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IIIrd Generation Solar Cell

Paweł Kwaśnicki

Abstract

Light harvesting for generation of electric energy is one of the most important research topics in applied sciences. First, for an efficient harvesting one needs a material with a broad light absorption window having a strong overlap with the sunlight spectrum. Second, one needs an efficient conversion of photoexcited carriers into produced current or voltage which can be used for applied purposes. The maximum light conversion coefficient in semiconductor systems is designated by so called Shockley-Queisser law, which is around 32% for an optimal bandgap value of 1,2–1,3 eV. However the efficiency may be increased using a solutions based on semiconductor nano materials such as quantum dots. Solar cells based on such a structures are included in the group of 3rd generation solar sell. 3rd generation solar cell encompasses multiple materials as a base of cell, such as: perovskite, organic, polymers and biomimetics. The most promising and in the same time most discussed are quantum dots and perovskite. Both material has a potential to revolutionize the solar cell industry due to their wide absorption range and high conversion coefficient. Nonetheless before the most common used material in photovoltaic namely silicon is replace one must overcome few major issues such as: stability and lifetime for at least 5 to 10 years or more, manufacturing process for a large surfaces and low production cost as well as recycling after the time of optimal use.

Keywords: DSSC, QDSC, transparent PV

1. Introduction

Light harvesting for generation of electric energy is one of the most important research topics in applied sciences. First, for an efficient harvesting one needs a material with a broad light absorption window having a strong overlap with the sunlight spectrum. Second, one needs an efficient conversion of photoexcited carriers into produced current or voltage which can be used for applied purposes. The maximum light conversion coefficient in semiconductor systems is designated by so called Shockley-Queisser law, which is around 32% for an optimal bandgap value of 1.2–1.3 eV. However the efficiency may be increased using a solutions based on semiconductor nano materials such as quantum dots. Solar cells based on such a structures are included in the group of 3rd generation solar cell. 3rd generation solar cell encompasses multiple materials as a base of cell, such as: perovskite, organic, polymers and biomimetics. The most promising and in the same time most discussed are quantum dots and perovskite. Both material has a potential to revolutionize the solar cell industry due to their wide absorption range and high conversion coefficient. Nonetheless before the most common used material in photovoltaic namely silicon is replace one must overcome few major issues such as: stability and lifetime for at least 5 to 10 years or more, manufacturing process for a large surfaces and low production cost as well as recycling after the time of optimal use.

In this chapter we focus on two most promising material for photovoltaic application. The basic overview of organometallic properties of perovskites and quantum dots from the point of view of photovoltaics and formulation description of the electronic structure in the form of a simplified effective Hamiltonian as an approximation of a tight tie will be presented. The electronic structure plays a key role in the photovoltaic effect and is responsible for the high efficiency of the effect. Additionally perovskites or quantum dots show the spin-orbit coupling in the general form, this coupling can increase the carrier's lifetime - the quantity important for solar cell applications.

