Dear Editor,

We like to express our appreciation to the reviewers for their comments. We are resubmitting the revised version of the paper number SST-108576. We have studied the comments of the reviewer carefully, and have changed the text according to the comments they have listed. The location of revisions is highlighted by yellow in "Marked-SST-108576.pdf". Below we refer to each of the reviewer's comments.

Response to Reviewer #1

Comment 1. Details that how the microwave treatments affect the semiconductors are not clear. The author claimed that it was not only heating up, and several possibilities were suggested. But still the physics of the microwave treatment is very unclear for the reviewer.

Reply: Let's consider the possible ways microwave treatments affect defects in structures under investigation. The primary process, which determines defect production under irradiation, is a displacement of the atoms from the sites of the lattice. But the energy of a photon with frequency 2.45 GHz is about 10^{-5} eV only; the threshold displacement energies in GaAs and SiC are (8-28) eV [1] and (20-35) eV [2], respectively. Therefore such a channel of microwave-induced modification of defect subsystem is unreal.

Microwave energy is known to transform into heat inside the material. Processes based on microwave heating find many industrial applications. But the used experimental procedure (pulsed microwave radiation with a period of 500 s and a duty cycle of 1%) allowed prevention of essential heating. The calculation according to Bacherikov et al. [3] shows that the maximum possible heating temperature of the sample ΔT is about 1 K. The ΔT magnitude was confirmed by measurements using a T-type thermocouple. As a result, the influence of microwave heating can be neglected.

There are many experimental observations that suggest non-thermal influence of microwave fields [4, 5]. These phenomena can be related to various physical reasons. First, it is known [6, 7] that a free charged particle in an electromagnetic field performs a drift in parallel to the electric component. The drift velocity magnitude is given by $v_{\perp} \propto (E_0/m\nu)$ (where E_0 is the amplitude of the electric field, m is the mass of the particle) and velocity direction depends on the phase of the field at the initial time. Consequently, along with the systematic drift of individual charged particles, directional movement of the entire set of particles is absent. Besides, the particle drifts in the direction of wave propagation with the velocity $v_{\parallel} \propto (E_0/m\nu)^2$. However, the charged point defects in semiconductor crystal are not free and have to overcome potential barriers when moving. It can be taken into account by using effective mass $m_{\rm eff}$, which exponentially depends on barrier height [7]. In our opinion, the mentioned features testify that such MW-induced movement is not responsible for revealed effects.

Second, the ponderomotive forces can arise under MW action. Under inhomogeneous microwave electromagnetic field conditions, the induced oscillatory defect fluxes are rectified, leading to directional, macroscopic mass transport [5, 8, 9]. The ponderomotive force can be described as follows [8, 7]:

$$F_p(x) = \frac{q^2 \beta E_0^2}{8m_{\text{eff}} \pi^2 \nu^2} \exp(-2\beta x), \qquad (1)$$

where q is the charge of the defect, and β is the coefficient of electromagnetic wave absorption, the axis x is along the direction of wave propagation. On the one hand,

the both the MW attenuation and ponderomotive force are essential when $d \ge \beta^{-1}$ (where d is the thickness of the semiconductor crystal). On the other hand, the following expression can be used to estimate β [7]:

$$\beta = \frac{1}{c} \left(\frac{\sigma \pi \nu}{\varepsilon_0} \right)^{\frac{1}{2}},\tag{2}$$

where $\sigma = en\mu_n$ is crystal conductivity; μ_n is the electron mobility, 8500 cm²/sV for GaAs, 400 cm²/sV for SiC. According to [7], the Eq. (2) is correct in the case of $(\sigma/2\pi\varepsilon_0\varepsilon\nu)\gg 1$ (where ε is the dielectric perminity; 12.9 for GaAs, 10.03 for SiC), which corresponds to the samples under investigation. The calculations show that β^{-1} is $(57-90)~\mu m$ for SiC crystals, 138 μm for GAS2, 20 μm for GAS1 and substrate of epitaxial structures, $(100-470)~\mu m$ for epi-layers. Thus the ponderomotive forces are able to cause the movement of the charged point defects both in the single-crystal samples and substrate of epitaxial structures, and the effect is maximal in the near–surface region. Though ponderomotive influence can not be the only reason for observed effects. In fact $\beta({\rm GAS1}) > \beta({\rm GAS2})$, however, MWT with $t_{\rm MWT} = 20~{\rm s}$ does not lead to defect transformation in GAS1, unlike in GAS1 — see table 1. Similar results are observed for SIC1(2) and SIC3.

