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Analytical description of hopping electrical conductivity of compensated semiconductors and calculations on the example of p-Ge : Ga

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Analytical expressions are proposed for the prefactor σ_{03} and the thermal activation energy of direct current electrical ε_3-conductivity $\sigma_h = \sigma_{03}\exp(-\varepsilon_3/k_BT)$ of compensated n- and p-type semiconductors for hydrogen-like impurities. The obtained formulas are applicable to describe the hopping migration of both holes via acceptors and electrons via donors. For certainty, we considered p-type crystalline semiconductors in the range of doping levels corresponding to the insulator side of the insulator–metal (Mott) concentration phase transition. For simplicity, it was assumed that the majority and compensating impurities form a single simple nonstoichiometric cubic lattice in the crystal matrix. The calculation of σ_{03} and ε_3 values is based on the preliminary determination of the characteristic temperature T_3, in the region of which phonon-assisted tunnel hopping of holes via nearest neighbor acceptors is observed. The shift of the top of the v-band deep into the band gap due to the formation of a quasi-continuous band of allowed energy values for v-band holes from the excited states of neutral acceptors is taken into account. The distribution of the density of hole states in the acceptor band was assumed to be Gaussian. The influence of the configurational entropy and thermal entropy of holes in the acceptor band on the values of σ_{03} and ε_3 was also taken into account. The values of σ_{03} and ε_3 calculated from the obtained formulas for moderately compensated p-Ge : Ga are in quantitative agreement with the known experimental data on the entire insulator side of the Mott transition.

Keywords: doped and moderately compensated semiconductor, hydrogen-like impurities, hopping migration of charge carriers via impurities, stationary hopping electrical conductivity, thermal activation energy and prefactor of electrical ε_3-conductivity, mobility edge, p-Ge : Ga crystals.

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Introduction

Temperature dependence of hopping conduction σ_h in case of hole hops between the closest by distance hydrogen-like acceptors in covalent semiconductors (for certainty let us consider the crystalline semiconductors of p-type) is represented in the form [1–5]:

$$\sigma_h = \frac{1}{\rho_h} = \sigma_{03}\exp\left(-\frac{\varepsilon_3}{k_BT}\right), \qquad (1)$$

where $\sigma_{03} \equiv 1/\rho_{03}$ — prefactor determined by extrapolation of dependence $\ln\rho_h(1/T)$ to zero of reciprocal temperature $1/T \to 0$ (Fig. 1), ε_3 — energy of thermal activation of hopping transfer of holes by acceptors, k_B — Boltzmann constant, T — absolute temperature.

Derivation of analytical expressions are of interest and represents a major challenge using equation (1) for values of prefactor σ_{03} and energy of thermal activation ε_3 depending on concentration of N_a acceptors and degree of their compensation K by hydrogen-like donors. Thus, for lightly

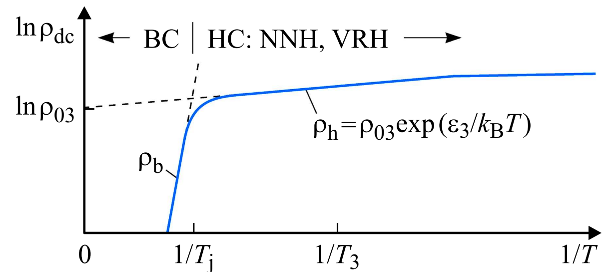

Figure 1. DC electrical resistivity of crystalline semiconductor of p-type ρ_{dc} at Arrhenius scale. At temperature $T = T_j$ band $\sigma_{bj} = 1/\rho_{bj}$ and hopping $\sigma_{hj} = 1/\rho_{hj}$ electroconductivities are equal; $T_3 \approx T_j/3$ — temperature, at which thermal energy of hopping electroconductivity activation is determined; BC (band conduction) — band electroconductivity, HC (hopping conduction) — hopping electroconductivity, NNH (nearest neighbor hopping) — phonon-assisted tunnel hops of holes between closest by distance acceptors in charge states (0) and (−1) with activation energy ε_3; VRH (variable range hopping) — hops of holes between acceptors optimized by energy of activation and length.

doped semiconductors according to Shklovskii and Efros [3]:

$$\rho_{03} = \rho_0 \exp\left(\frac{\delta}{a_i N_a^{1/3}}\right), \quad (2)$$

where ρ_0 — unknown power function of N_a and T; $\delta(K)$ — parameter depending on the compensation ratio (for example, $\delta(0.2) \approx 1.78$, $\delta(0.5) \approx 1.81$, $\delta(0.8) \approx 1.98$); a_i — radius of localization of a light hole on a single acceptor.

At temperatures and levels of doping satisfying the inequation $0.3 < e^2 N_a^{2/3} a_i / 4\pi\varepsilon_r\varepsilon_0 T < 1$, dependence of energy of thermal activation ε_3 on concentration of acceptors N_a and their compensation ratio K for lightly doped semiconductors has the form [3]:

$$\varepsilon_3 = \frac{e^2 N_a^{1/3}}{4\pi\varepsilon_r\varepsilon_0} F(K), \quad (3)$$

where e — elementary charge, ε_r — relative static dielectric permittivity (determined by electrons of v-band on the background of ion cores of the crystal matrix), ε_0 — electric constant, $F(K)$ — function of compensation ratio K (for example, $F(0.2) \approx 0.71$, $F(0.5) \approx 0.73$, $F(0.8) \approx 1.12$).

However, calculations using equation (3) give values of ε_3 that are higher than the ones experimentally observed. Besides, this equation is only applicable in the region of low concentrations of the majority impurities, which yet does not show the overlapping of their wave functions, i. e. it does not take into account the contribution to ε_3-conductance from the splitting of the energy levels of closely located majority impurities („molecular pairs").

Previously in [6–8] the model was proposed for calculation of concentration dependences of prefactor σ_{03} and energy of thermal activation ε_3 in crystalline semiconductors of p- and n-type. It was believed that atoms of impurities create in a crystal matrix a single simple cubic nonstoichiometric lattice. Hops of holes take place at thermally activated „alignment" of energy levels of acceptors in charge states (0) and (-1), while their compensating donors block some of the sites in the impurity lattice.

However, in papers [6–8], when values of σ_{03} and ε_3 were calculated, the selection of the characteristic temperature was not justified, at which hops of holes are implemented between the nearest atoms of the majority impurity. Later in [9] the analytical expression was obtained for temperature $T_3 = T_j/3$ as the characteristic temperature, at which the hops are implemented in the nearest neighbors (NNH). Here T_j — temperature of transition from „free" motion of holes of v-band to their hopping migration between acceptors in charge states (0) and (-1). Besides, [6–8] did not take into account the shift of the top of v-band deep into the band gap due to the formation of a quasi-continuous energy spectrum due to the overlap of the excited states of acceptors with their concentration quite high. Note that the contribution to values of σ_{03} and ε_3 may be made by configurational and thermal entropies (see, for example, [10–12]). Note that in [6–8,13] the impact of compensation ratio on values of ε_3 and σ_{03} was not studied.