2. Perovskites for photovoltaics

Some perovskite-structured oxides have an internal electrical field, which plays an important role as it leads to the separation of electrons and holes generated in the process of light absorption. These oxides have the general structure of the ABO_3 type. In general, there are quite a few different materials called perovskites, but the crystalline structure for all perovskites is similar. Perovskite oxides and, above all, organometallic halogen perovskites play an important role for photoelectronics and photovoltaics. Nonetheless perovskite oxides turned out to be inefficient in terms of photovoltaics. The interest in perovskite materials increased significantly towards the end of the last year a decade, when a series of works appeared showing the possibility of increasing efficiency in organometallic perovskites [1]. It turned out that there was a fairly broad class organometallic halide perovskites of the type $CH_3NH_3PbX_3$ (X = I, Br, Cl), which show promising properties from the photovoltaic point of view. Although the first results gave relatively low photovoltaic efficiency, however this efficiency is quite fast it grew with new research. Besides, the conducted research did not show any significant restriction on the upper limit of the photovoltaic efficiency organometallic perovskites, which now reaches over 20%, which in turn gives hope for its further growth. The main advantages of organometallic halide perovskites are their relatively low levels price and relatively simple technology, which makes these materials competitive. Recent research results show that the efficiency of the laudatory prototypes of perovskite solar cell are already equalled and even exceeded the silicon based solar cell. Hence the great interest these materials from the point of view of application in photovoltaic cells [2, 3]. Of course, these materials also have weaknesses. One of these weaknesses is the lead toxicity they contain. The second is quite rapid degradation resulting from the sensitivity of photovoltaic cells based on them on humidity and the effect of ultraviolet radiation to which they are exposed. Therefore, the main lines of current research are not only aimed at further increasing efficiency photovoltaic, but also removing these undesirable weaknesses. As mentioned for photovoltaic the most interesting and promising are halide perovskites, the crystal structure of these materials has the general form ABX_3, where A is the cation of the methylammonium group CH3NH3 for organometallic halide perovskites (metal cation for oxides), B is the metal cation Pb or Sn (the smaller metal cation in the case of oxides), while X is a Cl, Br or I anion for halide perovskites (O for oxides). The unit cell of the ABX3 perovskite crystal in the cubic phase is shown in **Figure 1**.

One of the most promising materials is a perovskite with the chemical composition $CH_3NH_3PbI_3$, because in this case the photovoltaic efficiency turned out to be the highest in this class of materials. It is worth noting, however, that the class of organometallic perovskites is in fact quite rich and contains many elements, which allows the use not only of single perovskites, but their more complex structures, e.g. double perovskites or systems composed of various materials [4]. The high photoelectric efficiency of organometallic perovskites is related to their electronic properties.

This material is a semiconductor with a band gap width of about 1.6 eV. The light absorption coefficient is very high while energy losses associated with the possibility of non-radiative electron processes (e.g., electron–hole recombination by phonons) are relatively low. Moreover, the mobility of the carriers (electrons and holes) in these perovskite materials is quite high due to the low effective mass of the carriers. All these properties underlie high photovoltaic efficiency. On the other hand, the physical mechanisms underlying these properties are not yet fully researched and elucidated.

The excellent photovoltaic properties of perovskites are related to their electronic structure, in particular to the quantum states of electrons and holes in the conduction and valence bands, respectively. In the case of organometallic halide perovskites these properties are related to the organic CH_3NH_3 positive ion and its orientation with respect to the crystallographic axes [5].

Even better results using perovskite material from energy harvesting point of view may be achieved using hybrid structure. Recent discovery by the group of Prof. Miyasaka of a highly efficient light-to-voltage conversion in hybrid organic–inorganic perovskites [6] made these material promising elements for photovoltaics, especially taking into account simple low-cost fabrication technology. The basic structure of the perovskite-based solar cell is presented in **Figure 2**.
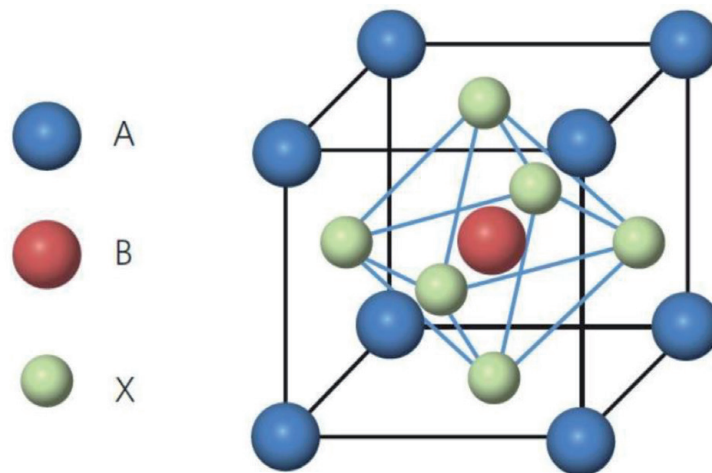

