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ELECTRONIC AND OPTICAL PROPERTIES OF SEMICONDUCTORS

The Spread of Cross Section for Electron Capture by a Trap with a Discrete Energy Level in γ-La_2S_3 Crystals

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Abstract—The data on a new phenomenon (a formation of the range of values for an electron-capture cross section) are reported by the example of an electron trap with a discrete level of $E_c - 0.2$ eV in γ-La_2S_3 crystals; the data were obtained by employing the thermally stimulated luminescence. The range of variations in the cross section is as large as four orders of magnitude (10^{-23}–10^{-19} cm^2). A model, according to which the electron trap at $E_c - 0.2$ eV is a donor involved in the donor–acceptor pairs distributed in interatomic distances and localized in the vicinity of a negatively charged dislocation, is suggested. It is shown that the formation of a range of electron-capture cross sections is a result of a spread of the cross-section modulation factor at points with different values of potential of the dislocation electric field. *© 2001 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

Impurity-photoconductivity studies of fast electron traps related to donor–acceptor pairs (DAPs) and to single-, double-, and multiple-donor particles in II–VI compounds [2–9] have shown that these traps possess large electron capture cross sections ($S_t \geq 10^{-15}$ cm^2) characteristic of point defects with an attractive potential; in addition, it has been shown that the cross sections S_t decrease with increasing ionization energy (E_t) of the center in accordance with the theory of the charge-carrier nonradiative capture [10].[1]

An analysis of the known data on the kinetic properties of slow electron traps ($R \ll 1$) makes it possible to state that there are two groups of electron traps of this type. Slow electron traps of the first group are characterized by theoretically predictable kinetic properties, whereas the electron traps of the second group possess anomalous kinetic properties and are photoelectrically inactive [9, 11]. As the results of studying the thermally stimulated phenomena showed, these slow traps may feature the cross sections S_t that (irrespective of the electron-trap origin) decrease exponentially to very small values on the order of 10^{-25} cm^2 as the energy released during electron localization decreases (see, for example, [6, 9, 11–16]). If we hold to the conventional concepts of individual properties of both kinetic parameters and slow electron traps belonging to the second group, then their interpretation meets serious difficulties from the standpoint of both the statistical aspect of the center's charge state and the dynamic aspect of the mechanism of capturing the charge carriers by these centers.

Previously [6, 11], one of us suggested a model according to which the electron traps with anomalous kinetic properties are point defects located in the region of collective electric fields within large-scale imperfections in a crystal. If the cross sections S_t for both the fast and normal slow electron traps are intrinsic (unperturbed by other defects), then the cross sections of electron traps localized in the region of macroinhomogeneities become effective because of modulation by collective potential fields; to the first approximation, these cross sections may be represented as $S_t = S_{t0}\exp(-\varphi/kT)$. Here, S_{t0} is the intrinsic cross section, and φ is the recombination-macrobarrier height. This model not only accounts for the known anomalous properties of slow electron traps (for example, the exponential increase in the cross sections S_t with increasing E_t), but also leads to a number of other inferences [11]. In particular, a discrete cross section S_t of a separate electron trap may transform into a range of values without changing the energy spectrum. A necessary condition for this is the distribution of electron-trap atoms over the entire volume of the macroinhomogeneity, within which the field potential changes.

In this paper, we report for the first time the experimental evidence for the existence of cross section S_t expansion into the range of values; this evidence was obtained by the method of thermally stimulated luminescence (TSL) for an electron trap with a discrete level of $E_c - 0.2$ eV in γ-La_2S_3 crystals.

[1] According to the accepted concepts, the fast electron traps are the centers for which the ratio of the probability of electron capture to the probability of electron recombination is $R \gg 1$ [1].

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2. PHOTOELECTRIC AND THERMOLUMINESCENT PROPERTIES OF γ-La_2S_3

The photoconductivity spectrum of γ-La_2S_3 crystals at 295 K consists of two bands, the intrinsic and impurity-related (Fig. 1, curve *a*) bands. The photosensitivity to the light in the fundamental-absorption region increases with decreasing temperature, whereas the photosensitivity to impurity-absorption light first increases, attains a maximum in the vicinity of T = 190 K, and then decreases with a deactivation energy of 0.06 eV. At 90 K, only an intrinsic band is observed in the photoconductivity spectrum (Fig. 1, curve *b*).

