

Dipole relaxation, dielectric and electric properties of PbWO_4 crystals

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Experimental dependences of permittivity for undoped PbWO_4 single crystals at 1 kHz frequency in 290–550 K temperature range are considered. The $\varepsilon(T)$ curve runs under heating and cooling are different. The $\varepsilon(T)$ function under heating is characterized by groups of narrow maxima at 290–330 K and 330–400 K, the former being predominating. At 400–470 K, the $\varepsilon(T)$ dependence is linear, while above 470 K, the function is nearly exponential. After the high-temperature heat treatment of a sample, a multistage relaxation process of ε values within limits of 25 to 30 is observed at 290 K. The features of $\varepsilon(T)$ at low temperatures are defined by the dipole polarization and hopping mechanism of the charge exchange between complex dipole associates. The pairs of lead and oxygen vacancies (dipolons) are predominating intrinsic defects of the crystal structure; those defects form more complex dipole associates.

Экспериментальные зависимости диэлектрической проницаемости нелегированных кристаллов PbWO_4 на частоте 1 кГц рассматриваются при температурах $T = 290\text{--}550$ К. Ход кривой $\varepsilon(T)$ при нагревании и при охлаждении различен. Функция $\varepsilon(T)$ при нагревании характеризуется наличием групп узких максимумов при 290–330 К и 330–400 К с доминированием первых. При 400–470 К наблюдается линейная зависимость $\varepsilon(T)$, выше 470 К закон изменения ε близок к экспоненциальному. После высокотемпературного нагревания образца наблюдается при 290 К многостадийный процесс восстановительной релаксации значений ε в пределах 25–30. Дипольная поляризация и прыжковый механизм обмена зарядами между сложными дипольными ассоциатами определяют особенности $\varepsilon(T)$ при низких температурах. Доминирующими дефектами структуры являются пары вакансий свинца и кислорода — диполонны, на основании которых образуются более сложные дипольные комплексы.

Lead tungstate PbWO_4 (PWO) crystals are under intense study in connection with the use thereof in high-energy photon and elementary particle physics as an effective self-activated scintillation material with short afterglow time [1]. The luminescence properties of PWO crystals and their time characteristics depend on the structure defects, the nature of the latter is complex and is not established unambiguously to date. The electric and dielectric studies of that compound are fragmentary and insufficient, in spite of the fact that such data are highly informative as to the defects and state changes thereof during the crystal heat treatment.

Our works [2–4] on the studies of PWO crystals by thermostimulated polarization and depolarization currents within the temperature range of 290 to 600 K under a constant external field have revealed a series of local energy levels and relaxation processes of various nature. The previous studies [5] of the permittivity ε under variations of the PWO crystal temperature within the 290–600 K range have shown a close connection of such dependences with the depolarization processes in the compound.

The work [6] aimed at the study of lead tungstate and molybdate in variable and constant electric fields has provided some

dielectric characteristics of those crystals at temperature above 540 K. In [7–11], the regularities of Nb, Y, La, Gd, and F entering into PWO matrix have been elucidated by dielectric response method at frequencies ranging from 10 to 10^6 Hz in 413–673 K temperature region. The frequency dependences of dielectric losses in PWO crystals are considered in [12] basing on the vacancy dipole model. The temperature dependences of dielectric characteristics have been determined for CuWO_4 [13]. The permittivity and dielectric losses at 1 kHz frequency for some polycrystalline tungstates are reported in [14]. Recently [15], a relaxation character anomaly of the permittivity has been found for ceramic PWO samples near 650 K at 1 kHz frequency.

In this work, considered are experimental temperature and isothermal dependences of the permittivity in undoped PWO crystals (at the sample transition into a quasi-equilibrium state in the latter case) with the purpose to elucidate the regularities of defect formation and ion-electron processes. The $\varepsilon(T)$ study results are compared with the measured thermal depolarization currents for the same crystals.

