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Electroluminescence spectra of an STM-tip-induced quantum dot

M. D. Croitoru,* V. N. Gladilin,* V. M. Fomin,*1 and J. T. Devreese‡
Theoretische Fysica van de Vaste Stoffen (TFVS), Universiteit Antwerpen, Belgium

M. Kemerink, P. M. Koenraad, K. Sauthoff,‡ and J. H. Wolter
COBRA Inter-University Research Institute, Eindhoven University of Technology, The Netherlands
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We analyze the electroluminescence spectrum of an STM-tip-induced quantum dot in a GaAs surface layer. A flexible model has been developed that combines analytical and numerical methods and describes the key features of many-particle states in the STM-tip-induced quantum dot. The dot is characterized by its depth and lateral width, which are experimentally controlled by the bias and current. We find, in agreement with experiment, that increasing the voltage on the STM tip results in a redshift of the electroluminescence peaks, while the peak positions as a function of electron tunneling current through the STM tip reveal a blueshift.

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I. INTRODUCTION

Semiconductor quantum dot (QD) systems are of continuing scientific interest due to their three-dimensional carrier confinement, resulting in atomlike characteristics1–4 that make them valuable for applications. In addition to allowing the study of fundamental physics in a zero-dimensional semiconductor system, the discrete energy levels in QD’s are expected to result in a number of advantageous properties for optoelectronics5,6 and memory devices.7–9 Various fabrication technologies of quantum dots have been proposed, among them chemical methods, such as metal-organic chemical-vapor deposition (MOCVD) and molecular-beam epitaxy (MBE), and electrostatic patterning.

Despite years of intensive research into the optical properties of quantum dots, there are still several problems hindering device applications of the zero-dimensional properties of the dots. These include control of the carrier confinement and energy level separation, areal density and optical quality, uniformity in size and shape, and ordering of the dots in an ensemble. The complexity of a system containing an ensemble of QD’s, caused by variations in the dot size, shape, composition, and the nearest-neighbor distance, makes predictions of the optical properties problematic. With this in mind, a study of the electroluminescence of quantum dots, which have the possibility of tuning their optical characteristics by choosing of parameters like voltage, is of strong interest.

A special and unique type of quantum dot that can only be studied by using a scanning-tunneling microscope (STM) is the so-called tip-induced quantum dot.10–13 When a bias is applied between the metallic STM tip and semiconductor sample, the electric field extends into the semiconductor structure and a hole or an electron accumulation layer can be formed under the tip. For sufficiently small tip apex radii, quantization occurs both in the radial direction and along the growth axis of the structure. It should be noted that the tip-induced band bending confines only a single type of carrier—holes in our case. The electrons are repelled from the region under the tip. Thus the tip forms an antidot for electrons. By putting a barrier just below the surface injected electrons can be confined to the surface layer and radiative recombination with holes in the dot becomes possible. We analyze the electroluminescence of a quantum dot induced by an STM tip in the GaAs surface layer of a GaAs/A10.25Ga0.75As multilayer structure.13 To describe the energy spectra of charge carriers the Poisson and Schrödinger equations are solved self-consistently within the framework of the Hartree approximation, by using a finite-difference scheme on a composite grid. The experimentally observed positions of electroluminescence peaks, attributed to electron-hole recombination in the quantum dot, are very sensitive to the electron tunneling current, even in the case when the current is weak and the effect of electron charge on the electrostatic potential profile in the structure is negligible.

We show that this puzzling sensitivity can be linked to variations of the STM-tip contact area with varying current. The developed model also allows us to calculate the electroluminescence spectrum as a function of the applied tip voltage.

The remaining part of the paper is organized as follows: in Sec. II, the results of optical spectroscopy measurements, performed on a STM-tip-induced quantum dot, are presented. In Sec. III, a quantum mechanical description of the system of holes and electrons, as well as the numerical method of the self-consistent solution of the Poisson and Schrödinger equations are given. Calculation of the electroluminescence spectrum is presented in Sec. IV. In Sec. V, the results of the calculations are discussed.

II. EXPERIMENTAL SETUP AND RESULTS

The experiments are performed in a low-temperature STM setup with light collection facilities.14 The sample that was used in the experiments is grown by molecular-beam epitaxy. The surface layer is a 17-nm-thick undoped GaAs layer, the barrier material is A10.25Ga0.75As. The first doping layer is a δ layer of beryllium acceptors, placed 200 Å below the GaAs/A10.25Ga0.75As interface. The deeper layers of the sample contain, among others, a quantum well and a superlattice, 62 and 107 nm below the surface, respectively. Their contribution to the optical spectra is discussed in Ref. 15. Prior to mounting, the samples are sulfur passivated, which

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results in a surface that is covered with a thin sulfur layer—typically one or a few monolayers thick—and is free of gap states. Because of the presence of this insulating top layer, the STM tip is in physical contact with the sample surface under typical experimental conditions. However, the resulting deformation of the tip apex is found to be the totally elastic.

Since the GaAs surface is free of gap states, a surface hole accumulation layer is formed under the tip at positive sample bias. The light that is emitted when the holes in this layer recombine with electrons that are injected from the tip is collected by two large-diameter fibers and projected on a cooled Si charge-coupled device (CCD) by a 30-cm monochromator. The typical integration time was 30–60 sec.

