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# Optical properties of multi-layer type II InP/GaAs quantum dots studied by surface photovoltage spectroscopy

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We present a low-temperature (73 K) study of the optical properties of multi-layer type II InP/GaAs self-assembled quantum dots by means of surface photovoltage (SPV) spectroscopy, taking advantage of its high sensitivity and contactless nature. The samples contain 10 periods of InP quantum dot planes separated by 5 nm GaAs spacers. The SPV amplitude spectra reveal two major broad peaks, situated at low and high energies, respectively. These features are analyzed taking into account the type II character of the structure, the quantum coupling effects, the spectral behavior of the SPV phase, and the photoluminescence spectra. As a result they have been attributed to optical transitions in the quantum dots and the wetting layers, respectively. The main mechanism for carrier separation in the SPV generation process is clarified via the analysis of the SPV phase spectra. The influence of the substrate absorption on the SPV spectra is discussed in details. © *2011 American Institute of Physics*. [doi:10.1063/1.3638705]

## I. INTRODUCTION

The InP/GaAs quantum dots (QDs) are type II heterostructures, where the electron is confined in the InP material while the hole remains in the GaAs layer around the QDs, due to the Coulomb attraction toward the electron.[1,2] The spatial separation of the carriers results in a weaker exciton with a more extended wave function as compared to the type I QD exciton, where both carriers are strongly confined in the QD. The electron-hole wave function spatial overlap is also much smaller for type II QDs, resulting in relatively long carrier lifetimes.[3,4] Due to these properties and their near-infrared (∼0.95 pm) optical transition energies, InP/GaAs QDs are promising candidates for optical memories and optical communication devices.

These type II QD heterostructures are more complex systems as compared to type I QDs, because of the different possibilities for hole wave function distribution around the QD, which is very sensitive to the QD geometry.[5] The study of multi-layer type II QDs (i.e., sequences of QD planes separated by narrow spacer layers) is further complicated, because of the structural and electronic couplings between dots adjacent along the growth direction, which depends on the spacer layer parameters. So far the published works devoted to the optical properties of InP/GaAs QDs are mainly based on photoluminescence (PL) investigations.[2,5–7] Few results are available in the literature concerning optical absorption measurements of type II QD systems,[8] probably due to the relative novelty of the research field and the general difficulties related to the optical absorption spectroscopy in nanostructures. A promising alternative to the optical absorption is the surface photovoltage (SPV) spectroscopy, which is a high-sensitive semiconductor characterization technique successfully applied in the study of complex nanostructures in recent years.[9–16] It is contactless, nondestructive and can provide information for both the optical absorption transitions and the electronic transport in bulk materials and nanostructures even at room temperature.

Concerning the room temperature SPV studies of the nanostructures described above, we would like to note the following: Due to the novelty of these complex nanostructures and of their SPV studies, a comparison with the results of other experimental techniques, for example PL, is desirable for the correct interpretation of the experimental SPV data. However no PL signal could be detected from the samples at room temperature, because of the carrier escape toward the barriers due to the weak electron-hole coupling. That is why in this work we have used the SPV spectroscopy to investigate the optical properties of the above-mentioned nanostructures at 73 K, where a reasonable PL signal has been obtained.

The main goals of the paper are as follows: (i) To observe and discuss the InP/GaAs QD and wetting layer optical transitions in optical absorption (SPV); (ii) to study the mechanisms of carrier separation in the SPV generation process; and (iii) to describe the substrate contribution to the SPV spectrum. To our knowledge this has not been achieved until now.

In order to achieve an express and reliable analysis of the SPV amplitude and phase spectra, our recently developed vector model for representation of the SPV signal[17] has been applied.