The PWO (scheelite structure) crystals were prepared by Czochralski technique. The samples were cut out perpendicular to the growth axis [001] and shaped as plane-parallel plates of $10 \times 10 \times 0.4$ mm³ size. Aquadag electrodes were used in the studies. A quartz cell thermostated to within ± 1 K was used in the experiments. The ε values were calculated assuming negligible edge effects under the flat condenser approximation using the known relationship [16]:

$$\varepsilon = Cd / \varepsilon_0 S, \quad (1)$$

where C is the sample capacity to measure; d and S are the sample thickness and area, respectively; ε_0 , is the dielectric constant. The sample capacity was recorded using a CLR E7-13 instrument (the working frequency 1 kHz). The r.m.s. value of voltage drop across the sample did not exceed 0.2 V. For correctness sake, the correction for the connecting system capacity was taken into account. Other features of the experiment did not differ substantially from those described before [17]. The temperature was measured using a chromel/alumel thermocouple. The measurements were done under linear heating or cooling at the rate of 0.1 K/s within the

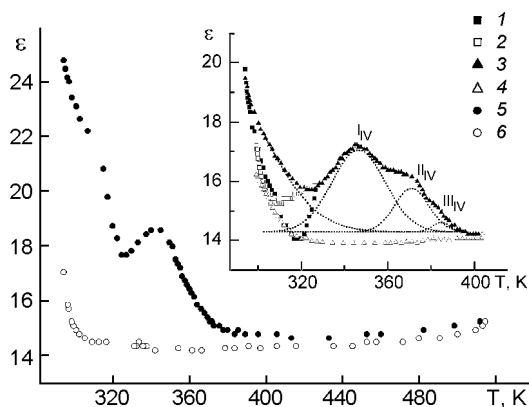


Fig. 1. Temperature variations of dielectric permittivity of PWO crystals under heating (1, 3, 5) and cooling (2, 4, 6) depending on the maximum measurement temperature T_h within one heating/cooling cycle. T_h (K) values: 335 (1, 2), 405 (3, 4), 515 (5, 6). Measurements in each heating/cooling cycle were done after the sample keeping in normal conditions for at least 24 h. The components I_{IV} – III_{IV} (dotted lines) have been obtained by expansion of curve (3).

290–550 K range. The sample transition into quasi-equilibrium state at room temperature following heating was monitored at time steps t by measuring its capacity. To establish the influence regularities of the previous excitation in an external field, a constant voltage U_p (up to 50 V) was applied to the crystal for t_p of 5 to 15 min at various fixed temperatures T_p of the sample holding in the field. The thermostimulated depolarization (TSD) currents I_{TSD} were measured under linear heating using a V7-30 voltmeter and the procedure described in [2, 17]. The experimental data processing and the complex maxima expansion into Gaussians were carried out using the Origin standard software.

Temperature dependences of permittivity for PWO crystals and the effect of the maximum sample heating temperature T_h thereon in each experiment are shown in Fig. 1 (curves 1 through 6). The $\varepsilon(T)$ dependences under heating (1, 3, 5) and cooling (2, 4, 6) within one heating/cooling cycle are presented or for several sequential cycles. To discriminate the individual components of the relaxation process in the 320–400 K range, the curve 3 was expanded into its components I_{IV} – III_{IV} . The temperature T_h was selected taking into account the TSD currents of the same samples

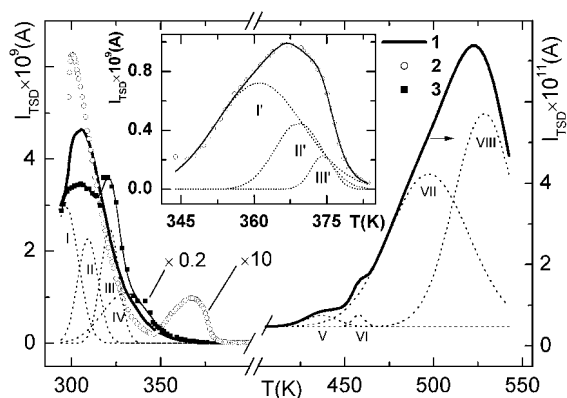


Fig. 2. TSD curves of PWO crystals at different polarization conditions. Curves 1, 2: $T_p = 335$ K and 320 K, respectively, $U_p = 50$ V, $t_p = 5$ min in both cases; Curve 3: $T_p = 290$ K, $U_p = 50$ V, $t_p = 5$ min. Inset: expansion of the 350–380 K TSD peak into elementary components: circles denote experimental values, elementary components are shown by dotted lines, thin solid line is envelope of I'–III' components. Similar notation for expansion of curves 1 and 3 into I–VIII components.

(Fig. 2). The thermal depolarization method has shown that series of the depolarization maximum currents are formed at certain conditions in the temperature ranges 290–350 (A), 350–380 (B), and 400–600 K (C). As is seen in Fig. 2, those maxima are non-elementary (expansion into components I–VIII), thus evidencing the complex character of the depolarization processes. The experimental maxima have been shown [2–4] to be connected with the relaxation of a polarization charge having dipole nature (A and B regions) and with formation of volume charges in the sample by equilibrium charge carriers (C region). The active relaxation processes of dipole nature seen in the TSD spectra at 290–380 K are also of importance for the features of $\varepsilon(T)$ curve shape in the same temperature range.