The TSL of γ-La_2S_3 features a number of nontrivial properties. These properties manifest themselves most clearly in the measurements allowing for a variation in the photon energy of the preliminary excitation radiation. If integral light from the impurity-photoconductivity region ($h\nu \cong 2.0$–2.6 eV) is used for low-temperature preliminary excitation, a single broad band with a poorly resolved structure is observed in the TSL spectrum (Fig. 2, curve *a*). This band decomposes into a series of daughter bands if photons belonging to the impurity-photoconductivity region and having a certain energy are used for excitation. The daughter band located at the highest temperature corresponds to the photons with the lowest energy and shifts to lower temperatures as the energy $h\nu$ increases within the impurity-photoconductivity range (Fig. 2, curves *a*'–*e*'). The largest TSL-band shift amounts to 85 K. Preliminary excitation with light with a wavelength in the fundamental-absorption range gives rise to a TSL spectrum with a single discrete band at T_m = 110 K of another origin (Fig. 2, the dashed curve). Under the conditions of combined excitation with light corresponding to fundamental and impurity absorption, this band is adjacent to the broad TSL band on the low-temperature side.

The γ-La_2S_3 TSL possesses other properties as well. Dependence of the TSL intensity on the photon energy within the impurity-absorption range at the peaks of the amplitude-saturated daughter bands replicates the profile of the impurity-photoconductivity band (cf. curves *a* and *c* in Fig. 1). Preliminary excitation of γ-La_2S_3 with impurity-absorption light at 90 K, which is necessary for the observation of TSL, is not accompanied by photoconductivity and photoluminescence.

The thermally stimulated currents in γ-La_2S_3 possess similar properties. This is also evidenced by the results of pioneering studies of thermally stimulated currents in γ-La_2S_3 [17]; unfortunately, the kinetic parameters of electron traps were not estimated.

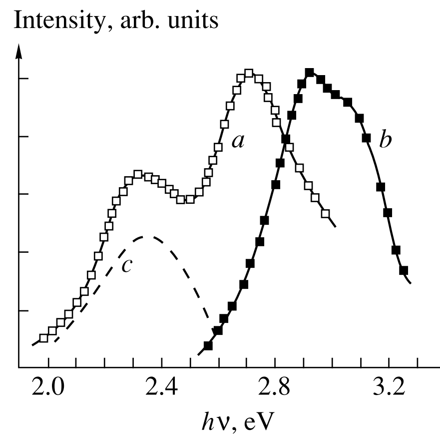

Fig. 1. Photoconductivity spectra of γ-La_2S_3 crystals at (*a*) 295 and (*b*) 90 K. Curve *c* represents the dependence of amplitude values of intensities of the daughter TSL bands on the photon energy of preliminary excitation.