Third, it was shown [10, 11, 12] that under resonance conditions (the coincidence of eigenfrequencies of the dislocation segment vibrations and electrical component of the microwave radiation), multiple dislocation loops occur. Besides, the MWT causes the movement of dislocations. In particular, at $\nu=2.45$ GHz for GaAs, resonant detachment of numerous dislocations with the length $L \leq 4~\mu \rm m$ becomes possible [12]. The dislocation climb is accompanied by intrinsic defect generation. In addition, the behavior of the dislocation segment in a MW electric field may be strongly affected by impurity atoms, which decorate dislocations. Having accumulated at dislocations, they, on the one hand, decrease resonance frequency; on the other hand, impurity atoms may detach from dislocations at high oscillation amplitudes, and free impurity atoms may appear in the crystal [10, 11]. In turn, the appearance of free doping atoms can result in an intrinsic defect concentration increase. In our case, the dislocation generation is confirmed by a change in both curvature radius and deformation of the near-surface crystallographic planes after MWT.

Fourth, the MW-induced destruction of impurity complexes, united in clusters, is described [10, 11, 12]. This is resonant phenomenon as well, and it is expected if the irradiation frequency is close to ion–plasma frequency $\nu_r = \sqrt{e^2 N_{\text{com}}/4\pi^2 \varepsilon_0 \varepsilon \mu}$ (where N_{com} is the complex concentration in the cluster, μ is the reduced mass of complex ions). For example, ν_r equals to 2.01 GHz for Te⁺–Cu⁻ complex with $N_{\text{com}} = 5 \cdot 10^{16} \text{ cm}^{-3}$ in GaAs [10]. But since the revealed transformation of deep levels relates to intrinsic defects, this mechanism seems unlikely. Besides, data about defect clusters in the samples under investigation are absent.

In our opinion, the process of MWT influence was two-stage. Initially, the resonant movement of the dislocation segment has caused an increase in the concentration of point defects, which are mostly intrinsic interstitial atoms. The approach of primary vacancy-related defects and secondary defects under the ponderomotive forces action and subsequent defect reactions occurred at the final stage. If the time of MW processing is not long enough for an essential increase in interstitial atom concentration or effective mass transport, the defect transformation does not occur, and the energy level does not change. In this case, the modification

of electron capture cross-section is able under a field of newly formed dislocations (or distant point defects).

The additional information was added to the revised manuscript (page 5, last paragraph; page 6; page 7, left column paragraph 1, 4, and 5).

Comment 2. The author's suggested modifications of the defects are too drastic. Such the defect modifications change the deep level parameters more. For example, deep level parameters for $V_Si\ V_C$ and V_C in silicon carbide have been identified by both the experiments and theoretical calculations, and their parameters are very different. The small changes in deep level parameters should be other reasons. They should be experimental errors or very small modifications of the defect structures.

Reply: The reviewer is quite right and vacancy—related defects in silicon carbide are investigated extensively — see, for instance, [13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24]. In particular, it is known [15] that 6H–SiC gives rise to 3 configurations for carbon vacancy and 6 configurations for the divacancy. The further increase in deep levels, which are relevant to one defect, are connected to the different charge states.

The defect configurations were identified by using level location in the gap. The primary indicator was the maximum agreement between determined values of $(E_c - E_t)$ and data given in previous publications (see References). If the change in $(E_c - E_t)$ after MWT has exceeded the experimental errors limit, we supposed the microwave-induced configuration change. The level locations were determined from the dependency of the TAV relaxation time versus inverse temperature with enough high precision (about 10 meV). The typical dependencies are shown in Fig. 3. In our opinion, the MWT-induced changes in deep level parameters ware not so small in most cases. For example, the $(E_c - E_t)$ changes were 70 meV and 90 meV for SIC1-3 and GAT samples, respectively. The minimal level shift, which was used as evidence of defect transformation, was 40 meV.

Comment 3. The adopted deep level observation technique is not so common, and it is difficult to judge measurement accuracy. For confirmation of the results, conventional techniques such as DLTS should be employed simultaneously.

Reply: In fact, DLTS-related techniques, designed for deep level characterization, are numerous and most widely used. Unfortunately, we are currently unable to apply the DLTS method. But it should be noted the transverse acoustoelectric voltage (TAV) has been extensively performed to characterize variety of semiconductors and their interfaces [25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38]. The most important studies include the characterization of the interface between the GaAs and its anodic oxide, and detection of defect levels in GaAs, InP, InAs, CdS, Si crystals, $\text{Hg}_{1-x}\text{Cd}_x\text{Tel/CdTe}$, ZnSe/GaAs, Si/SiO_2 heterostructures, Si MOS-structures, epi-GaAs, and $\text{Al}_{1-x}\text{Ga}_x\text{As/GaAs}$ quantum wells [31, 25, 29, 37, 33, 32]. The principle of TAV spectroscopy closely coincides with the idea of the original DLTS technique; as a result, the names "acoustic deep-level transient spectroscopy (A-DLTS)" [26, 32, 33, 34] or "acousto-electric deep level transient spectroscopy (AEDLTS)" [35] are used this method as well. Besides, the acoustoelectric transient spectroscopy has some strengths over the original DLTS. First, DLTS requires the measurement over a wide temperature range in the case of the presence of defects,

