

## *Study of the optical properties in charged CdTe quantum dots*

### *Estudo das propriedades ópticas em pontos quânticos de CdTe carregados*

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#### **Abstract**

In this article we theoretically study how the presence of charge carriers added to the conduction and valence band of a spherical CdTe quantum dot affects the optical properties, especially the absorption coefficient. For this study, we simulated the interband optical absorption spectrum for incident light with circular polarizations on the left ( $\hat{e}^-$ ), on the right ( $\hat{e}^+$ ) and linear ( $\hat{e}^z$ ). We also consider Coulomb's interaction between optically excited and added carriers. Our results show that the appropriate choice of incident light polarization is an important factor in the analysis of charged quantum dots and the light with circular polarization on the left ( $\hat{e}^-$ ) was the one that presented the best results regarding the clarity of the identification of the optical absorption peaks.

**Keywords:** Quantum dots. Added charge. Optic absorption. Coulomb interaction.

#### **Resumo**

Neste artigo estudamos teoricamente como a presença de portadores de carga adicionados a banda de condução e de valência de um ponto quântico esférico de CdTe afeta as propriedades ópticas, principalmente o coeficiente de absorção. Para tal estudo, simulamos o espectro de absorção óptica interbanda, ou seja, entre a banda de valência e a banda de condução para a luz incidente com polarizações circular à esquerda ( $\hat{e}^-$ ), à direita ( $\hat{e}^+$ ) e linear ( $\hat{e}^z$ ). Consideramos também a interação de Coulomb entre os portadores opticamente excitados e os portadores adicionados. Nossos resultados mostram que a escolha apropriada da polarização da luz incidente é um fator importante na análise dos pontos quânticos carregados e a luz com polarização circular à esquerda ( $\hat{e}^-$ ) foi a que apresentou os melhores resultados quanto a clareza da identificação dos picos de absorção óptica.

**Palavras-chave:** Pontos quânticos. Carga adicionada. Absorção óptica. Interação de Coulomb.

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## Introduction

Semiconductor quantum dots (QDs) have been proposed for new and promising applications as building blocks in quantum information systems, advanced photon sources and spintronics (HOLTKEPPER; REITER; KUHN, 2018; KAIFENG *et al.*, 2017; KAIFENG; JAEHOON; KLIMOV, 2017; KOZLOV *et al.*, 2019; PRADO, 2015). In this context, the physical properties of the conduction and valence bands, such as spin relaxation and optical excitations, came into the focus of current research (HOLTKEPPER; REITER; KUHN, 2018; PRABHAKAR; MELNIK, 2016).

The present study demonstrates that the control and the precise identification of interband optical transitions can be useful for the design and characterization of these new systems. Two important problems related to interband transitions are the dependence of absorption on the polarization of incident light and the effects of electrons and holes added.

The precise control of the added charge in the QD plays an important role in the identification of the spin relaxation mechanism, in the performance of infrared detectors, and in the analysis of the magnetic properties of the diluted magnetic semiconductor QDs (PRADO *et al.*, 2003).

In this work, we consider the effects of the population of electrons and holes on the interband optical absorption spectrum in spherical QDs, considering three different polarizations of incident light, which allows us to verify which of these polarizations provides the best results in the characterization of the charged QD.

The analysis of the results will be made in terms of factors such as; the resolution of the optical absorption peaks, the added charge and the energy range involved. We present a discussion of the selection rules obtained for each polarization of the incident light and its relationship with the electronic levels involved in optical transitions.

This study provides an overview of the system's response to different polarizations of incident light. Thus, we highlight the importance of an adequate choice of factors that control the rules for selecting transitions, which, in turn, can increase or reduce the sensitivity of the response to the addition charge in QD.

We derive the interband optical selection rules and calculate the absorption spectra for the three polarizations of the incident light, providing a compact and general analysis of the system response.

