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A LETTERS JOURNAL EXPLORING THE FRONTIERS OF PHYSICS

April 2012

EPL, **98** (2012) 27012
doi: 10.1209/0295-5075/98/27012

www.epljournal.org

Light emission and spin-polarised hole injection in InAs/GaAs quantum dot heterostructures with Schottky contact

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received 19 November 2011; accepted in final form 26 March 2012
published online 30 April 2012

PACS 78.67.-n – Optical properties of low-dimensional, mesoscopic, and nanoscale materials and structures
PACS 72.25.Hg – Electrical injection of spin polarized carriers
PACS 72.25.Mk – Spin transport through interfaces

Abstract – We demonstrate the feasibility to obtain electroluminescence (EL), up to room temperature, from InGaAs self-assembled quantum dots (QDs) included in a forward-biased Schottky diode. Moreover, using a ferromagnet (FM) as the contact layer, sizable circular polarization of the EL emission in the presence of an external magnetic field is obtained. A resonant behavior of the degree of circular polarization (P) as a function of the applied voltage (V), for a given value of magnetic field, is observed. We explain our findings using a model including tunneling of (spin-polarised) holes through the metal-semiconductor interface, transport in the near-surface region of the heterostructure and out-of-equilibrium statistics of the injected carriers occupying the available states in the QD heterostructure. In particular, the resonant $P(V)$ dependence is related to the splitting of the quasi-Fermi level for two spin orientations in the FM.

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Introduction. – Adding the spin degree of freedom to conventional semiconductor charge-based electronics or using the spin degree of freedom alone can add substantially more capability and performance to electronic devices [1]. One of the most widely used semiconductor spintronic devices is the spin light-emitting diode (LED) where a diluted magnetic semiconductor is used as spin aligner [2–5]. In a spin LED circularly polarized light is emitted after the recombination of spin-polarized carriers that are electrically injected into a semiconductor heterostructure. A possible way of improving the spin alignment of the injected carriers is to use a Schottky contact. Single-element ferromagnetic (FM) metals are attractive spin injectors as they possess a high Curie temperature and exhibit well-understood thin-film magnetism [6]. There is a common belief that low injection efficiency of minority carriers is inherent to Schottky diodes (SDs). However, it has been shown that SDs based on n-type GaAs [6] or InAs/GaAs quantum well (QW) heterostructures [7–10] can serve as a good light source (we shall call it light-emitting Schottky diode, LESD). The injection of non-equilibrium electrons for radiative recombination can be achieved in a *reverse*-biased SD where the depletion width is made small by heavily doping the near-surface region of the semiconductor [7,8]. Recently, an efficient injection of spin-polarised electrons from a FM contact through a MgO tunnel barrier at room temperature was demonstrated [9]. Alternatively, one can obtain light emission by applying a sufficiently high *forward* bias and the LESD efficiency can be increased by placing a thin (∼1 nm) oxide layer between the metal and the semiconductor [10,11]. Using a FM Schottky contact and applying a sufficiently strong magnetic field, it is possible to obtain a significant degree of circular polarisation of the emitted light (up to 40% for an Au-Ni/InAs/GaAs QW LESD [10]).

Incorporation of self-assembled quantum dots (SAQDs) into LESD offers a number of potentially interesting applications. First, it makes possible to realize a light-emitting device with SAQDs covered with a very thin capping layer and emitting at a wavelength up to 1.6 μm [12], interesting for telecommunications. Secondly, owing to the higher