## II. EXPERIMENTAL DETAILS

The self-assembled InP/GaAs QDs are grown by the Stranski-Krastanov mode in a chemical beam epitaxy (CBE) system. The growth details are similar to the ones given in Ref. 2 for single layer QD structures. The present sample contains 10 periods of InP/GaAs QDs layers, separated by

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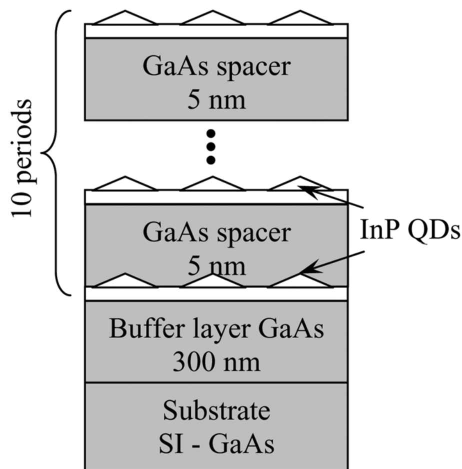


FIG. 1. Layer sequence in the sample.

GaAs spacer layers with nominal thickness of 5 nm. The top QD layer is left uncovered in order to perform atomic force microscopy (AFM) measurements. The structure is grown on a 300 nm GaAs buffer layer deposited on a (001) oriented semi-insulating (SI) GaAs substrate. A schematic of the sample structure is shown in Fig. 1. The CBE grown multi-layer structure and GaAs buffer layer reveal a p-type residual doping of around $10^{16}$ $cm^{-3}$

The SPV measurements were performed at 73 K, using the metal-insulator-semiconductor (MIS) operation mode of the SPV technique.[11] In this mode the SPV is measured as the photo-induced voltage change between the MIS capacitor terminals caused by periodic light excitation. The probe electrode was a semitransparent $SnO_2$ film evaporated on the bottom surface of a quartz glass. During the measurements, the probe was used to gently press the sample against a grounded copper platform. The optical excitation was performed using a 100 W halogen lamp along with a SPEX grating monochromator (f = 0.25 m, 600 g/mm) and an optical chopper. The probe signal with respect to the ground was fed to a high-impedance unity gain buffer and then measured by an EG&G 5207 Lock-in amplifier. Normally incident light chopped at 94 Hz was used. The light wavelength was scanned from the higher to the lower values keeping the photon flux constant ($\approx 2 \times 10^{14}$ $cm^{-2}s^{-1}$) within $\pm$ 2% for all the wavelengths. The SPV amplitude and phase were calculated from the signal, which is in phase with the reference signal from the optical chopper and the signal, shifted in phase by 90$^o$. The reference signal from the optical chopper defines the zero value of the SPV phase. For that purpose we took all precautions to eliminate undesired phase shifts, following the procedures described in Ref. 18. More details about the SPV experimental set-up and measurement procedure can be found elsewhere.[18]

PL measurements were performed at 73 K using a 200 mW diode laser for excitation with 532 nm light, a SPEX single monochromator (1200 g/mm) and a Ge detector connected to a Lock-in amplifier. The excitation intensity was adjusted by means of a neutral density filter.

## III. RESULTS AND DISCUSSION

The QD size distribution is obtained by AFM measurements[4] on a $1 \times 1$ $\mu m^2$ area of the top layer. The QD height ($H$) varies from $\sim$1.5 to more than 7 nm with an average value of (3.2 $\pm$ 0.9) nm and the corresponding QD radius ($R$) is between $\sim$12 and $\sim$24 nm with an average value of (18 $\pm$ 3) nm. The dot density is $3.5 \times 10^{10}$ $cm^{-2}$. Transmission electron microscopy (TEM) images reveal that the QDs in the first layer are smaller than the measured thickness of the GaAs spacer layer, which is $\sim$3 nm.[4] Therefore these QDs are physically separated from the next InP layer. This separation decreases for the subsequent layers and the dots after the fourth layer coalesce into a quantum post (QP)-like[19] structure. More details about the structural characterization of the samples are given elsewhere.[4]

Typical SPV amplitude and phase spectra of the QD sample, measured at 73 K, are presented in Fig. 2. For the sake of convenience of the analysis, they can be divided into two parts: A low-energy region for photon energies $h\nu < 1.38$ eV and a high-energy one for $h\nu > 1.38$ eV. In order to explain the SPV behavior, we perform a simultaneous analysis of the SPV amplitude and phase making use of our recently developed vector model for representation of the SPV signal.[17] In this model the ac SPV signal is represented as a radial vector with magnitude equal to the SPV amplitude and angle with respect to the X-axis equal to the SPV phase. Thus, the superposition of two or more SPV processes is represented by summation of the corresponding SPV vectors.