Figure 1.
Perovskite crystal unit cell, a - large cation (methylammonium group CH3NH3), B - smaller cation (Pb or Sn), X - anion (I, Br or Cl).
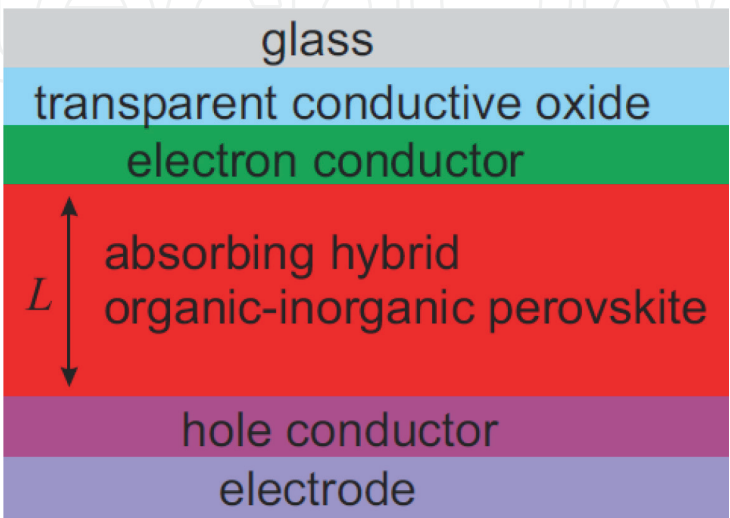

Figure 2.
Schematic picture of a hybrid organic–inorganic perovskite solar cell. (figure source: USA). Department of Energy website.

The first two compounds of this family investigated by Prof. Miyasaka group, that is $CH_3NH_3PbBr_3$ and $CH_3NH_3PbI_3$, deposited on the TiO_2 surface, demonstrated a good power conversion efficiency η higher than 3% (now reaching as high as 20%, similar to that in best conventional semiconductor-based solar cell) accompanied by the open-circle voltage Voc higher that 0.6 V and generated current density Jsc higher than 5.5 mA/cm^2. These results of Prof. Miyasaka group attracted a great deal of attention and caused a tide of research in this field.

These compounds belong to the family of perovskite structures, similar to the high- temperature superconductors, where the main element is represented by Cu-O octahedrons. Although some structure elements of these groups of materials are similar, their physical properties are very different. In general, all the perovskites are known for formation of different structures and a variety of temperature-induced structural transitions.

Due to a large variety of the organic cations, the entire family of hybrid organic-inorganic perovskites potentially contains more than 1000 members [7], all different in their properties. Structure-wise, the main element of these compounds as presented in two-dimensional projection in **Figure 3** is an octahedron built by metal and halogen ions, these elements are surrounded by organic layers.