III. MODEL

In our model we consider the structure sketched in Fig. 1. The shape of the STM tip is modeled by a truncated cone, because when the tip made of a soft metal (Pt) is pressed to the semiconductor layer, its apex becomes flat. The STM tip and GaAs layer are separated from each other by an insulating sulfur passivation layer of thickness \( l_1 \) (Ref. 13). The holes and electrons are confined in the cap layer of width \( l_2 \). The width of the \( \text{Al}_{0.25}\text{Ga}_{0.75}\text{As} \) barrier is \( l_3 \). The \( \text{Al}_{0.25}\text{Ga}_{0.75}\text{As} \) layer borders on the \( \text{In}_{0.25}\text{Ga}_{0.75}\text{As} \) quantum well. The contact area of the STM tip with the multilayer structure is a circle of radius \( R \).

The contact area of the STM tip with the multilayer structure is a circle of radius \( R_{\text{tip}} \). Owing to the axial symmetry of the system, the cylindrical coordinates \( r, \varphi, \) and \( z \) are used. The \( z \) axis is directed along the symmetry axis of the tip, which is chosen parallel to the growth axis of the structure. While the hole motion is confined in all directions by the potential well, the electron motion is confined only along the \( z \) axis.

A. Hole states in the quantum dot

In order to find the energy spectra of holes, trapped in the quantum dot, we solve self-consistently the Poisson equation

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial U(r,z)}{\partial r} \right) + \frac{\partial^2 U(r,z)}{\partial z^2} = -\frac{\rho_h(r,z)}{\varepsilon_0 \varepsilon_i},
\]

where \( \rho_h(r,z) = e n_h(r,z) \) and \( \varepsilon_i \) are the hole charge density and dielectric constant of the material, respectively, and the Schrödinger equation, which governs the hole motion,

\[
\frac{\hbar^2}{2} \left[ \frac{1}{m_j(z)} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{1}{m_j(z)} \frac{\partial^2}{\partial \varphi^2} + \frac{1}{m_j(z)} \frac{\partial^2}{\partial z^2} \right] \Psi_{j,s,n,m}(r,\varphi,z) + V(r,z) \Psi_{j,s,n,m}(r,\varphi,z) = E_{j,s,n,m} \Psi_{j,s,n,m}(r,\varphi,z),
\]

Here the index \( j \) labels the hole band type: \( j = 1 \) for a hole which is heavy for motion along the \( z \) axis and light in the plane of the GaAs layer and \( j = 2 \) for a hole which is light for motion along the \( z \) axis and heavy in the plane of the GaAs layer. For the case of a quantum well the light and heavy holes exactly decouple at \( |k_i| = 0 \). Since the lateral size of the dot is much larger than the GaAs unit cell, only states around \( |k_i| = 0 \) contribute to the wave functions in the dot. The effect of coupling is very weak in the close vicinity of the center of the Brillouin zone \( k_i = 0 \) (see, e.g., Refs. 17 and 18). The index \( s \) labels subbands due to the size quantization of the hole motion along the \( z \) axis. The index \( n \) is the radial quantum number and \( m \) is the angular quantum number of the lateral (in the plane of the GaAs layer) motion of the hole. Since the holes are accumulated in a rather thin layer near the surface, the hole motion along the \( z \) axis is expected to be faster than the lateral motion. Hence the adiabatic approach can be used: i.e., it is assumed that the lateral motion of a hole takes place in the potential formed by the fast motion along the \( z \) axis. Therefore, the hole wave function

---

**FIG. 1.** Scheme of the tip/insulator/GaAs/\( \text{Al}_{0.25}\text{Ga}_{0.75}\text{As}/\text{quantum well} \) structure. The shown calculated hole density distribution corresponds to the STM-tip voltage −3.1 V and the radius of the tip, 20 nm. 

---
can be represented as a product: \( \Psi_{j,s,n,m}(r, \varphi, z) = \Psi_{j,s,n,m}^r(z; r) e^{im\varphi} \), where \( \Psi_{j,s,n,m}^r(z; r) \) and \( \Psi_{j,s,n,m}^s(r)e^{im\varphi} \) are the wave functions of the transverse (along the \( z \) axis) and lateral motion, respectively. The Schrödinger equation governing the transverse motion of a hole reads

\[
- \frac{\hbar^2}{2m_j} \frac{\partial^2}{\partial z^2} \Psi_{j,s}^r(z; r) + V(r, z) \Psi_{j,s}^r(z; r) = E_{j,s}(r) \Psi_{j,s}^r(z; r),
\]

where \( V(r, z) = eU(r, z) + V_{\text{bar}}(z) \) with \( U(r, z) \) the electrostatic potential. \( V_{\text{bar}}(z) \) describes band offsets for a hole. We adopt the following rule for the band-gap energy difference between GaAs and Al\(_{0.25}\)Ga\(_{0.75}\)As: \( \Delta E_c/\Delta E_g = 67\% \) and \( \Delta E_c/\Delta E_g = 33\% \) (Ref. 19). So

\[
V_{\text{bar}}(z) = \begin{cases} 
0 \text{ meV}, & \text{if } z \in \text{GaAs layer}, \\
102 \text{ meV}, & \text{if } z \in \text{Al}_{0.25}\text{Ga}_{0.75}\text{As layer}, \\
\infty, & \text{if } z \in \text{sulfur layer}.
\end{cases}
\]

Where solving the Schrödinger equation (3), the total potential energy \( V(r, z) \) is modeled by a piecewise linear function\(^\text{10}\)

\[
V_{\text{p}}(r, z) = -\left[A_i(r) + a_i(r)z\right], \quad z_{l-1} < z < z_l, \quad l = 1, \ldots, n_z,
\]

where \( z_l \) are the nodes of the partition of the segment \([0, L] = \sum z_l \) in \( n_z \) parts. The functions \( A_i(r) \) and \( a_i(r) \) are determined by

\[
a_i^2(r) = -\frac{V(r, z_l) - V(r, z_{l-1})}{z_l - z_{l-1}}, \\
A_i(r) = \frac{V(r, z_l)z_l - V(r, z_{l-1})z_{l-1}}{z_l - z_{l-1}}.
\]

