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N.B.: When citing this work, cite the original article.

Original Publication:

Jan Beyer, Irina A Buyanova, Suwaree Suraprapapich, Charles Tu and Weimin Chen, Spin injection in lateral InAs quantum dot structures by optical orientation spectroscopy, 2009, Nanotechnology, (20), 37, 375401.

<http://dx.doi.org/10.1088/0957-4484/20/37/375401>

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Postprint available at: Linköping University Electronic Press

<http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-20249>

# Spin injection in lateral InAs quantum dot structures by optical orientation spectroscopy

J Beyer<sup>1</sup>, I A Buyanova<sup>1</sup>, S Suraprapapich<sup>2</sup>, C W Tu<sup>2</sup>, and W M Chen<sup>1</sup>

<sup>1</sup> Department of Physics, Chemistry and Biology, Linköping University, 58183 Linköping, Sweden

<sup>2</sup> Department of Electrical and Computer Engineering, University of California at San Diego, La Jolla, USA

**Abstract.** Optical spin injection is studied in novel laterally arranged self-assembled InAs/GaAs quantum dot structures, by using optical orientation measurements in combination with tuneable laser spectroscopy. It is shown that spins of uncorrelated free carriers are better conserved during the spin injection than the spins of correlated electrons and holes in an exciton. This is attributed to efficient spin relaxation promoted by the electron-hole exchange interaction of the excitons. Our finding suggests that separate carrier injection, such as that employed in electrical spin injection devices, can be advantageous for spin conserving injection. It is also found that spin injection efficiency decreases for free carriers with high momentum, due to acceleration of spin relaxation processes.

PACS numbers: 72.25.Fe, 72.25.Rb, 78.67.Hc

## 1. Introduction

Quantum dots (QDs) are often considered as a promising candidate for applications in a variety of novel devices with spin-enabling functionality, e.g. in spin light-emitting devices<sup>1</sup>, for spin filtering, as well as in quantum information technology<sup>2-4</sup>. Strong quantum confinement in these zero-dimensional structures allows tailoring at will their energy structure and also leads to high efficiency of optical transitions. Furthermore, discreteness of energy states in the QD systems causes suppression of spin relaxation as spin relaxation mechanisms based on the spin-orbit interaction become inhibited<sup>5</sup>. This results in extremely long spin lifetimes of carriers<sup>6-9</sup>, which can exceed 20 ms at 1K for electron spins in the InGaAs QD structures<sup>7</sup>. In(Ga)As QDs fabricated, e.g., by strain driven self-assembly are particularly attractive for these applications as they can be controllably positioned<sup>10</sup> and embedded into active device structures<sup>1,11-14</sup>. Moreover, due to major advances in fabrication, implementation of complex QD structures containing either vertically stacked or laterally aligned self-assembled QDs has recently become possible with a high degree of precision.

Among prerequisites for nanospintronic devices is the ability to prepare the confined carriers in a desired spin state. This is often accomplished non-resonantly, i.e. either by electrical injection of spin-polarized carriers<sup>1, 11-14</sup> or by optical orientation of carrier spins<sup>5</sup>. Non-resonantly refers to the fact that spin-polarized carrier populations are supplied to the QDs from the surrounding layers. Efficiency of spin conservation during this process will likely depend on the exact mechanism responsible for the injection, i.e. injection of individual carriers or exciton transfer<sup>15</sup>. The purpose of this work is to characterize spin injection efficiency of these processes in novel laterally assembled InAs/GaAs QD structures, by using optical orientation combined with photoluminescence excitation (PLE) spectroscopy. This technique allows detection of spin orientation of carriers confined in the QDs via circular polarization of photoluminescence (PL) under optical pumping of spin-oriented carriers in the surrounding layers<sup>16</sup>. It also enables a comparison of various spin depolarization mechanisms during spin

injection processes by choosing an appropriate excitation photon energy and, thus, the energy of the photo-excited carriers.