## Material and methods

The  $\mathbf{k}, \mathbf{p}$  Kane-Weiler method, in Prado *et al.* (2003), was used to describe the electronic structure of a QD with a spherical confinement potential. The effects of spatial confinement on the eight-component spinors ( $\psi$ ) and on the eigenvalues ( $E$ ) are studied by solving the equation  $H_{\mathbf{k}, \mathbf{p}} \psi = E \psi$ . In Hamiltonian  $\mathbf{k}, \mathbf{p}$ , states with different symmetries are coupled even in the absence of external fields and the angular momentum  $L$  is not a good quantum number. However, neglecting the warping term, proportional to  $\mu = (\gamma_3 - \gamma_2)/2$ , the space of the Kane-Weiler Hamiltonian solution can be separated into two independent Hilbert subspaces, ( $H = \text{I}, \text{II}$ ). The state of the calculated spinor,  $\psi_{\text{I(II)}}^M$ , can be indexed using two quantum numbers:

- (i) The  $z$ -component of the angular momentum  $L_z = \hbar M$ ;
- (ii) the ordering index  $N$  for the increasing energy (LÓPEZ-RICHARD *et al.*, 2005).

The energy  $\Delta E_{h,e}(N_{e_a})$  necessary to optically excite an electron from the valence band (hole  $h$ ) to a state of the conduction band (electron  $e$ ) in the presence of added electrons ( $e_a$ ) is given by (FRANCESCHETTI; ZUNGER, 2000):

$$\Delta E_{h,e}(N_{e_a}) = (E_e - E_h - J_{h,e}) + \sum_{e_a=1}^{N_{e_a}} (J_{e,e_a} - J_{h,e_a}), \quad (1)$$

where  $E_e$  and  $E_h$  are the electron and hole energies obtained using the  $\mathbf{k}, \mathbf{p}$  Hamiltonian,  $J_{h,e}$  is the Coulomb interaction between electron and hole,  $J_{e,e_a}$  is the repulsive Coulomb interaction between the optically excited electron and the added electron and  $J_{h,e_a}$  is the attractive Coulomb interaction between the optically excited hole and the added electron.

Similarly we have for the added holes ( $h_a$ ),

$$\Delta E_{h,e}(N_{h_a}) = (E_e - E_h - J_{h,e}) + \sum_{h_a=1}^{N_{h_a}} (J_{h,h_a} - J_{h,h_a}). \quad (2)$$

Coulomb's interactions were calculated through expression,

$$V_C(r_h, r_e) = \frac{e^2}{\epsilon} \sum_{\sigma_e, \sigma_h} \iint \frac{|\psi_h(r_h, \sigma_h)|^2 |\psi_e(r_e, \sigma_e)|^2}{|r_h - r_e|} d^3 r_e d^3 r_h. \quad (3)$$

The rules of interband optical selection allow only transitions between states belonging to different Hilbert subspaces,  $I \rightarrow II$  or  $II \rightarrow I$  and can be summarized as: incident light with  $\hat{\mathbf{e}}^\pm$ ,  $\Delta M = \pm 1$  polarization and light with linear  $\hat{\mathbf{e}}^z$  polarization,  $\Delta M = 0$ . The optical absorption coefficient, in the electrical dipole approximation, can be written as

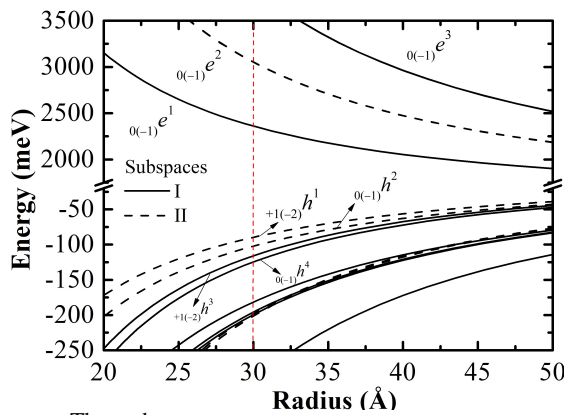
$$\alpha(\hat{\mathbf{e}}^{\pm,z}, \omega) = \frac{\alpha_0 \Gamma}{\pi} \sum_{N_h, N_e, M} \frac{|F_{n_e, M}^{n_h, M}(I, II)|^2}{(E_{N_e, M}(I) - E_{N_h, M}(II) - \hbar\omega)^2 + \Gamma^2} \times P_{E_{n_i}}^{(II)} (1 - P_{E_{n_f, M \pm 1}}^{(I)}) + \sum_{N_h, N_e, M} \{I \leftrightarrow II\}, \quad (4)$$

