Shallow acceptors in strained multiquantum-well Ge/$Ge_{1-x}Si_x$ heterostructures

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Far infrared photoconductivity spectra due to excitation of shallow acceptors in strained multiquantum well Ge/$Ge_{1-x}Si_x$ ($x \approx 0.1$) heterostuctures are investigated. It is shown that these spectra are shifted toward longer wavelengths in the far infrared region compared with those of bulk *p*-Ge, owing to ″built-in″ strain and size quantization, which lead to splitting of the light- and heavy-hole subbands in the Ge layers. Shallow acceptor spectra are calculated variationally for bulk germanium under uniaxial tension, which is ″equivalent″ to the strained Ge layers in the heterostructures. Although this method is only appropriate for wide quantum wells ($d_{Ge} \approx 800$ Å), the calculations are shown to qualitatively account for photoconductivity spectra obtained from narrower wells ($d_{Ge} \approx 200$ Å) as well. © *1998 American Institute of Physics.* [S1063-7826(98)02010-9]

Strain is one of the techniques for controlling the energy spectrum of both free and bound charge carriers. By lowering the crystal symmetry, uniaxial strain in semiconductors with degenerate valence bands at $\mathbf{k}=0$ (e.g., in Ge and Si) causes splitting of the heavy- and light-hole subbands and reconstruction of the shallow acceptor spectrum. The influence of $\mathbf{P} \| [001]$ and $\mathbf{P} \| [111]$ uniaxial strains on the spectrum of shallow acceptors in bulk germanium and silicon has been investigated previously, both theoretically[1–3] and experimentally.[4–6] In these papers it was shown that uniaxial strain causes splitting of the ground and excited fourfold-degenerate states into two twofold-degenerate states. Since the distance between light- and heavy-hole subbands increases linearly with pressure,[1] as the strain increases the spectrum of acceptor bound states is determined primarily by the subband that forms the top of the valence band. It is known[1] that for $P \neq 0$ the dispersion relation for holes becomes anisotropic in both subbands. The problem of motion of a particle near the top of a simple anisotropic band in a Coulomb potential was investigated in Refs. 2 and 7, where the dependences of the ground and excited state energies of an impurity center on the anisotropy parameter of the effective mass was calculated. In Ref. 3 the authors calculated the spectrum of a shallow acceptor in uniaxially strained germanium for arbitrary values of the strain.

In this paper we will investigate shallow acceptors in wide strained Ge/$Ge_{1-x}Si_x$ heterostructure quantum wells grown on a (111) germanium substrate. By ''wide'' quantum wells we mean wells whose width exceeds the characteristic size of the wave function in the acceptor ground state. In such quantum wells, the spectrum of shallow acceptors is primarily determined by the strain of the quantum well material, i.e., essentially the magnitude of the splitting of the heavy- and light- hole subbands. Therefore, in calculating bound-state energies quantum-well effects can to first approximation be included through their influence on the magnitude of the subband splitting of the light and heavy holes.

It is well known that the germanium layers in strained Ge/$Ge_{1-x}Si_x$ heterostructures are potential wells for holes.[8] If the total thickness of the heterostructure exceeds a critical value, then the germanium layers undergo compression in the growth plane of the heterostructure. This strain may be considered as hydrostatic compression plus uniaxial tension along the [111] axis. The hydrostatic compression, which does not decrease the crystal symmetry, leads to an insignificant change the hole mass and therefore has only a slight effect on the binding energy of acceptors. Thus, the spectrum of shallow acceptors in wide germanium layers in the strained Ge/$Ge_{1-x}Si_x$ heterostructures can be approximately treated as the spectrum of shallow acceptors in bulk germanium under uniaxial tension. The results of Ref. 3 cannot be used to analyze the impurity spectra in such heterostructures, because these calculations were made for material under compression. In this paper we calculate the spectrum of shallow acceptors in germanium under uniaxial tension in the direction [111] for an arbitrary value of the strain using the variational method discussed in Ref. 9 for calculating acceptor levels in unstrained germanium.

