Electroluminescence and injection of spin-polarised holes in InAs/GaAs quantum dot heterostructures

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There is a common belief that low minor carrier injection efficiency is inherent to Schottky barriers (SBs). In this work we show that the Schottky diode (SD) based on an InAs/GaAs quantum-size heterostructure (HS) can serve as a good light source. The injection of minority carriers can be achieved by applying a sufficiently high direct bias to the SB and the efficiency can be increased by placing a thin (~ 1 nm) oxide layer between the metal and the semiconductor. Using a ferromagnetic metal (FM) for the SB and applying a sufficiently strong magnetic field, it is possible to obtain a significant degree of circular polarisation of the emitted light.

During the last years, reverse biased SBs were used as a tool for studies of the spin injection from the ferromagnetic metal to the semiconductor [1, 2]. In such structures the injection occurs via tunneling through the barrier that can be made rather thin by using a high doping level of the semiconductor. Another type of SB structure is a forward biased diode where tunneling through the intermediate oxide layer can prevent spin coherence losses due to the conductivity mismatch problem [3].

The theory of the minor carriers' injection in silicon based MOS structures was developed in the 70s [4] and optimum conditions for achieving a high injection efficiency have been established. Such conditions for SBs based on III-V semiconductors may differ because of (i) a higher density of surface states owing to the lower interface quality, and (ii) smaller lifetime and diffusion length of the minority carriers. Nevertheless, a rather intense electro-luminescence (EL) from quantum dot (QD) and quantum well (QW) InAs/GaAs HSs with gold contacts has been demonstrated in our previous publications [5, 6]. It means that the limitations (i) and (ii) have been bypassed by using, respectively, the anode oxide layer and thin capping layers (i.e. placing the light-emitting region closer to the interface).

Owing to the high confinement potential, self-assembled QDs can emit, in principle, at room temperature. However, several studies revealed a temperature quenching effect in the QD photoluminescence (PL), taking place due to the carriers' escape from the quantum levels to the barriers where they can recombine non-radiatively. A simple way to solve this problem is to eliminate the non-radiative recombination centers (i.e. defects) in the GaAs barriers. PL intensity of the QD HSs described in this work does not show any thermal quenching thanks to some technological treatments used during the growth, decreasing the defect concentration in the QD and capping layers [7].

In this communication, we describe the results of our study of the EL properties of near surface InAs/GaAs QDs incorporated into a SD. We compare the experimental data to the results of our modelling of the electron and hole transport across the SB that determines the minority carriers' population in the QDs out of equilibrium and their radiative recombination as described in Ref. [8]. Based on this, we believe that the hole injection from the metal to the (*n*-type doped) semiconductor heterostructure occurs from the states in the vicinity of the Fermi level in the metal into the valence band states in the GaAs capping layer through the thin oxide barrier as shown in Fig. 1. We also demonstrate the possibility of injection of spin polarized holes from the FM contact into the QDs by measuring the degree of circular polarization of the light emitted by a Ni-InAs/GaAs SD placed into magnetic field of several Tesla in magnitude. Although the degree of circular polarization measured for the QD emission is lower than we have previously obtained for QW HSs [9], we hope that it can be improved in the future.

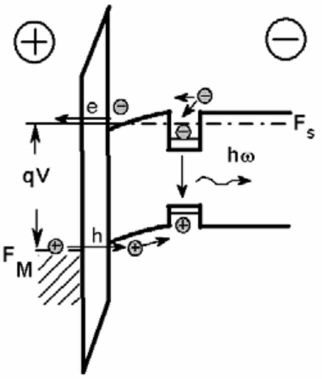

Fig.1 Schematic energy band diagram for the studied HS under applied bias of the order of 1.5 V.

Samples and measurements

We studied MOVPE grown InAs/GaAs HSs with self-assembled QDs covered with an InGaAs layer (10-30 nm in thickness) that plays the role of a QW. Thanks to the CCl4 treatment during the growth of the QD layer, the PL of these HSs does not show temperature quenching up to room temperature. Usual SDs were made by vacuum deposition of Au contacts. The temperature dependence of the EL was measured using the standard lock-in technique in a close cycle helium cryostat. Ni and combined Au-Ni-Au layers were used as ferromagnetic contacts. Circular polarised EL was measured at 4K in a helium bath cryostat in Faraday geometry with magnetic field up to 10 T. A quarter-wave plate in combination with a linear polarizer was used to select the EL with left and right circular polarizations. The signal was dispersed with a monochromator and detected with a liquid nitrogen-cooled InGaAs based photodiode array. The degree of circular polarization was calculated according to $\delta = (I_+ - I_-)/(I_- + I_+)$ where I_- and I_+ are the intensities of the left and right-hand polarised emission, respectively.