where  $F_{N_e, M}^{N_h, M}$  is the strength of the optical oscillator for the allowed transitions,  $\alpha_0$  is the normalization constant and  $\omega$  is the frequency of the incident light.  $P_{E_{N, M}}$  is the probability of finding an electron or hole in the state  $(N, M)$  with energy  $E(N, M)$  and  $\Gamma$  is the homogeneous scattering factor, whose the used value in this work is 30 meV.

## Results

Initially, we calculated the energy levels of a spherical CdTe QD using the Kane-Weiler  $\mathbf{k} \cdot \mathbf{p}$   $8 \times 8$  method, as a function of the radius. The results are shown in Figure 1, which shows the expected behavior of the increase in energy with the decrease in radius.

**Figure 1** – Electron and hole spectra of a CdTe spherical QD, plotted as a function of radius  $R$ . Solid (dash) line represent states arising from the Hilbert subspace I (II). Some levels have been identified considering the quantum number  $M$  and the energy ordering index  $N$  (see text for details).

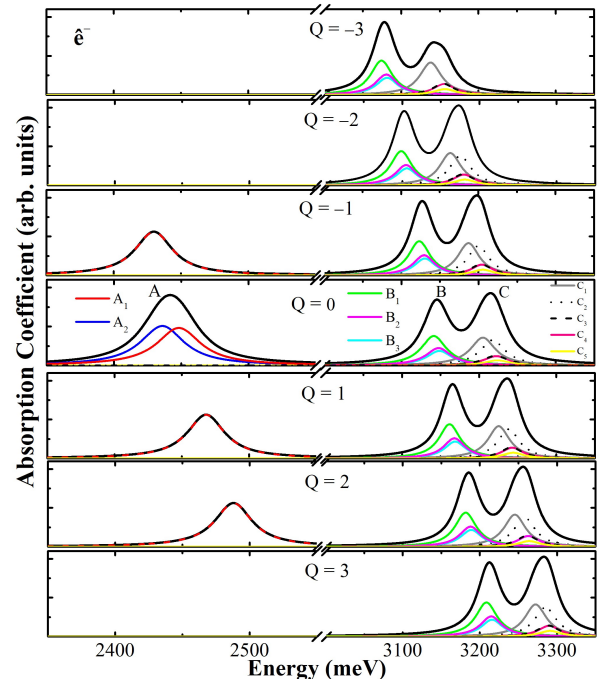


Source: The author.

The energy levels are indexed as follows,  ${}_M X^N(H)$ , where  $X$  represents the carrier, electron or hole,  $N$  and  $M$  are the quantum numbers already mentioned and  $H$  represents the two independent Hilbert subspaces. Subspace I is represented in Figure 1 by a solid line and subspace II by a dotted line. As the energy levels are degenerated in the absence of an external magnetic field, two  $M$  values appear in the indexation, for example, the first level of the conduction band we have; one energy level with  $M = 0$  and spin up and another level with the same energy,  $M = -1$  and spin down, represented by  ${}_{0(-1)}e^1(I)$ , and in the same way for the other energy levels.