Despite several years of extensive research efforts, many microscopic properties, which can help in the understanding of the high photovoltaic efficiency in these compounds, remain unknown. This holds true even for $CH_3NH_3PbI_3$ and
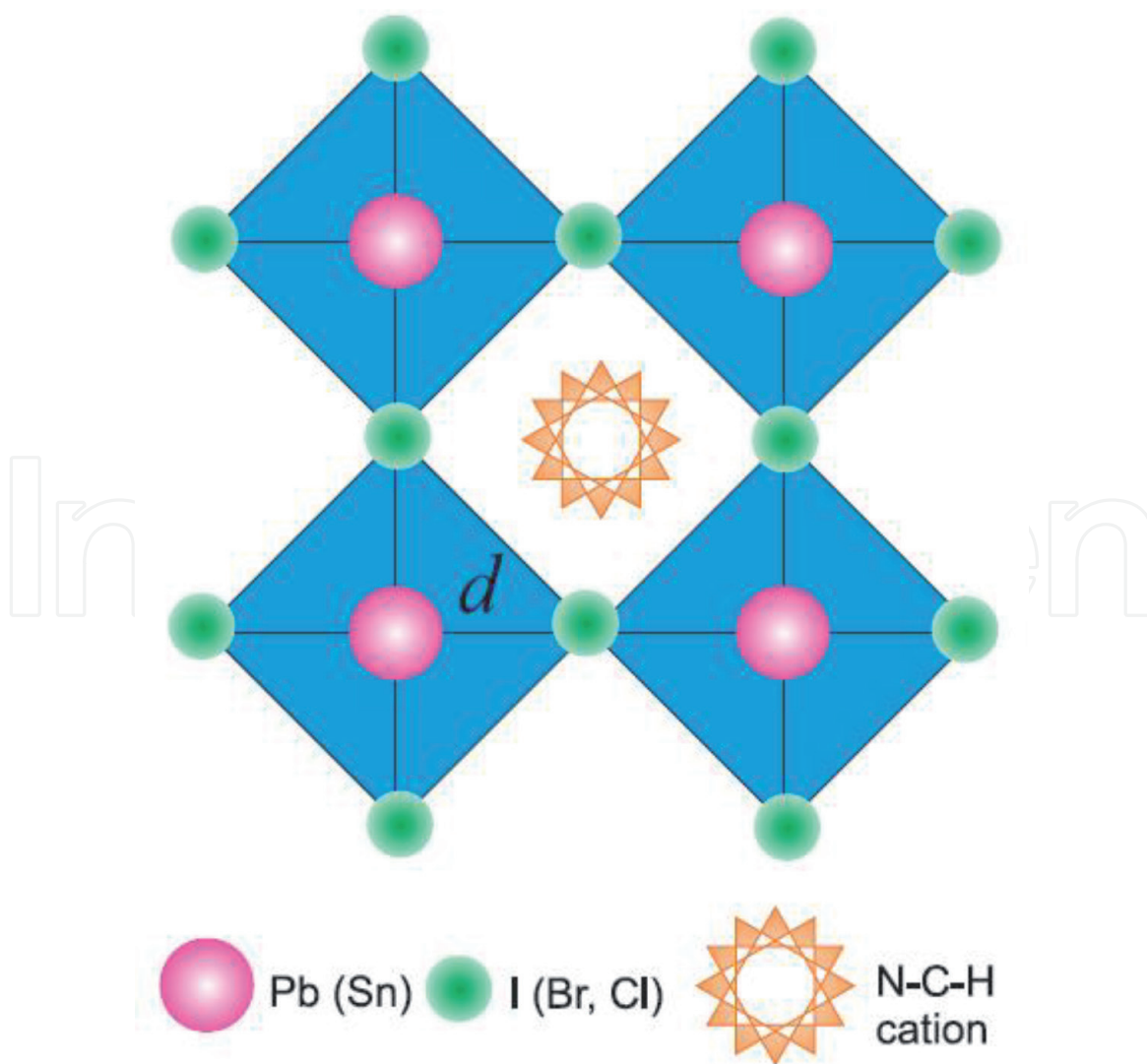

Figure 3.
Typical crystal structure of hybrid organic–inorganic perovskite compounds.

$HC(NH_2)_2PbI_3$ - the leaders of the race for the low-cost high photovoltaic efficiency. Electrical properties such as conductivity of these compounds can strongly depend on the temperature since due to a relatively soft lattice, various structural phase transitions occur in the temperature range of the order of 100 K - the property common for all perovskite structures, as mentioned above. Here we review and analyze properties of these materials in the context of their applicability for photovoltaics and connect these properties to the processes related to their possibly high efficiency.

Typical hybrid perovskite structure has the known form of a vertex-sharing networks of BX6 octahedrons as shown in **Figure 3**, which can be modeled as negatively charged PbI^-_3 elements. The bandstructure and optical properties are due mainly to the metal and halogen orbitals. Mutual overlap of these orbitals determines the matrix elements of interatomic hopping and, in turn, the band structure, corresponding to direct-band semiconductors with the bandgap Eg. This gap can be evaluated by different techniques. The overlap of the orbitals forms a relatively small effective mass of the carriers as well as the optical properties of these systems.