The purpose of this paper is to propose the model for quantitative description of the hopping conductivity in the nearest impurities, including behavior values of σ_{03} prefactor and energy of thermal activation ε_3 with the change of the doping level and compensation ratio in the moderately compensated semiconductors on the insulator side of the insulator–metal (Mott) phase transition, and also to compare the analytical and numerical calculations performed on its basis with experimental data [14–23] for crystals p-Ge:Ga. Selection of the material for comparison is caused by the fact that homogeneous and controlled introduction of the majority impurities may be carried out in it — of acceptors (gallium) and compensating donors by neutron transmutation doping with thermal reactor neutrons with subsequent annealing of radiation-induced defects. Physical and technological aspects of such doping in respect to germanium of various isotope composition are presented in [24–26].

1. Model of impurity lattice in crystal matrix for calculation of ε_3-electroconductivity

Let us consider a crystalline semiconductor of p-type, doped with hydrogen-like acceptors with concentration of $N_a = N_{a,0} + N_{a,-1}$ and compensated by hydrogen-like donors with concentration of $N_d = KN_a$. Here $N_{a,0}$ and $N_{a,-1}$ — concentrations of acceptors in charge states (0) and (-1) respectively, $0 < K < 1$ — degree of acceptor compensation by donors (compensation ratio). All compensating donors are in charge state $(+1)$. The condition of electrical neutrality of the crystal at concentration of holes of v-band $p \ll K(1-K)N_a$ has the following form: $N_{a,-1} = KN_a$. Therefore: $N_{a,0} = (1-K)N_a$.

In a crystalline semiconductor of p-type the density of stationary hopping current $J_{hp} = J_{0,-1}$ of holes between neighbor acceptors in charge states (0) and (-1) has the form [7]:

$$J_{hp} = eN_{hp}\left[M_h E - D_h \frac{d}{dx}\ln\left(\frac{N_{a,0}}{N_{a,-1}}\right)\right] = \sigma_h E - eD_h \frac{dN_{a,0}}{dx},$$

where $N_{hp} = N_{a,0}N_{a,-1}/N_a = K(1-K)N_a$ — effective concentration of holes hopping between acceptors, M_h — hopping drift mobility of holes, E — external electric field strength directed along x axis in a semiconductor, D_h — coefficient of hopping diffusion of holes; $\sigma_h = eN_{hp}M_h$.

In a crystalline semiconductor of n-type the density of stationary hopping current $J_{hn} = J_{0,+1}$ of electrons between neighbor donors in charge states (0) and $(+1)$ has the form [6]:

$$J_{hn} = eN_{hn}\left[M_h E + D_h \frac{d}{dx}\ln\left(\frac{N_{d,0}}{N_{d,+1}}\right)\right] = \sigma_h E + eD_h \frac{dN_{d,0}}{dx},$$

where $N_{hn} = N_{d,0}N_{d,+1}/N_d$ — effective concentration of electrons hopping between donors, M_h and D_h — hopping

drift mobility and coefficient of hopping diffusion of electrons; $\sigma_h = eN_{hn}M_h$.

Note that expressions for densities of hopping currents of holes via acceptors (J_{hp}) and electrons via donors (J_{hn}) are in general identical to expressions (see, for example, [27,28]) for densities of currents of delocalized holes of v-band and electrons of c-band.

DC hopping electroconductivity σ_h is measured at constant external pressure P, temperature T and constant number of acceptors N_aV and donors KN_aV in a crystalline sample with volume of V. Hopping migration of holes in the acceptor band is characterized by activation energy ε_3. Under these conditions the expression for σ_h similarly to Arrhenius equation (see, for example, [29]) may be presented as

$$\sigma_h = \tilde{\sigma}_{03} \exp[-(h_3 - s_3 T)/k_B T], \quad (4)$$

where $h_3(T) = \varepsilon_3(T) + Pv_3(T)$ — enthalpy averaged over volume V of sample, $\varepsilon_3(T)$ — average energy necessary for implementation of an elementary act of thermally activated tunelling of a hole between two acceptors in charge states (0) and (−1), $v_3(T)$ — average change of the total „volume“ of two acceptors when a hole hops between them, $s_3(T) = s_{3t}(T) + s_{3m}$ — sum of the average thermal entropy $s_{3t}(T)$ of hole hops activation and configurational entropy s_{3m} of holes on acceptors (its value practically does not depend on temperature).

Temperature dependences of quantities $\varepsilon_3(T)$, $v_3(T)$ and $s_3(T)$ are related by an equation (see, for example, [30,31]):

$$T(\partial s_{3t}/\partial T)_{P,NV} = (\partial \varepsilon_3/\partial T)_{P,NV} + P(\partial v_3/\partial T)_{P,NV}, \quad (5)$$

where $NV = (N_a + N_d)V$ — number of impurities in a sample with volume of V.

From (4) with account of (5) we obtain

$$-k_B[\partial \ln(\sigma_h/\tilde{\sigma}_{03})/\partial(1/T)]_{P,NV} = h_3(T) = \varepsilon_3(T) + Pv_3(T), \quad (6)$$

i.e. derivative with respect to $1/T$ from logarithm of ratio $\sigma_h/\tilde{\sigma}_{03}$ is proportional to enthalpy $h_3(T)$ of thermal activation of hopping ε_3-electroconductivity.

With high accuracy the average change of the total „volume“ $v_3(T)$ of two acceptors in charge states (0) and (−1) when the hole hops between them is negligibly low, so from (4) we have

$$\sigma_h = \sigma_{03} \exp[-(\varepsilon_3 - s_{3t}T)/k_B T], \quad (7)$$

where configurational entropy s_{3m} is included in prefactor $\sigma_{03} = \tilde{\sigma}_{03} \exp(s_{3m}/k_B)$.

