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Ambipolar tunneling in near-surface quantum wells

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We study the photoluminescence from a near-surface quantum well in the regime of ambipolar tunneling to the surface states. Under steady-state excitation an electric field develops self-consistently due to the condition of equal tunneling currents for electrons and holes. The field induces a Stark shift of the photoluminescence signal which compares well with experimental data from near-surface GaAs/AlGaAs single quantum wells.

For a quantum well built in proximity of an unpassivated surface, tunneling to surface states can be a nonradiative recombination channel competitive with photoluminescence. The importance of this effect in determining the emission efficiency has been demonstrated experimentally in various papers [1–4]. Recently we have proposed a quantitative model based on ambipolar tunneling of electrons and holes which is applicable to many-well systems in the bulk or single wells coupled to surface state [5]. In steady-state situations the ambipolar regime, with equal tunneling currents for electrons and holes, imposes an electric field to develop [6–9]. The field induces a peak shift of the excitonic recombination via the quantum confined Stark effect [10]. Here we specialize the discussion to the case of a quantum well coupled to surface states and we compare the theoretical results with photoluminescence experimental data in GaAs/AlGaAs material.

We will use the following notation. The width of the quantum well is a and the width of the surface barrier is b. The bottom of the $e1$ and $hh1$ bands of the well are E_{e1} and E_{hh1} and G is the generation current density of electron-hole pairs in the well. We assume that no pairs are generated within the barrier or at the surface. The pairs generated into the well relax almost instantaneously, compared to the other relevant time scales, to the lowest band of the well. Electron-hole interaction leads to exciton formation. Tunneling from the well to the surface states is due to free electrons and holes only [6]. On the other hand, photoluminescence is restricted only to excitonic recombination in the well. If n_w and p_w are the steady-state concentrations (number of particles per unit area) of electrons and holes in the well, the following rate equations hold:

$$0 = G - J_e - \lambda n_w p_w \tag{1a}$$

$$0 = G - J_h - \lambda n_w p_w \tag{1b}$$

$$0 = \lambda n_w p_w - I. \tag{1c}$$

The bimolecular generation rate of excitons is assumed proportional to the electron and hole concentrations [11]. The photoluminescence current density I is proportional to the exciton concentration in the well. Transfer of electrons (holes) from the well to the surface states is realized in a non-coherent two-step process. Quantum coherent tunneling of electrons (holes) from an occupied

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state of the $e1$ ($hh1$) band of the well to an equal-energy empty state at the surface is followed by relaxation toward the lower energy states. When the barrier width b is not too small, the phonon relaxation process at the surface is much faster than the tunneling process (current densities J_e and J_h) and can be neglected.

The tunneling current densities are approximately proportional to the charge concentrations in the well and the proportionality factor, namely the tunneling probability, is generally quite different for electrons and holes. Therefore, in a steady-state situation when $J_e = J_h$, the concentrations of electrons and holes in the well must be different. The resulting electric field, in turn, affects the electron and hole tunneling probabilities.

Since the tunneling rate depends both on the effective mass of the carriers and the density of states at the surface, two cases are possible. When the density of states of the donor-like band at the surface is not sufficiently smaller than that of the acceptor-like band, the electron tunneling rate is larger than the hole tunneling rate. In this case electrons accumulate at the surface and in a steady-state situation $p_w > n_w$. The electric field is directed from the well to the surface and its value is given by

$$F = \frac{en}{\varepsilon_0 \varepsilon_r} \tag{2}$$

where $n = p_w - n_w$ and ε_r is the permittivity of the barrier material. A reversed situation, however, may happen when the effective mass difference between electrons and holes is overcompensated by the difference in the surface densities.