## 2. Samples and Methods

Three different structures were studied, including self-assembled single QDs (SQDs), lateral double QDs (DQDs) and QD rings (QDRs) with five to seven dots per ring – see Figs.1 (a)-(c). All structures were grown by molecular beam epitaxy (MBE) on a (001) semi-insulating GaAs substrate starting from a 300 nm GaAs buffer layer. For the SQD sample, 1.8 monolayers (ML) of InAs were deposited at 500 °C, leading to Stranski-Krastanov growth of single InAs QDs on a thin wetting layer (WL). After capping of the QDs with a 150 nm-thick GaAs layer, the quantum dot growth was repeated. The top QD layer served for atomic force microscopy (AFM) measurements whereas the embedded one was mainly responsible for the PL signal. An AFM image confirming the single dot formation is shown in figure 1(a). In the DQD sample, both InAs layers were partially capped at 470 °C by a 6 ML-thick GaAs layer. It was then followed by deposition of another 1.8 ML-thick InAs layer at the same temperature. This led to the DQDs formation, see figure 1(b). Raising growth temperature of the partial capping layer and the additional InAs-layer to 490 °C resulted in the growth of rings of dots, which was the case for the QDR sample - figure 1(c). Details of the growth procedure can be found elsewhere<sup>17</sup>. All structures were nominally undoped.

PL and PLE spectra were measured at sample temperatures between 6 K and 180 K using a tunable Ti:sapphire solid state laser as an excitation source. The PL signal was dispersed by a grating monochromator and detected by a liquid-nitrogen-cooled Ge-detector. Control of excitation and detection polarization was achieved by using quarter-wave-plates in conjunction with linear polarizers in the respective light paths. The direction of the propagating light was along the growth direction. A circular polarization degree  $P_\alpha$  for a specific polarization state  $\alpha$  of the excitation light was defined as  $P_\alpha = (I_\alpha^{\sigma^+} - I_\alpha^{\sigma^-}) / (I_\alpha^{\sigma^+} + I_\alpha^{\sigma^-})$ , where  $I_\alpha^{\sigma^+}$  and  $I_\alpha^{\sigma^-}$  denote the

PL intensities of  $\sigma^+$  and  $\sigma^-$ -polarized emission, respectively.  $\alpha$  can be one of  $\sigma^+$ ,  $\sigma^-$  or  $\sigma^x$  (linearly polarized).

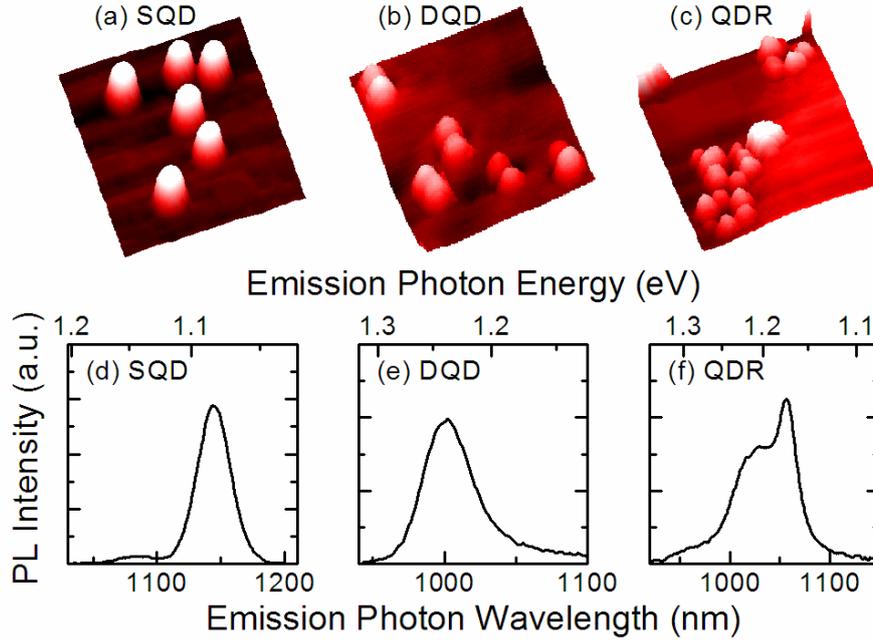


FIG 1. AFM images (a)-(c) and PL spectra (d)-(f) of the studied InAs/GaAs QD structures, as specified in the Figure. The AFM images were taken within areas of approximately  $400 \times 400 \text{ nm}^2$ . For the PL spectra, the excitation photon energy was chosen just above the WL bandgap at a measurement temperature of 6K. The PL intensity is not scaled for comparison between different structures.