Following [8,32], let us suggest for simplification of calculations that doping and compensating impurities form a nonstoichiometric simple cubic lattice with translation period d_{im} in a crystal matrix. Let d_{im} be the length of hole hopping between acceptors, and consider two options of the impurity lattice with the translation periods:

$$1)\ d_{im1} = 2R_{im1} = 2[4\pi(1+K)N_a/3]^{-1/3} \approx 1.24[(1+K)N_a]^{-1/3},$$

where R_{im1} — radius of spherical area per one atom of impurity in the lattice;

$$2)\ d_{im2} = B_c^{1/3} R_{im1} = B_c^{1/3}[4\pi(1+K)N_a/3]^{-1/3} \approx 0.867[(1+K)N_a]^{-1/3}$$

— distance between acceptors equal to percolation radius of spherical area per one acceptor with account of donors blocking the hole hops. Here $B_c \approx 2.735$ — dimensionless parameter — average number of „hopping“ bonds per one acceptor [3,33–36]. At percolation radius d_{im2} the charge state (−1) of the acceptor, being activated and „detached“ from the donor ion, may migrate in a hopping manner via acceptors through a crystal. In general quantity d_{im2} reflects effect of self-avoiding walk (by terminology [37]) of holes via acceptors.

For impurities forming in a crystalline semiconductor a nonstoichiometric simple cubic „lattice“ with translation period d_{im}, the temperature T_j of transition from migration of holes by states of v-band to hopping migration of holes between acceptors (similarly to paper [9]) let us determine from the virial theorem at concentration of holes of v-band $p \ll KN_a$ in the following form

$$T_j = \frac{1}{3k_B} \frac{e^2}{4\pi\varepsilon_r\varepsilon_0 R_{ch}}, \quad (8)$$

where R_{ch} — radius of spherical area per ion of impurity in a crystal. According to option 1) the value of $R_{ch1} = [(4\pi/3)2KN_a]^{-1/3} \approx 0.62(2KN_a)^{-1/3}$; option 2) $R_{ch2} = (B_c^{1/3}/2)[(4\pi/3)2KN_a]^{-1/3} \approx 0.434(2KN_a)^{-1/3}$; $2KN_a = N_{a,-1} + N_d$ — concentration of ions of hydrogen-like impurities in a crystal matrix.

At temperatures $T < T_j$ the condition of electric neutrality of crystal has the form

$$N_{a,-1} = N_a \int_{-\infty}^{+\infty} f_{-1} G_a \, d(E_a - I_a) = N_a \langle f_{-1} \rangle = KN_a, \quad (9)$$

where f_{-1} — probability of finding an acceptor in the charge state (−1) with energy level $E_a > 0$ in acceptor band, G_a — density of distribution of energy levels E_a relative to the thermal ionization energy $I_a = e^2/8\pi\varepsilon_r\varepsilon_0 a_p$ of a single acceptor with Bohr radius a_p of the hole orbit.

For concentration of electrically neutral acceptors at $T < T_j$ we have

$$N_{a,0} = N_a \int_{-\infty}^{+\infty} f_0 G_a \, d(E_a - I_a) = N_a \langle f_0 \rangle = (1-K)N_a, \quad (10)$$

where f_0 — probability of finding an arbitrary acceptor with energy level E_a in charge state (0).

Then, following [38], we assume that energy levels of acceptors in the band gap have normal (Gaussian) density of distribution

$$G_a = \frac{1}{\sqrt{2\pi}W_a} \exp\left(\frac{-(E_a - I_a)^2}{2W_a^2}\right), \quad (11)$$

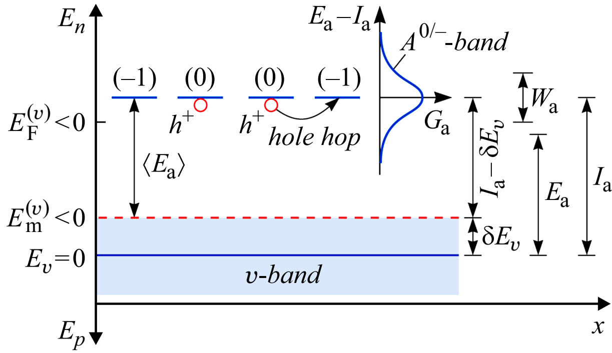

Figure 2. Energy of electron E_n and energy of hole E_p depending on coordinate x in a single-electron band diagram of a semiconductor of p-type: $E_v = 0$ — top of valence band of undoped crystal; „hole hop" — thermally activated hole (h^+) hop between acceptors (0) and (−1) in the center of acceptor $A^{0/-}$ band; $I_a > 0$ — energy of thermal ionization of a single acceptor, $E_F^{(v)} < 0$ — Fermi level, $E_m^{(v)} < 0$ — mobility edge for holes of v-band, $\delta E_v = -E_m^{(v)} > 0$ — shift of the top of v-band deep into the band gap of the semiconductor due to the merging of excited states of impurities, W_a — root mean square fluctuation of acceptor energy levels.

where W_a — effective width of acceptor band (Fig. 2).

Probability that an arbitratory acceptor with energy level $E_a > 0$ above the top of v-band ($E_v = 0$) is ionized

$$f_{-1} = 1 - f_0 = \left[1 + \beta_a \exp\left(\frac{E_a + E_F^{(v)}}{k_B T}\right)\right]^{-1}. \quad (12)$$

Here β_a — degeneracy factor of energy level of a hydrogen-like acceptor, $E_F^{(v)} < 0$ — Fermi level in band gap, counted from the top of v-band of undoped crystal, $\xi - k_B T \ln\beta_a = E_F^{(v)} + I_a$ — Fermi level counted from energy level I_a of a single acceptor.

With account of Coulomb interaction of ionized acceptor (in charge state (−1)) only with ions in the first coordination sphere of a nonstochiometric cubic impurity lattice, the width of acceptor band is [39]

$$W_a = \left(\sum_{i=1}^{6} P_i U_i^2\right)^{1/2} = \frac{e^2}{4\pi\varepsilon_r\varepsilon_0 d_{im}}\left(\frac{12K}{1+K}\right)^{1/2}, \quad (13)$$

where $P_i = 2K/(1+K)$ — probability that near a selected impurity ion any of the six sites of the impurity lattice in the first coordination sphere is occupied by an ionized acceptor or donor; $1/(1+K)$ — fraction of acceptors in the sites of the impurity lattice; $|U_i| = e^2/4\pi\varepsilon_r\varepsilon_0 d_{im}$ — module of Coulomb energy of interaction of the selected ion with one of the nearest ions located at the distance of $d_{im} = 2R_{im}$ in the lattice of doping and compensating impurities. When deriving equation (13) it was taken into account that the crystal-average energy of Coulomb interaction between the selected impurity ion and ions in the nearest six stres of the impurity lattice is equal to zero:

$$\sum_{i=1}^{6} P_i U_i = 0.$$

Average value of thermal ionization energy of an average acceptor in charge state (0), i.e. from the center of the acceptor band (Fig. 2) [40]:

$$\langle E_a \rangle = I_a\left(1 - \frac{a_p}{R_{im}}\right) = I_a - \delta E_v, \quad (14)$$

where I_a — energy level of a single hydrogen-like acceptor, $R_{im} = d_{im}/2$ — radius of spherical area in a semiconductor per one atom of impurity, $\delta E_v = I_a a_p/R_{im} > 0$ — decrease in the thermal ionization energy of acceptor in the charge state (0) due to overlap of excited states of neutral acceptors with increase of their concentration and formation of a quasi-continuous band of the allowed energy values for holes of v-band.