In the interval \((z_l, z_{l-1})\) and at a fixed value of \( r \), the equation for the transverse wave function \( \Psi_{j,s}^r(z; r) \) transforms into

\[
\frac{\partial^2}{\partial x_l^2} \Psi_{j,s}^r(x_l) + x_l \Psi_{j,s}^r(x_l) = 0,
\]

where

\[
x_l = - \left[ x + \frac{e + A_i^2}{a_i^2} \right] (a_i^2m_j^2)^{1/3},
\]

\[
a_i^2 = \frac{2m_j^2}{h^2}a_i, \quad A_i^2 = \frac{2m_j^2}{h^2}A_i, \quad e = \frac{2m_j^2}{h^2}E(r).
\]

A general solution of this equation is

\[
\Psi_{j,s}^r(x_l) = c_i^1 \text{Ai}(x_l) + c_i^2 \text{Bi}(x_l),
\]

where \( \text{Ai}(x) \) and \( \text{Bi}(x) \) are Airy functions.\(^\text{21}\) Using the condition of continuity of the wave function and of its derivative at the nodes, we obtain a homogeneous set of equations for the coefficients \( c_i^j \):

\[
c_i^1 \text{Ai}((m_j^2)^{1/3}x_l^0) + c_i^2 \text{Bi}((m_j^2)^{1/3}x_l^0) = 0,
\]

\[
c_i^1 \text{Ai}((m_j^2)^{1/3}x_l^{0}) + c_i^2 \text{Bi}((m_j^2)^{1/3}x_l^{0}) = 0,
\]

\[
- c_i^{l+1} \text{Ai}((m_j^2)^{1/3}x_l^{0}) - c_i^{l+1} \text{Bi}((m_j^2)^{1/3}x_l^{0}) = 0,
\]

\[
- c_i^{l+1} \text{Ai}((m_j^2)^{1/3}x_l^{0}) - c_i^{l+1} \text{Bi}((m_j^2)^{1/3}x_l^{0}) = 0,
\]

\[
- c_i^{n} \text{Ai}((m_j^2)^{1/3}x_l^{0}) + c_i^{n} \text{Bi}((m_j^2)^{1/3}x_l^{0}) = 0.
\]

Each of the energies \( E_{j,s}(r) \) determines the top of a hole subband in the multilayer structure and plays the role of an adiabatic potential for the lateral motion.

Consequently, the equation, which describes the motion along the \( r \) coordinate, is as follows:

\[
- \frac{\hbar^2}{2m_j^2(z)} \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) - \frac{m_j^2}{r^2} \right] \Psi_{j,s,n,m}^s(r) + E_{j,s}(r) \Psi_{j,s,n,m}^s(r) = E_{j,s,n,m}^s(0).
\]

Our numerical approach to solving the Schrödinger equation, which governs the lateral motion of the holes, is to model the effective potential \( E_{j,s}(r) \) by a steplike ring function

\[
U_{j,s}^k(r) = \frac{E_{j,s}^k + E_{j,s}^{k-1}}{2}, \quad r_{k-1} < r < r_k, \quad k = 1, \ldots, n_r,
\]

where \( r_k \) are the nodes of the partition of the segment \([0, R_H]\) in \( n_r \) parts and \( E_{j,s}^k = E_{j,s}(r_k) \). In the interval \((r_{k-1}, r_k)\) and at a fixed value of \( r \), the equation for the wave function \( \Psi_{j,s,n,m}(r) \) takes the form

\[
- \frac{\hbar^2}{2m_j^2(z)} \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) - \frac{m_j^2}{r^2} \right] \Psi_{j,s,n,m}^s(r) + E_{j,s}(r) \Psi_{j,s,n,m}^s(r)
\]

\[
= E_{j,s,n,m}^s(0).
\]
where \( r_{k-1} < r < r_k, \ k = 1, \ldots, n_p \). A general solution of this equation is

\[
\Psi^{l,k}_{j,s,n,m}(r) = b^1_k A_m(\sqrt{\lambda_k} r) + b^2_k B_m(\sqrt{\lambda_k} r)
\]

\[
= \begin{cases} 
\lambda_k^{-1} J_m(\sqrt{\lambda_k} r) + b^2_k Y_m(\sqrt{\lambda_k} r), & \text{if } \lambda_k > 0, \\
\frac{b^1_k}{\lambda_k} J_m(\sqrt{\lambda_k} r) + b^2_k K_m(\sqrt{\lambda_k} r), & \text{if } \lambda_k < 0,
\end{cases}
\]

(15)

where \( J_m(r) \) and \( Y_m(r) \) are Bessel functions of integer order \( m \), and \( I_m(r) \) and \( K_m(r) \) are modified Bessel functions of integer order \( m \) (Ref. 21). Here

\[
\lambda_k^l = \frac{2m_j^l}{\hbar^2} \left( E_{j,s,n,m} - \frac{E^k_j + E_j^{k-1}}{2} \right).
\]

(16)

Using the condition of continuity of the wave function and of its derivative at the nodes, we obtain a homogeneous set of equations for the coefficients \( b^l_k \), which reduces to

\[
\begin{pmatrix} b^1_{n+1} \\ b^2_{n+1} \end{pmatrix} = \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix} \begin{pmatrix} b^1_n \\ 0 \end{pmatrix}.
\]

(17)