It follows from Figs. 1 and 2 that for the studied PWO samples, the maxima in $\varepsilon(T)$ curves are due to polarization effects {regions A-B}. At the same time, an usual run of $\varepsilon(T)$ dependence is seen in the 400–550 K region. The non-linear ε increase at high temperatures is due to the exponential conductance increase at the activation energy of 0.7 to 0.9 eV [2–4] in the respective temperature ranges. Thus, under heating, the $\varepsilon(T)$ curves (Fig. 1) in the 290–400 K exhibit complex maxima positioned close to each other are observed that are grouped by

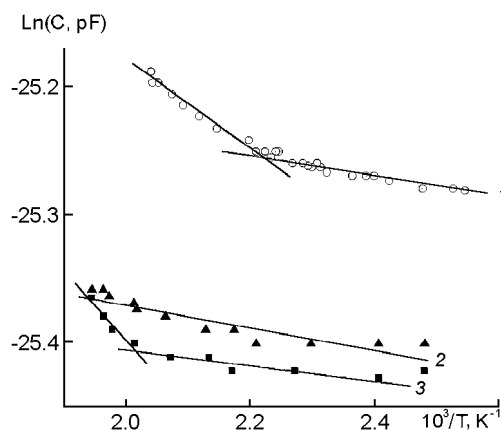


Fig. 3. Capacity dependences of inverse temperature for heating of a polarized PWO sample ($T_p = 500$ K, $t_p = 5$ min) (curve 1); heating of a non-polarized sample (2) and its cooling (3).

the relative intensity values within two ranges, 290–330 K and 330–400 K. The $\varepsilon(T)$ curves taken under heating and cooling differ in shape. A considerable temperature hysteresis is observed at $T_h \geq 400$ K where the values obtained under heating exceed those obtained under cooling. No maxima are observed in the $\varepsilon(T)$ curve under cooling. The non-linear character of $\varepsilon(T)$ curves taken under heating remains in repeated experiments independent of T_h or the sample keeping in normal conditions prior to measurements. At $T_h \leq 400$ K (Fig. 1, curve 1), the ε values obtained under cooling exceed those observed under heating, the temperatures being the same.

At room temperature, ε value was as a rule within limits of 25 to 30. These data agree with those of [8, 18]. To restore fully the ε value (to attain the crystal quasi-equilibrium state) after high-temperature treatment, the sample should be kept in normal conditions for about 24 h. During that time, the initial conductance of the crystal is restored, too [4]. In the range of 400 to 470 K, the $\varepsilon(T)$ function under heating is linear (Fig. 1, curve 5)

$$\varepsilon = 10.72 + 1.67 \cdot 10^{-3}T. \quad (2)$$

Under cooling, the $\varepsilon(T)$ is also linear in the range of 470 to 320 K (Fig. 1, curves 4 and 6):

$$\varepsilon = 10.71 + 9.07 \cdot 10^{-4}T. \quad (3)$$

The non-linear section of the experimental $\varepsilon(T)$ dependence both under heating and

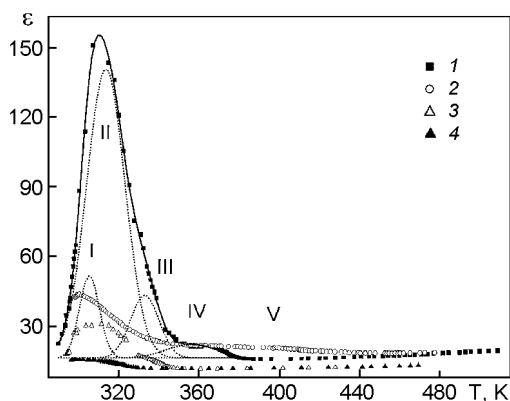


Fig. 4. Temperature dependence of permittivity for a previously polarized PWO sample ($T_p = 300$ K, $t_p = 10$ min) (curve 1) and at its subsequent ($T_p = 500$ K, $t_p = 5$ min) heating (curve 2). Curves 3 and 4 show the measurement results for the same sample but non-polarized in two subsequent heating/cooling cycles. Dotted lines show elementary components (I–IV) obtained by expansion of curve (1) into Gaussians. Black squares denote experimental data and the solid line is the total of components (I–IV).

under cooling is approximated satisfactorily by an exponential law:

$$\varepsilon = \text{const} \cdot \exp(-\Delta H/kT). \quad (4)$$

The pre-exponential factor in Eq.(4) amounts about 30 and about 29 under heating and under cooling, respectively. Thus, the experimental $C(T)$ curve can be approximated by one or two straight segments in coordinates $\ln C = f(1/T)$ at $T > 470$ K (Fig. 3), the slopes of the segments defining the activation energy ΔH . Under heating, ΔH at $T > 400$ K is low (about 0.014 eV), while under cooling, about 0.033 eV ($T > 500$ K) and about 0.011 eV ($T < 500$ K).