At temperature $T_3 = T_j/3$, when $W_a \gg k_B T_3$ and

$$f_0 f_{-1} \to k_B T_3\, \delta(E_a + k_B T_3 \ln\beta_a + E_F^{(v)}),$$

where $\delta(E_a + k_B T_3 \ln\beta_a + E_F^{(v)})$ — Dirac delta-function, quantity $\xi = k_B T_3 \ln\beta_a + I_a + E_F^{(v)}$ is found from electric neutrality equation (9) in the form

$$2K \approx 1 - \mathrm{erf}(\xi/\sqrt{2}W_a), \quad (15)$$

where $\mathrm{erf}(\cdot)$ — error function.

Hopping heat capacity $C_h = -d\langle E_h \rangle/dT$ per one hole in the acceptor $A^{0/-}$ band is the derivative with respect to absolute temperature T from average energy $\langle E_h \rangle$ of the electrically neutral acceptor and has the following form (see Appendix):

$$C_h = \frac{1}{(1-K)k_B T^2}\left[\int_{-\infty}^{+\infty} E_a^2 G_a f_0 f_{-1}\, d(E_a - I_a) - \frac{\xi_h}{K(1-K)}\left(\int_{-\infty}^{+\infty} E_a G_a f_0 f_{-1}\, d(E_a - I_a)\right)^2\right], \quad (16)$$

where $\xi_h \geq 1$ — dimensionless parameter [38]:

$$\frac{1}{\xi_h} = \frac{k_B T M_h}{e D_h} = \frac{1}{K(1-K)}\int_{-\infty}^{+\infty} G_a f_0 f_{-1}\, d(E_a - I_a). \quad (17)$$

For a narrow acceptor band, when $W_a \ll k_B T_3$ and $G_a \to \delta(E_a - I_a)$, from equation (9) with account of (11), (12) we obtain $\xi = k_B T_3 \ln\beta_a + E_F^{(v)} + I_a \approx \approx -k_B T_3 \ln[K/(1-K)]$. In this case from (17) with account of (11)−(13) it follows that $\xi_h \approx 1$ and $C_h = 0$.

For a wide acceptor band, when $W_a \gg k_B T_3$ and

$$f_0 f_{-1} \to k_B T\, \delta(E_a + k_B T_3 \ln\beta_a + E_F^{(v)}),$$

equation (9) with account of (11), (12) takes the form $2K \approx 1 - \mathrm{erf}(\xi/\sqrt{2}W_a)$. In this case from (17) we have $\xi_h \approx K(1-K)\gamma\sqrt{2\pi}\exp(\xi^2/2W_a^2)$ and $C_h = qT_3$, where $\gamma = W_a/k_BT_3 \gg 1$ and q — a certain constant.

Dimensionless parameter $\xi_h \geq 1$ characterizes the difference in the degree of impact from fluctuations of electrostatic potential energy in a crystal due to its doping on the coefficient of hopping diffusion and on drift hopping mobility of holes in the acceptor band. For a narrow acceptor band ($W_a \ll k_BT_3$) parameter $\xi_h = 1$ and $D_h/M_h = k_BT_3/e$.

Average thermal entropy $s_{3t} = s_{3t}(T)$ of activation of hole hops between acceptors is (see equation (A6) from Appendix)

$$s_{3t} = \int_0^T \frac{C_h(T')}{T'}\,dT', \tag{18}$$

where C_h — hopping heat capacity per one hole in the acceptor band using equation (16).

Configurational entropy of distribution of electrically neutral states of hydrogen-like acceptors with number of $N_{a,0}V = (1-K)N_aV$ per one acceptor in the crystal matrix of a semiconductor with volume of V over all acceptors is (see equation (A9) from Appendix):

$$s_{3m} = -k_B \ln[K^K(1-K)^{1-K}], \tag{19}$$

where $0 < K < 1$.

Note that configurational entropy s_{3m} by equation (19) matches the equation for entropy proposed in paper [12] that describes the number of ways to distribute „free" holes of v-band over acceptors at hole concentration $p \ll K(1-K)N_a$.

2. Equations for prefactor σ_{03} and activation energy ε_3

Hopping conductivity averaged by all possible orientations of a nonstoichiometric impurity lattice relative to direction of external electric field strength $\sigma_h = 1/\rho_h$ with account of thermal entropy $s_{3t} = s_{3t}(T)$ by equation (18) and configurational entropy s_{3m} by equation (19) is given by expression (compare [6–8]):

$$\sigma_h = \frac{e^2K^{1-K}(1-K)^KN_ad_{im}^2\Gamma_h}{12(1+K)\xi_hk_BT}$$
$$= \tilde{\sigma}_{03}\exp\left(-\frac{h_3 - s_3T}{k_BT}\right) = \sigma_{03}\exp\left(-\frac{\varepsilon_3 - s_{3t}T}{k_BT}\right), \tag{20}$$

where $\sigma_h = eN_{hp}M_h$ — hopping electroconductivity in direction of external electric field, $N_{hp} = K(1-K)N_a$ — effective concentration of holes hopping between acceptors, d_{im} — length of hole hopping; here it accounts that $\sigma_{03} = \tilde{\sigma}_{03}\exp(s_{3m}/k_B)$ (see equations (4) and (7)), and also $D_h/M_h = \xi_hk_BT/e$ — ratio of diffusion constant of hopping holes to their drift hopping mobility [38]; $s_3(T) = s_{3m} + s_{3t}(T)$.

We specifically note that equation (20) accounts hops of holes only along the edges of the impurity cubic lattice with its arbitrary orientation in relation to the direction of external electric field strength in a macroscopic semiconductor sample [6,7].

Average frequency of hole hops included in equation (20):

$$\Gamma_h \approx \frac{1}{\tau_3K(1-K)}\int_{-\infty}^{+\infty} G_af_0f_{-1}\,d(E_a - I_a)$$
$$= \frac{1}{\tau_3\xi_h} \equiv \Gamma_3\exp\left(-\frac{\varepsilon_3 - s_{3t}T}{k_BT}\right), \tag{21}$$

where $\Gamma_3 = 1/\tau_F \equiv 1/\tau_3$ — frequency of hole tunneling between acceptors in charge states (0) and (−1). Equation (21) includes Fermi level $E_F^{(v)}$. To find it, the condition of electrical neutrality (9) may be resolved relative to the quantity $\xi - k_BT\ln\beta_a = E_F^{(v)} + I_a$. Then we obtain the value of Fermi level $E_F^{(v)} = \xi - k_BT\ln\beta_a - I_a < 0$ relative to the top of v-band in an undoped crystalline semiconductor of p-type (Fig. 2).