Here the transmission matrix is defined by

\[
\hat{S} = \prod_{k=1}^{n_p} \hat{W}^{-1}(r_k) \hat{W}(r_{k-1}),
\]

where

\[
\hat{W}(r_j) = \begin{pmatrix} A_m(\sqrt{\lambda^l_k} r_j) & B_m(\sqrt{\lambda^l_k} r_j) \\ \sqrt{\lambda^l_k} A'_m(\sqrt{\lambda^l_k} r_j) & \sqrt{\lambda^l_k} B'_m(\sqrt{\lambda^l_k} r_j) \end{pmatrix},
\]

(19)

\[
\hat{W}^{-1}(r_j) = \begin{pmatrix} \frac{\sqrt{\lambda^l_k}}{m_j} B'_m(\sqrt{\lambda^l_k} r_j) & -B_m(\sqrt{\lambda^l_k} r_j) \\ -\frac{\sqrt{\lambda^l_k}}{m_j} A'_m(\sqrt{\lambda^l_k} r_j) & \frac{\sqrt{\lambda^l_k}}{m_j} A_m(\sqrt{\lambda^l_k} r_j) \end{pmatrix},
\]

(20)

and

\[
\Delta_k^l = \begin{cases} \frac{2}{m_j^l \pi r_k}, & \text{if } \lambda_k^l > 0, \\
\frac{1}{m_j^l r_k}, & \text{if } \lambda_k^l < 0.
\end{cases}
\]

(21)

The secular equation corresponding to the above set of equations yields the energy spectrum of holes, confined in the quantum dot.

Given a particular number of holes in the quantum dot, we find the quasi-Fermi level \( E^F_h \) for holes from the equation

\[
N_h = \sum_{j=1,2} \sum_{s,n,m} f_h(j,s,n,m,E^F_h).
\]

(22)

where \( f_h(j,s,n,m,E^F_h) \) is the probability of occupation by a hole of the quantum state \((j, s, n, m)\). Then, within the Hartree approximation scheme, the hole charge density may be written as

\[
\rho_h(r,z) = e \sum_{j=1,2} \sum_{s,n,m} |\Psi_{j,s,n,m}(r,\varphi,z)|^2 f_h(j,s,n,m,E^F_h).
\]

(23)

This is the basic formula employed to invoke a self-consistent solution to the Schrödinger and Poisson equations.

The procedure, involved to calculate the hole states and the corresponding electrostatic potential, is divided into three steps. At the first step, the electrostatic potential \( U(r,z) \) in the whole structure is calculated in the absence of holes. Then the obtained potential is used as the initial guess in the iteration procedure. In order to solve the Poisson equation numerically we choose to work in the domain \( \Omega = [0, l_1 + l_2 + l_3] \times [0, R_H] \) with the following boundary conditions:

\[
\frac{\partial U(r,z)}{\partial r} \bigg|_{r=0} = 0, \quad U(r,z) \bigg|_{r=R_H} = 0,
\]

(24)

\[
U(r,0) = V_{\text{tip}} \quad \text{for } r \in [0,R_{\text{tip}}], \quad U(r,l_1+l_2+l_3) = 0.
\]

(25)

The potential at the interface between the Al_{0.25}Ga_{0.75}As barrier and In_{0.25}Ga_{0.75}As quantum well, which has a high concentration of charge carriers, is assumed to be equipotential with \( U = 0 \). The mesh on the domain \( \Omega \) is defined as follows:

\[
\Omega_{ij} = \{(r,z) \in \Omega \left| r_i = \sum_{k=1}^{i} h_p(k), \quad z_j = \sum_{k=1}^{j} h_z(k), \quad 0 \leq i \leq n_p, \quad 0 \leq j \leq n_z \right. \}.
\]

(26)
which establishes the measure of the accuracy. When increasing the number of holes in the quantum dot, the electrostatic potential, obtained earlier for a smaller number of holes, is used as an initial guess for calculations.

We introduce the value \( N_h^{(0)} \), which is the maximal number of holes, trapped in the tip-induced quantum dot, at a given potential of the STM tip. The value \( N_h^{(0)} \) is the largest integer number (the “equilibrium” number of holes), which satisfies the following inequality:

\[
E(N_h^{(0)}) \leq E(N_h^{(0)} - 1) + E_n(1),
\]

where \( E_n(1) \) is the energy of one nontrapped hole and \( E(N_h^{(0)}) \) is the energy of \( N_h^{(0)} \) trapped holes.

**B. Electron states in the quantum dot**

Given the electrostatic potential formed by the hole charge, we solve the Schrödinger equation for the electrons:

\[
-\frac{\hbar^2}{2m_e^L(z)} \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \psi_e(z,r)}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \psi_e(z,r)}{\partial \varphi^2} \right)
+ V(z,r) \psi_e(z,r)
= E_{e,s_e,n_e,m_e} \psi_e(z,r),
\]

The index \( s_e \) labels subbands due to the size quantization of the electron transverse motion. The index \( n_e \) is the radial quantum number and \( m_e \) is the angular quantum number of the lateral motion of the electron. The electron transverse motion is faster than the lateral motion; consequently, we use the adiabatic approach similarly to the above case of holes. The electron wave function can be represented in a product form \( \psi_e(z,r) = \psi_{e,s_e,n_e,m_e}(z) \psi_{e,s_e,n_e,m_e}(r) \), where \( \psi_{e,s_e,n_e,m_e}(z) \) and \( \psi_{e,s_e,n_e,m_e}(r) \) are the wave functions of the transverse and lateral motion, respectively. The Schrödinger equation governing the transverse motion of an electron is

\[
-\frac{\hbar^2}{2m_e^L(z)} \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \psi_e(z,r)}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \psi_e(z,r)}{\partial \varphi^2} \right)
+ V(z,r) \psi_e(z,r)
= E_{e,s_e}(z) \psi_e(z,r),
\]

where \( V(z,r) = -eU(z,r) + V_{ba}(z) \). Here \( V_{ba}(z) \) describes band offsets for an electron. Consequently, the equation, which describes the motion along the \( r \) coordinate, is as follows:

\[
-\frac{\hbar^2}{2m_e^L(z)} \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \psi_e(z,r)}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \psi_e(z,r)}{\partial \varphi^2} \right)
+ V^e(z,r) \psi_e(z,r)
= E_{e,s_e}(r) \psi_e(z,r),
\]