For a given value of the electric field F, in the first order perturbation theory the tunneling current densities are $J_e = n_w/\tau_e$ and $J_h = p_w/\tau_h$ where

$$\frac{1}{\tau_e} = \frac{2\pi}{\hbar} |\langle \Phi_0^s | V_e | \Phi_0^w \rangle|^2 \; A \; v_e(0) f \left(\frac{\epsilon_F^e}{k_B T} \right) \tag{3}$$

$$\frac{1}{\tau_h} = \frac{2\pi}{\hbar} |\langle \Phi_0^s | V_h | \Phi_0^w \rangle|^2 \; A \; v_h(0) f \left(\frac{\epsilon_F^h}{k_B T} \right). \tag{4}$$

$v_e(\epsilon)$ and $v_h(\epsilon)$ are the densities of states in the donor-like and acceptor-like surface bands respectively. Energies are measured from the bottom of the $e1$ and $hh1$ bands. The electron and hole Fermi energies ϵ_F^e and ϵ_F^h which appear in the Fermi function f are related to the respective electron and hole concentrations at the surface, n_s and p_s, as explained in the following. Finally, A is the relevant transverse area and the matrix elements are evaluated in the Appendix.

When the electric field and the tunneling rates are known, the solution of the rate equations is

$$n_w = \sqrt{\left(\frac{n}{2} + \frac{1}{2\lambda\tau_e} \right)^2 + \frac{G}{\lambda}} - \left(\frac{n}{2} + \frac{1}{2\lambda\tau_e} \right) \tag{5a}$$

$$p_w = n_w + n. \tag{5b}$$

Moreover, $n_s = n$ and $p_s = 0$ if $F > 0$ and $n_s = 0$ and $p_s = -n$ if $F < 0$.

This result allows one to find the steady-state values of the electric field, of the charge concentrations and of the tunneling rates by a recursive method. Starting from some trial value, the electric field, i.e. the charge concentration n, is changed until the condition $J_e = J_h$ is reached. At this point the luminescence current density I is obtained from the equilibrium concentrations of electrons and holes in the well.

Carrying out explicitly the calculations implies the knowledge of the energy distribution of the surface states. Inversely we can try to get information on the surface states by fitting experimental

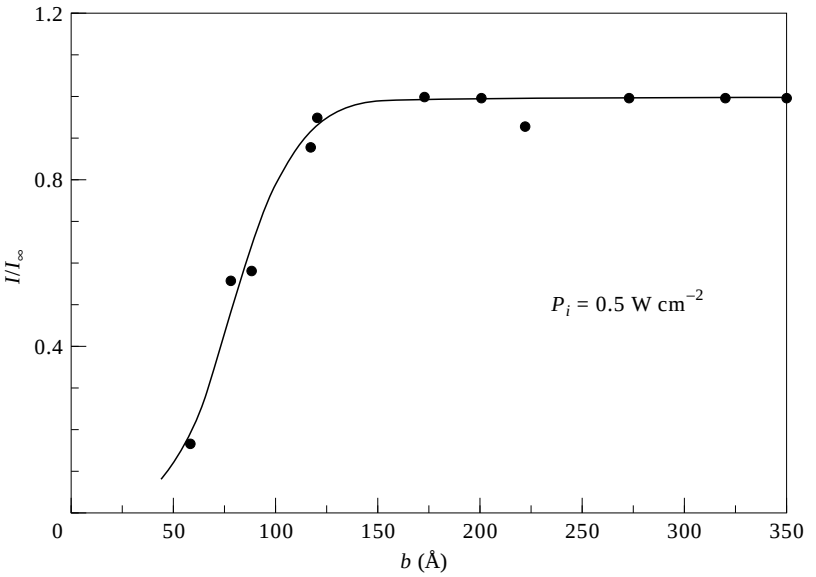

Fig. 1. Normalized photoluminescence ratio I/I_∞ of a near-surface well versus the surface-barrier thickness b. Dots: experimental data from Ref. [1]; solid line: best fitting in terms of the self-consistent model. Incident power density is $P_i = 0.5$ W cm^{-2}.

photoluminescence data. We concentrate on the specific example of an $Al_{0.3}Ga_{0.7}As$ surface with a nearby GaAs quantum well [1,6].