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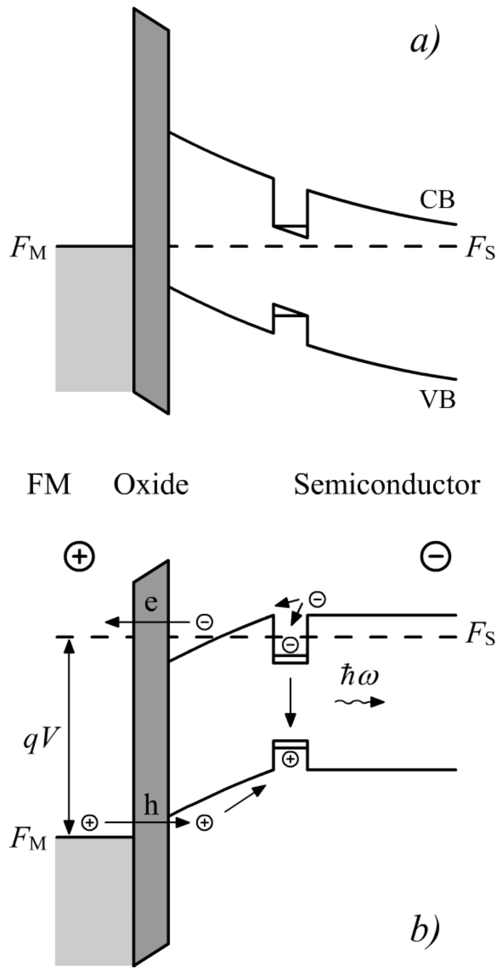

Fig. 1: Schematic energy band diagram for the studied HS at equilibrium in an open circuit (a) and under an applied bias (b). F_M (F_S) depicts the Fermi level of the ferromagnet (semiconductor). CB (VB) is the conduction (valence) band profile of the semiconductor and $\hbar\omega$ indicates the energy of the photon emitted after e-h recombination.

confinement potential (compared to QWs), SAQDs can emit at room temperature and even above it [13]. Thirdly, this is a promising way of spin injection into the dots, which is of great importance for spintronics [14]. During the last years, reverse-biased SDs have been used as a tool for studies of the injection of spin-polarized electrons from a FM metal into a semiconductor [15,16]. In such structures, the injection occurs via tunneling through the barrier produced by the depletion layer, depending on the doping level of the semiconductor [17]. However, we pursue a different approach by considering a *forward*-biased LESD based on an n-type doped InAs/GaAs quantum size heterostructure (HS) [18]. Figure 1 shows a band diagram of the SD studied in this work, with a tunnel-thin oxide layer separating the Schottky contact from the semiconductor HS. Under a sufficiently high forward bias, the band alignment is such that the Fermi level in the metal lies below the top of the valence band (VB) in the semiconductor making the tunneling of the VB electrons into the metal possible (see fig. 1(b)). Alternatively, this process can be thought of as a *hole* tunneling into the semiconductor HS, providing the necessary minority carriers for the recombination via photon emission. The injection of spin-polarized minority carriers can be achieved in a LESD with a FM contact, as it has been shown for a QW HS [10]. In this work we demonstrate the feasibility to obtain this for quantum dots. The central result is that the LESD emission due to the carrier recombination in the SAQDs is circularly polarized. However, this occurs only for a relatively narrow range of bias voltages. The effect is explained by the tunneling of spin-polarized holes through the oxide layer. The Fermi levels for two spin orientations in the metal are different in the presence of magnetic field. For a certain range of applied voltages, only holes with favorably oriented spin can tunnel into the semiconductor, and this orientation is not completely lost during the tunneling processes, transport to the SAQD layer and relaxation inside the dots.

Experiment. – InAs/GaAs SAQD heterostructures were grown on n-type GaAs substrates by atmospheric-pressure metal-organic vapor-phase epitaxy. The SAQDs were overgrown by an InGaAs QW layer (2 nm in thickness) and covered by a GaAs capping layer of variable thickness (10–30 nm) with a thin (1–2 nm) oxide on top of it. Two kinds of metallic layers were used for the Schottky contacts: Au (non-magnetic) and Au-Ni-Au (FM). The current voltage characteristics of similar diodes have been described before [19] and allowed for the determination of the Schottky barrier height (in the range 0.5–0.9 eV).