In order to numerically solve the Schrödinger equation, which governs the lateral motion of the electrons, we enclose the domain of electron motion, which is much larger than the induced quantum dot, inside a cylinder of radius \( R_E \) and height \( l_2 + l_3 \), on the surface of which the wave functions are forced to vanish. The domain of numerical modeling, characterized by the radius \( R_E \), is chosen to be much larger than the radius of the contact area.

To calculate the electron wave function \( \psi_{e,s_e,n_e,m_e}(r) \), we express it as

\[
\psi_{e,s_e,n_e,m_e}(r) = \sum_l b_{l,m_e} e^{im_e \varphi} \frac{\sqrt{2} J_m^e \left( \frac{\mu_{l,m_e} R_E}{\rho} \right)}{R_E} (r),
\]

and finally obtain a set of linear equations for \( b_{l,m_e} \):

\[
\sum_l \left[ \left( \frac{\hbar^2}{2m_e^L(z)} \right)^2 E_{e,s_e,n_e,m_e} - \frac{\hbar^2}{2m_e^L(z)} \delta_{l,m_e} + v_{s_e,m_e,l} \right] b_{l,m_e} = 0,
\]

where

\[
v_{s_e,m_e,l} = \int_0^{R_E} \frac{dr}{R_E} \frac{2 J_m^e \left( \frac{\mu_{l,m_e} R_E}{\rho} \right) J_m^e \left( \frac{\mu_{l,m_e} R_E}{\rho} \right)}{J_m^{e*} \left( \frac{\mu_{l,m_e} R_E}{\rho} \right) J_m^e \left( \frac{\mu_{l,m_e} R_E}{\rho} \right)},
\]

The found set of energies \( E_{e,s_e,n_e,m_e} \) and wave functions \( \psi_{e,s_e,n_e,m_e}(r,\varphi,z) \) together with the set of hole energies \( E_{j,x,n,m} \) and wave functions \( \psi_{j,x,n,m}(r,\varphi,z) \) are used for the analysis of the electron-hole radiative recombination.

Let us estimate the average number of electrons in the proximity of the quantum dot. In the absence of current the number of electrons in this region can be described by the following differential equation:

\[
\frac{dN_e^{(0)}}{dt} = g_{eh} N_e^{(0)} - \frac{N_e^{(0)}}{\tau_{dr}},
\]

where \( g_{eh} \) is the rate of thermal excitations of electron-hole pairs. The time \( \tau_{dr} \) characterizes the electron drift away from the quantum dot in the absence of current. In a stationary situation we obtain

\[
N_e^{(0)} = g_{eh} \tau_{dr}.
\]

When a current \( I_{up} \) is turned on, the time evolution of the number of electrons in the proximity of the quantum dot is given by
Hence, in a stationary situation, we obtain

$$\Delta N_e = \frac{I_{\text{tip}} \tau_{\text{dr}}}{e},$$

(38)

where $\Delta N_e = N_e - N_e^{(0)}$. The characteristic $\tau_{\text{dr}}$ appears to be of the order of 0.5 ps (Ref. 22). Consequently, the average number of additional electrons due to the current through the STM tip $I_{\text{tip}} \approx 10$ nA, is of the order of $10^{-3}$. Since the experiments are performed at $T=4.2$ K, also the average number of electrons in the absence of current, $N_e^{(0)}$, is small ($\sim 10^{-3}$). It is obvious that the probability to find simultaneously two or more electrons in the proximity of the quantum dot is negligible and many-electron effects play no role.

IV. INTENSITY OF THE ELECTROLUMINESCENCE

The intensity of the electroluminescence at a frequency $\Omega$ in the quantum dot created by the STM tip can be described by the following expression: 17

$$I(h\Omega) \sim \sum_{j,s,n,m} \sum_{s_e,n_e,m_e} f_h(j,s,n,m,E_F^h) f_e(s_e,n_e,m_e) \times P(j,s,n,m,s_e,n_e,m_e,h\Omega),$$

(39)

where $f_e(s_e,n_e,m_e)$ is the probability of occupation by an electron of the quantum state $(s_e,n_e,m_e)$. The probability of electron-hole recombination,

$$P(j,s,n,m,s_e,n_e,m_e,h\Omega) \sim \left| O_{j,s,n,m,s_e,n_e,m_e} \right|^2 \delta(E_{j,s,n,m} + E_{e,s_e,n_e,m_e} + E_h - h\Omega),$$

(40)

is proportional to the squared modulus of the overlap integral of the electron and hole wave functions:

$$O_{j,s,n,m,s_e,n_e,m_e} = \int \Psi_{j,s,n,m}(r,\varphi,z) \Psi^*_{e,s_e,n_e,m_e}(r,\varphi,z)$$

$$= \delta_{m,m_e} \int d\rho d\varphi \Psi^*_{j,s,n,m}(r)$$

$$\times \sum_{l_e} b_{l_e,m_e} R_{E,l_e,m_e} \left( \mu_{l_e,m_e} \rho \right)$$

$$\times \int dz \Psi^*_{j,s,n,m}(z;r) \Psi_{e,s_e,n_e,m_e}(z;r).$$

(41)