Experimental results

HSs show good luminescence properties, with the emission bands located at 1.0 -1.1 μm for the QW and at 1.3 -1.5 μm for the QDs. The current-voltage characteristics of similar diodes were described before [6] and allowed for the determination of the SB height (0.7 - 0.9 eV). The presence of the intermediate oxide provides the ideality factor $n \approx$ 1.3 - 1.5. The EL spectra (not presented here, see Ref. [5]) show several peaks corresponding to the electron-hole transitions to the ground and excited states in QDs. The first EL peak (corresponding to the ground state transition) is seen even at very low pump current values. The ratio between the second and first peaks' intensities changes very slowly with temperature. This implies that the second peak does not appear because of the saturation of the ground state in the dots, rather it can be manifest of the phonon bottleneck.

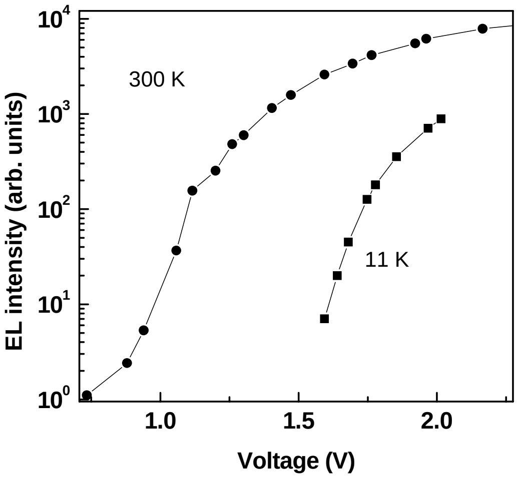

Fig. 2 Integrated intensity of the first EL peak *versus* applied voltage measured at 300 K and 11 K.

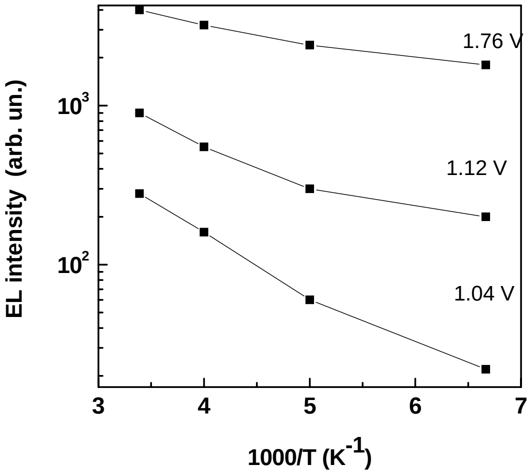

Fig. 3 Temperature dependence of the intensity of the first EL intensity measured for different bias values.

Fig. 2 shows the dependence of the integrated intensity of the first EL peak on applied bias for two temperatures. One can distinguish three different regions in the room temperature curve, with an exponential growth of the intensity with the bias in the middle (between 0.9 and 1.4 V) and a slower (power law) increase for smaller and larger values of the voltage. At the low temperature, the pre-exponential region is not observed. The temperature dependence of the EL intensity measured at a fixed bias also changes (Fig. 3) from an Arrenius-type law, characteristic of a bias of the order of 1 V, to a more moderate increase with temperature at high bias. These trends will be interpreted below in terms of our modelling results. SDs with FM contacts, of which Au-Ni-Au produces the best light emission properties, show a similar behavior. The exponential dependence of the EL intensity on the applied voltage (measured at 4K) changes to an approximately quadratic one above ≈ 1.5 V. It should be noted that decreasing the capping layer thickness, d_c, below 20 nm leads to a considerable decrease of the EL intensity, most likely due to the surface recombination. The shape of the EL spectra is also changing: the peaks corresponding to the optical transitions involving excited states become stronger.

Let us turn to the measurements performed in magnetic field. Sample with Au contact (considered as a reference diode) shows a slight domination of the left handed polarization in high magnetic fields ($\delta \approx$ -3% at 10 T, see Fig. 4). This behavior most likely is a consequence of the Zeeman splitting of the hole levels in the QDs. For the diodes with FM contacts, the strong magnetic field leads to the opposite, right-handed circular polarisation (positive value of δ in Fig. 4). We interpret this as the result of the injection of spin-polarised holes from the Schottky contact. This is confirmed by the fact that δ is higher for the second emission peak. Relaxation from the excited states to the ground states involves an extra process and, consequently, an additional loss of spin coherence. However, we have to note that the degree of circular polarisation of the EL of SDs with FM contacts in magnetic field shows a complex behavior that depends on several parameters, mainly on the capping layer thickness and applied voltage.