### 3. Experimental results and discussion

Figures 1(d)-(f) show representative PL spectra from the three samples. Emission from the QD ground state is seen as the dominant PL peak in all spectra. The weaker PL components at the higher energies, most pronounced for the QDR sample (figure 1(f)), are attributed to recombination from the QD excited states based on state filling spectroscopy (not shown in the figure).

In the PLE measurements, the excitation wavelength was scanned continuously within an energy range corresponding to light absorption within the InGaAs WL. The spectra were detected at the energies corresponding to the maximum of the PL emission from the QD ground state and are shown by solid lines in figure 2. Two types of excitation transitions can be clearly distinguished for all structures. Free exciton absorption in the InGaAs WL involving strain-split

light hole (lh) and heavy holes (hh) valence band (VB) states gives rise to the sharp PLE peaks denoted as XL and XH, respectively. In addition, the PLE spectra contain step-like components due to band-to-band absorption transitions related to photo-excitation of free electrons and holes in the WL of a 2D character.

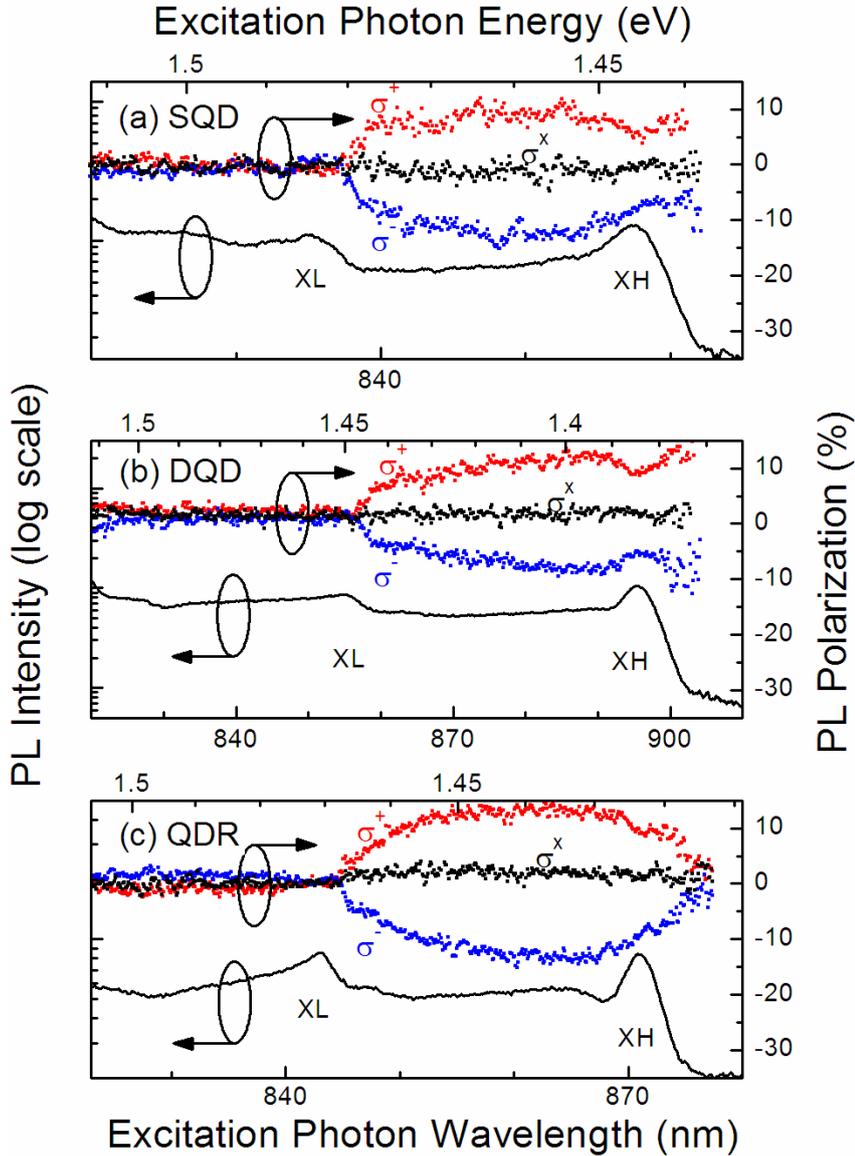


FIG. 2. PLE spectra (solid black lines) and the PL polarization (dotted lines) at 6K, detected at the peak positions of the QD ground state for the SQD (a), DQD (b) and QDR (c) samples, respectively. The polarization of the excitation light is indicated for each polarization curve. The PL intensity is not scaled for comparison between different structures.