The experimental data on the effect of an external electric field on the temperature dependence of the PWO permittivity are presented in Fig. 4 (curves 1, 2). For comparison, the curves (3 and 4) for a non-polarized sample are presented. At the moment of the external field switch-off at 290 K, fast relaxation processes are observed with C decreasing in time. Therefore, the $C(T)$ measurements were carried out after the relaxation was over. In the 290–430 K range, the $\varepsilon(T)$ curve for polarized samples exhibits several maxima (I–V) at 305, 314, 332, 362, and 400 K, respectively (Fig. 4). The maxima I–III near 320 K form an intense relatively narrow

Table. Temperatures of the $\varepsilon(T)$ curve maxima (T_m^ε) and the TSD current maxima (T_m) for PbWO_4 crystals. The peaks are numbered according to Figs. 1, 4, and 2, respectively

$\varepsilon(T)$	T_m^ε , K	TSD	T_m , K
–	–	I	295
I	305	II	309
II	314	III	322
III	332	IV	331
I _{IV}	349	I'	360
II _{IV}	370	II'	369
III _{IV}	384	III'	375
V	400	V	435
–	–	VI	458
–	–	VII	500
–	–	VIII	532

complex peak. At the first measurement under heating, this peak exceeds that for non-polarized samples by several times. The intensities of the I–IV peaks drop down to the initial values after several heating/cooling cycles followed by spontaneous relaxation of the sample in normal conditions for 4 or 5 days. A sample polarized at $T_p = 300$ K, when being not heated, restores high ε values (150 to 200) for a prolonged time. The high-temperature capacity increase at $T > 400$ K for polarized samples is approximated satisfactorily in $\ln C = f(1/T)$ coordinates by two straight segments with a knee at $T = 450$ K (Fig. 3, curve 1). In this case, the ΔH values are low and amount 0.008 eV at $400 \text{ K} < T < 450 \text{ K}$ and 0.027 eV at $T > 450 \text{ K}$.

The low-intensity blurred maximum V in the $\varepsilon(T)$ dependence (Fig. 4, curve 2) is seen only for PWO samples polarized at $400 \text{ K} < T < 500 \text{ K}$. At the same time, the I–III peaks are of low intensity. At $T \geq 420$ K, the ε values for non-polarized samples increase slightly as T rises. The sample polarization results also in a faster (as compared to non-polarized sample) increase of ε at 400–550 K. A comparison of $\varepsilon(T)$ and $I_{TSD}(T)$ within the same temperature range 290–470 K (see Table) shows that the number of maxima and the temperature positions thereof are similar in both cases.

The non-polarized sample returns to the quasi-equilibrium state after one or two heating ($T > 400$ K) and cooling (to room

temperature) cycles, the ε value being restored. The kinetics of the crystal transition to the initial state is illustrated in Fig. 5. The measured results are presented here in coordinates $\ln[\Delta C/(C_\infty - C)] = f(t)$ where $\Delta C = C_\infty - C_0$ is the capacity value by which the initial capacity C_0 changes as the sample attains the quasi-stationary state and the C_∞ capacity becomes attained at $t \rightarrow \infty$. It is obvious that at $t = 0$, the capacity is $C = C_0$. That is, to describe the sample capacity variation in time the relation