The samples were placed in a helium bath cryostat at 2.5 K and its electroluminescence (EL) together with the degree of circular polarisation, under forward bias ranging from 1 to 2 V, were measured in Faraday geometry, for magnetic fields up to 10 T. For regular EL experiments, the samples were mounted on the cold finger of a variable-temperature cryostat. The signal was dispersed with a spectrometer and detected with a liquid-nitrogen–cooled InGaAs photodiode array. A quarter-wave plate in combination with a linear polarizer was used to select the EL with left and right circular polarizations. Our LESDs show good light emission properties, with the EL bands located at 1.2–1.3 eV for the QW and at 0.8–0.95 eV for the QDs (see inset in fig. 2) [18]. The EL spectra are qualitatively alike to the photoluminescence spectra of similar HSs without Schottky contact [20] and show several peaks corresponding to the electron-hole transitions involving the ground and excited states in the dots. The EL intensity depends strongly on the injecting current (*i.e.*, applied bias) and temperature. It exhibits an exponential growth with increasing bias (see fig. 2), at low voltages, followed by a power law above a threshold (V_t), which is of the order of the band gap energy of GaAs. Moreover, the EL intensity rises markedly with increasing the temperature for voltages below V_t, while a much weaker temperature

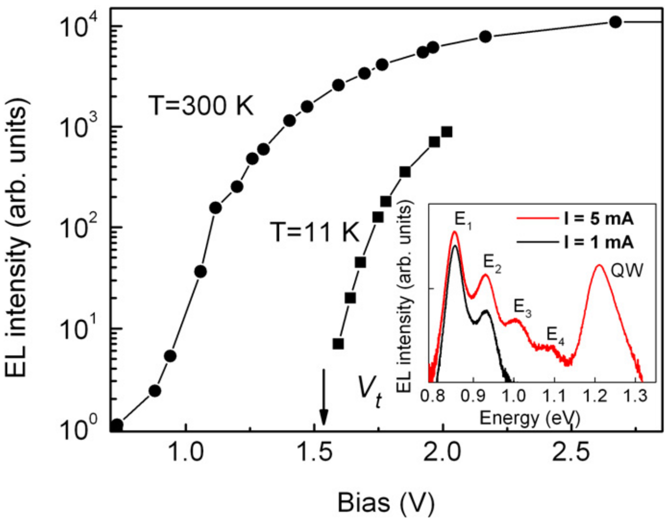

Fig. 2: (Color online) Dependence of the intensity of the first SAQD emission peak (E_1) on the applied bias for a SD with Au contact measured at two temperatures. Inset: EL spectra of the structure measured for two different values of injecting current ($T = 77\,\mathrm{K}$), showing several emission peaks originating from the ground (E_1) and excited (E_2–E_4) states of the SAQDs and the QW covering the dots. The capping layer thickness is 20 nm.