The distinct appearance of excitonic peaks besides the free carrier continua enables us to separately study spin injection processes related to correlated and free carriers from the WL to the

QDs. Here, generation of a preferred spin orientation of excitons and charge carriers in the WL was accomplished by using a corresponding circularly polarized light excitation, i.e. by utilizing the optical spin orientation approach<sup>5</sup>. For example,  $\sigma^+$ -polarized light will create spin-down electrons and spin-up holes in the case of optical transitions involving the hh VB states, governed by the optical selection rules. This is in contrast to linearly polarized excitation when both spin orientations of carriers are simultaneously generated. Under  $\sigma^+$  (or  $\sigma^-$ ) excitation when spin polarized carriers are generated in the WL and subsequently injected into the QDs, a corresponding circular polarization can be observed in the QD PL. The polarization degree is found to strongly depend on the excitation energy and reaches approximately 10% when exciting within the WL continuum involving the VB hh states – see dotted lines in figure 2.

The observed polarization of the QD PL is a direct result of spin injection from the WL into the charged QDs. Indeed, at low temperatures PL in QDs is known to be dominated by transition of neutral and/or charged excitons. As shown by experiments<sup>6, 18</sup> and theory<sup>19</sup>, the ground state of neutral excitons in the QDs is characterized by two linearly polarized eigenstates, due to an anisotropic electron-hole exchange interaction. Under conditions of spin injection, this will lead to circular polarization quantum beats, that are quickly damped in ensemble measurements<sup>20, 21</sup>, resulting in unpolarized PL in continuous-wave experiments as presented here. Therefore, the observed PL polarization must arise from the QDs with strongly reduced exchange interaction so that the circularly polarized ground states are recovered. The probable candidates are charged dots<sup>21, 22</sup>, which may exist within the QD ensemble due to unintentional doping during the MBE growth. In the charged dots, the injected electron-hole pair forms, together with the resident carrier, a trion in which the exchange interaction is cancelled<sup>23</sup>. This restores spin polarized eigenstates, making circularly polarized luminescence possible. Moreover, the single (resident or injected) electron's spin in the charged dots can polarize nuclear spins of the host atoms in the dot via hyperfine interaction. This will create a nuclear magnetic (Overhauser) field which may in turn polarize the electron/excition spin, leading to the circularly polarized ground states<sup>24,25</sup>.

Using the circularly polarized luminescence from the trions as a measure for the degree of spin polarization of the QD ground state, which should be zero without spin injection, we can now analyze the spin injection efficiency of different injection processes. Several important conclusions, which are common for all investigated structures and are therefore of a general character, can be deduced. These conclusions are valid in spite of different volumes of QDs in the studied structures. Indeed, this difference in volumes of the QDs may lead to different carrier injection efficiency from the WL as compared with the resonant excitation of the QDs themselves, leading to a difference in PLE intensity between optical excitation at below and above WL bandgap energy. However, it does not affect the conclusions on spin injection efficiency when only the free carrier and exciton injection from the WL is considered.

First of all, spin injection efficiency of the excitons from the WL to the QDs is lower than that of free carriers. This is based on the fact that the PL polarization degree significantly decreases each time when the excitation energy is tuned resonant with the XH or XL excitonic peaks as compared with that detected under optical excitation of the corresponding free carrier continua (see figure 2). This suggests that the spins of the correlated carriers in an exciton supplied to the QDs from the InGaAs WL are less conserved than the spins of the free uncorrelated carriers. A likely cause for the observed decrease in the spin injection efficiency during the exciton transfer is fast spin relaxation between the exciton spin states mediated via exchange Coulomb interaction<sup>26</sup>. The process may be viewed as being a result of a fluctuating effective-magnetic field, which is created due to exciton motion and vanishes when the exciton center of mass momentum  $\mathbf{K}$  is equal to zero. Even though the excitons are created in our experiments under resonant excitation, i.e. close to  $\mathbf{K}=\mathbf{0}$ , scattering processes involving acoustic phonons, impurities and interfaces will populate the  $\mathbf{K}\neq\mathbf{0}$  states within a finite homogeneous linewidth. This will allow spin relaxation via the exchange interaction. The process is likely dominated by the long-range exchange interaction as the contribution of short-range exchange requires coupling between heavy and light holes<sup>26</sup> and is reduced due to a large VB splitting