METHOD OF CALCULATION AND RESULTS OBTAINED

The Luttinger Hamiltonian,[10] which describes light and heavy holes in a Cartesian system of coordinates $\hat{x}[110]$, $\hat{y}[112]$, $\hat{z}[111]$, has the form:

$$H_L = \frac{\hbar^2}{2m_0} \begin{pmatrix} F & H & I & 0 \\ H^* & G & 0 & I \\ I^* & 0 & G & -H \\ 0 & I^* & -H^* & F \end{pmatrix},$$

$$F = A(k_x^2+k_y^2+k_z^2)+\frac{D}{2\sqrt{3}}(k_x^2+k_y^2-2k_z^2)+\left\{a\varepsilon+\frac{d}{2\sqrt{3}}(\varepsilon_{xx}+\varepsilon_{yy}-2\varepsilon_{zz})\right\}\frac{2m_0}{\hbar^2},$$

$$G = A(k_x^2+k_y^2+k_z^2)-\frac{D}{2\sqrt{3}}(k_x^2+k_y^2-2k_z^2)+\left\{a\varepsilon-\frac{d}{2\sqrt{3}}(\varepsilon_{xx}+\varepsilon_{yy}-2\varepsilon_{zz})\right\}\frac{2m_0}{\hbar^2},$$

$$H=\frac{i}{\sqrt{3}}\left(B-\frac{D}{\sqrt{3}}\right)k_+^2-\frac{2i}{\sqrt{3}}\left(B+\frac{D}{2\sqrt{3}}\right)k_zk_-,$$

$$I=\frac{1}{2\sqrt{3}}\left(B+\frac{2D}{\sqrt{3}}\right)k_-^2-\sqrt{\frac{2}{3}}\left(B-\frac{D}{\sqrt{3}}\right)k_zk_+,$$

where A, B, and D are constants that determine the hole dispersion relations, m_0 is the free electron mass, a and d are constants that determine the change in the hole spectrum under strain (see Ref. 1), $k_\pm=k_x\pm ik_y$; ε_{ij} are components of the strain tensor, and $\varepsilon=\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}$. All the nonzero components of the strain tensor can be expressed in terms of ε_{xx}:

$$\varepsilon_{xx}=\varepsilon_{yy},$$

$$\varepsilon_{zz}=-\frac{2(C_{11}+2C_{12}-2C_{44})}{C_{11}+2C_{12}+4C_{44}}\varepsilon_{xx}=-0.369\varepsilon_{xx},$$

where C_{ij} are elements of the elastic stiffness tensor.[11] The quantity ε_{xx} is linearly related to the tensile stress (pressure)

$$P=-\frac{6C_{44}(C_{11}+2C_{12})}{C_{11}+2C_{12}4C_{44}}\varepsilon_{xx}=183.3\varepsilon_{xx}\text{GPa}.$$

In germanium uniaxially strained along the direction [111], the center of the Brillouin zone corresponds to the symmetry point group D_{3d}. In unstrained germanium, the L-point possesses this symmetry group. Therefore, wave functions of acceptors in the strained material will transform according to a complementary representation of the double group at the L point. There exist six such irreducible representations of the group D_{3d}: $L_4^\pm$, $L_5^\pm$, and $L_6^\pm$ (Ref. 12). The wave function of the acceptor ground state transforms according to an irreducible representation corresponding to the top of the valence band. In unstrained germanium the top of the valence band corresponds to the four-dimensional representation Γ_8^+ (Ref. 12). This representation decomposes into irreducible representations under the group D_{3d} as follows:

$$\Gamma_8^+=L_4^++L_5^++L_6^+.$$

Representations L_4^+ and L_5^+ are one- dimensional and complex conjugates of one another; they should therefore be treated as the single, two-dimensional, irreducible representation $L_{4,5}^+$ for transforming the wave functions.[13] Representation L_6^+ is two-dimensional. As we already noted, uniaxial strain lifts the degeneracy of the light- and heavy-hole subbands at the point $\mathbf{k}=0$. In this case, wave functions corresponding to states with projections $\pm3/2$ of total angular momentum on the strain axis (heavy holes) transform according to the representation $L_{4,5}^+$. Wave functions of states with the projections $\pm1/2$ of total angular momentum on the strain axis (light holes) transform according to the representation L_6^+. Note that for those holes determined to be ''heavy'' the mass in the direction [111] is larger than that of the light holes (by a factor of almost 10). In general, this assertion is incorrect for other directions. In uniaxial strained germanium, the top of the valence band is made up of heavy holes, while under uniaxial compression it is made up of light holes.