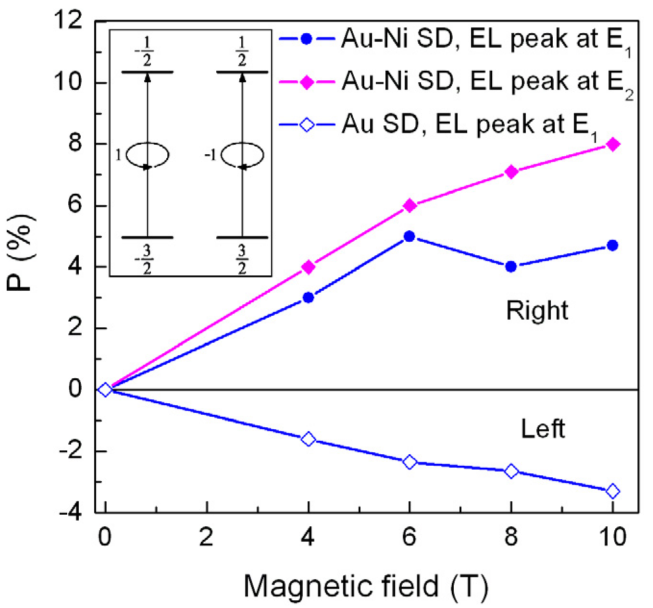

Fig. 3: (Color online) Degree of circular polarisation as a function of the applied magnetic field, measured for Schottky diodes (SDs) with FM (Au-Ni-Au) and non-magnetic (Au) contacts. The inset shows the electron-hole recombination processes occurring in the QDs, leading to the emission of light with left and right polarisation.

dependence is obtained for higher voltages. The capping layer thickness plays also an important role for the EL characteristics. 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. On the other hand, increasing d_c diminishes the tunneling current and, consequently, also the EL intensity.

In the presence of an external magnetic field, the degree of circular polarisation ($P = (I_+ - I_-)/(I_+ + I_-)$, where I_+ (I_-) denotes the intensity of the left (right) polarised emission) is determined by the nature of the contact metal. Samples with Au contacts display a left-handed polarisation ($P = -3\%$ at 10 T), while for the diodes with ferromagnetic contacts, a right-handed polarisation is obtained (see fig. 3). The former effect can be attributed to the Zeeman splitting of the hole levels in the QDs, while the latter must be due to the injection of spin-polarized holes from the FM Schottky contact, as confirmed by the larger values of P for the second emission peak ($P = +8\%$ at 10 T). The lower values of P obtained for QDs, as compared with those measured in similar experiments in QWs [10], may be attributed to a different ordering of the Zeeman-split levels in both systems. The degree of circular polarization for the samples with FM contacts shows a nontrivial behavior that depends mainly on the capping layer thickness and voltage. At 10 T, P reaches a maximum at a bias slightly above V_t and then decreases (see fig. 4), as has been mentioned in the introduction. The capping layer thickness also plays a critical role for the observation of the spin injection. In fact, the right-hand polarization was observed only for LESDs with $d_c \leqslant 16\,\mathrm{nm}$.

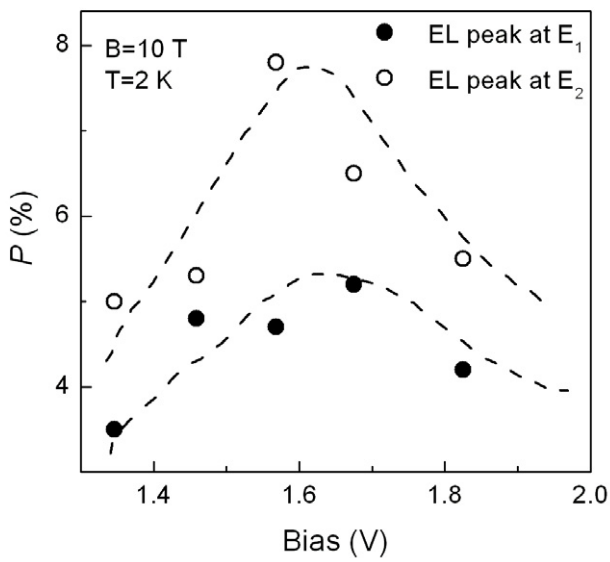

Fig. 4: Degree of circular polarisation of the SAQDs' emission *vs.* bias applied to the Schottky diode with FM contact (same as in fig. 3). The lines are guides to the eye.

Model. – In order to understand these experimental findings we developed a model outlined in fig. 1(b), whose implementation includes the following steps:

i) calculation of the electron and hole tunneling currents in the SD structure with intermediate oxide layer;

ii) determination of the minority carrier's generation rate in the SAQDs/QW region of the semiconductor

HS by a simplified analysis of the diffusion-drift transport in the near-surface region and using the tunneling injection rate i);

iii) calculation of the statistical distributions of the minority carriers out of equilibrium, under steady bias using the generation rate ii), and

iv) Determination of the light emission rates.

