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Strongly Optical Absorbing Nanostructures Containing Metal Quantum Dots: Theory

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Abstract

In framework of dipole approximation it is shown, that the oscillator strengths of transitions as well as the dipole moments for transitions for one-particle electron Coulomb states emerging above the spherical surface quantum dot of metal assume giant values considerably (by two orders of magnitude) exceeding the typical values of the corresponding quantities for dielectrics. It has been established that the giant values of the light absorption cross section in the nanosystems under investigation make it possible to use such nanosystems as strongly absorbing nanomaterials in a wide range of infrared waves with a wavelength that can be varied in a wide range depending in the type of contacting materials.

Keywords: One-particle electron Coulomb states; Quantum dots; Light absorption

Introduction

At present, the optical and electro optical [1-6] properties of quasi-zero-dimensional structures are extensively studied. Such structures commonly consist of spherical semiconductor, metal and insulator nanocrystals (the so-called quantum dots (QDs)) with a radius $a \approx 1\text{-}10^2$ grown in dielectric (or semiconductor) matrices. The studies in this field are motivated by the fact that such nanoheterosystems represent new promising materials for the development of new elements of nano-optoelectronics to be used, specifically, for controlling optical signals in optical computers or for manufacturing active layers of optical lasers [1] as well as new strongly absorbing nanomaterials [7]. Special attention is paid to analysis of optical properties of such nanosystems in view of its unique photoluminescence properties and the ability to effectively emit light in the visible or near infrared ranges at room temperatures [1].

Previously [8], the conditions for the localization of charge carriers near the spherical interface between the two dielectric media were analyzed. In this case, the polarization interaction of a charge carrier with the surface charge induced at the spherical interface, $U(r,a)$ depends on the relative permittivity $\varepsilon=(\varepsilon_1/\varepsilon_2)$. Here, r is the spacing between the charge carrier and the center of the dielectric QD; a is the radius of the QD; and ε_1 and ε_2 are the permittivities of the surrounding medium and of the dielectric QD embedded in the medium, respectively. For the charge carriers in motion near the dielectric QD, there are two possibilities: due to the polarization interaction $U(r,a)$, the carriers can be attracted to the QD surface (to the outer or inner surface at $\varepsilon<1$or $\varepsilon>1$, respectively), with the formation of outer [9] or inner surface states [9].

It has been show [8,9] that the formation of the above-mentioned local states is of a threshold – type nature and is possible if the radius of the dielectric QD a is large than a certain critical radius a_c:

$$a \geq a_c > b_i = 6 \left| \beta \right|^{-1} a_{Bi} \text{ where} \quad (1)$$

$$a_{Bi} = \varepsilon_i h^2 / m_i e^2, \quad (2)$$

is the Bohr radius of a charge carrier (m_i is the effective mass of the charge carrier) in a medium with the permittivity; ε_i $(i=1,2)$ is the average distance from a charge carrier localized over the planar interface in the ground state to this surface, parameter $\beta=(\varepsilon_2-\varepsilon_1)/(\varepsilon_1+\varepsilon_2)$. The interaction of the electromagnetic field with one-particle localized states of charge carriers emerging near the spherical QD–matrix interface [9,10] was studied in [7,8]. It was shown that localization of charge carriers on a spherical surface and in the bulk of QDs was manifested in different ways in the size and frequency dependences of light absorption and scattering. This paved new ways for spectroscopic investigations of such localized states in nanosystems [2-5].

Investigations in the theory of absorption and scattering of light at outer surface Coulomb states in nanosystems have not been performed as of yet; to fill this gap, a theory of interaction of the electromagnetic field with the Coulomb states of charge carriers emerging in nanosystems on the outer surface of metal QDs is developed in this study. In present work in framework of dipole approximation it is shown, that the oscillator strengths of transitions as well as the dipole moments for transitions for one-particle electron Coulomb states emerging above the spherical surface QDs of metal assume giant values considerably (by two orders of magnitude) exceeding the typical values of the corresponding quantities for dielectrics. It has been established that the giant values of the light absorption cross section in the nanosystems under investigation make it possible to use such nanosystems as strongly absorbing nanomaterials in a wide range of infrared waves with a.