In this paper we use a variational method to calculate the spectra of shallow acceptor states. Starting from the symmetry of the problem and using group theory analogous to Ref. 9, we can minimize the number of variational parameters and choose the following trial wave functions for the split ground state that transform according to representations $L_{4,5}^+$ and L_6^+:

$$\Psi_h=c_1\begin{pmatrix}1\\0\\0\\i\end{pmatrix}\exp\left(\frac{-\sqrt{\rho^2+(qz)^2}}{f}\right)+\left\{c_2[\rho^2-2(qz)^2]\begin{pmatrix}1\\0\\0\\i\end{pmatrix}+c_3\rho^2\begin{pmatrix}0\\ie^{-2i\varphi}\\e^{2i\varphi}\\0\end{pmatrix}+c_4\rho z\begin{pmatrix}0\\-ie^{i\varphi}\\e^{-i\varphi}\\0\end{pmatrix}\right\}\exp\left(\frac{-\sqrt{\rho^2+(qz)^2}}{b}\right), \quad (1)$$

$$\Psi_l=c_1\begin{pmatrix}0\\1\\0\\0\end{pmatrix}\exp\left(\frac{-\sqrt{\rho^2+(qz)^2}}{f}\right)+\left\{c_2[\rho^2-(qz)^2]\begin{pmatrix}0\\1\\0\\0\end{pmatrix}+i\rho z\begin{pmatrix}c_3e^{i\varphi}+c_4e^{-i\varphi}\\0\\0\\0\end{pmatrix}+i\rho^2\begin{pmatrix}c_5e^{2i\varphi}+c_6e^{-2i\varphi}\\0\\0\\0\end{pmatrix}\right\}\exp\left(\frac{-\sqrt{\rho^2+(qz)^2}}{b}\right). \quad (2)$$

Here Ψ_h and Ψ_l transform according to the representations $L_{4,5}^+$ and L_6^+, respectively; $\rho^2=x^2+y^2$, φ is the angle of the radius vector in the plane xy, and c_i, q, f, and b are variational parameters.

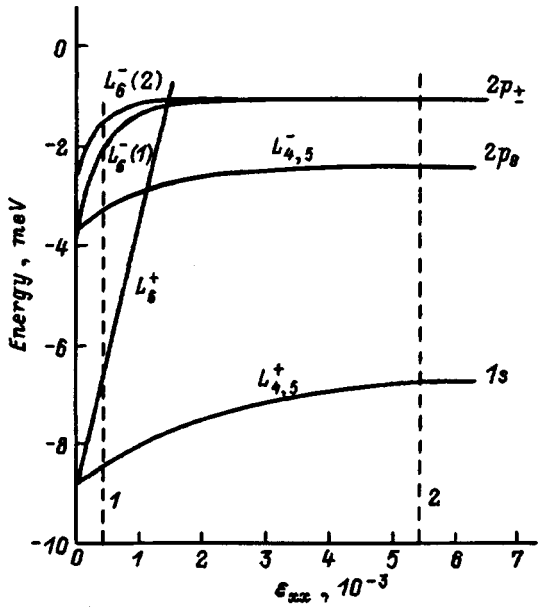

FIG. 1. Dependence of the binding energy of the ground and lowest excited states of an acceptor in germanium under uniaxial tensile strain along the (111) direction on the magnitude of the strain. The vertical lines indicate values of the effective strain corresponding to the splitting of the light- and heavy-hole subbands in $Ge/Ge_{1-x}Si_x$ heterostructures 309 (*1*) and 306 (*2*) including size quantization.