$$C = C_0 + \Delta C(1 - e^{-t/\tau}) \quad (5)$$

is used. Here, τ is the time constant in the model of a homogeneous electrically neutral dielectrics with deep energy levels [19]. When several exponential relaxation processes take place, the $\ln[\Delta C/(C_\infty - C)] = f(t)$ curve presents several linear segments with slopes defining the corresponding τ values. As is seen in Fig. 5 (curve 1), the $C(t)$ dependence is approximated in $\ln[\Delta C/(C_\infty - C)] = f(t)$ coordinates by at least two straight segments (I) and (II). At the final stage, a relaxation process with large τ values is observed (Fig. 5). At $t \rightarrow 24$ h, the quasi-equilibrium state is attained and the $C(t)$ curve becomes parallel to the time axis. The corresponding time constants for the 1st and 2nd components of the initial C reinstatement kinetics at the initial relaxation stage (Fig. 1, curve 5) amount approximately $\tau_1 = 4.3$ h and $\tau_2 = 6.8$ h. The repeated multiple heating/cooling cycles induce irreversible changes in the sample that are equivalent to its annealing at elevated temperatures. This results in decreased ε values at 295 K and the total relaxation time. In this case, one straight segment with the time constant of the initial relaxation stage about 6.9 h and close to τ_2 predominates in the $C(t)$ plot of the reinstatement relaxation (Fig. 5, curve 2).

As the measurements have shown, the studied PWO samples after cooling from $T > 500$ K to room temperatures at rates used in experiment (about 0.1 K/s) keep the non-equilibrium state for a prolonged time. This feature of the crystals causes the hysteresis phenomena (Fig. 1) that, as is known [20] are explained by the "asymmetry" effects in the temperature-induced evolution of electrically active defects under heating as compared to the cooling. In particular, the dissociation energy of complex neutral

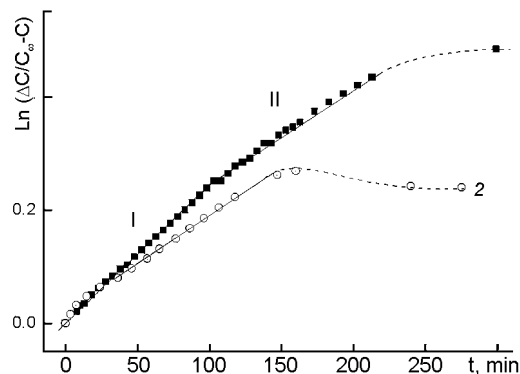
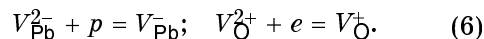


Fig. 5. Capacity relaxation of a PWO sample at 290 K. Curve (1) has been obtained after the first heating up to 550 K; (2), after subsequent several measurements in the 290–550 K range.

accumulations of defects may differ from the association energy thereof. That is why, even at low cooling rates, the crystal has no time to attain the quasi-equilibrium (the initial state of defects and initial concentration of the defect complexes are not restored, so the sample return to the equilibrium requires a prolonged relaxation). Said processes take place in the case when the Schottky defects dominate [20, 21]. In PWO that is a typical non-stoichiometric system, the Schottky defects are mainly double-charged vacancies of lead V_{Pb}^{2-} and oxygen V_O^{2+} . The energy minimization and the static interaction between V_O^{2+} and V_{Pb}^{2-} cause formation of neutral associates of those vacancies called bivacancies or dipolons. The latter form the base for formation of more complex dipole complexes of defects with electrons (holes) localized at the oxygen (lead) vacancies, respectively. The hole (p) localization at V_{Pb}^{2-} and electron (e) capturing by V_O^{2+} weaken the electrostatic interaction between the dipolon components and this may result in the binding breakdown. Thus, in the crystal, there are centers formed due to processes that can be written in simplified form as



The electrically neutral associates $V_{Pb}^{2-} + 2p$ and $V_O^{2+} + 2e$ are energetically hardly probable at the experimental temperatures. The concentration ratios between the double vacancies (dipolons), isolated single vacancies, the charged centers of (6) type, and $(V_{Pb}^- + V_O^+)$ with zero effective charge will be defined by the electric neutrality condition and the completeness measure of the

corresponding quasi-chemical reactions in the sample bulk under excitation. The concentration of uncontrollable impurities is assumed to be insignificant. The above schemes of the dominating disordering in PWO crystals make it possible also to consider in a non-contradictory manner the electric conductance and the depolarization currents [2–4].

The works aimed at the PWO examination using optical methods [22, 23], EPR [24], and theoretical calculations [25] consider various models of defects that may be involved in the electron exchange. So, in [22], it is assumed that it is just the centers of O^- , F , F^+ , Pb^+ , Pb^{3+} induced by irradiation and high-temperature annealing due to the charge re-localization that are responsible for the changes in the absorption spectra of PWO crystals. In [23], when studying the additional absorption spectra, the vacancy pairs $V_O^{2+}-V_{Pb}^{2-}$ are considered as well as electron processes associated with the transformation of those pairs due to UV irradiation. The low-temperature EPR made it possible to identify the electron centers formed by the complex anion WO_3^- that contains an oxygen vacancy and is associated with a defect in the lead sublattice [24]. The distribution of electronic state density in PWO is modeled [25] when V_O^{2+} and $V_O^{2+}-V_{Pb}^{2-}$ -pairs are dominating.