caused by strain and quantum confinement in the WL. Furthermore, the higher degree of the PL polarization detected under injection of individual carriers also provides clear evidence that the uncorrelated carriers are injected directly into the QD without forming excitons first. Otherwise, the PL polarization should decrease when exciting the free carrier continuum due to additional spin loss accompanying momentum relaxation and trapping to the free exciton states, which is opposite to our experimental observations.

Secondly, spin injection efficiency decreases for hot carriers due to accelerated spin relaxation at high  $\mathbf{K}$ . This conclusion is based on the following experimental observations. (i) We notice that for excitations above the free exciton peaks, the QD PL polarization decreases gradually with increasing excitation photon energy or, in other words, when the free carriers are generated with high kinetic energy. The trend is probably most pronounced for optical transitions between the hh VB states and conduction band (CB) of the InGaAs WL – figure 2. (ii) The QD PL polarization reduces almost to zero or even reverses its sign (for the QDR structure) when the excitation energy is tuned above the XL excitonic peak (figure 2). This is instead of  $\sim 2$  times reduction in the PL polarization expected in this case as both the hh- and lh VB states participate in the optical absorption transitions with the 3:1 ratio of the corresponding oscillator strengths. The likely reason for this behavior is as follows. Due to the large valence band splitting caused by strain and quantum confinement in the WL, the electrons excited from the lh-VB states will be generated close to the bottom of the conduction band, whereas the electrons from the hh-VB states are excited with high  $\mathbf{K}$ . Therefore, even though optical orientation using e.g.  $\sigma^+$  light generates 3 times more spin-down electrons, their contribution to the total electron polarization will be significantly diminished by the accelerated spin relaxation. This tends to equalize concentrations of spin-up and spin-down electrons in the WL and results in a weak QD PL polarization as is observed experimentally. Fast spin relaxation of hot carriers is probably not surprising viewing that spin relaxation of free carriers in two-dimensional structures is usually governed by the D'yakonov-Perel' mechanism<sup>5, 27</sup>. In this case, the spin flip is assured by lifting spin degeneracy of the CB states in a crystal that lacks inversion symmetry. Due to the energy

dependence of the CB splitting, the efficiency of this mechanism substantially enhances with increasing kinetic energy (and therefore momentum  $\mathbf{K}$ )<sup>28</sup>.

In order to characterize spin injection processes at elevated temperatures, we carried out temperature-dependent optical spin orientation measurements. Representative results from the QDR sample are shown in figure 3. For the excitation photon energies below the XL peak, an overall decrease of the QD PL polarization with increasing temperature is observed, due to acceleration of spin relaxation both in the WL<sup>29</sup> and the QDs<sup>30</sup>. Nonetheless, the same trend of the enhanced spin loss during exciton transfer as compared with the free carrier injection is obeyed, at least within the investigated temperature range up to 180K, as the QD PL becomes less polarized when the XH and XL excitons were resonantly excited – see figure 3.

In the case of free carrier injection, on the other hand, with increasing temperature the QD PL polarization restores its values expected from the optical selection rules. Indeed, for  $T \geq 90\text{K}$  the detected PL polarization degree has the same sign and is about 2 times higher when spin generation is accomplished via the hh-VB – CB transitions, as compared with that observed when both lh and hh-VB states are involved - figures 3(b) and (c). This may be interpreted in terms of decreased spin conservation of carriers excited from the lh-VB states at high temperatures, which effectively restores the 3:1 ratio of the oriented spin populations in the WL CB. This decrease of spin conservation likely occurs because of the thermal population of the higher-lying CB states, which facilitates spin relaxation.