For obtaining the stationary distribution functions $f_h(j,s,n,m,E_F^h)$ and $f_e(s_e,n_e,m_e)$ in the presence of a non-zero tunneling current through the tip, in the case of a rigorous consideration, it is necessary to consider a nonequilibrium situation. However, since the current through the tip is weak and the radiative lifetime of holes is expected to be much longer than the time of relaxation between the hole energy levels, we assume that the distribution functions $f_h(j,s,n,m,E_F^h)$ can be described by the Fermi-Dirac function with quasi-Fermi level for holes $E_F^h$:

$$f_h(j,s,n,m,E_F^h) = \frac{1}{\exp \left( \frac{E_{j,s,n,m} - E_F^h}{kT} \right) + 1}.$$  

(42)

The energies of electrons injected from the tip into the GaAs layer are extremely high ($\sim 2.5 \text{--} 3.5$ eV with respect to the bottom of the conduction band in the GaAs layer). On the other hand, our estimations show (see next section) that the recombination probability, Eq. (40), takes appreciable values only for electrons whose energies lie within a relatively narrow interval with a width of $\sim 150$ meV. We assume that as a result of the energy relaxation of electrons injected from the tip, the occupation probability does not depend on the energy of an electron.

The electrons injected in the GaAs layer from the STM tip have zero angular momentum $m_e$. After injection the electron can further acquire a nonzero angular momentum $m_e$ by scattering. However, the scattering probability decreases with increasing the kinetic energy of an electron due to a radial accelerating electric field. With strengthening this field, the radial electron motion approaches the ballistic regime. Therefore, we assume that due to a strong radial accelerating field, the occupation probability for states with the angular quantum number $|m_e|>0$ is negligible as compared to that for $m_e=0$:

$$f_e(s_e,n_e,m_e) \sim \delta_{m,0}.$$  

(43)

We will also consider the opposite limiting case when due to a strong scattering the electron motion is rather diffusive than ballistic so that the decrease of $f_e$ with increasing $|m_e|$ is negligible.

Let us analyze how change of the energy of the Coulomb interaction between holes due to the electron-hole recombination (the electrostatic potential for the remaining holes) affects the electroluminescence spectrum of the dot. Transition energies can be represented in the form

$$\Delta E = \tilde{E}(N_h,1_e) - \tilde{E}(N_h-1,0_e),$$

(44)

where $\tilde{E}(N_h,1_e)$ is the energy of the system, which consists of $N_h$ interacting holes and one electron, and $\tilde{E}(N_h-1,0_e)$ is the energy of the system after the electron-hole recombination. These terms are expressed as follows:

$$\tilde{E}(N_h,1_e) = E(N_h) + E_g + E_e + \Delta,$$

(45)

where $\Delta$ describes the Coulomb energy due to the interaction between $N_h$ holes and one electron,

$$\tilde{E}(N_h-1,0_e) = E(N_h-1) + E_F^h(N_h) - E_h,$$

(46)

where $E_F^h(N_h)$ is the quasi-Fermi level of $N_h$ holes and $E_h$ is the energy of the hole, involved in the radiative recombination. Substituting Eqs. (45) and (46) into Eq. (44), we obtain
\[ \Delta E = \Delta e + \eta, \]  
\[ \Delta e = E_h + E_g + E_e \]  
\[ \eta = E(N_h) - E(N_h - 1) - E_h^0(N_h) + \Delta. \]

The quantity \( \eta \) describes a correction to the transition energy due to the recombination-induced change of the electrostatic potential for the remaining holes. As a result of the electron-hole recombination, the total charge in the quantum dot domain does not change and, moreover, the charge density distribution changes only slightly. Therefore, the quantity \( \eta \) is negligible as compared to the transition energy.

V. RESULTS AND DISCUSSION

The experimental sample is characterized by the following parameters. The thicknesses of the GaAs and Al\(_{0.25}\)Ga\(_{0.75}\)As layers are, respectively, \( l_2 = 17 \) nm and \( l_3 = 46 \) nm. The dielectric constants of these layers are \( \varepsilon_2 = 13.2 \) and \( \varepsilon_3 = 4.2 \). In our model we consider two representative sets of parameters, which characterize the insulator layer: sulfur case, where the insulator layer width \( l_1 = 0.25 \) nm and the dielectric constant \( \varepsilon_1 = 4 \) (surface of the GaAs cap is covered with one monolayer of Ga-bound S [Ref. 16]), and vacuum case, where \( l_1 = 0.5 \) nm, \( \varepsilon_1 = 1 \) (surface of the GaAs cap is separated from the STM tip by a vacuum barrier). The hole masses are

\[ m_1^+ = \frac{m_0}{\gamma_1 - 2 \gamma_2}, \quad m_2^+ = \frac{m_0}{\gamma_1 + 2 \gamma_2}, \]

with the Luttinger parameters for Al\(_{x}\)Ga\(_{1-x}\)As Ref. 17. Here \( m_0 \) is the free electron mass. The electron effective mass is taken as follows:

\[ m_e = (0.067 + 0.083 x) m_0. \]

The work functions of a pure Pt and GaAs are approximately the same within uncertainties due to faceting and contaminations.\(^{23}\) Therefore, we assume the difference between those work functions to be zero in our numerical calculations.

The calculations were performed with the following parameters of our model: \( R_H = 200 \) nm, \( R_E = 1000 \) nm, \( l = 1, \ldots, 1000 \) [see Eq. (32)], \( n_p = 800 \), and \( n_z = 200 \). When calculating the electroluminescence spectra, we have replaced the \( \delta \) function in Eq. (40) by a Lorentzian function with the half width \( \Gamma = 5 \) meV. This allows us to smooth out electroluminescence intensity oscillations, related to a finite size of the domain of numerical modeling of the electron motion, characterized by \( R_E \). The results of numerical calculations are presented in Figs. 1–8.

