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Capture coefficients of free excitons by shallow acceptors and donors in gallium arsenide

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Abstract. An analysis of the 4.2 K exciton luminescence spectra of semi-insulating GaAs crystals with different concentrations of shallow acceptors (C) and donors (Si) is given. As a result, the 4.2 K capture coefficients of free excitons by shallow neutral acceptors [b_{A^0X} = $= (4 \pm 2)\cdot 10^{-8}$ cm^3/s] and donors [$b_{D^0X} = (1.5 \pm 0.8)\cdot 10^{-7}$ cm^3/s] are found and also an estimate of the capture coefficient of free excitons by ionized shallow donors was made [$b_{D^+X} >> b_{D^0X}$].

Keywords: gallium arsenide, shallow acceptors and donors, exciton luminescence, capture coefficients of free excitons.

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1. Introduction

As is known, in semiconductors effective processes of binding free excitons (their concentration n_X) by shallow neutral acceptors A^0, shallow ionized D^+ and neutral D^0 donors (further neutral acceptors, ionized and neutral donors, their concentrations accordingly N_{A^0}, N_{D^+} and N_{D^0}) take place [these processes lead to formation of bound exciton-neutral acceptor (A^0X), bound exciton-ionized donor (D^+X) and bound exciton – neutral donor (D^0X) complexes]. The important characteristics of the rate of above-pointed binding processes are capture coefficients of free excitons by neutral acceptors b_{A^0X}, ionized b_{D^+X} and neutral b_{D^0X} donors. But in the literature practically no data about b_{A^0X}, b_{D^+X} and b_{D^0X} values in semiconductors exist. Really, only in [1] a theoretical examination of free exciton binding by neutral impurities in semiconductors was carried out (unfortunately, the expression for b_{A^0X} is not given in the cited paper), and in [2,3] an attempt to evaluate the free exciton capture coefficient by shallow neutral and ionized traps in CdS was made (b_{A^0X} $= 3\cdot 10^{-10}$ cm^3/s or $3\cdot 10^{-7}$ cm^3/s according to [2], and b_{D^+X} $= 6\cdot 10^{-8}$ cm^3/s, b_{D^0X} = $= 6\cdot 10^{-9}$ cm^3/s according to [3]). In the present work, as a result of the analysis of the excitonic luminescence spectra of semi-insulating GaAs crystals at $T = 4.2$ K (at low temperatures they are photoconductors [4]) with different concentrations of shallow acceptors N_A and donors N_D, we will give the 4.2 K b_{A^0X} and b_{D^0X} values and evaluate the 4.2 K b_{D^+X} value for gallium arsenide.

2. Method for obtaining b_{A^0X}, b_{D^+X} and b_{D^0X} values

A method to obtain the b_{A^0X}, b_{D^+X} and b_{D^0X} values follows from an analysis of intensities of excitonic emission bands in photoconductors [their conductivity is determined by photoelectrons (concentration δn) and photoholes (concentration δp), in them at $T = 4.2$ K $N_{A^0} = N_{D^0} = 0$, $N_{D^+} = N_D$ in darkness and $N_{A^0} \approx N_A$, $N_{D^0} \approx N_D >> N_{D^+}$ when illuminated (the given N_{A^0}, N_{D^+} and N_{D^0} values in illuminated photoconductors are correct for isolated acceptors and donors [4], they predominantly bind free excitons in semi-insulating gallium

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arsenide [5])]. Evidently, in photoconductors luminescence intensities, induced by annihilation of exciton-impurity complexes $A^0X(I_{A^0X})$, $D^+X(I_{D^+X})$ and $D^0X(I_{D^0X})$ and also by recombination of free excitons $X(I_X)$, are given by the following relations {when writing Eqs (1)–(4) we assumed that, firstly, the concentrations of acceptors and donors that bound excitons is small, and secondly, annihilation of free and bound excitons proceeds mainly with the photon emission} [4]:

$$I_{A^0X} = b_{A^0X} N_{A^0} n_X \approx b_{A^0X} N_A n_X , \quad (1)$$

$$I_{D^+X} = b_{D^+X} N_{D^+} n_X , \quad (2)$$

$$I_{D^0X} = b_{D^0X} N_{D^0} n_X \approx b_{D^0X} N_D n_X , \quad (3)$$

$$I_X = \alpha_X n_X , \quad (4)$$

where α_X is the probability of the free exciton radiative annihilation {the 4.2 K α_X value is $\alpha_X = 2.5\cdot10^8$ s^{-1}, as follows from the 4.2 K decay time of the free exciton-induced luminescence $\tau_X = 1/\alpha_X \approx 4$ ns [6]; note here that if at low temperatures free excitons have the Maxwellian distribution, then theoretically $\alpha_X = 3\cdot10^9$ s^{-1} [7]; a difference in experimental and theoretical α_X values testifies that the distribution of free excitons at low temperatures is not the Maxwellian one}. So, as one could see from Eqs (1)–(4), the 4.2 K b_{A^0X}, b_{D^+X} and b_{D^0X} values could be found from the following obvious relations:

$$b_{A^0X} = \frac{I_{A^0X}}{I_X}\frac{\alpha_X}{N_A}, \quad b_{D^+X} = \frac{I_{D^+X}}{I_X}\frac{\alpha_X}{N_{D^+}},$$

$$b_{D^0X} = \frac{I_{D^0X}}{I_X}\frac{\alpha_X}{N_D}, \quad (5)$$

if we use the available experimental I_{A^0X}/I_X, I_{D^+X}/I_X, I_{D^0X}/I_X, N_A, N_{D^+} and N_D values and the known α_X value (see Appendix).

3. Experimental

In our experiments, semi-insulating GaAs crystals were used (the specific resistivity $\rho \approx 4\cdot10^7$ Ohm·cm at 300 K and $\rho \to \infty$ at $T = 4.2$ K). Their dark 300 K conductivity was determined by ionization of deep native donor defects EL2 (concentration $N_{EL2} \approx 2\cdot10^{16}$ cm^{-3}) partly compensated by shallow acceptors (carbon atoms, concentration N_A) and donors (silicon atoms, concentration N_D), i.e. $N_{EL2} > N_A - N_D > 0$ [8] {the concentration of $EL2$ defects and equilibrium room-temperature concentration of ionized $EL2$ defects $EL2^+$ $N^+_{EL2} = N_A - N_D > 0$ were found (with the accuracy ± 10 %) from 300 K infrared absorption spectrum associated with the deep donor $EL2$ [8–10]}. When excited (at $T = 4.2$ K) their conductivity was determined by excess electrons and holes [5,8]. The 4.2 K luminescence spectra (excitation intensity L) of semi-insulating GaAs crystals with different concentrations of carbon and silicon were studied {the carbon concentration (with the accuracy ± 20 %) was found from the usual 77 K infrared absorption measurements at $\lambda = 17.2$ µm (wavenumber 580 cm^{-1}); the silicon concentration (with the accuracy ± 30 %) was estimated from the mass – spectroscopy analysis and the known equilibrium concentration of positively charged $EL2$ defects N^+_{EL2} and carbon content: obviously $N_D = N_A - N^+_{EL2}$ at $T = 300$ K as $N_{D^+} \approx N_D$ and $N_{A^0} = 0$ at room temperature [4,8]}. In the 4.2 K GaAs luminescence spectrum (after its decomposition by individual components with the accuracy ± 20 %), emission bands induced by annihilation of bound excitons A^0X (peak position $h\nu_m = 1.512$ eV), D^+X ($h\nu_m = 1.5133$ eV) and D^0X ($h\nu_m = 1.5141$ eV) and also by recombination of free excitons X ($h\nu_m = 1.5153$ eV) were clearly seen (Fig. 1) [4]. The emission intensities I_{A^0X}, I_{D^+X}, I_{D^0X} and I_X as functions of carbon and silicon concentration were found. At low excitation powers, the used intensities I_{A^0X}, I_{D^+X}, I_{D^0X} and I_X changed quadratically with L (I_{A^0X}, I_{D^+X}, I_{D^0X}, $I_X \sim L^2$, see [4,5]); the same I_{A^0X}, I_{D^+X}, I_{D^0X}, I_X vs. L dependencies testify that in crystals studied N_{A^0}, N_{D^+}, $N_{D^0} \neq \varphi(L)$ [4]. As expected (see [4]) normalized bound exciton A^0X induced emission intensity I_{A^0X}/I_X changed linearly with N_A and normalized bound exciton D^0X induced emission intensity I_{D^0X}/I_X changed linearly with N_D i.e. $I_{A^0X}/I_X \sim N_A$ and $I_{D^0X}/I_X \sim N_D$ (see Fig. 2).