Calculation of the electron and hole currents

In order to calculate the electron and hole currents in the SD we applied the standard theory [10] to the system shown schematically in Fig. 1. This leads to the following expressions for the current densities:

$$j_e = -\frac{4\pi q m_e}{(2\pi\hbar)^3}\int_0^\infty \left[n_F(\varepsilon + q\varphi_b) - n_F(\varepsilon + q\varphi_b + qV)\right]F(\varepsilon)d\varepsilon \tag{1}$$

$$j_h = \frac{4\pi q m_h}{(2\pi\hbar)^3}\int_0^\infty \left[\tilde{n}_F(\varepsilon - q\varphi_b - qV) - \tilde{n}_F(\varepsilon - q\varphi_b + \Delta F_h)\right]F(\varepsilon)d\varepsilon \tag{2}$$

where m_e and m_h denote the electron and hole effective masses in the semiconductor, φ_b is the Schottky barrier height, V the applied bias, $n_F(\varepsilon)$ the Fermi function,

$$\tilde{n}_F(\varepsilon) = \left[1 + \exp\left(\frac{\varepsilon + E_g + E_F}{kT}\right)\right]^{-1},$$

E_F is the Fermi level in the semiconductor (determined by the doping level), E_g the band gap energy, $\Delta F_h = F_h - E_F$, F_h is the quasi-Fermi level for holes,

$$F(\varepsilon) = \int_0^\varepsilon D(E_z)dE_z$$

and $D(E_z)$ is the transparency of the trapezium-shape barrier formed by the oxide layer.

The applied bias is distributed between the oxide layer and the semiconductor. The distribution between the oxide and the semiconductor is obtained by solving the Poisson equation for the latter and matching the potential and its derivative at the interface. The sum of the potential drop on the semiconductor (V_1) and the *external* potential drop on the oxide (V_2) is equal to the applied voltage, $V_1 + (V_2 - V_2^{(0)}) = V$. This condition determines self-consistently φ_b and, through Eq. (1), the (majority) electron current. In principle, the quasi-Fermi level should be calculated self-consistently by considering the diffusion and recombination of the injected holes in the semiconductor. However, in the first approximation, the second term in the integral in Eq. (2) can be neglected, at least for large V. Then the hole current is completely determined by the tunneling through the barrier.

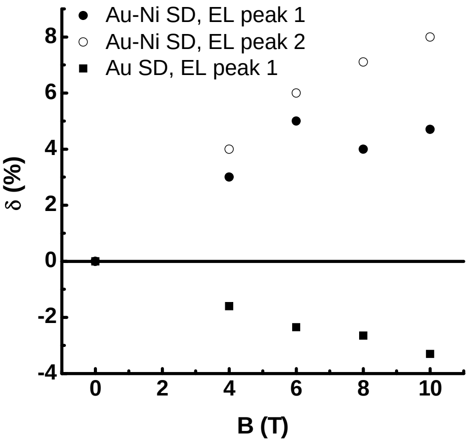

Fig. 4 Degree of circular polarization measured at 4K and bias of 1.58 V, versus magnetic field, for two SDs with different contacts. For the FM SD, data for the first and the second EL peaks (corresponding to the ground and exited states electron-hole states in the QDs) are presented.

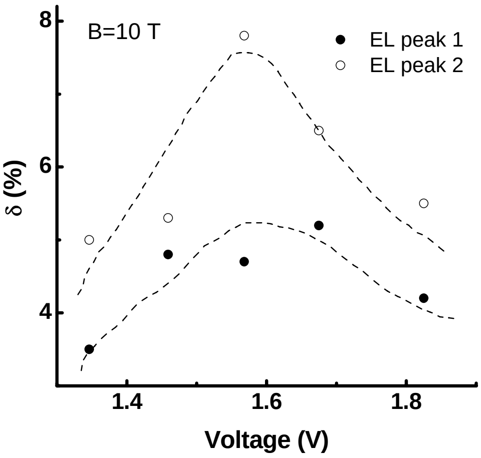

Fig. 5 Dependence of the degree of circular polarisation on bias at 10 T for two EL peaks of the same Au/Ni SD as in Fig. 4.