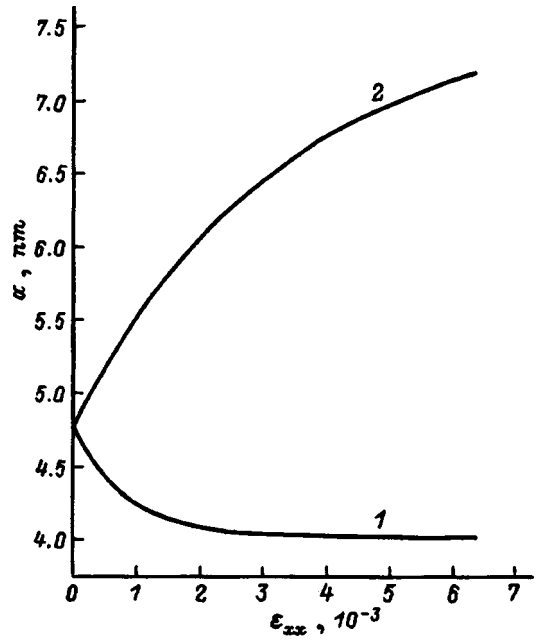

FIG. 2. Dependence of the longitudinal (along the tension axis *1*) and transverse (*2*) localization scales *a* for the wave function of the acceptor ground state in germanium under tensile uniaxial strain along (111) on the magnitude of the strain.

In Fig. 1 we plot the calculated energies of the components of the split acceptor ground state, i.e., $L_{4,5}^+$ [with wave function (*1*)] and L_6^+ [with wave function (*2*)], versus the strain ε_{xx}. The energy origin corresponds to the edge of the heavy-hole subband. The ionization energy obtained in this paper for the acceptor ground state when $P=0$ equals 8.87 meV, which is in good agreement with the value 8.83 meV obtained in Ref. 9. At large strains ($\varepsilon_{xx}>15\times10^{-3}$) the ionization of the ground state approaches an asymptotic value of 5.57 meV, which is in good agreement with calculations using the single-band model with an anisotropic mass.[14]

Figure 2 shows how the scales of localization of the ground-state wave function along the strain axis and in the perpendicular direction depend on the magnitude of the strain. It is easy to see that the localization radius (or ''Bohr'' radius) along the direction of tension decreases with increasing strain down to 4.0 nm, while in the perpendicular direction it increases. This behavior is explained by the change in mass in the corresponding directions.[1]

Let us now consider *p*-type excited states. The wave functions of these states are odd under inversion, and therefore they correspond to irreducible representations $L_{4,5}^-$ or L_6^-. Note that dipole transitions from the ground state are allowed in odd states and forbidden in even states.

In accordance with the symmetry of the problem (see Ref. 9) trial wave functions that transform according to representations $L_{4,5}^-$ and L_6^- can be written in the following form. For $L_{4,5}^-$:

$$\Psi_{4,5}=c_1 z\begin{pmatrix}1\\0\\0\\i\end{pmatrix}\exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{f}\right]+ic_2\rho\begin{pmatrix}0\\e^{i\varphi}\\ie^{-i\varphi}\\0\end{pmatrix}$$

$$\times\exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{f}\right]+c_3 z\left(z^2-\frac{1}{5}r^2\right)\begin{pmatrix}1\\0\\0\\i\end{pmatrix}$$

$$\times\exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right]+ic_4\rho\left(z^2-\frac{3}{5}r^2\right)\begin{pmatrix}0\\e^{i\varphi}\\ie^{-i\varphi}\\0\end{pmatrix}$$

$$\times\exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right]+ic_5\rho^2 z\begin{pmatrix}0\\e^{-2i\varphi}\\ie^{2i\varphi}\\0\end{pmatrix}$$

$$\times\exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right]+c_6\rho^3\begin{pmatrix}e^{3i\varphi}\\0\\0\\ie^{-3i\varphi}\end{pmatrix}$$

$$\times\exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right]+c_7\rho^3\begin{pmatrix}e^{-3i\varphi}\\0\\0\\ie^{3i\varphi}\end{pmatrix}$$

$$\times \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right]. \quad (3)$$

For L_6^-:

$$\Psi_6 = c_1 z \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{f}\right] + \rho \begin{pmatrix} i(c_2 e^{i\varphi} + c_3 e^{-i\varphi}) \\ c_4 e^{-i\varphi} \\ 0 \\ 0 \end{pmatrix}$$

$$\times \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{f}\right] + c_5 z\left(z^2 - \frac{1}{5}r^2\right)\begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}$$

$$\times \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right] + \rho\left(z^2 - \frac{3}{5}r^2\right)$$

$$\times \begin{pmatrix} i(c_6 e^{i\varphi} + c_7 e^{-i\varphi}) \\ c_8 e^{-i\varphi} \\ 0 \\ 0 \end{pmatrix}$$

$$\times \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right] + i\rho^2 z \begin{pmatrix} c_9 e^{2i\varphi} + c_{10} e^{-2i\varphi} \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