The calculation of the tunneling currents is based on the approach similar to the one proposed in the past for silicon SDs [21], based on the tunneling Hamiltonian. This approach assuming that the energy, spin ($\sigma = \uparrow, \downarrow$) and the component of the electron wave vector parallel to the interface are conserved, has been used in a number of works studying spin injection via tunneling [17,22–24]). Assuming further that the Fermi surface in the metal is spherical, the electron current density per spin component is

$$j_{e\sigma} = \frac{qm_e}{(2\pi)^2\hbar^3}\int_0^\infty \mathrm{d}E \int_0^E \mathrm{d}E_z D_e\left(E_z\right) \times \left[n(E-F_{S\sigma}) - n(E-F_{M\sigma})\right], \quad (1)$$

where q (m_e) is the electron charge (effective mass), D_e is the transparency of the barrier seen by the conduction band (CB) electrons (see fig. 1), $n(\epsilon) = [1+\exp(\epsilon/kT)]^{-1}$ is the Fermi function and $F_{S\sigma}$ ($F_{M\sigma}$) is the quasi-Fermi level in the semiconductor (metal) counted with respect to the CB bottom in the semiconductor far from the contact. The applied bias (V) is divided between the oxide layer and the depletion layer of the semiconductor (although for high V the latter vanishes). Solving self-consistently the standard electrostatic equations [25] yields the height of the contact barrier in the semiconductor *vs.* V for both depletion (fig. 1(a)) and accumulation (fig. 1(b)) regimes which depends on its value (φ_{b0}) at $V = 0$ and the oxide thickness. This defines the shape of the CB barrier for a given V and its transparency can be calculated numerically. The current voltage characteristics of the SD calculated using eq. (1) (see footnote [1]) are in qualitative agreement with our experimental data (to be published elsewhere). Here we are concerned with the hole current, which determines the EL intensity. It can be calculated in the same way as $j_{e\sigma}$ but considering the tunneling of VB electrons. We take into account only the heavy hole subband (with the effective mass m_{hh}), then the tunneling current density per spin component is

$$j_{h\sigma} = \frac{qm_{hh}}{(2\pi)^2\hbar^3}\int_0^\infty \mathrm{d}E_h \int_0^{E_h} \mathrm{d}E_z D_h\left(E_z\right) \times \left[n(E_h+E_g+F_{M\sigma}) - n(E_h+E_g+F_{S\sigma})\right], \quad (2)$$

where E_h is the hole kinetic energy, E_g is the band gap energy and D_h is the transparency of the barrier seen by the VB electrons. We notice that the second term in the second line of eq. (2) can be dropped for any $V > 0$ since $|F_{S\sigma}| \ll E_g$. It is clear from eq. (2) that, within the approximation used, $j_{h\uparrow}$ is not equal to $j_{h\downarrow}$ only if $F_{M\uparrow} \neq F_{M\downarrow}$. It is usually assumed that the rate of scattering events without spin flip of spin-up and spin-down electrons is much larger than the spin-flip rate implying that the quasi-Fermi levels for $\sigma = \uparrow, \downarrow$ may not be equal [23,26–28]. Therefore, we shall assume that in eqs. (1) and (2)

$$F_{M\sigma} = E_F - qV \pm \Delta F/2,$$

where E_F is the Fermi level and ΔF is proportional to the magnetic field in the FM.