At energy close to the bottom of the $e1$ band of the well the $Al_{0.3}Ga_{0.7}As$ surface has only donor-like states belonging to the exponentially vanishing Urbach tail

$$v_e(\epsilon) = \frac{m_e^s}{\pi\hbar^2} \exp\left(-\frac{\Delta E_c + eFb - E_{e1} - \epsilon}{\epsilon_e} \right). \tag{6}$$

Note that energy is measured from the bottom of the $e1$ band. Such states are assumed to be nodal hydrogenic wavefunctions [12] with radius r_e fixed by their depth into the gap. Their explicit expression is given in the Appendix. We assume that at the top of the gap the state density is the two dimensional density of free $Al_{0.3}Ga_{0.7}As$ electrons with effective mass m_e^s. The parameter ϵ_e will be considered as a fitting parameter. According to eqn (6) the Fermi energy for the donor-like surface band containing n_s electrons is

$$\epsilon_F^e = \Delta E_c + eFb - E_{e1} + \epsilon_e \ln\left(\frac{\pi\hbar^2 n_s}{m_e^s \epsilon_e} \right). \tag{7}$$

On the other hand, at energy close to the bottom of the $hh1$ band of the well the $Al_{0.3}Ga_{0.7}As$ surface has a very high concentration of acceptor-like defect states [13]. We schematize them again by nodal hydrogenic wavefunctions [12] but with radius r_h to be considered as a second fitting parameter. These states are assumed to be distributed in energy with constant density v_h over an interval ΔE_h into the gap. The Fermi energy for the acceptor-like surface band is then

$$\epsilon_F^h = p_s/v_h + \Delta E_v - eFb - E_{hh1} - \Delta E_h. \tag{8}$$

Due to the high ratio between the hole and the electron surface-state density [13] holes accumulate at the surface and in a steady-state situation we have $F < 0$.

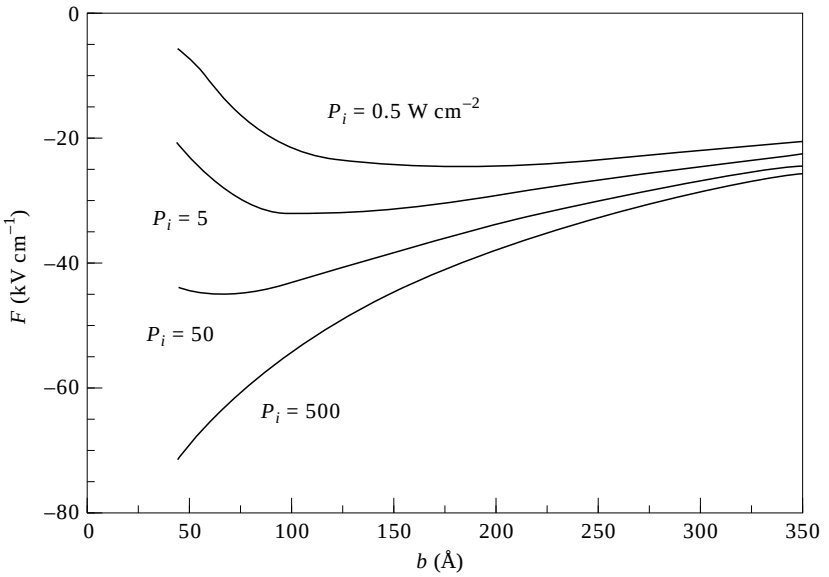

Fig. 2. Calculated electric field F across the surface barrier versus the surface barrier thickness b for different incident power densities P_i.