### A. Low-energy region ($h\nu < 1.38$ eV) of the SPV spectra

Due to the p-type residual doping of the multi-layer structure and buffer layer, it is expected that the energy bands at the surface are bent downward with respect to the bulk. Thus, a space charge region (SCR) arises at the sample surface with a built-in electric field oriented toward the bulk. The width of the SCR is assessed to be $\sim$215 nm, assuming a residual p-type doping of $1 \times 10^{16}$ $cm^{-3}$ and a surface potential of 0.65 eV. The whole QD structure is around 40 nm thick according to the TEM results. Therefore, the QD structure and a part of the GaAs buffer layer are situated in the SCR.

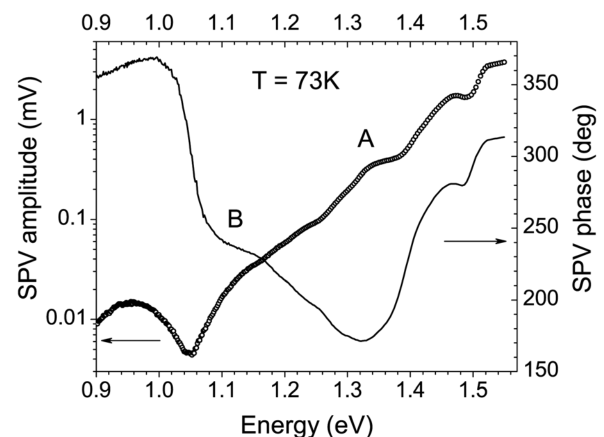


FIG. 2. SPV amplitude (open circles and left scale) and phase (solid line and right scale) spectra of the QD sample, measured at 73 K.

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As explained in Refs. 17 and 18 in the case where predominantly free electrons (holes) move toward the substrate and/or free holes (electrons) move toward the surface, the SPV phase is in the IV quadrant (II quadrant). For hv below 1.0 eV the SPV signal is probably due to deep level (DL) related transitions in the SI GaAs substrate. Such transitions in SI GaAs are known to produce SPV signals with different phases depending on the different DL emission and recombination rates for electron and holes.[20] In the present case the SPV phase is near 360°, which indicates predominant movement of free electrons toward the substrate, as mentioned above.

In the region $1.0 < h\nu < 1.16$ eV the SPV amplitude reveals a minimum at 1.05 eV, while the SPV phase drastically changes clockwise by more than 100° and approaches 240°. Such SPV behavior corresponds to the appearance and increase of a new SPV process with a phase in the II quadrant, close to 180°, which is superimposed on the DL-related process. This is explained by means of the vector diagram in Fig. 3, where the SPV vectors of this new process and that of the DL related process are labeled QP and DL, respectively. The vector QP increases with increasing h$\nu$, forming an obtuse angle with vector DL. As a result the overall SPV vector rotates clockwise, while its magnitude reveals a minimum at $h\nu = 1.05$ eV. The phase of the new SPV process indicates hole movement toward the substrate and/or electron movement toward the surface, which is in accordance with the downward band bending in the QD structure.[17,18] This implies that the new SPV process originates from the QD structure. Its signal overcomes the one from the DL transitions in the substrate and leads to the step in the amplitude spectrum in the range 1.05–1.16 eV. Further on, this step is labeled "B".

For $h\nu > 1.16$ eV the SPV amplitude continuously increases and reveals another step in the range 1.25–1.38 eV labeled "A". In the same time the SPV phase continues its change in clockwise direction, forming a minimum at 1.32 eV. In terms of the vector model this behavior is explained by the appearance and increase of another SPV vector in the II quadrant, which is added to the above depicted vectors. This new vector should have an orientation similar to that of the QP vector. Indeed, with the help of Fig. 3 one can guess, that if it was much closer to 90° the magnitude of the resulting overall SPV vector would decrease, while if it was closer to 180° the phase of the resulting overall SPV vector would rotate anticlockwise. Both these scenarios contradict the experimental SPV spectrum. The phase of the SPV process, corresponding to step A, implies that it takes place in the QD structure in accordance with the downward energy band bending there.[17,18] Its magnitude is larger as compared to the process, corresponding to step B. A detailed examination of step A reveals a weak shoulder on its low-energy side around 1.29 eV.