Oscillator Strengths and Dipole Moments of Transitions in Nanosystems

Let us consider model of a quasi-zero-dimensional system, viz., a neutral spherical insulator QD of radius a with permittivity ε_2 surrounded by a medium with permittivity ε_1 (such that relative permittivity is $\varepsilon=(\varepsilon_1/\varepsilon_2) << 1$). An electron (e) with an effective mass m_1 is localized over a spherical interface (QD-dielectric matrix) (the electron moves in a dielectric matrix with permittivity ε_1). The fact that

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all characteristic sizes of the problem (a and b_1) are considerably larger than atomic spacing a_0 makes it possible to consider the motion of quasi-particles in the nanoparticle in the effective mass approximation [8]. In ref. [8], the energy spectrum of outer surface states of a quasi-particle, which appear over a spherical interface (QD-dielectric matrix) (for ε<<1) was investigated, as well as its dependence on radius a of the QD under the conditions when the polarization interaction of the charge carrier with the spherical interface between the two media plays the leading role. It was shown that the spectrum of the outer surface states of the quasi-particle upon an increase in nanoparticle radius a such that

$$S=(a/b_1) >> n^2 \quad (3)$$

is transformed into the spectrum of the Coulomb form

$$E_{nl}(S) = -\frac{9}{4n^2} + \frac{L^2}{S^2} \quad (4)$$

where n and l are principal and orbital quantum numbers, $L^2=l(l+1)$. Here, we are using the energy units $(Ry/36)=(/2(\mathrm{h}^2/2\, m_1\, b_1^2)$.

In the frequency range $\omega_{nl}(S)=(E_{nl}(S)/\hbar)$ corresponding to Coulomb states (n, l) (4) an electron of a QD localized above the surface in the QD of radius S (3), the wavelength of the light wave considerably exceeds the sizes of these states ($\approx b_1$ (1)). Therefore, the behavior of such Coulomb states in the electromagnetic field is successfully described by the dipole approximation [7]. In this case, the dipole moment operator for a charge carrier in the QD has the form [10]

$$\boldsymbol{D}(r)=(1\text{-}a/r)^3)\, e\, r \quad (5)$$

where $\boldsymbol{r}$ is the radius vector determining the distance between the charge carrier and the center of the QD.

To estimate the dipole moment $D_{2,1,1,0}(a)$ it is sufficient to consider the transition between the lowermost Coulomb states (4) (e.g., between Coulomb ground state $1s\rangle=(n=1,\ l=0)$ and $2p\rangle=(n=2,\ l=1)$ Coulomb state). A transition between such states is allowed by the selection rules in the Coulomb field (in this case, the principal quantum number n changes arbitrarily, while the orbital quantum number l changes by unity) [7].

Using relations (5), we can write the expression of the dipole moment of the transition [7]:

$$D_{2,1;1,0}(S)=\langle 1_S \mid D(r) \mid 2p\rangle=((27S^2+36S+20)/2\,(27S^2+54S+36))\, eb_1 \quad (6)$$

in QDs with radii S satisfying inequality (3).

The oscillator strength of the transition of a charge carrier with effective mass m_1 from ground state $1s$ to state $2p$ assumes the form [7,8]

$$f_{2,1;1,0}(S) = \frac{2m_1}{\hbar e^2}\left[\omega_{2,1}(S)-\omega_{1,0}(S)\right]\left|D_{2,1;1,0}(S)\right|^2 \quad (7)$$

where $\hbar\omega_{2,1}(S) = E_{2,1}(S)$ and $\hbar\omega_{1,0}(S) = E_{1.0}(S)$ are the energies of Coulomb levels $2p$ and $1s$, respectively. With allowance for formulas (4) and (6), we can write the oscillator strength (7) of the transition in the form

$$f_{2,1;1,0}(S)=4^{-1}((27/16)+(2/S^2))\,((27S^2+36S+20)/(27S^2+54S+36))^2 \quad (8)$$

Absorption of Light at Coulomb States in Nanosystems

The cross section of light absorption on the spherical surface of a QD of radius a can be expressed in terms of its polarizability $A''(\omega,a)$ [8]:

$$\sigma_{abs}(\omega,a)=4\pi(\omega/c)A''(\omega,a) \quad (9)$$

where ω is the frequency of the external electromagnetic field and c is the speed of light in vacuum. At temperatures

$$T<(E_b/k) \quad (10)$$

lower than the binding energy $E_b(S)=E_{nl}=(S)$ (4) of the Coulomb states (n, l) (4) (where k is the Boltzmann constant), the polarizability of a charged QD can be determined if we treat the QD as a giant ion [7]. The main contribution to polarizability $A''(\omega,a)$ Ain this case comes from transitions in the discrete spectrum of such Coulomb states. Separating in polarizability $A''(\omega,a)$ the contribution from only one resonant term corresponding to the transition between the ground $1s$ and $2p$ Coulomb states, we can write polarizability $A''(\omega,a)$ of the QD in the form [7]

$$A''(\omega,a) = \frac{e^2}{m_1}\frac{f_{2,1;1,0}(a)}{\omega_{2,1}^2(a)-\omega^2-i\omega\Gamma_{2,1}(a)}, \quad (11)$$

where $\Gamma_{2,1}(a)$ is the width of the Coulomb $2p$ level.