Within the framework of the theory of a molecular hydrogen ion H_2^+ the time of hole tunneling between two acceptors (indices 1 and 2), located at the distance d_{im}, given the difference between their energy levels $\Delta_{a12} = E_{a2} - E_{a1} = \sqrt{3}\,\delta E_{at}$, can be estimated according to [41], as follows:

$$\tau_3 = \tau_F = \frac{\pi\hbar}{\delta E_{at}}\sqrt{1 + \left(\frac{\Delta_{a12}}{\delta E_{at}}\right)^2} = \frac{2\pi\hbar}{\delta E_{at}}, \tag{22}$$

where $2\pi\hbar = h$ — Planck's constant, $\delta E_{at}(E_F^{(v)})$ — broadening (splitting) of energy levels $E_{at} = E_m^{(v)} - E_F^{(v)}$ of two acceptors due to hole tunneling between them:

$$\delta E_{at} = 4E_{at}\frac{A - BS}{1 - S^2},$$
$$A = (1+\rho)\exp(-\rho); \quad B = [1 - (1+\rho)\exp(-2\rho)]/\rho,$$
$$\rho = d_{im}/a_t, \quad S = (1 + \rho + \rho^2/3)\exp(-\rho). \tag{23}$$

Here for option 1) $d_{im1} \approx 1.24[(1+K)N_a]^{-1/3}$ and for option 2) $d_{im2} \approx 0.867[(1+K)N_a]^{-1/3}$; $a_t = e^2/8\pi\varepsilon_r\varepsilon_0E_{at}$.

For a prefactor (pre-exponential factor) in temperature dependence (7) from expression (20) with account of (21) we obtain

$$\sigma_{03} = \frac{\tilde{\sigma}_{03}}{K^K(1-K)^{1-K}} = \frac{e^2K^{1-K}(1-K)^KN_ad_{im}^2\Gamma_3}{12(1+K)\xi_hk_BT}, \tag{24}$$

where the time of hole tunneling $1/\Gamma_3 = \tau_3 = \tau_F$ is determined by (22), coefficient $\xi_h \geq 1$ is given by equation (17).

Integral activation energy ε_3 for hopping electroconductivity is obtained from (21) with account of (20) in the form

$$\varepsilon_3 = -k_BT\ln(\Gamma_h/\Gamma_3) + s_{3t}T = k_BT\ln\xi_h + s_{3t}T > 0. \tag{25}$$

From (25) with account of (17) it follows that $\varepsilon_3 \to 0$ in the limit of zero temperature ($T \to 0$), which is, however, evident from physical assumptions [42].

Note that from (25) an expression for differential thermal activation energy ε_3' of hopping electroconductivity is obtained in the form

$$\varepsilon_3' = -k_B \frac{d\ln(\sigma_h/\sigma_{03})}{d(1/T)} = \frac{d(\varepsilon_3/T)}{d(1/T)},$$

so $\varepsilon_3' = \varepsilon_3$, if ε_3 does not depend on temperature in the vicinity of $T_3 \approx T_j/3$ (Fig. 1).

Further let us assume that quantity $2\Delta_{at}/\sqrt{3} = \Delta_{a12}/2$ is approximately equal to splitting of energy levels in two acceptors δE_{at} by (23) at resonance, i.e. $2\Delta_{at}/\sqrt{3} = \delta E_{at}$. Then the frequency of hole hops between quasi-steady-states of acceptor energy levels with account of (21) has the form

$$\Gamma_h(\Delta_{at}) \approx \frac{\Gamma_3}{K(1-K)} \int\limits_{-\infty}^{+\infty} G_a f_0(E_a + \Delta_{at}) f_{-1}(E_a - \Delta_{at}) \times d(E_a - I_a) = \Gamma_3 \exp\left(-\frac{\varepsilon_3(\Delta_{at}) - s_{3t}T}{k_B T}\right), \quad (26)$$

where

$$f_0(E_a + \Delta_{at}) = \{1 + \beta_a^{-1} \exp[-(E_F^{(v)} + E_a + \Delta_{at})/k_B T]\}^{-1}$$

— probability of filling the acceptor with a hole at energy level $E_a + \Delta_{at}$;

$$f_{-1}(E_a - \Delta_{at}) = \{1 + \beta_a \exp[(E_F^{(v)} + E_a - \Delta_{at})/k_B T]\}^{-1}$$

— probability that the acceptor with energy level $E_a - \Delta_{at}$ is ionized; at $\Delta_{at} \to 0$ we have: $f_{-1} = 1 - f_0$ for all values of E_a. From equation (26) the activation energy follows

$$\varepsilon_3(\Delta_{at}) = -k_B T \ln[\Gamma_h(\Delta_{at})/\Gamma_3] + s_{3t}T. \quad (27)$$

Equation (27) for a lightly doped crystal ($2\Delta_{at}/\sqrt{3} = \delta E_{at} \ll k_B T_3$) changes into equation (25). Note that in heavily doped moderately compensated semiconductors near the Mott concentration phase transition the holes migrate in the energy range of the acceptor band with width of $2\Delta_{at} = \delta E_{at}$ in the vicinity of Fermi level $E_F^{(v)}$ between (quasi)resonant pairs of acceptors [43]. Then, similarly to Drude–Lorentz approach (see, for example, [27,28]), the tunneling electric conductivity σ_{tun} by acceptors in the limit of the wide acceptor band W_a and low temperature, i.e. when $W_a \gg k_B T$, has the form [43]:

$$\sigma_{tun} = \frac{e^2 K(1-K) N_a \tau_{tun}}{2m_{p\sigma}} \Xi_a \Theta_{tun} = \frac{1}{\rho_{tun}}, \quad (28)$$

where $\tau_{tun} = \pi\hbar/\delta E_{at}$ — time of hole tunneling between two acceptors in charge states (0) and (−1), located at the distance d_{im} when their energy levels coincide ($E_{a1} = E_{a2}$) (see equation (23)); $\Xi_a = 1/(1+K)$ — fraction of acceptors in the impurity lattice sites, $m_{p\sigma}$ — effective mass of hole electroconductivity in v-band; Θ_{tun} — fraction of pairs of acceptors in charge states (0) and (−1), whose energy levels E_a are $\pm\Delta_{at} = \pm 0.5\,\delta E_{at}$ away from Fermi level ($-E_F^{(v)} > 0$) i.e. $(-E_F^{(v)} - \Delta_{at} - I_a) \leq E_a - I_a \leq (-E_F^{(v)} + \Delta_{at} - I_a)$, is given by the relation:

$$\Theta_{tun} = \frac{1}{2K(1-K)} \left[\operatorname{erf}\left(\frac{E_F^{(v)} + \Delta_{at} + I_a}{\sqrt{2}W_a}\right) - \operatorname{erf}\left(\frac{E_F^{(v)} - \Delta_{at} + I_a}{\sqrt{2}W_a}\right)\right] < 1. \quad (29)$$

Expression (29) takes into account the fact that some $(1-K)$ acceptors are occupied by holes, and some K are free.