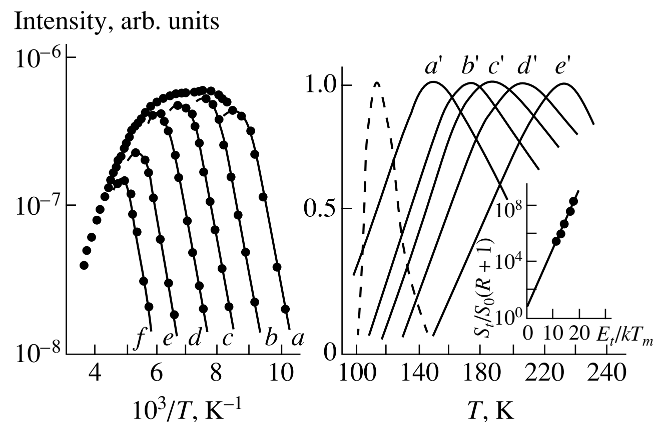

Fig. 2. (*a*) Integral TSL spectrum of the γ-La_2S_3 crystal measured with preliminary excitation with integral light belonging to the impurity band of photoconductivity ($h\nu \cong$ 2.0–2.6 eV). Curves (*b*–*f*) represent the calculated TSL spectra. The points correspond to experimental values of TSL intensities measured using thermal purification. Curves *a*'–*e*' represent a series of normalized daughter TSL bands for γ-La_2S_3. These bands are observed under preliminary irradiation with photons with energies $h\nu$ = (*a*') 2.58, (*b*') 2.48, (*c*') 2.3, (*d*') 2.13, and (*e*') 2.07 eV. The dashed line represents the TSL observed under preliminary exposure to the light corresponding to fundamental absorption ($h\nu \cong$ 3.0 eV). The recording rate for the TSL spectra was β = 0.16 K/s. The inset shows the straight line corresponding to the universal plot of $S_t/S_0(R + 1)$ vs. E_t/kT_m [24]; the points correspond to characteristic parameters of the electron trap with $E_c - 0.2$ eV and the daughter *a*'–*e*' bands.

3. EFFECT OF EXPANDING OF THE CROSS SECTION S_t TO A RANGE VALUES

As a rule, structurally complex thermally stimulated spectra are interpreted on the assumption that these spectra are related to an electron-trap set corresponding to the number of bands; each of the traps is characterized by the discrete parameters E_t and S_t. The γ-La_2S_3 TSL spectrum observed under excitation with integral light corresponding to the impurity absorption (Fig. 2, curve *a*) may be regarded as a structurally complex

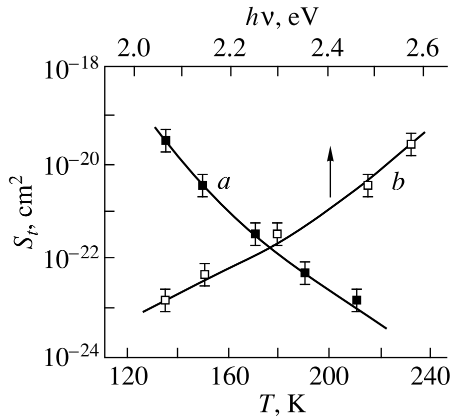

Fig. 3. Dependences of the cross section S_t of the electron trap with the level of $E_c - 0.2$ eV on (*a*) the temperature corresponding to the peaks of the daughter TSL bands in γ-La_2S_3 and (*b*) the energies of excitation photons.

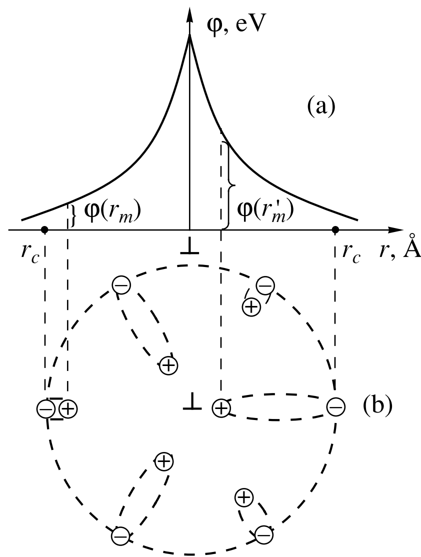

Fig. 4. (a) Hypothetical curves describing the variation in the electric-field potential of a negatively charged dislocation as the distance from its core increases; r_c is the distance from the dislocation core to the DAP acceptor centers; and $\varphi(r_m)$ and $\varphi(r'_m)$ are the heights of recombination barriers for electrons trapped by DAPs with r_m and r'_m. (b) A section of dislocation tube; for the sake of clarity, six arbitrary dipoles with different interatomic distances r_m are shown in the cross-section plane.

spectrum. However, analyzing it by "thermal purification" gives rise to the concept of a new type of electron trap with a single level $E_c - 0.2$ eV and a cross section S_t expanded to the range of 10^{-23}–10^{-19} cm^2.