The change in the charge state of the defect associates at the crystal temperature variation in an applied field is accompanied by a jump-like displacement of holes (electrons) from one defect site to another. This is confirmed by low ΔH values. That process is equivalent to a change in the orientation state and/or dipole moment of the associate and causes an additional contribution to the $\varepsilon(T)$ dependences due to polarization.

The polarization and other dipole effects take place often in solids at the jump-like electron transfer over the defect lattice sites [12, 26, 27]. In particular, such processes may cause high intensity maxima in the $\varepsilon(T)$ curve as the temperature rises; this has been revealed in semiconductors [28, 29]. The localization of non-equilibrium charge carriers at vacancy dipole associates in gadolinium-gallium garnet single crystals [30] causes the photoinduced reorientation of such dipoles at $T_p = 80$ K and the dipole polarization and the depolarization current peaks at about 320 and 450 K in those crystals.

The discussed jump-like mechanism of the center recharging can be due obviously

to ionizing radiation [23, 24] and cause thermal glow; this has been studied in experiment [21, 23]. It is to note that the number of thermal glow maxima (see, e.g., [22, 24]), the temperature positions, the half-width, and relative intensity thereof within the studied temperature range are close to both the same parameters of the thermal depolarization maxima (290 to 470 K region) and to the maxima of $\varepsilon(T)$ curves presented in this work (see Table).

Thus, within temperature range of 290 to 550 K, three regions (290–380 K, 400–470 K, and 470–550 K) can be discriminated where the changes in ε (or C) of PWO crystals are defined by domination of different components in the temperature-induced and/or isothermal evolution of the ion-electron processes. The weak $\varepsilon(T)$ dependence of PWO crystals within the 400–470 K range following the linear law (2) or (3) under heating or cooling, respectively, evidences the electron polarization with characteristic time of 10^{-15} to 10^{-13} s. This type of $\varepsilon(T)$ dependence is realized against the background of an insignificant contribution to $\varepsilon(T)$ due to a weak conductance increase. A significant conductance increase of PWO has been observed only at $T > 470$ K [2–4]. The difference between the $\varepsilon(T)$ curves taken under heating and cooling is minimum in the 400–470 K temperature range (Figs. 1, 3, 4).

The large ε values of PWO crystals at room temperature are thus seen to be defined to a large extent by intrinsic electrically active structure defects (vacancies and associates including such vacancies) and by the jump-like charge exchanges between those defects. The temperature dependence of permittivity is influenced also by thermal polarization and elevated temperatures (annealing at $T > 500$ K). The heating in air can be used to vary the trend of PWO crystals to a prolonged relaxation at room temperature after the high-temperature treatment, the relaxation time (the process of attainment of quasi-stable ε values) being shortened mainly due to elimination its fast components. The latter seem to be associated with more mobile elementary electrically active defects of p (or e) type, the domination of one of those being defined, in particular, by the operating temperature region. The heating of PWO crystals to elevated temperatures favors the dissociation of the point defect dipoles and provides an increased concentration of free vacancies.

The vacancies, when migrating towards dislocations or other crystal irregularities, become fixed thereon and excluded from active processes, thus defining in part the irreversible changes in the samples under study.

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Дипольна релаксація, діелектричні та електричні властивості кристалів PbWO₄

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Експериментальні залежності діелектричної проникності нелегованих кристалів PbWO₄ на частоті 1 кГц розглядаються при температурах $T = 290-550$ К. Хід кривої $\epsilon(T)$ при нагріванні та при охолодженні відмінний. Функція $\epsilon(T)$ при нагріванні характеризується наявністю груп вузьких максимумів при 290–330 К та 330–400 К з домінуванням перших. При 400–470 К спостерігається лінійна залежність $\epsilon(T)$, вище 470 К закон зміни ϵ близький до експоненційного. Процес відновної релаксації значень ϵ у межах 25–30 при 290 К після високотемпературного нагрівання здійснюється в декілька стадій. Дипольна поляризація та стрибковий механізм обміну зарядами між складними дипольними асоціатами визначають особливості $\epsilon(T)$ при низьких температурах. Домінуючими дефектами структури є пари вакансій свинцю та кисню — диполони, на основі яких утворюються складніші дипольні комплекси.