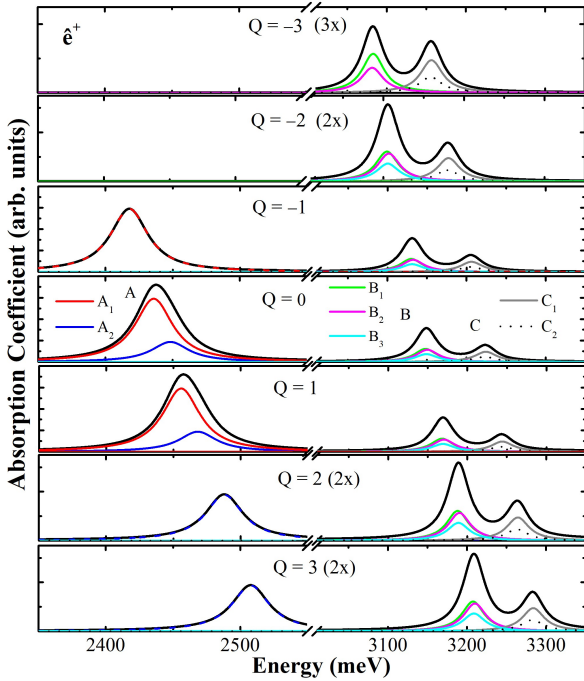
The effects of adding charge on the PQ, are studied by the term  $P_{E_{N_i, M}}^{(H_i)} (1 - P_{E_{N_f, M}}^{(H_f)})$ , on the absorption coefficient;  $P_{E_{N, M}}^{(H)}$ , being the occupation function. These statistical terms determine the probability of finding a occupied state  $(N, M)$  with  $E_{N, M}$  energy, in the initial state ( $i$ ) and an empty state ( $f$ ), which are connected according to the optical selection rules summarized above. The discussion will focus on the general characteristics of the optical response due to the addition of charges. In Figures 2, 3 and 4 illustrates the absorption spectra calculated with the equation (4), for a QD with radius 30Å (highlighted in dashed red line in Figure 1), for the polarizations of the incident light  $\hat{\mathbf{e}}^-$ ,  $\hat{\mathbf{e}}^+$  and  $\hat{\mathbf{e}}^z$ , respectively. The details of the calculation of the optical absorption are in Prado (2003).

**Figure 2** – Interband optical absorption for  $\hat{\mathbf{e}}^-$  polarization of a  $R = 30\text{Å}$  CdTe charged QD.



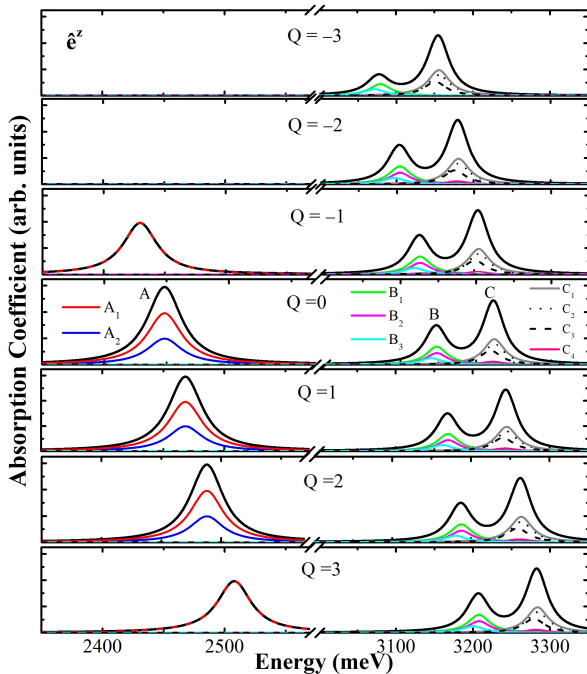
Source: The author.

**Figure 3** – Interband optical absorption for  $\hat{e}^+$  polarization of a  $R=30\text{\AA}$  CdTe charged QD. The symbols 2 $\times$  and 3 $\times$ , indicate that the absorption spectrum has been enlarged two and three times, respectively, for a better visualization.



Source: The author.