which differ significantly in a level location. TAV spectroscopy needs a much narrower temperature range. Second, the acoustoelectric transient spectroscopy does not require barrier formation. On the one hand, the rectified contact formation can influence the defect subsystem in the near-surface region before (after) microwave processing. On the other hand, the presence of metal contact before irradiation significantly affects the penetration of electromagnetic waves into the semiconductor. Third, the TAV method permits the detection of traps at the epi-layer and substrate interface [36, 37, 38]. The last two features were notably essential in our case.

The description of observation technique was modified (page 3, left column, paragraph 3)

Comment 4. The materials observed are not the state of the art materials in industries. Therefore, even if the results and physics are truth, the impacts of this manuscript are limited. If the microwave treatment has surely advantages compared with conventional processes, the author can apply it to industry important materials, such as 4H-SiC or GaN.

Reply:

Allow us to say a few words in favor of GaAs and 6H-SiC.

Gallium arsenide is a material widely used mainly in semiconductor technologies due to its attractive properties. Compared with silicon, GaAs has a wider energy gap, substantially higher electron mobility, higher dielectric strength, shorter carrier lifetimes, and better radiation hardness. One more advantage of GaAs is that it is a direct-band semiconductor material for which solid solutions can be obtained [39]. Gallium arsenide can be used to manufacture devices such as monolithic microwave integrated circuits, microwave frequency integrated circuits, infrared light-emitting diodes, solar cells, laser diodes and optical windows.

As a result, the GaAs market is growing constantly and quickly — see figure 1. Another evaluation of the market is given in [40]: "GaAs Markets is \$3.8 Billion in 2020, Promise to Grow to \$22 Billion by 2026".

GaAs was invented and developed as a basic RF electronics material, but GaAs market development trends are changing now. The development of photonics and microwave engineering industry has attained a level suggesting that it will consume most of fabricated GaAs by 2025 [41]. In particular, next generation GaAs support the signal speed that is needed to implement 5G [40]. Simultaneously, performance of semiconductor electronic devices is ultimately dictated by the presence and behavior of point defects. Therefore detailed knowledge of the properties of those defects under MW irradiation is crucial to the engineering of robust devices that operate effectively.

Besides, after silicon, GaAs is perhaps the most studied semiconductor. (Although notwithstanding intense effort over decades, definitive knowledge concerning even simple intrinsic defects in GaAs remains scanty [42]) In particular, microwave treatment influence on various parameters of GaAs structure has been investigated [43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 3, 54, 55]. Consequently, the GaAs is a good model material to study the microwave-induced transformation of defects in semiconductors.

SiC, a most promising wide band gap semiconductor is widely in use and progress for high power, high-frequency device applications due to high saturated electron drift velocity, high mobility, and extreme hardness. SiC is also a promising semiconductor

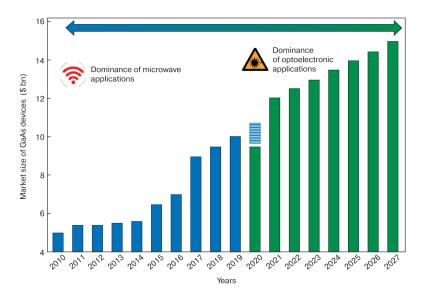


Figure 1. World GaAs device market development and prediction. Sources [41]

for spintronics and photonics due to the spin coherence property of some of its lattice defects. Both 4H and 6H polytypes are the most commonly used hexagonal polytypes of SiC with wafer size samples and high quality [15]. 4H-SiC is more suitable for manufacturing high-power electronic devices because of its large forbidden bandwidth, good thermal conductivity, and relatively small anisotropy. But both 4H- and 6H-SiC are used for power and opto-electronics, and as growth substrates for graphene and gallium nitride [56].