Assuming that frequency ω of the light wave differs significantly from resonance frequency $\omega_{2,1}(a)$ (4) of the Coulomb $2p$ state and that broadening $\Gamma_{2,1}(a)$ of level $2p$ is small $\Gamma_{2,1}(a)/\omega_{2,1}(a)<<1$ ([8]), we obtain the following expression for the qualitative estimate of polarizability $A''(\omega,a)$ (11) of the QD with allowance for eqn.(4).

$$A^*(S) = \frac{24\varepsilon_1 f_{2,1;1,0}(S)}{(-(9/16)+(2/S^2))^2}b_1^3 \quad (12)$$

We can now write the expression for the cross section of elastic scattering of an electromagnetic wave of frequency an QD of radius S (3) [8]:

$$\sigma_{SC}(\omega)=2^7\cdot 3^{-1}\pi^3(\omega/c)^4\left|A''(\omega)\right|^2 \quad (13)$$

Results and Discussion

The outer surface Coulomb states of electrons under investigation, which are localized over a spherical of metal QDs of radii a (3), can be studied in the processes of absorption (and emission) on transitions (n',l') (n,l) with frequencies $\omega_{n,l}^{n',l'}(a)=\left|E_{n',l'}(a)-E_{n,l}(a)\right|/\hbar$, which lie in the infrared spectral region in accordance with (4). Let us estimate absorption cross sections $\sigma_{abs}(\omega,a)$ (9) and scattering cross sections $\sigma_{SC}(\omega,a)$ (13) for light at the above-mentioned Coulomb states of the electron localized over a spherical surface QDs of metal of radii a (3), in the case of singled-out transition ($|1s\rangle\rightarrow|2p\rangle$). The estimates of oscillator strength of the transition $f_{2,1;1,0}(S)$ (8), dipole moment of the transition $D_{2,1;1,0}(S)$ (6), polarizability $A''(\omega, a)$ (11), and cross section $\sigma_{abs}(\omega,a)$ (9) of absorption of a light wave with frequency ω (in this case, ratio $(\omega/\omega_{2,1}(S))^2=9\ 10^{-2}$ and the wave frequency ω lies in the infrared region) at the above Coulomb states of the electron appearing over a spherical surface (QD of metal – matrix silicate glass) are given in the table. If we take into account the fact (see the table) that the oscillator strength $f_{2,1;1,0}\approx 0.4$ and the dipole moment $D_{2,1;1,0}\approx 1.85$ (where $(D_0 = e\,\text{Å})$, (Debye)) of the transition over a spherical surface QDs of metal of radii a=10 nm assume giant values (exceeding the typical values of oscillator strength and dipole moments in matrix silicate glass by two orders of magnitude [1-5] and dipole transitions between the nearest Coulomb levels E_{nl} (a) (4) in QDs in the electromagnetic field are allowed by the selection rules with a change (or preservation) of principal quantum number n and with a change in orbital quantum number l by unity [7], the quasi-zero-dimensional nanosystems under investigation are obviously strongly absorbing nanostructures for infrared radiation.

The estimates given in the table lead to the conclusion that the

cross sections of light absorption in QD of radius a=10 nm attains giant values $\sigma_{abs}(,a) \approx 10^{-17}cm^2$. This value of $\sigma_{abs}(\omega,a)$ (9) is seven orders of magnitude higher than typical values of atomic absorption cross sections [7,8]. Since the scattering cross section $\sigma_{abs}(,a)$ (13) is negligibly small as compared to the corresponding value of absorption cross section $\sigma_{abs}(\omega,a)$ (9) ($(\sigma_{cs}/_{abs}) \approx 10^{-12}$), the value of $\sigma_{cs}(\omega,a)$ is not given in the table.

Conclusions

Thus, we have shown using the dipole approximation that the oscillator strengths of transitions as well as the dipole moments for transitions for one-particle electron Coulomb states emerging in over a spherical surface (QD of metal-matrix silicate glass) assume giant values considerably (by two orders of magnitude) exceeding the typical values of the corresponding quantities for dielectric matrices. It has been established that the giant values of the light absorption cross section in the nanosystems under investigation make it possible to use such nanosystems as strongly absorbing nanomaterials in a wide range of infrared waves with a wavelength that can be varied in a wide range depending in the type of contacting materials.

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