**Figure 4** – Interband optical absorption for  $\hat{e}^z$  polarization of a  $R=30\text{\AA}$  CdTe charged QD.



Source: The author.

The central panel in all figures corresponds to the absorption spectrum for a neutral QD ( $n = 0$ ), in which we can see the three peaks A, B and C, which appear in the considered energy range. The panels with  $n = -1$  (1),  $n = -2$  (2) and  $n = -3$  (3), correspond to the charged QD with one, two and three electrons (holes) in the conduction band (valence). The peaks represented in colors indicate that each of the absorption peaks is formed by more than one transition from the valence band to the conduction band, these transitions are shown in Table 1, and obey the selection rules for each polarization of the incident light.

As the energy levels are degenerate, they are occupied by carriers with spin-up and down in the conduction and valence band. When adding carriers blocking one of the allowed transitions, depending on the spin of the added carrier, so in this work we assuming that the first added carrier has spin-up polarization, the second spin-down and the third spin-up. Thus, when we add an electron ( $n = -1$ ) with spin-up in the first level of the conduction band, the QD becomes spin polarized, and the next electron to occupy this level must have spin-down, so we have a single peak of absorption, the peak  $A_1$  in Figures 2, 3 and 4. If the added carrier had spin down there would be two changes in the spectra presented, (i) in the height of the peak, that is, the intensity of the absorption light by the QD, (ii) the position of the peaks  $A_1$  and  $A_2$ , which, as can see in Figures 2 and 3, are slightly displaced when compared to  $\hat{e}^z$  polarization.

When two electrons are added in the first level of the conduction band, this state is completely occupied, corresponding to the QD without spin polarization. In this case the peak A disappears, and peaks B and C remains unchanged. For the case  $n = -3$ , the fundamental level is fully occupied and the first excited level will have an electron, the QD is again spin polarized, therefore this level can accepted an electron with spin-down and peak A also does not appear, in addition, for  $\hat{e}^-$  polarization, peak C loses  $C_2$  contribution and for  $\hat{e}^z$  polarization, peak  $C_4$  does not appear.

Similar behavior occurs when added holes in the valence band, whose influence is greater for the  $\hat{e}^-$  polarization, as can be seen in Figure 2, principally in peak A, which starts to have the  $A_1$  contribution when  $n = 1$  and 2, and when  $n = 3$ , this peak disappears, the same behavior does not happen for the other two polarizations.

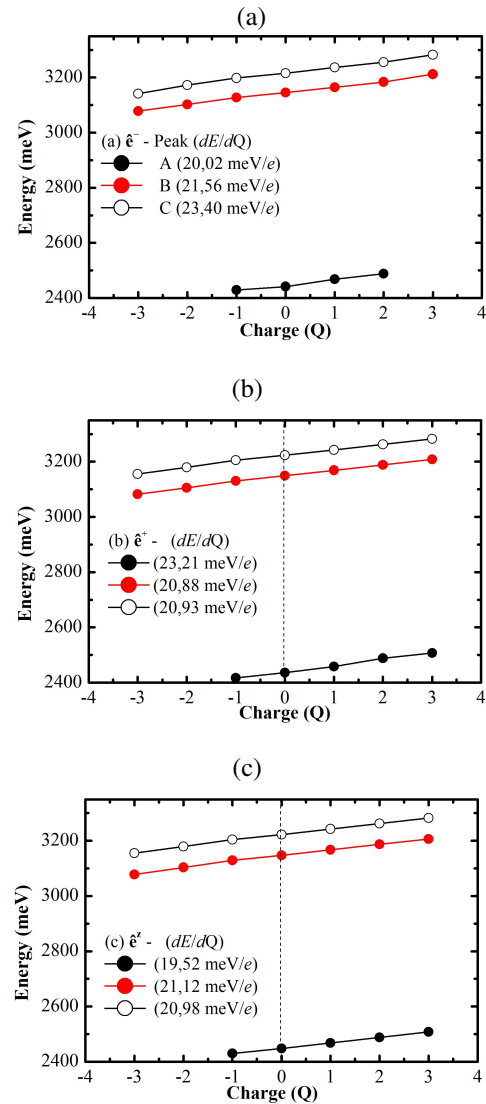
**Table 1** – Allowed transitions for the  $\hat{e}^-$ ,  $\hat{e}^+$  and  $\hat{e}^z$  incident light polarizations.