Moreover, in recent years, point defects in semiconductors have been suggested for implementing quantum bits (qubits) and single photon sources for quantum computation, quantum information processing, spintronics, and quantum sensing applications. Color centers in SiC, such as the silicon vacancy and divacancy, have recently been shown to be promising qubits for a variety of applications [15, 19, 57, 14]. Point defect qubits may have several nonequivalent configurations with different characteristics in each polytype which provide alternative tools for engineering qubit properties in SiC. The theoretical description and engineering of the defect centers require the assignment of each of the different microscopic configurations. This has been done for the silicon vacancy and the divacancy in 4H-SiC, but for 6H-SiC, the work is still in progress [15]. As a result, nowadays, defects in 6H-SiC are under intensive investigation [24, 15, 56, 22, 58, 59, 57, 60]

Thereby now the part of GaAs and 6H-SiC devices is still big enough in industries, and those materials are promising for future use. The reviewer is correct, and the use of wide bandgap materials gives the possibility to increase the blocking voltages for high power devices, as well as to make devices smaller and to reduce power losses. The reviewer's suggestion about 4H-SiC and GaN investigation is very interesting, and may be done in the future.

We have added some details in the research justification (page 2, left column, last paragraph, and right column, fist paragraph).

Comment 1. I suggest the authors concentrating on a specific semiconductor, digging deeply on the evolution of defects after MWT, and discussing the effect of doping on the process. The current work relates to the type of the host, the single crystal/epitaxial layers, doping concentrations. Readers lost easily during the introduction of results and discussions.

Reply: The various doping concentrations allowed us to conclude that the heating and ponderomotive forces are not the main reason for the revealed microwave-induced change in the defect subsystem. The set of GaAs epitaxial structures and single-crystals with different orientations allowed the investigation set of various vacancy-related defects (V_{As} , $V_{As}As_i$, $V_{Ga}Ga_iV_{As}$, $V_{Ga}Ga_{As}$, and $V_{Ga}V_{As}$). Besides, the difference in dose dependencies in epi-structures and single-crystals provided additional evidence that dislocation density influences microwave hardness. Finally, the revealed microwave-induced transformation of defects located in the near-surface layer in two materials testifies that this phenomenon is typical for semiconductors (at least binary). The next step after general features research may be deep digging.

To our shame, the reviewer is correct about some fog in Results and discussion. We hopefully rephrased the section to add sunlight.

Using two materials

The text was revised. In the present paper, we have concentrated on three specific and common mechanisms, which frequently take place in MSM or MIS structures, and could be verified experimentally. The reviewer's suggestion is very interesting, and may be done in the future.

This is very interesting suggestion, but this comparison requires an additional analysis, which can be done in the future. The reviewer's suggestion is very interesting, and may be done in the future.

We have added this interpretation in the revised text, in page 11 (second paragraph). We have revised the text accordingly. We rephrased this sentence (see page 7)

Comment 2. The authors carried out MWT experiments on differently doped SiC or GaAs. Did the concentration of dopants change the evolution of point defects? The Fermi level is different for differently doped hosts, which charge states of intrinsic defects and MWT-induced defects, and thus the interaction, may be different.

Reply: The text was revised.

Comment 3. The Pool-Frenkel effect is related to defect states of dislocations. Yes, some works dealt the interaction between irradiation-induced point defects and dislocations, such as Appl. Phys. Lett. 117, 023501 (2020) and J. Mater. Chem. C 9, 3177 (2021). But I didn't see any detailed discussion in this work.

Reply: The text was revised.

[61, 62, 63]

Comment 4. Typos throughout the manuscript should be corrected, including but not

limited to:

- (1) "and semiconducting compounds including [6, 8]"
- (2) "doping degree"
- (3) "0:31 ÷ 0:33"

Reply: The text was revised.

Response to Reviewer #3

Comment 1. The reviewed manuscript presents results on investigation of deep levels in various materials before and after microwave treatment with different durations (doses). Therefore, the investigation is not well focused, since the nature of defects is different for different materials, and they not necessarily should be governed by the same trends.

Reply: The text was revised.

Comment 2. The results could present interest, even though the investigated materials are not the state of the art ones in industries. However, the used experimental method is not widely approved, and the identification of the microscopic nature of defects is not an easy task even with widely used investigation methods such as DLTS or DLOS. As a result, the discussions about the microscopic nature of the observed defects and their reconfiguration during the microwave treatment as well as the made conclusions are not enough convincing (especially in cases when the deep level activation energies do not change very much under microwave treatment, as well as in cases when there is a large difference between the experimental results and those coming from theoretical consideration).

Reply: The text was revised.

Comment 3. I consider that the manuscript is not suitable for publication in the present form. However, the reliability of the obtained results may be significantly improved, if the study is complimented with other methods of investigation such as DLTS or DLOS, at least for some of the investigated materials. Moreover, the DLTS method also gives information about the concentration of deep levels. In such a case, a comparison of parameters of deep levels obtained by TAV and DLTS, combined with an analysis of the results of previously performed investigations, may provide enough arguments confirming the proposed microscopic nature of defects and their transformations.

Reply: The text was revised.

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