3. Comparison of calculations with experimental data on *p*-Ge:Ga

From experimental works on *p*-Ge:Ga the data was selected for samples with moderate compensation ratios $0.15 < K < 0.85$ and gallium concentrations $N_a < N_M$, where $N_M \approx 1.85 \cdot 10^{17}$ cm^{-3} — concentration corresponding to Mott transition at $K \approx 0.35$ (see, for example, [43,44]).

When calculating the energy of thermal activation ε_3 and prefactor ρ_{03} in crystals *p*-Ge:Ga the following values of parameters were used: relative dielectric permittivity $\varepsilon_r = 15.4$ [45]; gallium atom energy level degeneracy factor $\beta_a = 4$ [46]; energy of thermal ionization of a single gallium atom $I_a = 11.32$ meV [47,48]; effective mass of electroconductivity of hole of v-band $m_{p\sigma} = 0.26m_0$ [43,49]; m_0 — mass of electron in vacuum; $a_p = e^2/8\pi\varepsilon_r\varepsilon_0 I_a = 4.13$ nm.

Note that experimentally observed [50] temperature values T_{je} for crystals *p*-Ge:Ga with gallium compensation ratio K=0.3 are: T_{je}=4.03 K for concentration N_a=3.57 · 10^{14} cm^{-3}; T_{je}=5.02 K for N_a=7.59 · 10^{14} cm^{-3}; T_{je} = 8.03 K for N_a = 3.10 · 10^{15} cm^{-3}; T_{je} = 11.20 K for N_a = 1.49 · 10^{16} cm^{-3}. Calculations of temperatures T_{j1} and T_{j2} using equation (8) for options 1) and 2) provide values: T_{j1} = 3.49 K and T_{j2} = 4.99 K for N_a = 3.57 · 10^{14} cm^{-3}; T_{j1} = 4.49 K and T_{j2} = 6.41 K for N_a = 7.59 · 10^{14} cm^{-3}; T_{j1} = 7.17 K and T_{j2} = 10.25 K for N_a = 3.10 · 10^{15} cm^{-3}; T_{j1} = 12.10 K and T_{j2} = 17.30 K for N_a = 1.49 · 10^{16} cm^{-3}. So, the calculations T_j using equation (8) for two options of the impurity lattice periods generally agree with the experimental data [50].

Fig. 3 presents the dependence of the thermal activation energy ε_3 of hopping electroconductivity in crystals *p*-Ge:Ga on concentration of gallium at $K = 0.35$. The calculation was carried out using equation (27) at temperature $T_j/3$ for distances between impurities $d_{im1} \approx 1.24[(1+K)N_a]^{-1/3}$ (curve *1*);

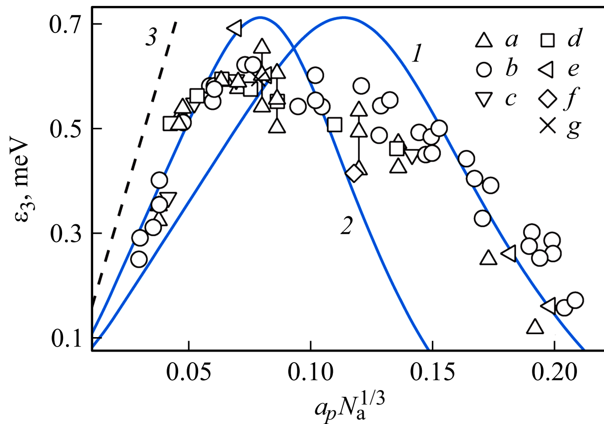

Figure 3. Dependence of activation energy ε_3 of hopping conductivity in p-Ge:Ga on concentration of acceptors N_a in units of $a_p N_\mathrm{a}^{1/3}$, where a_p — Bohr radius of the hole on the gallium atom. Solid lines — calculation using equation (27) for $K = 0.35$ at temperature $T_\mathrm{j}/3$ for $d_\mathrm{im1} \approx 1.24[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *1*) and $d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2*); dashed line *3* — calculation using model [3]; points — experiment: *a* — [14,15], *b* — [16–18], *c* — [19], *d* — [20], *e* — [21], *f* — [22], *g* — [23].

$d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2*). Dashed line *3* in Fig. 3 shows calculation $\varepsilon_3 \approx 0.7e^2 N_\mathrm{a}^{1/3}/4\pi\varepsilon_\mathrm{r}\varepsilon_0$ using model [3] for $K = 0.35$. It is seen that calculation by (27) generally agrees with the experimental data [14–23], and approximation d_im2 better describes the ascending part of experimental dependence in Fig. 3, while approximation d_im1 — the descending one. While the model [3] gives overestimated values of activation energy ε_3 of hopping conductivity and is applicable only for the ascending section of experimental dependence at low doping levels.

Fig. 4 shows with solid lines the calculated dependences of the pre-exponential factor (prefactor) $\rho_{03} = 1/\sigma_{03}$ by (24) for hopping resistivity $\rho_\mathrm{h} = 1/\sigma_\mathrm{h}$ on concentration N_a of gallium atoms in p-Ge:Ga. The calculation was carried out at compensation ratio $K = 0.35$ and temperature $T_\mathrm{j}/3$ for distances between all impurities $d_\mathrm{im1} \approx 1.24[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *1*) and $d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2*). Dashed lines in this figure show dependences of tunneling resistance ρ_tun using equation (28) of model [43] for distances between impurities $d_\mathrm{im1} \approx 1.24[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *1′*) and $d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2′*). It is seen that when Mott transition is approached, the experimental values of resistivity reach calculations using model [43].