Peaks	$\hat{e}^-$	$\hat{e}^+$	$\hat{e}^z$
A <sub>1</sub>	$0h^2(\text{II}) \rightarrow -1e^1(\text{I})$	$-2h^1(\text{II}) \rightarrow -1e^1(\text{I})$	$-1h^2(\text{II}) \rightarrow -1e^1(\text{I})$
A <sub>2</sub>	$+1h^1(\text{II}) \rightarrow 0e^1(\text{I})$	$-1h^2(\text{II}) \rightarrow 0e^1(\text{I})$	$0h^2(\text{II}) \rightarrow 0e^1(\text{I})$
B <sub>1</sub>	$+1h^9(\text{I}) \rightarrow 0e^2(\text{II})$	$-1h^4(\text{I}) \rightarrow 0e^2(\text{II})$	$0h^4(\text{I}) \rightarrow 0e^2(\text{II})$
B <sub>2</sub>	$0h^4(\text{I}) \rightarrow -1e^5(\text{II})$	$-1h^4(\text{I}) \rightarrow 0e^5(\text{II})$	$-1h^4(\text{I}) \rightarrow -1e^2(\text{II})$
B <sub>3</sub>	$-1h^4(\text{I}) \rightarrow -2e^2(\text{II})$	$0h^4(\text{I}) \rightarrow +1e^2(\text{II})$	$+1h^3(\text{I}) \rightarrow +1e^2(\text{II})$
C <sub>1</sub>	$+2h^6(\text{I}) \rightarrow +1e^6(\text{II})$	$-1h^{10}(\text{I}) \rightarrow 0e^6(\text{II})$	$0h^5(\text{I}) \rightarrow 0e^6(\text{II})$
C <sub>2</sub>	$+1h^4(\text{I}) \rightarrow 0e^6(\text{II})$	$0h^{10}(\text{I}) \rightarrow +1e^2(\text{II})$	$+1h^6(\text{I}) \rightarrow +1e^6(\text{II})$
C <sub>3</sub>	$0h^9(\text{I}) \rightarrow -1e^2(\text{II})$		$-1h^5(\text{I}) \rightarrow -1e^6(\text{II})$
C <sub>4</sub>	$0h^{10}(\text{I}) \rightarrow -1e^2(\text{II})$		$-1h^5(\text{I}) \rightarrow -1e^6(\text{II})$
C <sub>5</sub>	$-1h^{10}(\text{I}) \rightarrow -2e^2(\text{II})$		

Source: The authors.

The spin polarization of the QD affects the optical response, which depends heavily on the polarization of light. The choice of the optical configuration plays a decisive role if these spectra are used to characterize the added charge and the changes that this addition causes in the optical absorption spectrum of the QD.

Figure 5 shows the behavior of the absorption energies of peaks A, B and C as a function of the added carrier, for the three polarizations of the incident light,  $\hat{e}^-$ ,  $\hat{e}^+$  and  $\hat{e}^z$ , illustrated in Figures 5(a)-(c), respectively. As can be seen from these results, the  $dE/dQ$  derivative of the absorption energy as a function of the charge, has values that vary very little in a given polarization, as well as, when we compare the three polarizations of the incident light. In addition, these results show that, the absorption energies are independent of the polarization of the incident light and have a red shift when adding electrons and holes, this effect occurs due to Coulomb interaction, equation (3), which corresponds to the second term in equations (1) and (2). In this result, we can also observe that, for the  $\hat{e}^-$  polarized light we have four energy values for peak A, while for the other two polarizations we have five values, that is, we can use the  $\hat{e}^-$  polarization of the incident light to better characterize the addition of holes. For added electrons the absorption spectra of the three polarizations are similar, however we can also consider  $\hat{e}^-$  polarization as a good tool to characterize the presence of added electrons, even if in case  $n = 1$ , peak A does not present the greatest intensity, when compared to  $\hat{e}^+$  and  $\hat{e}^z$ , but presents the most intense peaks B and C, at  $n = 1, 2$  and 3.