In order to assess the possible influence of the SPV generated in the SI GaAs substrate for $1.0\ \text{eV} < h\nu < 1.38$ eV, we have measured the SPV spectrum at 73 K of a bare substrate sample. In the range 1.0–1.38 eV, where the signal from the QD structure is expected, the SPV amplitude exhibits no distinct spectral features; only a structureless very broad band is observed around 1.1 eV. In the same range the SPV phase changes smoothly, revealing a different behavior as compared to the QD sample (Fig. 2).

In Fig. 4 we present the PL spectrum of the same sample measured at 73 K. It is used for comparison with the SPV spectrum in order to better understand the origin of the SPV features, depicted above. Unfortunately, we could not detect PL with an excitation density as low as that used in the SPV experiment. That is why we have used one of the lowest possible excitation densities (3 $\text{Wcm}^{-2}$), which give a reasonable PL signal from the sample. Lowering the excitation density could lead to redshift of the optical transitions in type II QDs; however at such low densities this trend is likely to saturate, as observed in Ref. 21. This allows comparing the SPV and PL spectra presented here. In the range 1.05–1.18 eV the PL spectrum shows a broad band, labeled B, centered at 1.14 eV. This range is similar to the range of step B in the SPV spectrum. At higher energies the PL spectrum reveals a sharp peak at 1.36 eV, labeled A, preceded by a shoulder at around 1.29 eV. These energies fall within the spectral range of step A in the SPV spectrum.

The good correspondence in energy positions of the bands A and B in the SPV and PL spectra allows concluding that they are due to optical transitions between the same energy states. Following the discussion given in our recent PL study[4] on similar multilayer InP/GaAs QD samples, we

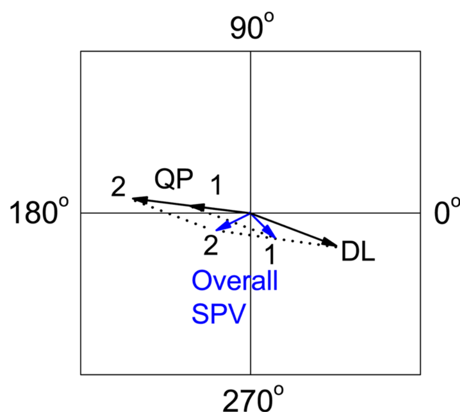


FIG. 3. (Color online) Vector diagram representation of the SPV processes for energies in the range 1.0–1.16 eV. Due to the appearance and growth of vector **QP** the overall **SPV** vector (which is the sum of vectors **DL** and **QP**) rotates clockwise revealing a minimum at 1.05 eV. Two consecutive states of the **QP** and the overall **SPV** vectors are shown and labeled "1" and "2".

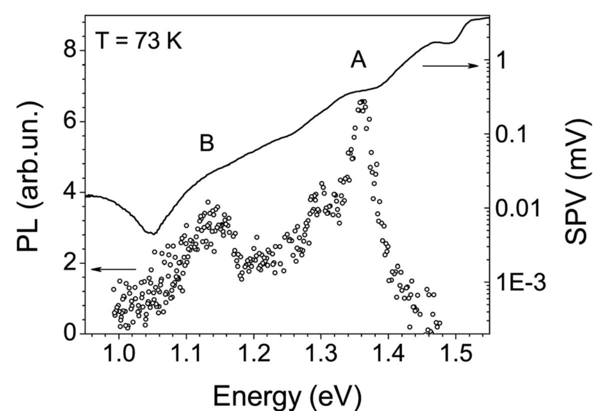


FIG. 4. Photoluminescence spectrum (open circles) of the QD sample measured at 73 K with an excitation density of 3 $\text{Wcm}^{-2}$. SPV amplitude spectrum (solid line) from Fig. 2, is added for comparison.