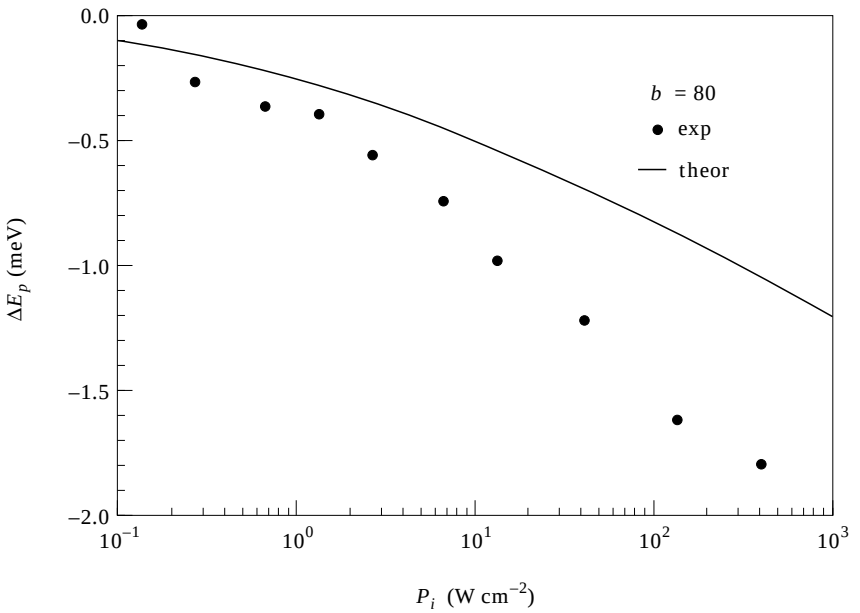

Fig. 3. Comparison between the Stark shift of the photoluminescence signal calculated from the model (solid line) and measured (dots) versus the incident power density P_i. The surface-barrier thickness is $b = 80$ Å.

Experimental photoluminescence data are available (see Ref. [1] for details) for a well width $a = 60$ Å, a temperature $T = 4.2$ K, a photon-pump energy $h\nu = 1.608$ eV and with an incident power density $P_i = 0.5$ W cm^{-2}. The absorption efficiency is estimated to be 1%, so we take $G = 0.01\ P_i/h\nu$.

The relevant material parameters are [14]: $\Delta E_c = 0.3$ eV, $\Delta E_v = 0.128$ eV, $m_e^s = 0.091\, m$, $m_e^w = 0.067\, m$, $m_h^w = 0.34\, m$, m being the free electron mass, and $\varepsilon_r = 12$. Moreover we put $\lambda = 6\ \mathrm{cm}^2\ \mathrm{s}^{-1}$ [11]. We assume an acceptor-like surface state density $\nu_h = 10^{14}\ \mathrm{cm}^{-2}\ \mathrm{eV}^{-1}$ [15] with $\Delta E_h \simeq 0.5$ eV (the results we found do not depend crucially on this particular value).

The free parameters, ϵ_e and r_h, are fixed by fitting the normalized photoluminescence intensity I/I_∞ to the experimental data [1] obtained for different values of the barrier width b (the normalization factor I_∞ is the photoluminescence current density for $b \to \infty$). A least-square-error procedure gives the unique solution $\epsilon_e = 12$ meV and $r_h = 11$ Å. In Fig. 1 we compare the ratio I/I_∞, calculated with these values, with the experimental data. The agreement is excellent.

In Fig. 2 we show the electric field value calculated in the situation of Fig. 1 as a function of the barrier thickness. In the same figure we show also the field obtained with different values of the incident power density P_i, all the other parameters being fixed. It is seen that, for high levels of excitation, the field approaches values of order $10^5\ \mathrm{V\ cm}^{-1}$ and keeps increasing when the barrier becomes thinner.

An important check of the validity of our model is given by the analysis of the Stark shift. The self-consistently estimated electric field induces a band bending which modifies the single particle levels $e1$ and $hh1$ and, therefore, the exciton recombination energy ΔE_p. The Stark shift calculated as a sum of the shifts of levels $e1$ and $hh1$ is shown in Fig. 3 as a function of the QW excitation (details of the calculation will be reported elsewhere). In Fig. 3 we show also the corresponding measured energy shifts (dots). The agreement is good in the region ($P_i \simeq 0.5$) where the fitting parameters were fixed as explained above and is fairly satisfactory over three orders of magnitude. An improvement should be possible if the surface-state spectrum were known *a priori*. This is a confirmation that the ambipolar tunneling approach provides a reasonably accurate description of the loss of efficiency in near-surface quantum wells.