The energy $E_t = 0.2$ eV is determined from the slope of the linear logarithmic dependence of the TSL intensity at the initial stage of its increase on the reciprocal temperature [18, 19]; i.e.,

$$\ln I = C - \frac{E_t}{kT}. \quad (1)$$

In accordance with the above statement that the electron-trap spectrum is monoenergetic ($E_t = E_c - 0.2$ eV), the slopes of the straight lines separated within the broad TSL band by thermal purification coincide (cf. curves *a–e* in Fig. 2) and are equal to 0.2 ± 0.01 eV. The cross sections were estimated with the formula [20, 21]

$$S_t = \frac{\beta I_{\text{ext}}}{v N_c \Delta T}, \quad (2)$$

however, formulas (1) and (2) may be used if the initial stage of an increase in TSL can be identified. In formula (2), β is the rate of sample heating in the course of recording the TSL spectra, N_c is the effective density of electron states in the conduction band, and v is the electron thermal velocity. The quantity I_{ext} is the extrapolated value of TSL intensity at the intersection point of the straight line given by (1) with the ordinate corresponding to $T^{-1} = 0$ under the condition that the amplitude value of TSL intensity is normalized and is equal to unity (in arbitrary units). In the case of complex spectra, the half-width ΔT may be determined to a reasonable accuracy as the doubled value of the half-width of the low-temperature component of the discrete band extracted by thermal purification. Under the same condition of amplitude value normalization, formula (2) can be used to estimate the S_t cross section using not only the TSL method but also other thermal stimulation methods.

The dependences of the cross section S_t on the photon energy in the preliminary excitation of the daughter TSL bands and on the temperature positions T_m of the peaks of these bands are shown in Fig. 3.

The reliability of the values of E_t and S_t measured using (1) and (2) is supported by good agreement between the contours of experimental TSL bands (they are represented by points in Fig. 2) and those of the TSL bands calculated with the formula [22, 23]

$$I(T) = \nu_t \exp\left(-\frac{E_t}{kT}\right) \times \exp\left[-\frac{\nu_t}{\beta}\left(\frac{kT^2}{E_t}\right)\exp\left(-\frac{E_t}{kT}\right)\left(1 + \frac{4kT}{E_t}\right)\right] \quad (3)$$

the results of calculations are shown by continuous lines *a–f* in Fig. 2. When calculating $I(T)$, we used experimental values E_t and the frequency factor $\nu_t = vN_cS_t = \frac{\beta I_{\text{ext}}}{\Delta T}$ [see formula (2)]. In order to prove that the parameters E_t and S_t are reliable and that the electron traps with $E_t = E_c - 0.2$ eV belong to the class of

slow electron traps, we demonstrated that these parameters along with the parameters of the daughter TSL bands (see the inset in Fig. 2, the points) fall on the previously [24] suggested universal plot [$S_t/S_0(R + 1)$ vs. E_t/kT_m] for the characteristic parameters of thermally stimulated spectra (T_m and β) and electron traps (E_t, S_t, and R) in semiconductors and insulators (see the inset in Fig. 2, the straight line) provided that the capture factor $R = 0$. The slope of this plot with respect to the horizontal axis E_t/kT_m is equal to 0.45. The coordinates of the origin point are (5, 0). The quantity S_0 is defined as $S_0 = \beta/nN_cT_m$.

4. A MODEL OF ELECTRON TRAP WITH $E_t = E_c - 0.2$ eV

As a rule, the electron-trap atoms and the TSL-excitation centers are spatially separated. Their wave functions do not overlap, which rules out the process of direct transfer of charge carriers between them. An attempt to interpret correctly the nontrivial special features of TSL in γ-La_2S_3 suggests that the corresponding thermoluminescence-active centers are structurally complex and that the nonequilibrium accumulation of electrons at these centers may be accomplished only by the extraband (interimpurity) mechanism.