The next step consists in calculating the rates of supply of non-equilibrium holes to the SAQDs and QW. For this, a diffusion-drift model can be employed [27,29] assuming again that the spin-flip characteristic time is long compared to the transport time from the interface to the SAQD/QW region. Indeed, the spin diffusion length of electrons is of the order of several hundred nanometers [29], much higher than the thickness of the capping layer plus the SAQD layer in our HSs. Assuming that it is so also for holes, we can write

$$j_{h\sigma}(z) = -q\left(\mu_h p_\sigma \frac{\mathrm{d}\varphi}{\mathrm{d}z} + d_h \frac{\mathrm{d}p_\sigma}{\mathrm{d}z}\right), \quad \frac{\mathrm{d}j_{h\sigma}}{\mathrm{d}z} = -\frac{qp_\sigma}{\tau_r}. \quad (3)$$

Here μ_h (d_h) is the hole mobility (diffusion coefficient), p_σ is the concentration of spin-polarised holes, φ is the electrostatic potential and τ_r is the lifetime (an average recombination time representative of the whole HS). A boundary condition for (3) is given by the value $j_{h\sigma}(0)$ at the semiconductor-oxide interface calculated by eq. (2).

Using a simplified analysis of the diffusion-drift model, similar to that presented in ref. [29] we can obtain the following expression for the average generation rate of holes in the near-surface region of the semiconductor HS:

$$g_\sigma = \frac{2j_{h\sigma}(0)}{q\left(L_E + \sqrt{L_E^2 + 4L_h^2}\right)}, \quad (4)$$

where $L_E = q\varphi_b L_h^2/(kTL_D)$, φ_b is the (V-dependent) contact barrier height, $L_h = \sqrt{d_h\tau_r}$ is the difusion length and L_D is the Debye length. Then we can make use of the statistical model previously developed by some of us in order to describe the steady-state emission of SAQD/QW heterostructures under optical excitation (*i.e.*, photoluminscence) [20]. The model considers an effective homogeneous system representing the HS, with certain energy levels due to SAQDs ($E_{QD}^{(e)}$, $E_{QD}^{(h)}$) (see footnote [2]), QW ($E_{QW}^{(e)}$, $E_{QW}^{(h)}$) and also a VB continuum due to the GaAs barriers. Under steady-state excitation

[1]It is obtained by summing over σ. We neglect any difference between $F_{M\uparrow}$ and $F_{M\downarrow}$ for electrons in the non-magnetic semiconductor.

[2]For the sake of simplicity, the small Zeeman splitting of these levels was neglected.

conditions, the quasi-Fermi level for holes is determined by the following equation [20]:

$$g_\sigma \tau_r^{(0)} = \xi p_{B,\sigma} + \eta_{QD} p_{QD,\sigma} + \eta_{QW} p_{QW,\sigma}\,, \qquad (5)$$

where $\tau_r^{(0)} = 3m_0^2\hbar^2c^3/(8\mathtt{n}e^2\mathcal{P}^2E_g)$ is a constant entering the expression for τ_r [20] (m_0 is the free-electron mass, c is the light velocity, $\mathtt{n}$ is the refractive index and $\mathcal{P}$ is the momentum matrix element of the dipole transition between VB and CB), $\xi = (\tau_r^{(0)}/\tau_{nr})$, τ_{nr} is the lifetime for non-radiative recombination via traps,

$$\eta_{QW} = \left(\frac{(m_e + m_{hh})E_g'}{m_e E_g K_{QW}^2}\right)\exp\left(\frac{E_F - E_{QW}^{(e)}}{kT}\right),$$

$$\eta_{QD} = \left(\frac{E_g'}{E_g K_{QD}^2}\right)\exp\left(\frac{E_F - E_{QD}^{(e)}}{kT}\right),$$