Figure 2 illustrates the profile of the electrostatic potential induced by the STM tip with the radius of the contact area, \( R_{tip} = 20 \) nm, at the tip voltage \( V_{tip} = -3.1 \) V. It is clearly seen that the hole charge accumulated in the quantum dot does not completely screen the electrostatic potential in the depth of the GaAs layer. The inset demonstrates the corresponding density of the holes, trapped by the STM-tip-induced quantum dot. The charge of holes is concentrated in a relatively thin layer near the interface insulator/GaAs, with
a lateral size that is almost equal to the tip diameter. The thick solid line shows the potential profile at the interface insulator/GaAs.

Figure 3 displays the “equilibrium” number of holes, \(N_h^{(0)}\), in the quantum dot, calculated using Eq. (27), as a function of the squared radius of the contact area (solid lines) and of the voltage applied between the STM tip and semiconductor structure (dashed lines). The “equilibrium” number of holes is approximately proportional to the voltage applied and to the contact area of the STM tip. Qualitatively, this result is in agreement with what we would expect from the classical theory of capacitance (linear increase in charge upon an incremental change of the applied voltage or the area of the contact between the STM tip and semiconductor structure).

In Fig. 4 we present (a) the typical experimentally observed electroluminescence spectrum of the tip-induced quantum dot; (b) the electroluminescence spectrum calculated for the voltage on the STM tip, \(V_{\text{tip}} = -3.1\) V, and the radius of the contact area, \(R_{\text{tip}} = 16\) nm, in the case of a negligibly weak dependence of \(f_e\) on \(m_e\); (c) the electroluminescence spectrum calculated for the voltage on the STM tip, \(V_{\text{tip}} = -3.1\) V, and the radius of the contact area, \(R_{\text{tip}} = 16\) nm, in the case of the electron distribution function given by Eq. (43). We see that the electroluminescence spectrum in panel (c) is much closer to the experimentally observed one, which supports our assumption that the electron distribution function rapidly decreases with increasing \(m_e\). Therefore we will use the electron distribution function in the form (43) in our further calculations.

Figure 5 shows the calculated positions of the electroluminescence peaks as a function of the number of holes, trapped in the quantum dot. The insets of these figure show the calculated electroluminescence spectra of the tip-induced quantum dot at the voltage on the STM tip, \(V_{\text{tip}} = -3.1\) V, and the radius of the contact area, \(R_{\text{tip}} = 20\) nm. The peaks in these spectra are marked with series of letters. It can be seen that positions of the electroluminescence peaks, attributed to the electron-hole recombination in the quantum dot, are very sensitive to the number of holes, trapped in the quantum dot. An increase of the number of holes in the quantum dot leads to a greater transition energy between hole and electron levels (blue shift of the electroluminescence peaks). The reason for such behavior is the fact that an increase of the number of holes in the quantum dot results in a narrowing of the quantum well for holes and in a widening quantum well for electrons in the \(z\) direction. This pushes both hole and electron levels down. For holes, the potential well in the \(z\) direction is much narrower than that for electrons. Correspondingly, the downward shift of energy levels is more pronounced for holes than for electrons.

An examination of different terms entering Eq. (39) shows that for the structure with the vacuum barrier, the lowest hole states \((j = 1, s = 1, n = 1, 2, 3)\) and the electron states in the energy range 850–1000 meV give the main contribution to the electroluminescence spectrum.

Figure 6 illustrates the position of the peaks in the electroluminescence spectrum as a function of the voltage \(V_{\text{tip}}\) applied between the STM tip and the semiconductor struc-
increasing transition energies

For instance, from

Indeed, when increasing the radius of the contact area, for

than the increase of the energy of the recombining electron.

wave function. The upward shift of the hole levels is smaller

order to have a considerable overlap integral with a hole

result in a rising potential barrier for electrons and a slight

shifts the conduction and valence bands upwards. This re-

explained by the fact that an enlargement of the contact area

when changing the voltage on the STM tip, for instance,

from $V_{\text{tip}} = -1.9$ to $-3.1$ V, the hole and electron levels

responsible for peak $B_2$ in Fig. 6(c) shift from $-974$ to $-530$

meV and from $593$ to $892$ meV, respectively. Consequently,

the upward shift of the hole energy levels with increasing

$V_{\text{tip}}$ is larger ($444$ meV, in our example) than that for

electrons ($299$ meV, in our example). Thus these effects result in

a redshift in peak positions in the electroluminescence spec-

trum.

In Fig. 7 we present the peak positions for the spectra

shown in the insets of Fig. 5 as a function of the radius of the

contact area between the STM tip and semiconductor struc-

ture at 4.2 K. Panel (a) corresponds to the experimentally

observed peaks. Panels (b) and (c) display numerical results

obtained in the sulfur and vacuum cases, respectively. Clearly,
in agreement with experiment, the peak positions as a

function of $V_{\text{tip}}$ reveal a redshift, which can be explained as

follows. An increase of the voltage shifts the bottom (top) of

the dot (antidot) upwards nearly proportionally to the applied

voltage. The width of the dot in the $z$ direction is reduced due
to a stronger screening. Hence an increase of $V_{\text{tip}}$ leads to a

depening of the potential well for holes and to a rise of the

barrier for electrons. This, in its turn, raises the energy of

the relevant electron state with respect to the bottom of the con-
duction band in the absence of applied voltage and shifts the

hole levels also upwards. This is because the rise of the hole

energy level due to the shift of the bottom of the dot is larger

than its lowering due to the stronger confinement. Indeed,

when changing the voltage on the STM tip, for instance,

from $V_{\text{tip}} = -1.9$ to $-3.1$ V, the hole and electron levels

responsible for peak $B_2$ in Fig. 6(c) shift from $-974$ to $-530$

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$V_{\text{tip}}$ is larger ($444$ meV, in our example) than that for

electrons ($299$ meV, in our example). Thus these effects result in

a redshift in peak positions in the electroluminescence spec-

trum.