$$\times \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right] + \rho^3 \begin{pmatrix} c_{11} e^{3i\varphi} + c_{12} e^{-3i\varphi} \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

$$\times \exp\left[-\frac{\sqrt{\rho^2+(qz)^2}}{b}\right]. \quad (4)$$

In this paper we find two excited states with wave function (4), which we denote by L_6^- (1) and L_6^- (2). In finding the second state we impose an additional condition on the variational parameters: the requirement that the upper state (4) be orthogonal to the lower state. For states of type (3) we find only the lower excited state, since the second rapidly departs into the continuum with increasing strain. Figure 1 also shows the dependences of the calculated excited acceptor state energies on the strain ε_{xx}. On the right we indicate the corresponding acceptor states in the limit of large strain for a simple anisotropic band ($1s$, $2p_0$, $2p_\pm$). It is clear that the energy of the excited levels approaches its asymptotic value more rapidly than the ground- state energy, which is obviously associated with the lower binding energy of the excited states.

EXPERIMENTAL RESULTS AND COMPARISON WITH CALCULATIONS

The strained multilayer heterostructures $Ge/Ge_{1-x}Si_x$ under study (with $x \approx 0.1$, germanium layer thickness $d_{Ge} \approx$ 200–800 Å, and solid solution layer thickness $d_{GeSi} \approx 200$ Å) were grown by the gas-phase hydride method on (111) Ge substrates. The number of periods was chosen to be rather large ($n = 80 \div 160$) so that the total thickness of the heterostructure would exceed its critical value. In this case, a relaxation of the elastic stresses takes place at the heterostructure-substrate boundary, and the period of the lattice in the heterostructure growth plane no longer coincides with the period of the substrate lattice (it is determined primarily by the average content of silicon in the heterostructure), leaving the germanium layers in the heterostructure under biaxial compression. Thus, we can control the strain in the germanium layers by varying the fraction of silicon x in the $Ge_{1-x}Si_x$ alloy or the layer thickness. In this paper we studied heterostructures of two types, with different quantum-well widths. The first type ($x = 0.07$, $d_{GeSi} = 200$ Å, $n = 83$), an example of which is sample 309, had thick germanium layers ($d_{Ge} = 800$ Å) that consequently were weakly strained ($\varepsilon_{xx} = 0.46 \times 10^{-3}$). The second series consisted of several samples ($x = 0.11 - 0.15$, $d_{GeSi} \approx 200$ Å, $n = 80 - 160$) with thinner germanium layers ($d_{Ge} \approx 200$ Å), causing them to be under larger strains ($\varepsilon_{xx} = (0.9 - 2.1) \times 10^{-3}$). Spectra of these samples were measured previously.[14] We measured submillimeter photoconductivity spectra of these heterostructures using a ''BOMEM DA3.36'' Fourier spectrometer at a temperature $T = 4.2$ K.

The impurity photoconductivity spectrum of a sample with thick germanium layers is shown in Fig. 3a. This spectrum is shifted as a whole towards longer wavelengths compared to the spectrum of bulk p-Ge (the maximum energy $\hbar\omega_{max} \approx$ 12 meV; see, e.g., Ref. 15) and consists of a line at $\hbar\omega \approx$ 5.3 meV and a broad band at $\hbar\omega =$ 7.4–10 meV. In this sample the quantum-well effects are unimportant, and the photoconductivity spectrum is in good agreement with the theoretical model described above (Fig. 1); the strain corresponding to sample 309 is indicated by the vertical line *1*). The line at $\hbar\omega \approx$ 5.3 meV corresponds to a transition from the $L_{4,5}^+$ ground state to the first excited $L_{4,5}^-$ state. The spacing (1) between levels $L_{4,5}^+$ and L_6^- amounts to 7.1 meV, which is in good agreement with the start of the short-wavelength band in the spectrum shown in Fig. 3a. The photoconductivity at higher frequencies can in this case be associated with transitions to higher-lying excited states and to the continuum, and also to the split-off light-hole subband (the splitting of the subbands in this sample is $\Delta =$ 3 meV).

