal current region reported by the producer towards ultrahigh (up to 50 times) current density region. Since the AE is a manifestation of the local structure imperfection rearrangement, this fact points to relaxation processes occurred [7].

The AE in aged GaPAs structures is observed only rarely at relatively low densities of forward current through LED ($0.02{\cdot}10^5$ to $0.6{\cdot}10^5$ A/m$^2$). At the same time, the reversible intensity redistribution between the green (576 nm) and red (720 nm) electroluminescence (EL) spectrum occurs, as in [9].

As the current density increases, at $J \approx (0.75$ to $9.0){\cdot}10^5$ A/m$^2$, AE is observed as well as the intensity redistribution between the maxima and a shift of both the maxima towards longer wavelengths by 7 to 10 nm (Fig. 2, curves 1 and 2). The further $J_i$ increase results in that the green maximum shifts by further 40 nm (Fig. 2, curves 2 and 3). The red maximum shifts also, but it is just a maximum in IR region (950 to 1000 nm) that becomes soon the predominant one, while that maximum was not observed at all in the initial samples.

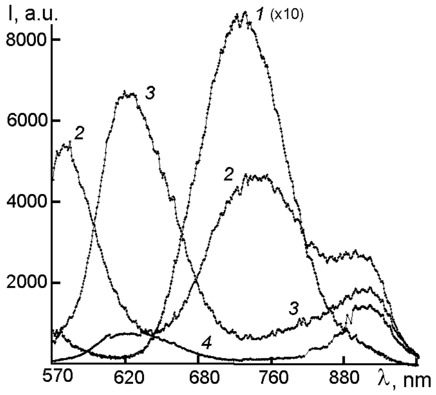

Fig. 2. EL spectra of $GaP_{0.85}As_{0.15}$ $n^+$-$n$-$p$ structures at the forward current density ($\cdot 10^5$ A/m$^2$): 0.75 (prior to AE) (*1*), 9 (*2*), 18 (*3*), 0.75 (after AE) (*4*).

The spectral changes at $J_i < 6{\cdot}10^5$ A/m$^2$ are fully or partially (in the case of AE) reversible. The AE causes the IR band appearance. A dramatic change occurs as $J_i$ exceeds $(6$ to $9){\cdot}10^5$ A/m$^2$, when an intense AE is recorded as well as an irreversible changes in the spectra (degradation in the both regions and IR band appearance at any $J_i$). For samples where the $J_i$ was adjusted somewhat lower than the failure current, $20 \cdot 10^5$ A/m$^2$, a characteristic IR maximum of a height comparable to that of the green maximum shifted to 620 nm was observed in the EL spectra as the current was decreased down to the nominal value $J_{nom}$ (Fig. 2, curve 4). The red maximum was absent. In fact, the AE onset corresponded to an accelerated degradation of the LED emissivity (by a factor up to 8 to 12).

Each AE pulse answers to different number of the AE sources actuated simultaneously, that number varies from 1 to $10^4$, depending on the defect nature. The AE sources are local regions where, at the structure rearrangement, the energy is released as a pulse of acoustic (and possibly also of electromagnetic and heat) emission. The kind of external force initiating AE depends on the FM nature.

Unfortunately, no consistent theory of acoustic emission was developed since the term itself exists. This is due obviously to that it is difficult to formulate exactly the general AE problem, that is, the problem of at least monotonous external action that, when attaining an (inaccurately defined) threshold, after a (difficult to forecast) time delay, causes a non-monotonous emission during a certain time (often being not characteristic even for samples of the same type) of acoustic pulses differing temporally in amplitude and duration, the pulse sequence is never reproducible.

The assumption that the AE source under consideration is independent of other ones is the condition of critical importance that makes it possible to analyze the process. In general, this answers to the FM state far from pre-failure one. In this case, the threshold (for AE) value of the external force means the onset of degradation processes (or acceleration of relaxation ones) in the FM. The increase in AE intensity corresponds to the acceleration of those processes, while the subsequent AE intensity decrease or stopping, the irreversible rearrangements occurred in certain local regions. A stable increase in the AE intensity evidences that the pre-failure sample state is attained.

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## Метод акустичної емісії для дослідження функціональних матеріалів

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Проведено аналіз експериментальних результатів при дослідженні акустичної емісії (АЕ) при динамічному та статичному впливі. Показано, що порогове виникнення АЕ супроводжується тимчасовими або постійними змінами в залежностях характеристик та властивостей функціональних матеріалів (ФМ) від величини даного впливу. Ці зміни можуть бути пояснені прискоренням процесів релаксації та деградації, що супроводжують виникнення АЕ при перебудові структури у локальних областях ФМ.