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ascribe the SPV band B to electronic transitions from the valence band (VB) states in the GaAs spacers to the conduction band (CB) states in the InP QPs, while the SPV band A is attributed to similar transitions, but involving the CB states of the wetting layer (WL), as illustrated in Fig. 5.

The energy of band B (1.14 eV) is considerably lower than the emission energy (1.32 eV) of single-layer capped InP/GaAs QDs observed at 7 K for excitation density similar to the one used in the current study.[21] The observed difference (180 meV) can originate from temperature related redshift, as well as from size and coupling effects. We have checked that with increasing the temperature from 7 K to 73 K the maximum of band B in the PL spectrum experiences a redshift of 55 meV. Therefore the lower energy position of band B with respect to the QD emission energy reported in Ref. 21 is mostly due to the vertical coupling of the QDs forming QPs with a size much larger as compared to the single QDs. The energy of band A (1.36 eV) is also lower than the energies ($\sim$1.41–1.45 eV) observed for WL emission at 10 K in single layer InP/GaAs QDs.[2,22] The maximum of band A in the PL spectrum experiences a redshift of 14 meV with increasing temperature from 10 K to 73 K. So, the main reason for the lower energy of the WL absorption/emission (band A) as compared to previous studies[2,22] is connected with the electronic coupling effect between the WL planes in the here-investigated multi-layers.

The shoulder around 1.29 eV on the low-energy side of band A, observed in both the SPV and PL spectrum, could be accounted for by electronic transitions involving the QDs from the first layer. As mentioned above, they are separated from the second InP layer and act as isolated QDs, with a size much smaller than that of the QPs. Therefore their transition energies may be between those of the QP and WL, as observed.

### B. Mechanisms of carrier separation in space

In our previous works,[17,18] we have demonstrated that the SPV phase gives information about the mechanisms, via which the photogenerated carriers are separated in space in order to produce a SPV signal. As explained above, the phase of the signals from the QPs and WL is in the II quadrant. This implies that they are generated by free holes moving toward the substrate and/or free electrons moving toward the surface. Carrier separation can occur via carrier drift in opposite directions or via different diffusion distances of electrons and holes. Concerning the drift, the above-depicted direction of carrier transport is in accordance with the downward energy band bending, i.e., with the direction of the built-in electric field in the QD structure. Concerning the carrier diffusion we note the following. In order to leave the QD structure and move toward the substrate, the electrons from the InP QPs have to overcome the barrier formed by the smaller QDs in the first 1–3 layers, as well as the barrier of the GaAs buffer layer. In the same time the holes, generated in the GaAs spacer layers, have much more freedom to move vertically toward the substrate, because the InP barriers of the WL are very thin and no additional barrier between the structure and the GaAs buffer layer exists. Besides, the hole diffusion toward the substrate has the same direction as its drift in the SCR electric field. Thus, the holes photogenerated in the QPs move predominantly toward the substrate via drift and diffusion, while the corresponding photogenerated electrons predominantly drift toward the surface. As the QD structure is very thin ($\sim$40 nm), the holes cover much longer distances (including part of the GaAs buffer layer) than the electrons.

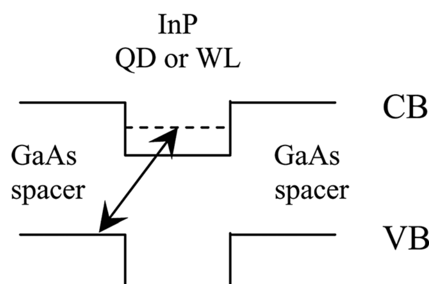


FIG. 5. Schematic diagram of the energy band line-up and electronic transitions in one period of the multi-layer InP/GaAs QD structure. The dashed line marks the energy level of the electron bound state in InP.

We mentioned above that the signals from the QPs and the WL are close in phase. Consequently, the way by means of which the photogenerated carriers separate in space after the WL related transitions is qualitatively similar to that for the QP related transitions. This result should be expected since in both cases the carrier separation is mainly due to the holes, photogenerated in the GaAs spacer layers and their movement toward the substrate, which is realized practically in the same way.