Spectra for the $Ge/Ge_{1-x}Si_x$ heterostructures with thinner germanium layers are shown in Figs. 3b and 3c. It is clear that these spectra are shifted even farther into the region of lower frequencies; features these spectra have in common are a line at $\hbar\omega \approx$ 6.9 meV and an intense band at $\hbar\omega =$ 3–5 meV. It is obvious that in heterostructures with narrow germanium layers the quantum-well effects are more important. As we have already noted, in these structures strain causes the masses of light and heavy holes along the [111] axis, i.e., the growth direction of the structure, to differ by an order of magnitude. This leads to additional splitting of the light- and heavy-hole subbands due to size quantization. In this paper this effect is included by introducing an effective strain ε_{eff} corresponding to the total splitting of the sub-

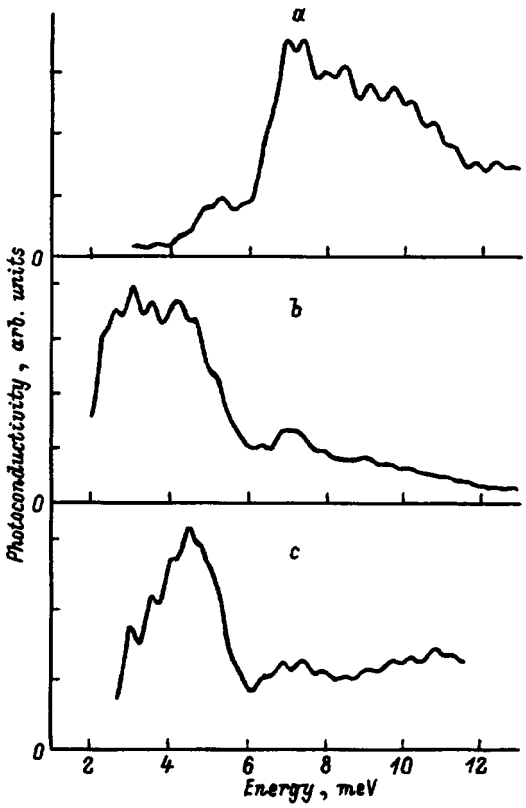

FIG. 3. Photoconductivity spectra for $Ge/Ge_{1-x}Si_x$ heterostructures at $T = 4.2$ K. *a*— heterostructure 309, undoped, $x=0.07$, $d_{Ge}=800$ Å, $d_{GeSi}\approx 200$ Å, $n=83$, $\varepsilon_{xx}=0.46\times 10^{-3}$; *b*—heterostructure 306, undoped, $x=0.12$, $d_{Ge}=200$ Å, $d_{GeSi}=\approx 200$ Å, $n=162$, $\varepsilon_{xx}=2.1\times 10^{-3}$; *c*—heterostructure 379, central quantum well doped with boron, $x=0.15$, $d_{Ge}=200$ Å, $d_{GeSi}\approx 200$ Å, $n=81$, $\varepsilon_{xx}=0.9\times 10^{-3}$.

bands calculated for given ε_{xx} and d_{Ge} (Ref. 16) (in Fig. 1 the vertical line *2* indicates the value of ε_{eff} for sample 306). From a comparison of Figs. 1, 3b, and 3c we see that the line at $\hbar\omega\approx 6.9$ meV is in good agreement with transitions from the ground state to the continuum. The most intense band in the spectrum probably should correspond to transitions from the ground state to states L_6^- (1) and L_6^- (2), i.e., transitions of type $1s\rightarrow 2p_\pm$ (since for large subband splittings the spectrum becomes similar to the spectrum of a donor, for which the transition to the lower-lying state $L_{4,5}^-$ $(2p_0)$ is forbidden for light at normal incidence). From Fig. 1 it is clear that this transition should correspond to the line at $\hbar\omega\approx 5.5$ meV, whereas in the experimental spectra the short-wavelength edge of the intense photoconductivity band corresponds to a photon energy of $\hbar\omega\approx 50$ meV. This discrepancy indicates the need for a more accurate inclusion of the quantum-well effect in structures with thin germanium layers. The long-wavelength tail of the band $\hbar\omega = 3$–5 meV can be explained by the dependence of the binding energy of an acceptor on its position in the well: At the well center the energy is a maximum and it decreases as the acceptor moves towards the barrier.[17] The additional structure in the region 1.8 to 3 meV observed in sample 306 can be related to photoexcitation of A^+ centers, which form when an additional hole is captured by a neutral acceptor in the quantum well.[18]

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