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Appendix

The surface is defined by the plane $z=0$ and the well is in $b<z<b+a$. Firstly, we consider the case of electrons. We assume a rectangular potential profile with left and right discontinuities $V_l=\Delta E_c+eFb/2$ and $V_r=\Delta E_c$ for the well where F is the electric field in the barrier region $0\le z\le b$. The electron wavefunction at energy $\epsilon=0$ from the bottom of the $e1$ band of the well is given by

$$\Phi^w_{\epsilon=0}=\frac{C}{\sqrt{A}}\begin{cases}\sin\delta\exp(k_1[z-b]) & z<b\\ \sin(k(z-b)+\delta) & b<z<b+a\\ \sin(ka+\delta)\exp(-k_0[z-b-a]) & z>b+a\end{cases} \tag{9}$$

where $\hbar k_1=\sqrt{2m^w_e(\Delta E_c+eFb/2-E_{e1})}$, $\hbar k=\sqrt{2m^w_e E_{e1}}$ and $\hbar k_0=\sqrt{2m^w_e(\Delta E_c-E_{e1})}$. The phase shift is $\delta=\tan^{-1}(k/k_1)$ and the energy is determined by solving

$$ka=\pi-\sin^{-1}\left(\frac{\hbar\mathrm{k}}{\sqrt{2m^w_e V_l}}\right)-\sin^{-1}\left(\frac{\hbar k}{\sqrt{2m^w_e V_r}}\right) \tag{10}$$

The constant C is fixed by normalization

$$C=\left\{\frac{\sin^2\delta}{2k_1}+\frac{\sin^2(ka+\delta)}{2k_0}+\frac{a}{2}-\frac{\sin(2(ka+\delta))-\sin(2\delta)}{4k}\right\}^{-1/2} \tag{11}$$

The donor-like surface state $\Phi^s_{\epsilon=0}$ is approximated by a truncated $2p$ hydrogenic wavefunction [12]

$$\Phi^s_{\epsilon=0}=\frac{z}{4\sqrt{\pi}r_e^{5/2}}\exp(r/2r_e)\begin{cases}0 & z<0\\ 1 & 0<z\end{cases} \tag{12}$$

where $r=\sqrt{x^2+y^2+z^2}$. The state is at energy $\hbar^2/(8m^s_e r_e^2)$ below the bottom of the conduction band for the barrier material where the electron effective mass is m^s_e. By imposing the condition that this energy corresponds to $\epsilon=0$, we determine the radius r_e

$$r_e=\frac{\hbar}{\sqrt{8m^s_e(\Delta E_c-E_{e1}+eFb)}} \tag{13}$$

Assuming that the perturbation potential V_e is of the order of the conduction band offset, the tunneling matrix element between the well and surface states at $\epsilon=0$ can be evaluated analytically

$$\langle\Phi^s_0|V_e|\Phi^w_0\rangle=\frac{\sqrt{\pi}r_e^{3/2}\Delta E_c C\sin\delta}{8\sqrt{A}}\left\{e^{-k_1 b}\left[\frac{1}{4(2k_1r_e-1)^2}-\frac{1}{32(2k_1r_e-1)^3}\right]\right.$$

$$\left.+e^{-b/2r_e}\left[\frac{(b/r_e)^2+2b/r_e}{2k_1r_e-1}-\frac{1+b/r_e}{4(2k_1r_e-1)^2}+\frac{1}{32(2k_1r_e-1)^3}\right]\right\} \tag{14}$$

In the case of holes we have a completely analogous situation where the relevant band in the well is $hh1$ instead of $e1$ and the acceptor-like surface state is given by eqn (12) with $r_e\to r_h$. Equation (14) gives the tunneling matrix element for holes with the substitutions $r_e\to r_h$, $\Delta E_c\to\Delta E_v$, $eFb\to -eFb$.