**Figure 5** – Interband optical absorption for  $\hat{e}^z$  polarization of a  $R = 30\text{\AA}$  CdTe charged QD.


Source: The author.

## Conclusion

In this work we show that the optical absorption measurements technique using three different polarizations of incident light can be used as a tool to study the number of electrons and holes added in a QD. We also show that the optical properties are modified by the addition of carriers in relation to the neutral QD, both in relation to the position of the peaks, and in the intensity of the absorption peaks. Coulomb's interaction between the added and the optically excited carriers explains the red shift of the peaks, and the oscillator strength values of the transitions allowed for each polarization of the incident light, that is, how strong the coupling between the levels the energy of conduction and valence band explains the intensity of the absorption peaks.

We can conclude that the appropriate choice of the incident light polarization is a key factor for the characterization of the charge in the QD and that of the optical absorption spectra calculated in this work, which presented the best results in general was for the incident light with  $\hat{e}^-$  polarization.

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## References

FRANCESCHETTI, A.; ZUNGER, A. Optical transitions in charged CdSe quantum dots. *Physical Review B*, College Park, v. 62, p. R16287, 2000.

HOLTKEMPER, M.; REITER, D. E.; KUHN, T. Influence of the quantum dot geometry on p-shell transitions in differently charged quantum dots. *Physical Review B*, College Park, v. 97, p. 075308, 2018.

KAIFENG, W.; YOUNG-SHIN, P.; JAEHOON, L.; KLIMOV, V. Towards zero-threshold optical gain using charged semiconductor quantum dots. *Nature Nanotechnology*, London, v. 12, n. 12, p. 1140-1147, 2017.

JAEHOON, L.; KLIMOV, V. Superposition Principle in Auger Recombination of Charged and Neutral Multicarrier States in Semiconductor Quantum Dots. *ACS Nano*, Washington, v. 11, n. 8, p. 8437-8447, 2017.

KOZLOV, O. V.; YOUNG-SHIN, P.; JEONGKYUN, R.; FEDIN, I.; NAKOTTE, T.; KLIMOV, V. I. Sub-single-exciton lasing using charged quantum dots coupled to a distributed feedback cavity. *Science*, [S. l.], v. 365, p. 672-675, 2019.

LÓPEZ-RICHARD, V.; ALCALDE, A. M.; PRADO, S. J.; MARQUES, G. E.; TRALLERO-GINER C. Intraband magnetoabsorption as a probing tool for the quantum dots charge. *Applied Physics Letter*, New York, v. 87, p. 231101, 2005.

PRABHAKAR, S., MELNIK, R. Spin relaxation in gas based quantum dots for security and quantum information processing applications. In: KRUCHININ, S.; BONČA, J. (ed.). *Nanomaterials for Security, NATO Science for Peace and Security Series A: chemistry and biology*. New York: Springer, 2016. p. 25-33.

PRADO, S. J.; TRALLERO-GINER, C.; ALCALDE, A. M.; LÓPEZ-RICHARD, V.; MARQUES, G. E. Optical transitions in a single CdTe spherical quantum dot. *Physical Review B*, College Park, v. 68, p. 235327, 2003.

PRADO, S. J. Influence of Strain and Spin-Orbit Interaction in a Cylindrical Quantum Point. *SEMINA Journal: Exact and Technological Sciences*, Londrina, v. 36, n. 1, p. 89-96, 2015. DOI: <http://dx.doi.org/10.5433/1679-0375.2015v36n1p89>.

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