Parameters of Deep Traps Responsible for Dark Conductivity of ZnSe Single Crystals

V.Ya. Degoda[a], G.P. Podust[a,*], N.Yu. Pavlova[b] and N.V. Martynyuk[c]

[a] *Taras Shevchenko National University of Kyiv, Physics Department, 03680 Kyiv, Ukraine*
[b] *National Pedagogical Dragomanov University, Kyiv, Ukraine*
[c] *Lviv Polytechnic National University, Lviv, Ukraine*

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Doi: 10.12693/APhysPolA.140.78 *e-mail: podust@univ.kiev.ua

The use of ZnSe single crystals as semiconductor detectors of ionizing radiation at room temperature requires determining the nature of their dark conductivity. Studies of the current–voltage characteristics of photo- and dark conductivity as well as temperature dependence of dark conductivity allowed us to establish that dark conductivity in ZnSe crystals is due to the thermal delocalization of electrons from deep traps. Electrons fall into these deep traps due to thermal ionization of shallow donors. A method for determining the concentrations of deep traps and their occupation degree is proposed. The paper demonstrates the use of this method for high-resistance ZnSe single crystals. In these crystals, the concentrations of deep electron traps reach 10^{15} cm^{-3} and are located at around 0.6–1 eV in the band gap.

topics: dark conductivity, deep electron traps, current–voltage characteristics, ZESe single crystals

1. Introduction

Zinc selenide (ZnSe) belongs to the wide-bandgap materials of $A^{II}B^{VI}$ type [1–7] and is used for the production of semiconductor electronic devices and information display systems. For the last decade, another promising direction for ZnSe application has been developed — ionizing radiation detectors with an indirect [4–7] and direct conversion of high-energy radiation into electric current [8, 9]. Today, due to the summation of its electrophysical, physicochemical, luminescent properties and radiation resistance, zinc selenide doped with tellurium, ZnSe (Te), is one of the most efficient scintillators when used in detectors of the "scintillator–photodiode" type [5–7].

The use of specially undoped ZnSe as a semiconductor detector became possible only after the development of technologies for growing sufficiently high-quality single crystals, i.e., crystals with low concentrations of uncontrolled impurities and high resistivity of the material at the level of the order of 10^{10}–10^{14} Ω cm at room temperature. However, the use of ultra-pure Si and Ge as classical semiconductor detectors at room temperature is impossible due to the high value of their own conductivity. The concentration of free carriers in these materials is much higher than the concentration of carriers generated by ionizing radiation. Although ZnSe-based semiconductor detectors run well under radiation exposure in the mode of direct current registration, only some samples allow detecting the current pulses. This is due to a significant concentration of traps in ZnSe crystals, which reduces the current pulse amplitude and increases its duration [2].

The aim of this work was to establish the concentration and occupation degree of deep traps which cause dark conductivity in high-resistance ZnSe single crystals. Unfortunately, to date, very few studies have examined the parameters of traps in zinc selenide [10–15].

2. Experimental method

The ZnSe single crystals were grown from the precleaned charge and not doped during their growth. As a result, we obtained crystals with a minimum concentration of point defects and maximum resistivity ($\geq 10^{12}$–10^{14} Ω cm). For the study purposes, the samples of $18 \times 9 \times 2$ mm^3 were cut from two different crystal boules, and polished. To study the conductivity, the indium electrical contacts in the form of two parallel bands (1×5 mm^2, i.e., $L = 5$ mm) were sprayed onto the single crystals. Copper electrodes were soldered to the contacts

distanced to $d = 5$ mm. The indium, selected as a contact, provided the ohmicity of the contact [16, 17]. Studies of current–voltage characteristics confirmed that such contacts are ohmic. A stabilized voltage in the range from 0 to 1000 V was applied to one electrode, while another contact was grounded via a nanoamperemeter. The nanoamperemeter allowed to measure the current (i) from 1 pA to 10 pA with an accuracy of 10%, (ii) from 10 pA to 100 pA with an accuracy of 3%, and (iii) from 100 pA to 1 mA with an accuracy of < 1%. For all values of the conductivity current, the condition was fulfilled that the input impedance of the nanoamperemeter is several orders of magnitude less than the electrical resistance of the ZnSe sample. The nanoamperemeter is a part of the specially developed measuring block which allows to choose the voltage change mode: stepped or monotonic with a possibility of smooth change of voltage with various speeds.