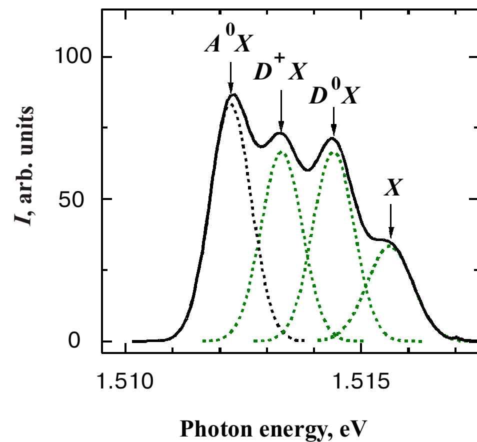

Fig. 1. The 4.2 K bound (A^0X, D^+X, D^0X) and free (X) exciton-induced emission bands in the luminescence spectrum of semi-insulating GaAs crystals.

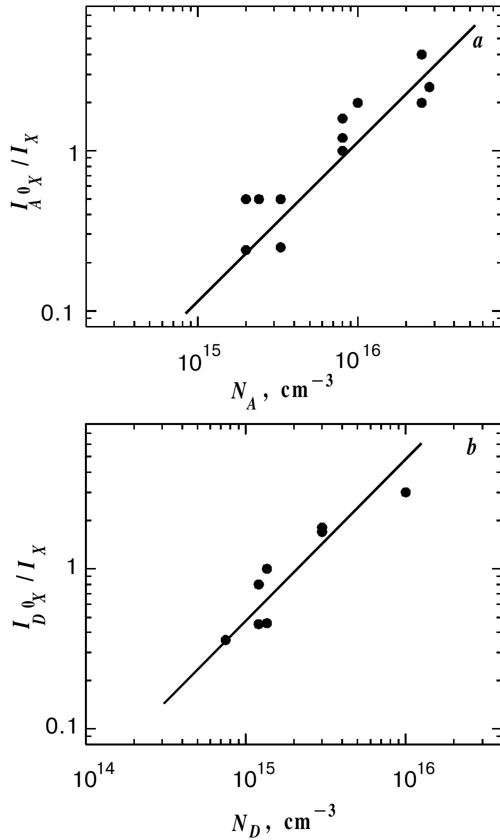

Fig. 2. I_{A^0X}/I_X vs. N_A (*a*) and I_{D^0X}/I_X vs. N_D (*b*) variations in semi-insulating GaAs at T = 4.2 K. The straight lines – theoretical dependences [they follow from Eqs (1), (3) and (4)] $I_{A^0X}/I_X = (b_{A^0X}/\alpha_X)N_A$ (*a*) and $I_{D^0X}/I_X = (b_{D^0X}/\alpha_X)N_D$ (*b*), where $b_{A^0X} = 4\cdot10^{-8}$ cm^3/s, $b_{D^0X} = 1.5\cdot10^{-7}$ cm^3/s and $\alpha_X = 3\cdot10^8$ s^{-1}. A deflection of experimental points from the theoretical straight lines shows the accuracy of b_{A^0X} and b_{D^0X} determination.