$E_g' = E_{QD(QW)}^{(e)} - E_{QD(QW)}^{(h)}$ and $K_{QD(QW)}$ is the overlap integral of the electron and hole envelope wave functions in the QD (QW) (see footnote [3]). The (effective) 3D concentrations of holes occupying different levels, $p_{B,\sigma}$, $p_{QW,\sigma}$ and $p_{QD,\sigma}$, are expressed through the corresponding quasi-Fermi level by the standard statistics formulae, for instance, $p_{B,\sigma} = N_v\exp[-(F_{S\sigma}^{(h)} + E_g)/kT]$ with $N_v = [(m_{hh}kT/(2\pi\hbar^2)]^{3/2}$. The quasi-Fermi levels $F_{S\sigma}^h$ are obtained by solving eq. (5) and the QD emission intensity is calculated using the following expression:

$$I_{QD}^{L(R)} = \frac{p_{QD,\uparrow(\downarrow)}}{\eta_{QD}\tau_r^{(0)}}\tilde{w}, \qquad (6)$$

where $\tilde{w}$ is the effective width of the considered system [20]. Therefore the degree of circular polarisation can be calculated as $P = (p_{QD,\uparrow} - p_{QD,\downarrow})/(p_{QD,\uparrow} + p_{QD,\downarrow})$. Material parameters used in the calculations are the same as those in ref. [20], the oxide thickness was taken equal to 1 nm and the contact barrier height at equilibrium is specified below. As it can be seen from fig. 5, the intensity of the QD emission at zero magnetic field rises above a threshold forward bias, $V_t \approx E_g/q$, as it has been observed experimentally (fig. 2). This threshold corresponds to the matching of the quasi-Fermi level in the metal and the VB top in the semiconductor. The smaller slope of the voltage dependence near V_t for higher temperature reflects the broadening of the Fermi function and the downward shift of the threshold reflects the decrease of the band gap energy with T. The enhancement of the injection rate with bias occurs mostly because of the increase of the transparency of the barrier for increasing kinetic energy of the tunneling particles. Experimentally, the dependence starts saturating at higher V, which is not quite reproduced in the calculated curves. This is probably due to the known limitations

[3]The model assumes quasi-equilibrium between different electronic states and should not be valid at very low temperatures where such equilibrium cannot take place.

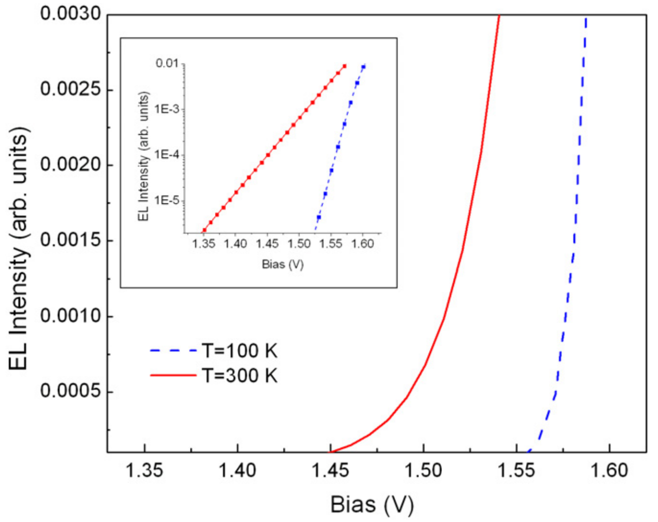

Fig. 5: (Color online) Calculated QD emission intensity *vs.* applied bias at $B = 0$ for two different temperatures ($\varphi_{b0} = 0.4\,\text{eV}$, $E_F = -0.04\,\text{eV}$). The inset shows the same dependences in semi-logarithmic scale.