For all ZnSe samples, the dark current–voltage characteristics (CVC) have been obtained at room temperature. As it is known, it is impossible to take the I–V curve at lower temperatures even at high applied voltages due to low current. Therefore, dark conductivity was investigated at higher temperatures (from 250 to 410 K). The sample was placed in a cryostat, hence we were able to use different temperatures in the range of 8–500 K. Heating of the sample was carried out using an electric heater up to 800 W built-in into the cryostat, and cooling was carried out with liquid nitrogen.

For excitation of photoconductivity, seven UV LEDs placed on one platform of Ø = 20 mm were used, having a common power supply unit. This setup allowed changing the value of stabilized current in the range of 30–180 mA. The LEDs were of UF-301 type (with radiation maximum at 395 nm, i.e., the energy of UV-quanta was greater than the width of the ZnSe bandgap). The radiation of each LED was directed to the sample through a quartz window of the cryostat from a distance of 55 mm. The intensity of incident UV radiation (IUV) was determined using an optical power meter IMO-2N (it measures the average power and energy of laser radiation $I_{\mathrm{UV}} = 0.156$ mW/cm^2).

3. Results and discussion

3.1. Dark conductivity

Undoped ZnSe crystals of high optical quality have a low dark electron conductivity, which is caused by thermo-activated delocalization of electrons from deep levels [18]. In the general case, the I–V characteristics of the dark conductivity of ZnSe crystals are nonlinear (see Fig. 1), but the initial sections of the I–V characteristics are linear. It was previously found that this nonlinearity of the I–V characteristics at room temperature is due to the Poole–Frenkel effect [18].

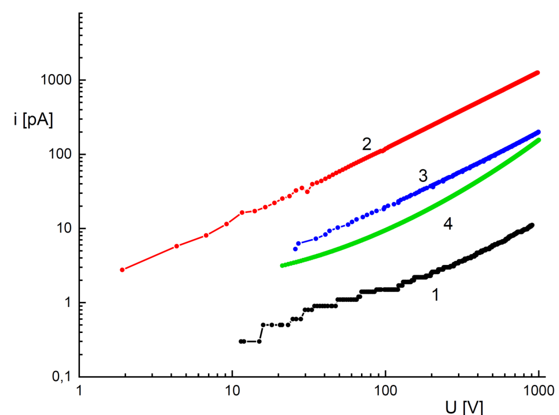

Fig. 1. I–V characteristics of dark conductivity of four ZnSe samples (denoted #1,..., #4) at 295 K.

From the I–V characteristics, the concentration of free electrons can be determined using the classical dependence for the current density in semiconductors. The current density j for an n-type semiconductor, providing the electrical contacts are ohmic, is defined as:

$$j = eN^- \mu^- E, \tag{1}$$

where N^- is the concentration of free electrons, μ^- is mobility of free electrons, E is the electric field strength ($E = U/d$, where U is the applied voltage, d is the distance between the electrical contacts). In the classical case of the contacts placing, $j = i/S$ and i is the magnitude of the conduction current, and S is the cross-section area. However, it is necessary to take into account the specific geometry of electrical contacts and their area [18]. In our case, when the ohmic contacts in the form of two parallel strips are on the same surface of the sample, the correcting geometric factor to be taken into consideration is $g = 0.0736$ cm. This factor shows the ratio of the contact area to the distance between the strips.

As a result, the concentration of free electrons that cause dark conductivity at room temperature is

$$N^- = \frac{i}{e\mu^- U}\frac{d}{S} = \frac{i_D}{e\mu^- U g} = \frac{1}{e\mu^- g}\left(\frac{\Delta i_D}{\Delta U}\right)_{\mathrm{init}}. \tag{2}$$

where i_D is the experimentally measured conduction current value taken from the current-voltage dependence.

We have evaluated the free charge carriers concentration in ideal ZnSe crystals at room temperature and found that $N_{\mathrm{id}} = P_{\mathrm{id}} = \sqrt{N_{\mathrm{C}} N_{\mathrm{V}}}\, \mathrm{e}^{-E_g/(2k_{\mathrm{B}}T)} = 5 \times 10^{-5}$ cm^{-3}. Here, N_{C} denotes the effective density of states for electrons in the conduction band, N_{V} is the effective density of states for holes, E_g is the band gap energy, k_{B} is the Boltzmann constant, and T is the temperature. Note that the concentration N_{id} in such ideal ZnSe crystals is equal to its own conductivity P_{id}.