Note that in calculations of tunneling resistivity ρ_tun the splitting of δE_at energy levels by equation (23) includes Bohr radius $a_\mathrm{t} = e^2/8\pi\varepsilon_\mathrm{r}\varepsilon_0 E_\mathrm{at}$. Radius a_t corresponds to energy level $E_\mathrm{at} = E_\mathrm{m}^{(v)} - E_\mathrm{F}^{(v)} > 0$ of hole tunneling between two acceptors, where $E_\mathrm{m}^{(v)} = -\delta E_v < 0$ (see equation (14)). Quantity a_t is somewhat different from the Bohr radius $a_p = e^2/8\pi\varepsilon_\mathrm{r}\varepsilon_0 I_\mathrm{a}$ of hole orbit in a single acceptor, used in calculations ρ_tun in article [43] for compensated

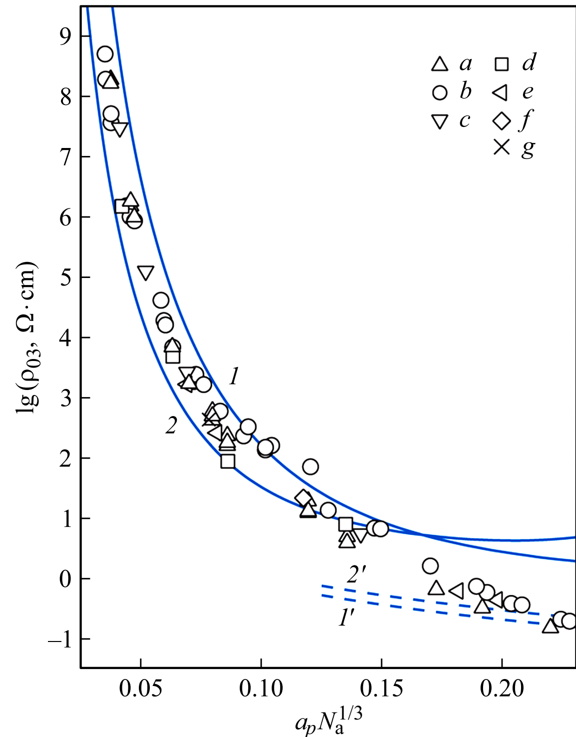

Figure 4. Dependence of resistivity $\rho_{03} = 1/\sigma_{03}$ of hopping conductivity in p-Ge:Ga on concentration of acceptors N_a in units of $a_p N_\mathrm{a}^{1/3}$, where a_p — Bohr radius of the hole on the gallium atom. Solid lines — calculation using equation (24) for $K = 0.35$ at temperature $T_\mathrm{j}/3$ for $d_\mathrm{im1} \approx 1.24[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *1*) and $d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2*); dashed lines — calculation using equation (28) of model [43] for $d_\mathrm{im1} \approx 1.24[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *1′*) and $d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2′*); points — experiment: *a* — [14,15], *b* — [16–18], *c* — [19], *d* — [20], *e* — [21], *f* — [22], *g* — [23].

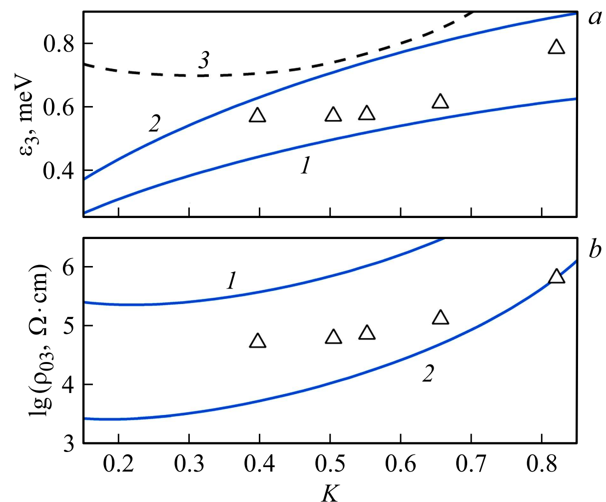

Figure 5. Dependences of activation energy ε_3 (*a*) and resistivity ρ_{03} (*b*) of hopping conductivity in p-Ge:Ga on compensation ratio K. Solid lines — calculation of ε_3 using equation (27) and ρ_{03} using equation (24) for $N_\mathrm{a} = 2.66 \cdot 10^{15}\,\mathrm{cm}^{-3}$ at temperature $T_\mathrm{j}/3$ for $d_\mathrm{im1} \approx 1.24[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *1*) and $d_\mathrm{im2} \approx 0.867[(1+K)N_\mathrm{a}]^{-1/3}$ (curve *2*); dashed line *3* — calculation using model [3]; points — experiment [15].

heavily doped semiconductors of p-type. However, the calculated values of ρ_{tun} at orbit radius a_t for moderately compensated crystals p-Ge:Ga near Mott transition practically do not differ from calculations ρ_{tun} at radius a_p.

Fig. 5 shows dependences of energy activation ε_3 (a) and pre-exponential factor $\rho_{03} = 1/\sigma_{03}$ (b) of hopping conductivity in germanium on compensation ratio K of gallium atoms with concentration $N_a = 2.66 \cdot 10^{15}\,\mathrm{cm}^{-3}$. Calculations of ε_3 using equation (27) and ρ_{03} using equation (24) at temperature $T_j/3$ for distances between impurities $d_{im1} \approx 1.24[(1+K)N_a]^{-1/3}$ (curve *1*) and $d_{im2} \approx 0.867[(1+K)N_a]^{-1/3}$ (curve *2*) are shown. Dashed line *3* in Fig. 5, *a* shows calculation ε_3 using model [3] for $N_a = 2.66 \cdot 10^{15}\,\mathrm{cm}^{-3}$. It is seen that calculation of ε_3 using (27) generally agrees with experimental data [15], while using model [3] gives overestimated values.

Conclusion

Analytical calculation of hopping electric conductivity parameters was conducted in crystalline semiconductors in NNH mode based on the example of p-type material. The principal difference of the proposed method for calculation of hopping conductivity parameters is the method to find the position of the drift mobility edge for holes of v-band. This mobility edge is caused by formation of a quasi-continuous band of allowed energy values from the excited states of electrically neutral acceptors for holes of v-band, which decreases the value of thermal energy of majority impurities ionization.

It was suggested, for simplicity, that in a crystal matrix the doping and compensating impurities form a single nonstoichiometric simple cubic lattice. Two variants of a cubic lattice are considered with different translation periods. It was also assumed that the width of the acceptor band is determined by Coulomb interaction of impurity ions in the first coordination sphere of the nonstoichiometric impurity lattice.

To determine values of σ_{03} and ε_3, first temperature T_3 was found, at which thermally activated tunnel hops of holes are observed between the neighbor acceptors in charge states (0) and (-1). It was taken into account that with the increase in the doping level in „molecular" pairs of acceptors in charge states (0) and (-1), energy levels of one pair split by value of δE_{at}. It was assumed that migration of holes by acceptor states occur in the energy band with width of $\sqrt{3}\,\delta E_{at}$ near Fermi level in the impurity band. For the first time the calculations of hopping ε_3-electroconductivity took into account the impact of thermal and configurational entropy on parameters σ_{03} and ε_3.