In the experiment, the STM feedback system increases the

electron tunneling current through the tip by moving the tip

forward. Since the tip is already in contact with the sample,

this leads to a flattening of the STM tip and, consequently, to

an enlargement of the contact area. Hence the experimentally

observed blueshift of the electroluminescence peaks with in-

creasing tunneling current can be attributed to an enlarge-

ment of the contact area. From the above analysis, we can

conclude that the applied STM-tip voltage governs the shift

of the peaks through the confinement along the symmetry

axis of the tip, while the tunneling current does this through

confinement along the $r$ coordinate.

In our model, we assume that the contact area is propor-
tional to the tunneling current:

$$I_{\text{tip}} = \alpha \pi R_{\text{tip}}^2,$$

where $\alpha$ is treated as a fitting parameter. The calculations are

performed for $\alpha = 0.0025$ nA/nm$^2$. Figure 8 displays experi-

mentally observed and calculated positions of the peaks in the

electroluminescence spectrum as a function of the tunnel-

ing current. From the panel (b) of this plot we can see that the
tendency in the behavior of the peak positions as a
A number of experimental parameters are unknown precisely, so it is of interest to consider the stability of the effects, calculated in this work, with respect to variations of these parameters. For example, from the above analysis we see that an increase of the effective thickness of the insulator $l_1/\varepsilon_1$ weakens the effect of tunneling current $I_{\text{tip}}$ on the electroluminescence peak positions. Similarly, the influence of the voltage $V_{\text{tip}}$ on the peak positions becomes less pronounced with increasing $l_1/\varepsilon_1$. At the same time, the direction of the electroluminescence peak shift with varying $I_{\text{tip}}$ or $V_{\text{tip}}$ does not depend on a particular choice of $l_1/\varepsilon_1$. A number of simulations prompt us to the conclusion that also a particular choice of the shape of the STM tip does not influence this direction. We have to distinguish between the “intrinsic” properties (i.e., properties that are not influenced by parameter choices) and “extrinsic” properties (which are influenced by parameter values) of the model. A blueshift of electroluminescence peaks with increasing $I_{\text{tip}}$, as well as a redshift of these peaks with increasing $V_{\text{tip}}$, can be considered as intrinsic properties of the tip-induced quantum dot, while the absolute peak positions as well as the absolute values of the electroluminescence-peak shifts with varying $I_{\text{tip}}$ or $V_{\text{tip}}$ are extrinsic properties.

VI. CONCLUSIONS

In order to describe the hole states that are confined in the quantum dot induced by an STM tip in the GaAs surface layer of a GaAs/Al$_{0.25}$Ga$_{0.75}$As multilayer structure, we have developed a model based on a self-consistent solution of the

<table>
<thead>
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<th>Transition energy</th>
<th>$B_1$</th>
<th>$B_2$</th>
<th>$B_3$</th>
<th>$B_4$</th>
<th>$B_5$</th>
<th>$B_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>without exchange corrections (meV)</td>
<td>1390</td>
<td>1406</td>
<td>1425</td>
<td>1453</td>
<td>1465</td>
<td>1493</td>
</tr>
<tr>
<td>with exchange corrections (meV)</td>
<td>1390</td>
<td>1410</td>
<td>1428</td>
<td>1453</td>
<td>1464</td>
<td>1494</td>
</tr>
</tbody>
</table>
Poisson and Schrödinger equations within the framework of the Hartree approximation. The hole states are obtained as a function of the voltage applied to the STM tip and the geometric and material parameters of the system under the investigation. We consider the influence of the electrostatic potential formed by the STM tip and the charge distribution of the trapped holes on the electron states. The electroluminescence spectra of the STM-tip-induced quantum dot are calculated as a function of the applied voltage and of the tunneling current through the tip. We have shown that the applied STM-tip voltage affects the electroluminescence peaks, positions through confinement along the growth direction of the semiconductor structure, while the tunneling current does this through lateral confinement. An increase of the voltage on the STM tip results in a redshift of the electroluminescence peaks. In contrast, an increase of the tunneling current shifts the electroluminescence peaks towards higher energies. Both these results are in agreement with experiments, demonstrating vast possibilities of tuning optical characteristics of the STM-tip-induced quantum dot by varying parameters like the applied voltage and tunneling current.

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As has been mentioned above, the energies of electrons injected from the tip are high. In order to recombine with holes, electrons have to relax to the energy interval, where their wave functions have considerable overlap integration with hole wave functions. In the course of nonradiative relaxation, electrons create a large number of phonons and heat up the domain of the structure, where the quantum dot is formed. It is of interest to investigate the effect of this heating on the electroluminescence peaks. The numerical simulation has shown that rising the effective hole temperature from 4.2 K until 77 K leads to an increase in the hole number $N_h^{(0)}$ trapped in the quantum dot. This increase in its turn leads to the blueshift of the electroluminescence peaks ($\sim 4-5$ meV). This result implies that if we take into account the effect of heating, caused by the electron current, on the distribution of holes in the quantum dot, these—more advanced—calculations would give a somewhat more pronounced blueshift of the electroluminescence peaks with increasing current.