TABLE I

Parameters of ZnSe samples.

Sample # of ZnSe	N_D [nm^{-3}] at 295 K	E_d [eV]	c_e	τ [s]	v_d [cm^{-3}]
#1	2.8×10^3	1.03	0.998	7.5×10^{-7}	5.9×10^{16}
#2	6.8×10^5	0.68	0.140	1.3×10^{-9}	2.2×10^{19}
#3	2.4×10^4	0.94	0.994	3.6×10^{-8}	1.1×10^{18}
#4	1.5×10^4	0.60	1.51×10^{-4}	1.2×10^{-7}	2.1×10^{17}

The electron concentrations obtained from the initial areas of the I–V characteristics of dark conductivity for four samples are given in Table I. These concentration values differ by orders of magnitude for different samples of ZnSe crystals, but the source of them is not indicated.

The results at this stage indicate that the dark conductivity of ZnSe crystals is due to uncontrolled impurities and structural defects. Therefore, the study of the temperature dependence of dark conductivity was carried out.

3.2. Temperature dependence of dark conductivity

The temperature dependence of dark conductivity makes it possible to determine from which levels the thermal delocalization of electrons occurs. Figure 2 shows the characteristic temperature dependence of the dark conductivity of four different samples of ZnSe single crystals at an applied voltage $U = 165$ V. The activation energy of dark conductivity was determined from the slope of the dependence of the conductivity on the inverse temperature in a logarithmic scale.

The following values of dark conductivity activation energy were obtained (see Fig. 2):

- $E_{\mathrm{T}} = 1.03$ eV for sample #1 (curve 1);
- $E_{\mathrm{T}} = 0.68$ eV for sample #2 (curve 2);
- $E_{\mathrm{T}} = 0.94$ eV for sample #3 (curve 3);
- $E_{\mathrm{T}} = 0.60$ eV for sample #4 (curve 4).

The activation energies close to the obtained values are observed in ZnSe in [19], and they are consistent with the data of thermostimulated luminescence and thermostimulated conductivity. Thus, we have the appropriate depths of traps, which cause dark conductivity at room temperature. The chance of the existence of deep donor levels in these crystals cannot be ruled out. In some ZnSe samples, two linear regions are observed in these coordinates. This means that at room temperature all deeper levels are filled and less deep traps are partially filled.

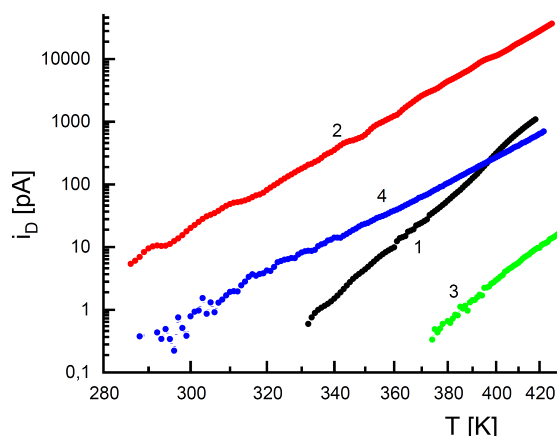

Fig. 2. Temperature dependence of dark conductivity of four different samples of ZnSe single crystals at the applied voltage $U = 165$ V.

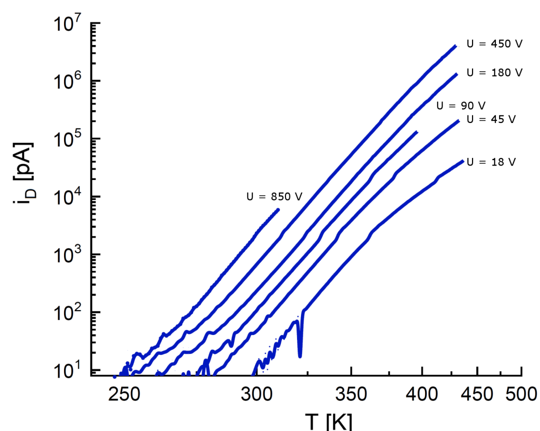

Fig. 3. Temperature dependence of dark conductivity of sample #2 of single crystalline ZnSe at different values of applied voltage..