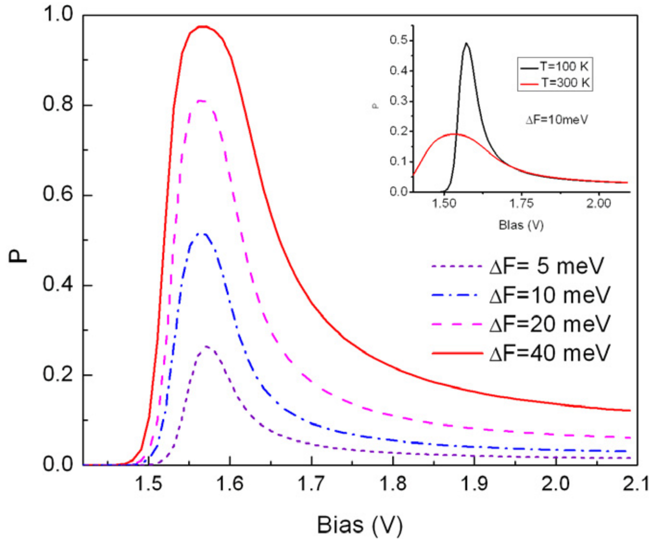

Fig. 6: (Color online) Calculated degree of circular polarisation of the QD emission *vs.* applied bias for different values of the quasi-Fermi level splitting between two spin orientations in the FM (T=100 K). The inset shows the effect of T raising from 100 to 300 K for $\Delta F = 10\,\text{meV}$.

of the tunneling Hamiltonian approach [30].Under an applied external magnetic field, $j_{h\uparrow} \neq j_{h\downarrow}$ and $P \neq 0$. Figure 6 shows the dependence of P on the applied bias for several values of ΔF treated as a phenomenological parameter. It qualitatively reproduces the experimentally observed non-monotonic behaviour of $P(V)$ (see fig. 4).

Discussion and conclusion. – Based on the presented experimental and modelling results, our understanding of the EL emission from SAQDs is the following. The majority carriers, electrons in this case, are supplied from the current flowing via the GaAs CB states into the metal by tunneling through the oxide layer. 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 begin to appear in the dots only when the quasi-Fermi level in the metal is approaching the top of the valence band in the semiconductor, so that tunneling through the oxide for some of the VB electrons in the semiconductor (or, in other words, holes in the metal) becomes possible. The probability of this process depends on the bias and temperature. The injected holes move towards the SAQD/QW 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 (figs. 2 and 5). At a given V, the injection rate increases exponentially with temperature because of the increase of the number of holes below the Fermi level in the metal.

Injection of spin-polarised holes from the FM Schottky contact in magnetic field occurs in the same way and manifests itself by the change of the sign of the degree of circular polarisation of the light emitted by the QDs when the gold contact is replaced by the Au/Ni one (fig. 3). P is higher for the second (higher-energy) emission peak. It could be understood by the fact that the relaxation from the excited states to the ground state in the dots involves an extra process and, consequently, an additional loss of spin coherence. Quite interestingly, P depends on the bias in a non-monotonic (almost resonant) way (fig. 4). Similar $P(V)$ dependences have been observed for AlGaAs/GaAs QW heterostructures with a Fe Schottky contact [31] where the light was emitted by means of electron's tunneling into the semiconductor from the Fe film through an AlGaAs barrier and their recombination with unpolarized holes from the substrate. According to our model, the maximum is reached when the top of the valence band lies between the quasi-Fermi levels for two spin polarisations in the FM (split by the magnetic field), $F_{M\uparrow} < E_v < F_{M\downarrow}$. The larger the splitting, the higher P can be achieved (fig. 6).

In conclusion, we have shown the feasibility of circularly polarised EL emission from InGaAs SAQDs incorporated into a forward-biased Schottky diode structure. The emission is caused by the tunneling-mediated injection of (spin-polarised) holes from the metal into the light-emitting zone of the heterostructure. Even though the values of P achieved for SAQD emission in this work are modest compared to those obtained for similar structures with QW [10] and for a SAQD HS combined with a diluted magnetic semiconductor injector [5], probably it can be increased by improving the FM layer and interface quality. The sharp dependence of P on the applied bias opens potentially interesting possibilities for applications.

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This work was supported by the FCT, Portugal (project POCI/FIS/58524/2004), the RFBR, Russia (grant 10-02-00501), MEC (grants MAT2008-01555, QOIT-CSD2006-00019) and CAM (S-2009/ESP-1503) (Spain).

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