4. Results and discussion

The following 4.2 K values for the capture coefficients of free excitons by neutral acceptors and donors were found [see Eq. (5) and also Fig. 2]: $b_{A^0X} = (4\pm2)\cdot10^{-8}$ cm^3/s and $b_{D^0X} = (1.5\pm0.8)\cdot10^{-7}$ cm^3/s [rather small accuracy of b_{A^0X} and b_{D^0X} measurements are connected with a fairly large inaccuracies in the N_A and N_D determination and in the decomposition of GaAs luminescence spectrum on individual A^0X, D^0X and X components (i.e. when determining of I_{A^0X}, I_{D^0X} and I_X values)]. As the 4.2 K N_{D^+} value is unknown (obviously, $N_{D^+} << N_D$), we could only evaluate the b_{D^+X} value – $b_{D^+X} >> b_{D^0X}$ (this follows from the usually observed in an experiment relation $I_{D^+X}/I_{D^0X} = b_{D^+X}N_{D^+}/b_{D^0X}N_{D^0} \le 1$). So, in gallium arsenide, the following relation between b_{A^0X}, b_{D^+X} and b_{D^0X} exists: $b_{A^0X} < b_{D^0X} << b_{D^+X}$. The found correlation between b_{A^0X}, b_{D^+X} and b_{D^0X} values could be explained as follows. A formation of bound excitons A^0X, D^+X and D^0X occurs due to attraction of free excitons to acceptors and donors as a result of dipole-dipole (this refers to X and A^0, X and D^0) or dipole-charge (this refers to X-D^+) interaction (obviously, dipoles are induced by the mutual polarization of excitons and neutral acceptors, donors). Undoubtedly, a dipole momentum of neutral acceptors is less than that of neutral donors due to a rather small Böhr radius of the acceptor-bound hole compared to that of donor-bound electron and a dipole momentum of ionized donors exceeds that of neutral donors. As a result, one expects that the relations $b_{A^0X} < b_{D^0X} << b_{D^+X}$ will be observed in semiconductors.

The obtained 4.2 K b_{A^0X}, b_{D^+X} and b_{D^0X} values in GaAs appear to be rather large, so they illustrate a relatively high efficiency of free exciton trapping by shallow acceptors and donors in gallium arsenide.

5. Conclusions

The 4.2 K capture coefficients of free excitons by neutral acceptors and ionized and neutral donors for gallium arsenide were found and analyzed. The obtained data are important for understanding of regularities of the bound exciton induced luminescence in intermetallic semiconductors, which are widely studied now.

Appendix

Evidently, Eqs (1) – (4) used as a basis for obtaining experimental b_{A^0X}, b_{D^+X} and b_{D^0X} values [see Eq.(5)] are valid for uniformly excited photoconductors [in them $\delta n, \delta p \neq \varphi(y)$, where y is the propagation direction of the exciting light]. Obviously, if excitation is non-uniform [i.e. $\delta n, \delta p = \varphi(y)$] then the excitonic emission intensities from a photoconductor are:

$$I_{A^0X} = b_{A^0X}\int N_{A^0}n_X dy \approx b_{A^0X}N_A\int n_X dy, \quad \text{(A1)}$$

$$I_{D^+X} = b_{D^+X}\int N_{D^+}n_X dy, \quad \text{(A2)}$$

$$I_{D^0X} = b_{D^0X}\int N_{D^0}n_X dy \approx b_{D^0X}N_D\int n_X dy, \quad \text{(A3)}$$

$$I_X = \alpha_X\int n_X dy, \quad \text{(A4)}$$

where integration is over the extent of the light-emitting region in the y-direction (in this region, as was pointed above, $N_{A^0} \approx N_A$ and $N_{D^0} \approx N_D >> N_{D^+}$).

So, as one could see from Eqs (A1)-(A4), in a discussed case (non-uniform excitation of photoelectrons and photoholes) the b_{A^0X}, b_{D^+X} {if $\delta n/\delta p \neq \varphi(y)$, then $N_{D^+} \neq$

$\neq \varphi(y)$ in a light-emitting region, see [4]}, and b_{D^0X} values could also be found from Eq. (5). From this follows that the proposed method of determination of b_{A^0X}, b_{D^+X} and b_{D^0X} values [from Eq. (5)] could be used for any kind (uniform or non-uniform) photoconductor excitation.

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