3.3. Influence of electric field magnitude on temperature dependence of dark conductivity

The temperature dependence of the dark conductivity of one of the samples (e.g., #2) of single crystalline ZnSe at different voltages in the range of 15–900 V was investigated. The obtained dependence is shown in Fig. 3. These results have shown that there is a slight decrease in activation energy as the applied voltage increases.

A slight change in the slope of the curves in these coordinates with increasing voltage is explained by the influence of the Poole–Frenkel effect [18]. As expected, the magnitude of the electric field does not fundamentally affect the temperature dependence of dark conductivity in ZnSe crystals.

High-resistance ZnSe single crystals have small concentrations of shallow donors ($n_{\text{sh-don}}$), as evidenced by the presence of the edge glow in photo- and X-ray luminescence spectra at low temperatures [20, 21]. At higher temperatures ($T > 200$ K), electrons pass into deep traps. It is the thermal delocalization of electrons from deep centers that actually causes ZnSe's dark conductivity at high temperatures. Increasing the concentration of shallow donors in single crystals can lead to complete filling of the deepest trap and partial filling of the shallower trap. Hence, the local level that causes dark conductivity becomes shallower.

It is obvious that there are more shallow levels between the deep level E_d in the band gap and the conduction band. Their dependence in the equilibrium state described as n_m can be determined by the Boltzmann distribution

$$\frac{n_m}{\nu_m} = \frac{n_d}{\nu_d} \exp\left(-\frac{E_d - E_m}{k_\mathrm{B}T}\right). \tag{3}$$

Index m corresponds to a given shallow level of energy E_m in the region $d < m$.

In fact, the trap frequency factors w_{0m} do not differ much from each other [22], and the concentration of free electrons in the conduction band will be determined by the thermal delocalization of electrons from all traps. Then

$$\sum_m w_m n_m = \sum_m w_{0m} \exp\left(-\frac{E_m}{k_\mathrm{B}T}\right) \frac{n_d \nu_m}{\nu_d} \exp\left(\frac{E_m - E_d}{k_\mathrm{B}T}\right) = w_{0d} n_d \,\mathrm{e}^{-E_d/k_\mathrm{B}T} \sum_m \frac{\nu_m}{\nu_d}, \tag{4}$$

which means that the concentration of a deep trap should be understood as the sum of the concentrations of all deep traps. This justifies the usage of a system of kinetic equations — two equations only — to analyze dark conductivity in such high-resistivity crystals. The first equation is for free electrons N^-, and the second equation is for localized electrons n_d at deep levels in the band gap with concentration v_d. Since other centers are shallow, their contribution to the system of kinetic equations can be neglected. Now, the system of differential equations has the form

$$\begin{cases} \frac{\mathrm{d}N^-}{\mathrm{d}t} = w_d n_d - N^- u^- \sigma_d^- \left(\nu_d - n_d\right) \\ \frac{\mathrm{d}n_d}{\mathrm{d}t} = -w_d n_d + N^- u^- \sigma_d^- \left(\nu_d - n_d\right) \end{cases}. \tag{5}$$

Here, $w_d = w_{0d} \exp\left(-E_d/k_\mathrm{B}T\right)$ is the probability of thermal delocalization of an electron from a deep trap, w_{0d} is the trap frequency factor, σ_d is the cross-section of the localization of the free electron to the deep trap. For a steady state, when $T = \text{const}$, $t \to \infty$ and time derivatives are zero, this system passes into two identical algebraic equations, i.e.:

$$N^- u^- \sigma_d^- \left(\nu_d - n_d\right) = w_{0d} n_d \,\mathrm{e}^{-E_d/k_\mathrm{B}T}, \tag{6}$$

or equivalently, $N^- u^- \sigma_d^- (1 - c_e) = w_{0d} c_e \,\mathrm{e}^{-E_d/k_\mathrm{B}T}$, where $c_e = n_d/v_d$ is the occupation degree of the deep trap. Using the relation obtained in [22] for σ_d:

$$\sigma_d^- = \frac{w_{0d}}{u^- N_\mathrm{C}}, \tag{7}$$

we could provide the occupation degree of the deep trap as

$$\frac{N^- u^- w_{0d}}{N_\mathrm{C} u^-} = \frac{c_e w_{0d}}{1 - c_e} \exp\left(-\frac{E_d}{k_\mathrm{B}T}\right) \Longrightarrow c_e = \frac{1}{1 + \frac{N_\mathrm{C}}{N^-} \exp\left(-\frac{E_d}{k_\mathrm{B}T}\right)}, \tag{8}$$

where $N_\mathrm{C} = 2\left(2\pi m_e^* k_\mathrm{B}T\right)^{3/2}/h^3$ is the effective density of states of electrons in the conduction band, m_e^* is the effective mass of the electron, and h is Planck's constant. Obviously, the condition $n_d > N^-$ will be met. The obtained values of c_e are given in Table I. They allow to verify what kind of the deep levels causes the dark conductivity of the crystal — the deep traps are partially filled with electrons. Also, having E_d and c_e it is possible to determine the position of the Fermi level in the band gap for each crystal.

Let us put together some facts: the depth of the trap is already known from the temperature dependence of dark conductivity as well as its frequency factor [23], and the thermal velocity of the free electron u^- is easily calculated as soon as its effective mass [24] is known. Only the concentrations of deep traps (ν_d) remain unknown. However, the value of ν_d can be determined from the average lifetime of an electron in the conduction band.

The average lifetime of a free electron turns out to be the same under excitation and without it. Therefore, the lifetime of a free electron can be estimated according to studies of the I–V characteristics of photoconductivity (PC), assuming the excitation intensity and electron mobility at room temperature are known [2]. The peculiarity of photoconductivity in contrast to dark conductivity is that the photoconductivity current does not flow through the entire sample, but only in the excitation region.

3.4. Volt–ampere characteristics of photoconductivity

The concentrations of free charge carriers generated by UV excitation are orders of magnitude higher than electron concentrations in the absence of excitation. The photoconductivity current (Fig. 4) significantly exceeds the dark current in ZnSe samples. Figure 4 shows the I–V characteristics of these ZnSe samples.

In [25], dependencies of photo- (PC) and X-ray conductivity (XRC) currents (respectively denoted as i_PC and i_XRC) were obtained, taking into account the absorption of excitation radiation. Thus

$$i_{\mathrm{PC}} = \frac{eI_{\mathrm{UV}}L}{h\nu_{\mathrm{UV}}}\mu^{-}\tau^{-}E \qquad (9)$$

and

$$i_{\mathrm{XRC}} = \frac{eI_{\mathrm{X}}L}{3E_g}\left(1 - C_{\mathrm{sc}}\right)\mu^{-}\tau^{-}E, \qquad (10)$$

where I_{UV}, I_{X} are the intensities of incident UV or X-ray excitation radiation, $h\nu_{\mathrm{UV}}$ is the UV quantum energy, C_{sc} is the scintillation efficiency of X-ray excitation, E_g is the ZnSe band gap width, μ^{-} is the mobility of a free electron, and τ^{-} is the lifetime of a free electron in the conduction band. If the intensity of the excitation radiation I_{UV} is known, one can calculate the number of free carriers generated per unit time in the excitation volume of the sample. The number of electrons that determines the photoconductivity current will be equal to the product of the number of generated electrons and their lifetime in the conduction band. With the transformation of (9), one gets

$$\tau^{-} = \frac{i_{\mathrm{PC}}}{U}\frac{h\nu_{\mathrm{UV}}d}{e\mu^{-}I_{\mathrm{UV}}L} = \frac{h\nu_{\mathrm{UV}}d}{e\mu^{-}I_{\mathrm{UV}}L}\left(\frac{\Delta i_{\mathrm{PC}}}{\Delta U}\right)_{\mathrm{init}}. \qquad (11)$$

Relying on the initial linear dependence of the I–V characteristics of PC, the values of τ^{-} at room temperature were determined (see Table I). From the photoluminescence and photoconductivity kinetics, the lifetime of free electrons in the conduction band is determined with

$$\tau^{-} = \frac{1}{u^{-}\sigma^{-}v_d}. \qquad (12)$$

Hence, using (7) and (12), we obtain the expression for the concentration of deep traps in the form

$$v_d = \frac{N_{\mathrm{C}}}{w_{0d}\tau^{-}}. \qquad (13)$$

The values of (13) for ZnSe samples are given in Table I. Determining the total number of electrons in deep traps, we obtain $n_{\mathrm{sh-don}} = c_e v_d$, i.e., the concentration of shallow donors that have provided these electrons.