Quantitative description was obtained for the behavior of prefactor σ_{03} and energy of thermal activation ε_3 of hopping electroconductivity with change of the doping level and degree of compensation of a semiconductor on the insulator side of Mott transition.

Numerical calculation of σ_{03} and ε_3 values using proposed equations (24) and (27) was carried out for crystals p-Ge:Ga. Results of the calculations for σ_{03} and ε_3 values agree with the known experimental data for moderately compensated and well-characterized crystals p-Ge:Ga, obtained in process of neutron transmulation doping.

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Conflict of interest

The authors declare that they have no conflict of interest.

Appendix

Hopping heat capacity of holes in acceptor band and thermal entropy

To calculate hopping heat capacity C_h using [39] we use relation

$$\frac{d\langle f_0\rangle}{dT} = \frac{\partial\langle f_0\rangle}{\partial T} + \frac{\partial\langle f_0\rangle}{\partial E_F^{(v)}}\frac{dE_F^{(v)}}{dT} = 0, \quad \text{(A1)}$$

where $\langle f_0\rangle = 1 - \langle f_{-1}\rangle = 1 - K$ — probability averaged over crystal volume that an acceptor randomly selected in the crystal matrix is in the charge state (0), it is given by equations (10)–(12); T — absolute temperature, $E_F^{(v)} < 0$ — Fermi level counted from the top of v-band.

Corresponding partial derivatives:

$$\frac{\partial f_0}{\partial T} = \frac{\partial}{\partial T}\left[1 + \beta_a^{-1}\exp\left(-\frac{E_a + E_F^{(v)}}{k_B T}\right)\right]^{-1} = -\frac{E_a + E_F^{(v)}}{k_B T^2} f_0 f_{-1},$$

$$\frac{\partial f_0}{\partial E_F^{(v)}} = \frac{\partial}{\partial E_F^{(v)}}\left[1+\beta_a^{-1}\exp\left(-\frac{E_a+E_F^{(v)}}{k_B T}\right)\right]^{-1} = \frac{f_0 f_{-1}}{k_B T}. \quad \text{(A2)}$$

From (A1) with account of (A2) we find

$$\frac{dE_F^{(v)}}{dT} = \frac{\xi_h}{K(1-K)T}\int_{-\infty}^{+\infty} E_a G_a f_0 f_{-1}\, d(E_a - I_a) + \frac{E_F^{(v)}}{T}, \quad \text{(A3)}$$

where quantity ξ_h is specified by equation (17).

Temperature dependence of average energy of an electrically neutral acceptor:

$$\langle E_h\rangle = \frac{1}{1-K}\int_{-\infty}^{+\infty} E_a G_a f_0\, d(E_a - I_a),$$

determines heat capacity (per one hole in acceptor $A^{0/-}$ band; see Fig. 2):

$$C_\mathrm{h} = -\frac{d\langle E_\mathrm{h}\rangle}{dT} = \frac{-1}{1-K}\left[\int_{-\infty}^{+\infty} E_\mathrm{a} G_\mathrm{a} \frac{\partial f_0}{\partial T}\, d(E_\mathrm{a} - I_\mathrm{a}) + \frac{dE_\mathrm{F}^{(v)}}{dT}\int_{-\infty}^{+\infty} E_\mathrm{a} G_\mathrm{a} \frac{\partial f_0}{\partial E_\mathrm{F}^{(v)}}\, d(E_\mathrm{a} - I_\mathrm{a})\right] > 0. \tag{A4}$$

Then, using ratios (A2) and (A3), from (A4) we obtain equation (16):

$$C_\mathrm{h} = \frac{1}{(1-K)k_\mathrm{B}T^2}\left[Q_1 - \frac{\xi_\mathrm{h} Q_2}{K(1-K)}\right], \tag{A5}$$

where

$$Q_1 = \int_{-\infty}^{+\infty} E_\mathrm{a}^2 G_\mathrm{a} f_0 f_{-1}\, d(E_\mathrm{a} - I_\mathrm{a}),$$

$$Q_2 = \left(\int_{-\infty}^{+\infty} E_\mathrm{a} G_\mathrm{a} f_0 f_{-1}\, d(E_\mathrm{a} - I_\mathrm{a})\right)^2.$$

Average thermal entropy $s_{3\mathrm{t}} = s_{3\mathrm{t}}(T)$ of activation of hole hops between two acceptors in charge states (0) and (-1) with account of (A5) is (see also [39]):

$$s_{3\mathrm{t}} = \int_0^T \frac{C_\mathrm{h}(T')}{T'}\, dT'. \tag{A6}$$

Configurational entropy of hole placement by acceptors

Let us consider a crystalline semiconductor of p-type of unit volume, containing $N_\mathrm{a} = N_{\mathrm{a},0} + N_{\mathrm{a},-1}$ hydrogen-like acceptors in charge states (0) and (-1) and $N_\mathrm{d} = KN_\mathrm{a}$ hydrogen-like donors in charge states $(+1)$, where $0 < K < 1$ — compensation ratio. The condition of electric neutrality of the crystalline sample has the form $N_{\mathrm{a},-1} = N_\mathrm{d} = KN_\mathrm{a}$. Calculation of the configurational entropy $S_{3\mathrm{m}}$ for distribution of electrically neutral states of acceptors $N_{\mathrm{a},0} = (1-K)N_\mathrm{a}$ over all acceptors provides (in units of Boltzmann constant k_B):

$$\frac{S_{3\mathrm{m}}}{k_\mathrm{B}} = \ln\left(\frac{N_\mathrm{a}!}{N_{\mathrm{a},0}!(N_\mathrm{a} - N_{\mathrm{a},0})!}\right). \tag{A7}$$

To calculate quantity $S_{3\mathrm{m}}$, we use Stirling formula $\ln(X!) = X\ln(X) - X$, where $X \gg 1$ and then from (A7) we obtain (compare [10–12]):

$$\frac{S_{3\mathrm{m}}}{k_\mathrm{B}} = N_\mathrm{a}\left[\ln\left(\frac{N_\mathrm{a}}{N_\mathrm{a} - N_{\mathrm{a},0}}\right) + \frac{N_{\mathrm{a},0}}{N_\mathrm{a}}\ln\left(\frac{N_\mathrm{a} - N_{\mathrm{a},0}}{N_{\mathrm{a},0}}\right)\right]. \tag{A8}$$

Note that $N_{\mathrm{a},0} = (1-K)N_\mathrm{a}$, and then from (A8) we get equation (19) for configurational entropy $s_{3\mathrm{m}} = S_{3\mathrm{m}}/N_\mathrm{a}$, per one acceptor in the crystal:

$$\frac{s_{3\mathrm{m}}}{k_\mathrm{B}} = -\ln[K^K(1-K)^{1-K}]. \tag{A9}$$

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