Taking into account the above discussion we can conclude that the main mechanisms of carrier separation include hole drift and diffusion toward the substrate. A smaller contribution is the electron drift toward the surface. These mechanisms give rise to an SPV signal from the QD structure with a phase in the II quadrant, which is superimposed on the DL related signal. The superposition of the two signals leads to the observed SPV signal with a phase in the III and II quadrants in the low-energy spectral region of the spectrum.

### C. High-energy region ($h\nu > 1.38$ eV) of the SPV spectra

Above 1.38 eV the amplitude increases and reveals a maximum at 1.47 eV, followed by a step with inflection point at 1.508 eV. In this range the phase changes anticlockwise and enters again in the IV quadrant where it virtually repeats the run of the amplitude, reaching values above 300° at 1.55 eV. Such SPV behavior can be explained by the consecutive appearance of two SPV processes with phases in the IV quadrant. Since the considered energy range corresponds to optical transitions in bulk GaAs, we concentrate on the substrate contribution to the SPV signal. To understand and explain the nature of the observed spectral peculiarities, we take into account that in SI GaAs the energy bands are practically flat and the dominant processes that give rise to SPV signal are the Dember voltage effect in the bulk as well as the different carriers emission and recombination rates of the sub-bandgap energy levels.[20,23] The Dember effect relies on the difference in the carrier mobilities and since the electrons diffuse faster than holes, the SPV phase is in the IV quadrant.

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Electron emission from a defect level (trapped hole) will results in a phase in the IV quadrant, too, while hole emission (trapped electron)—in a phase in the II quadrant.

Thus, the peak at 1.47 eV is attributed to transitions between shallow acceptors and the conduction band in the substrate, which provide free electrons and trapped holes. This assignment is supported by the fact that the energy difference (38 meV) between the peak position and the GaAs bandgap at 73 K (1.508 eV) is close to the ionization energy of the Si shallow acceptor (34 meV) in GaAs.[24] The SPV vector representing this process is in the IV quadrant. Its increase rotates the overall SPV vector from II through III toward the IV quadrant. At higher energies the band-to-band free carrier generation in the GaAs substrate starts, which is a strong and fast process with a phase in the IV quadrant closer to 360° due to the Dember effect. This process is revealed by the 1.508 eV step in the SPV spectra. The manifestation of the band-to-band transitions as a step in the SPV spectrum is in accordance with the theoretical calculations and experimental results of other authors.[25,26]

## IV. CONCLUSIONS

We present the first SPV spectroscopy study of multilayer self-assembled InP/GaAs QDs at 73 K. Thereby the following main results are obtained: (i) The SPV spectra have provided information about the optical absorption in the structure. In the region below 1.38 eV the SPV amplitude spectrum reveals two major absorption bands: (1.05–1.16 eV) and (1.25–1.38 eV). Their energy positions are in good correspondence with those of the two peaks observed in the 73 K PL spectrum. In accordance with the discussion of these PL peaks, presented in Ref. 4, the low-energy SPV band is ascribed to optical transitions in the InP QDs, coupled in QPs, due to the narrow spacers, while the high- energy one is attributed to optical transitions, related to the WL. This conclusion is supported by the simultaneous analysis of the SPV amplitude and phase spectra, performed by means of our vector model[17] for the representation of the SPV signal. (ii) The mechanisms of photocarrier separation in space are discussed, and it is concluded that the principal one is the predominant hole drift and diffusion from the QD structure toward the substrate. (iii) The contribution of the SI GaAs substrate absorption to the SPV spectrum has been revealed and discussed employing the vector model for the SPV signal. It is found that the substrate contribution dominates the spectrum for energies below 1.0 eV and above 1.38 eV, while at intermediate energies it is overpowered by the QD and WL contributions.

The obtained results contribute to the understanding of the optical properties of multi-layer InP/GaAs QDs in prospect of their optimization for device applications. Further, this study highlights the potential of the SPV spectroscopy in the investigation of complicated nanostructures. In particular it is evidenced that this method is a valuable alternative to the optical absorption in the case of such materials.

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