It is interesting to note that the spin injection efficiency remains nearly zero under the optical excitation at the lh-VB-CB transition energy at different measurement temperatures. This is despite of the fact that the degree in cancellation of spin polarization generated by the hh-VB-CB and lh-VB-CB excitations is known to vary with temperature, evident from the difference in spin injection efficiency by free carriers presented above. The exact physical mechanism for this observation is currently unknown, and requires further investigations.

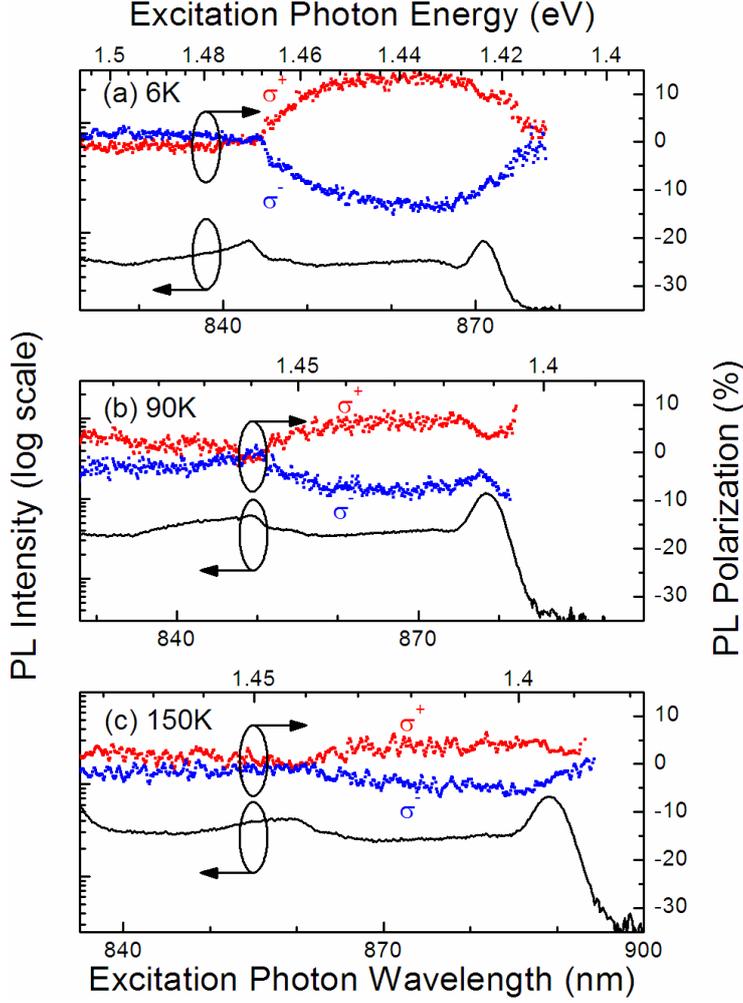


FIG. 3. PLE spectra (solid black lines) and the corresponding PL polarization (dotted lines) measured from the QDR sample at 6K (a), 90 K (b) and 150 K(c). The polarization of the excitation light is indicated for each polarization curve. The PL intensity is not scaled for comparison between different temperatures.

#### 4. Conclusions

In conclusion, we have employed optical orientation measurements combined with the PL excitation spectroscopy to evaluate efficiency of different spin injection processes from the InGaAs WL into the novel laterally-arranged InGaAs QDs. It was found that spin loss is less severe in the case of free carrier injection than that during the exciton transfer, evident from a higher degree of the QD PL polarization in the former case. This is attributed to the electron-hole exchange interaction within the exciton states that accelerates spin-flips. The same tendency was observed for all investigated structures within the investigated temperature range 6-180K, which suggests that it represents a general trend for the InGaAs-based QD systems. It was also found

that spin injection efficiency of the free carriers degrades for hot carriers with high momentum  $\mathbf{K}$ , which reflects severe spin loss driven by the physical mechanisms that are promoted at high momenta such as the D'yakonov-Perel' mechanism. Our findings suggest that separate carrier injection, such as that to be employed in electrical spin-injection devices, can be advantageous for spin conserving injection in novel quantum devices with spin functionality. They also demonstrate the need for careful optimization of nanostructure design to enhance spin performance.

### **Acknowledgements**

The financial support by the Swedish Research Council is greatly appreciated. We would like to thank V. Kalevich for useful discussion.

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