According to the multiparameter model we suggest, the roles of the electron trap with $E_t = E_c - 0.2$ eV and the corresponding excitation center (the TSL-excitation center) are played by the donor and acceptor, respectively, of the same DAPs that possess a number of unusual properties. These DAPs are distributed in interatomic distances (r_m) and are located around a negatively charged dislocation in such a manner that the acceptor atoms in all the pairs occupy the quasi-equidistant positions relative to the dislocation core, whereas the donor atoms are randomly distributed in the space between this core and the acceptor atoms. The DAPs, together with the dislocation core, form the space charge of the dislocation tube (Fig. 4). Kinetic parameters of the donor incorporated into the DAP are modulated by an electric field of dislocation. The modulation factor depends on the spatial extent of the DAPs; therefore, the latter are distributed not only in r_m and in the energy of interimpurity electron transition $h\nu_m$ but also in the cross section S_t. The donor that acts directly as an electron trap has an excited state E_c – 0.06 eV in addition to the ground trap state $E_c - 0.2$ eV. The distribution of the DAPs in r_m is such that only the wave functions of the donor excited state ($E_c - 0.06$ eV) and the acceptor ground state ($E_V + 0.54$ eV) overlap in the DAPs.

It is also worth noting here that, in the polycrystalline sample, the role of sources of macroscopic electric fields may be played not only by dislocations but also by intercrystallite boundaries.

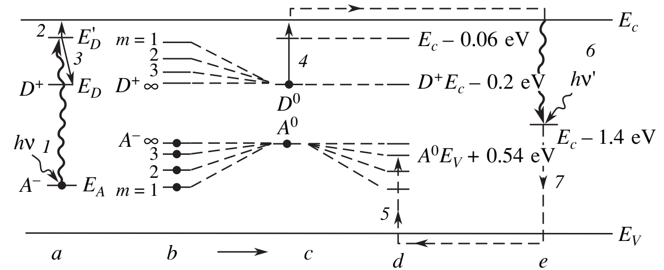

Fig. 5. (*a*) A scheme of energy levels of unexcited $(A^- - D^+)^0$ centers with a certain value of r_m. (*b*) The energy spectrum of the $(A^- - D^+)^0$ DAP ground states distributed in r_m. (*c*, *d*) The energy spectrum of a DAP in the excited state $(A^0 - D^0)^{0*}$ and in the semiexcited state $(A^0 - D^+)^+$. (*e*) The energy level of the recombination center. The electron transitions indicated by the arrows are described in the text.

5. INTERPRETATION OF PHOTOELECTRIC AND THERMOLUMINESCENT PROPERTIES

In terms of the model we suggest, the impurity photoconductivity of γ-La_2S_3 crystals is a result of two-stage ionization of DAPs. The first (optical) stage involves an intracenter transition of electrons from the acceptor's ground state to the donor's excited state (Fig. 5, scheme *a*, transition *1*). In the second stage, a thermal emission from the donor's excited state to the conduction band occurs (transition *2*).

Thermooptical processes "freeze" as the temperature is lowered. Simultaneously, the rate of transition of electrons (transferred optically to the donor's excited state) to the donor's ground state (transition *3*) increases. According to experimental data, the impurity photoconductivity of γ-La_2S_3 is thermally quenched with a rate governed by the thermal ionization energy of the donor's excited state ($E_c - 0.06$ eV).

The spectrum of DAPs distributed in r_m and $h\nu_m$ is polyenergetic (Fig. 5, scheme *b*). The absence of the relevant structure in the impurity-photoconductivity spectra (Fig. 1, curve *a*) indicates that the half-width of individual photoconductivity bands is much larger than the energy spacing between the neighboring levels of DAP.

As a result of the quantum-mechanical condition for the overlap of the wave functions of the acceptor's ground state only with those of the donor's excited state, the electrons transferred via an intracenter transition to the donor's ground state ($E_c - 0.2$ eV) at fairly low temperatures (Fig. 5, scheme *a*, transitions *1*, *3*) remain linked to the above states. These electrons can be neither involved in the transitions from the donor's ground state to the conduction band nor involved in the interimpurity donor ⟶ acceptor transitions. Since, in this temperature region, not only electrons but also holes appearing simultaneously at the acceptor atoms with their deeper state ($E_V + 0.54$ eV) remain bound, the lifetime of the excited (inversely populated) DAP

state may be infinitely long. Such three-level systems are of interest for the efficient "conservation" of optical energy.