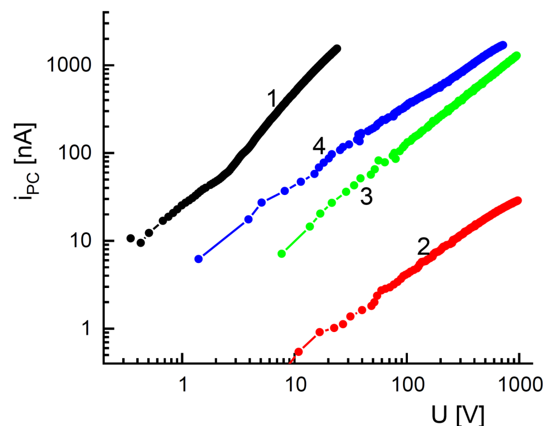

Fig. 4. I–V characteristics of the photoconductivity of ZnSe samples at room temperature ($I_{\mathrm{UV}} = 0.156$ mW/cm^2).

With the sequential studies of the I–V characteristics of dark conductivity, the temperature dependence of dark conductivity and the I–V characteristics of photoconductivity of samples, it is possible to determine the level of fill factors and concentrations of deep traps that cause the dark conductivity of ZnSe crystals. First, the concentration of free electrons at room temperature is determined from the CVC of dark conductivity. Then, the trap depths obtained from the temperature dependence of dark conductivity allow to determine their frequency factors from [22, 23] and to calculate the density of states in the conduction band. At this stage, the fill factor of deep traps is provided. Next, the I–V characteristics of the photoconductivity are measured and the lifetime of the free electron in the conduction band at room temperature is determined, which gives the concentration of these traps. From the total concentration of localized electrons in the deep trap at room temperature, the total concentration of small donors can be estimated.

Now, it is necessary to check how the current of dark conductivity i_d depends on temperature. It is known that $i_d \sim N^{-}\mu^{-}$, where $\mu^{-} \sim T^{-3/2}$ and $N^{-} \sim T^{3/2}\mathrm{e}^{-E_d/k_{\mathrm{B}}T}$ (and also $N^{-} \sim w_{0d}\mathrm{e}^{-E_d/k_{\mathrm{B}}T} \sim N_{\mathrm{C}}\mathrm{e}^{-E_d/k_{\mathrm{B}}T}$). As a result, the temperature dependence of dark conductivity $i_D \sim \exp(-E_d/k_{\mathrm{B}}T)$ is determined only by the exponent. If the temperature dependence of dark conductivity on the inverse temperature in a logarithmic scale gives two linear sections, i.e., there are two traps (with concentrations ν_{d1} and ν_{d2} and level depths E_{d1} and E_{d2}), then the concentration and occupation degree of the only less deep trap will be determined by this method. Obviously, for this case, the occupation degree of a deeper trap will be maximal. The concentration of this deeper trap can be estimated from the temperature dependence of dark conductivity, by approximating the total current by a function

$$i_D = i_{d1}\exp\left(-\frac{E_{d1}}{k_{\mathrm{B}}T}\right) + i_{d2}\exp\left(-\frac{E_{d2}}{k_{\mathrm{B}}T}\right). \qquad (14)$$

Then $i_{d1}/i_{d2} = c_{e1}\nu_{d1}w_{0d1}/(\nu_{d2}w_{0d2})$, which allows estimating the concentration of the filled deeper trap ν_{d2}.

When comparing the ZnSe samples with each other, it is obvious that sample #2 has the lowest resistivity, the shortest lifetime of free electrons in the conduction band and the lowest photoconductivity current due to the highest concentration of deep traps. Of course, such a ZnSe crystal cannot be used as a UV or X-ray detector.

4. Conclusions

Specially undoped high-resistance ZnSe crystals of high optical quality have a low dark electron conductivity, which is due to thermo-activated delocalization of electrons from deep traps. A method for determining the concentration of these centers, the

depth of their levels in the band gap and their occupation degree is proposed. It also allows to estimate the concentrations of shallow donors in ZnSe crystals. Different concentrations of deep traps in different ZnSe crystals indicate that the technology of growing ZnSe monocrystals can be further improved to obtain crystals with a lower concentration of defects and uncontrolled impurities.

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