Figure 6 shows the calculated current densities for the electron and the holes in the structure with the oxide thickness of 2 nm. Note that the dependence of the hole current on the bias is qualitatively different from that of $j_e(V)$ (which is, in fact, the current-voltage characteristics of the structure). It can be seen that the exponential increase of the injection begins when the SB for the electrons disappears and the voltage drops mostly on the oxide layer (region II). At a bias of ≈ 1.5 V, the dependence $j_h(V)$ changes, that corresponds to the matching between the Fermi level in the metal and the top of the valence band in the semiconductor. Further increase of the injection rate occurs mostly

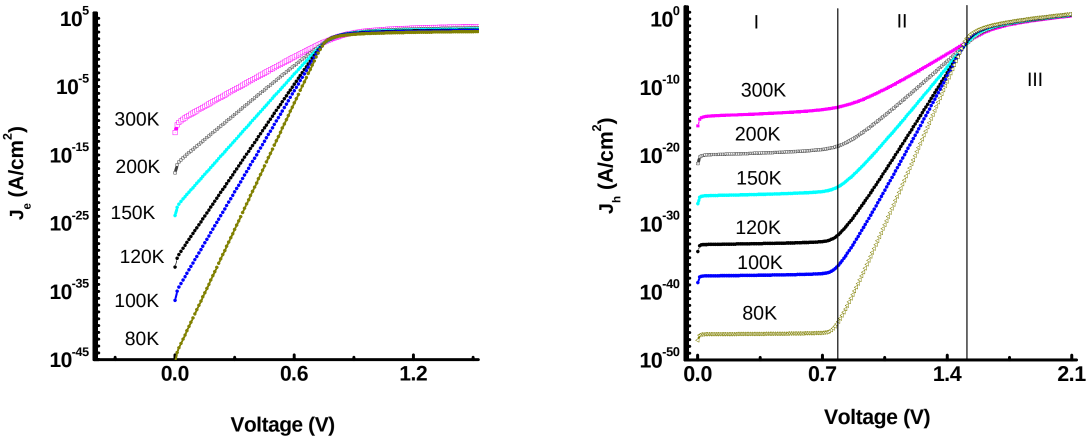

Fig. 6 Calculated electron (left) and hole (right) current densities versus voltage for different temperatures.

because of the increase of the number of states in the semiconductor, available for the tunneling. Analysis of the dependences calculated for different temperatures (Fig. 6) shows that the hole current grows exponentially with temperature in the bias region II, while it is almost temperature independent in the region III. Since the population of the QDs with holes (p_{QD}) under steady-state injection conditions must be proportional to j_h, we can say that the same trends should be characteristic of the QD emission intensity, $I_{EL} \propto p_{QD}/\tau_r$ where τ_r is the radiative recombination time that depends on E_F (we should note that not only τ_r but also the diffusion length affecting p_{QD} are temperature dependent and these effects, also in relation with the hole's spin, will be considered in the future work).

Discussion

Our understanding of the EL emission from self-assembled QDs is the following. The majority carriers, the electrons in this case, are supplied from the current flowing via the GaAs conduction band states into the metal by tunneling through the oxide. Since the tunneling current and the radiative recombination current (leading to the emission) are small, the population of the QD states with electrons is close to equilibrium and determined by the Fermi level. The minority carriers start appearing in the QDs only when the Fermi level in the metal is approaching the top of the valence band in the semiconductor (E_V), so that tunneling through the oxide for some of the holes in the metal becomes possible. The probability of this process depends on the temperature. At some threshold value of V, that is of the order of E_g, the Fermi level crosses over E_V and many holes can be injected into the capping layer. They move towards the QD layer thanks to the diffusion and drift acting in the same direction, and some of the holes are captured into the localized QD states where they eventually recombine radiatively with the electrons. Further increase of the bias leads to a steady increase of the EL intensity corresponding to the growth of the injection rate as depicted in Fig. 6.

Injection of spin-polarised holes from the FM contact in magnetic field occurs in the same way and manifests itself by the change of the sign of the degree of circular polarization of the light emitted by the QDs when the golden contact is replaced by the Au/Ni one (Fig. 4). However, the degree of circular polarisation depends on the bias in a non-monotonic way (Fig. 5), which is not completely understood by us yet. One possibility to explain this result is that the loss of the hole spin coherence during their thermalisation in the semiconductor increases when they are injected with a higher kinetic energy (higher V). The capping layer thickness also plays a critical role for the observation of the spin injection. In fact, the right-hand polarisation was observed only for HSs $d_c \le 16$ nm. This makes it necessary to attain a compromise between the EL intensity (lowering for small d_c) and the hole spin coherence.

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