The destruction of the inversely populated state by heating the γ-La_2S_3 crystals is accompanied by TSL, which is a result of the thermal emission of electrons from the DAP donor atoms to the conduction band and the subsequent radiative recombination of these electrons. Studies of spectral distribution of the TSL intensity suggest that the center with a level of E_c – 1.4 eV (presumably, the lanthanum vacancy), rather than the DAP directly responsible for the generation and accumulation of stored optical energy, acts as recombination center [25]. The charge carriers released from a DAP (Fig. 5, schemes *c*, *d*, transitions *4*, *5*) arrive at the center with E_c – 1.4 eV via the allowed bands (Fig. 5, scheme *e*, transitions *6*, *7*). The fact that DAPs are recombinationally inactive may be caused by the following two mechanisms. In the range of thermal ionization of the donor with the level of E_c – 0.2 eV, the probability of electron transitions from the excited donor state to the conduction band is higher than the probability of their interimpurity transition from this state to the acceptor's ground state. In addition, the thermal ionization and conversion of a donor to a charged state leads to a shift of the acceptor level towards the valence band and to a decrease in the lifetime of the holes localized at this level. The thermal ionization of an acceptor (Fig. 5, scheme *d*, transition *5*), made easier by interimpurity interaction [26], also rules out the electron transitions from the conduction band to the acceptor DAP. The short lifetimes of charge carriers captured separately by a DAP make it possible to understand why these carriers cannot be transferred to the excited (inversely populated) state under the conditions of γ-La_2S_3 irradiation with light corresponding to the fundamental-absorption region.

The suggested model, according to which TSL excitation is responsible for the low-temperature intracenter electron transitions in a DAP without generation of free charge carriers, can also be used to naturally interpret the "latent" mechanism of the TSL excitation.

A transition of a DAP to an excited state $(D^0 - A^0)^{0*}$ upon absorption of photons with energies corresponding to the impurity absorption results in the "condensation" of their *A* and *D* levels in the single-energy lines (Fig. 5, schemes *b*, *c*). Because of this, only a single unshifted (in depth) trap state E_c – 0.2 eV characteristic of an isolated donor is observed.

As we pass from compact to spatially extended pairs, the donor atoms become more and more involved in the electric field of dislocation with an ever increasing potential (Fig. 4). As a result, the height of the potential barrier for electrons passing to the DAP donor atoms increases, whereas the corresponding cross section S_t decreases. Obviously, the donor cross section S_t expands into a range of values. As r_m increases, it is not only the value of S_t, which governs the temperature position of the daughter TSL bands, that decreases. Simultaneously, the energy of interimpurity electron transition in a DAP decreases as well. Exposure of γ-La_2S_3 to photons with energies corresponding to the impurity absorption leads to the selective excitation of DAP with certain values of r_m, $h\nu_m$, and S_t. This circumstance explains the origin of the decomposition of integral TSL spectrum into daughter bands, the shape of the functional dependences $S_t = f(h\nu)$ and $S_t = f(T_m)$ (Fig. 3), and the consistency between spectral distribution of the impurity-photoconductivity intensity and the intensity amplitudes of the daughter TSL bands (Fig. 1, curves *a*, *c*).

The knowledge of cross sections limiting the range of the values of S_t (Fig. 3) makes it possible to assess the variation in the macrobarrier height Δφ when passing from the most compact of the observable DAPs to the most extended (Fig. 4). The corresponding calculations based on the assumption that $S_t = S_{t0}\exp(-\varphi/kT_m)$ and that the intrinsic cross section S_{t0} is in the range of 10^{-15}–10^{-17} cm^{-2} (as in the case of neutral donors) yield the difference Δφ = 0.21 ± 0.02 eV.

6. CONCLUSION

A number of implications follow from the suggested model of an electron trap with a level of E_c – 0.2 eV; when experimentally studied, these implications may additionally validate this model. In particular, we bear in mind the effect of the destruction of the DAP's inversely populated state by infrared illumination. New studies are also necessary to clarify the DAP origin and the cause of the quasi-equidistant arrangement of acceptor atoms around the dislocation core.

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