The sunlight has the following physical properties of our interest. Its spectrum corresponds to maximum in the green light region at photon energies $\hbar \omega$ close to 2.2 eV. This implies that the main contribution to the absorption in hybrid perovskites starts from the infrared part of the sunlight spectrum and its intense visible and ultraviolet parts can be used efficiently. The specific intensity of sunlight at the Earth's surface at a sunny day is of the order of 1000 W/m^2 corresponding to a relatively strong electric field of the order of 10^2–10^3 V/m. To understand the performance of a photovoltaic cell, one can establish that the energy flux of 1000 W/m^2 corresponds to the photon flux of the order of 10^{18} s^{-1} cm^{-2}. Being totally absorbed in a layer of the thickness of L $\sim$ 10 μm (10^{-3} cm), this photon flux corresponds to the free carrier bulk (3D) generation rate of the order of 10^{21} s^{-1} cm^{-3}; corresponding to two-dimensional (2D) concentration injection rate 10^{18} s^{-1} cm^{-2}. Although the exact electrodynamics of solar cells is very complex [8], by using the Gauss theorem for the electric field produced by charge distribution, one can obtain that this concentration corresponds to the rate of the voltage generation. The resulting bias V is of the order of $(dV/dt) \times G^{-1}$, where G^{-1} is the characteristic time corresponding to the relaxation of the optical injection, e.g. by electron–hole recombination or trapping carriers by lattice defects, leading to V $\sim 10^8) \times G^{-1}$. With the reasonable estimation of $G^{-1} \sim 10^{-9}$ s the value of V $\sim$ 0,1 V. Simultaneously, current J produced in such a sample of S = 1 cm^2 area, is of the order of 0,1 A. These numbers demonstrate that the first perovskite solar cells produced voltage close the maximal one, although the generated current was somewhat less than this basic estimate.

The light-induced transitions produce electron–hole pairs in the energy interval E > Eg, where Eg is the fundamental gap at the R − point of the Brillouin zone. The fundamental gap can be understood from the Coulomb energy arguments for the energy necessary to transfer electron from halogen to the IV-group heavy ion.

A qualitative plot of injected distribution of electrons in the energy representation is presented in **Figure 4**. This strongly nonequilibrium distribution then relaxes to the quasi equilibrium which, as we will see below, determines the performance of the photovoltaic devices. The relaxation process is mainly attributed to the multiple emissions of phonons.

The energy relaxation processes are understood even less sufficiently than the origin of the carrier's finite mobility. Indeed, due to a complex unit cell structure, crystals possess a large variety of phonon branches (of the order of 100) of different nature and symmetry. Here we propose a simple picture of the relaxation due to electron–phonon coupling. The analysis done in [9] shows that coupling to acoustic
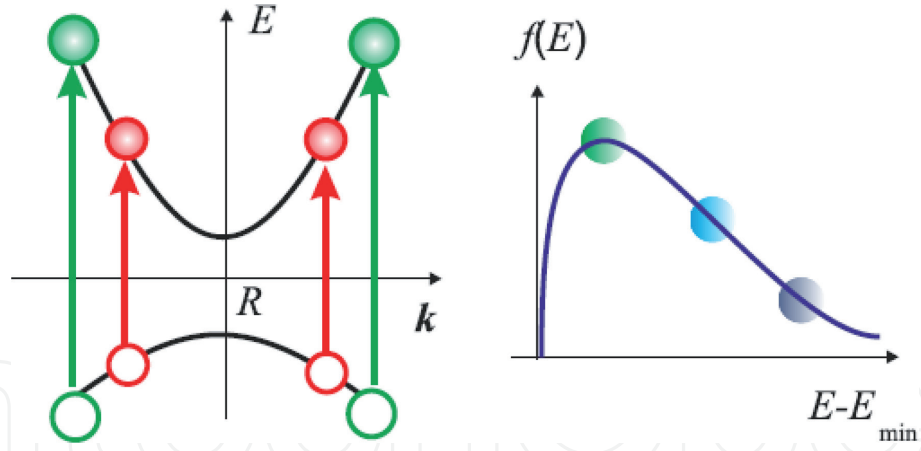
Figure 4.
Interband transitions caused by different photons, and electron distribution over the energy states, as injected. The behavior of the distribution at energies close to the minimum of the conduction band Ec is due to small density of states $\sim \sqrt{E - E_c}$ while the high-energy behavior is mainly determined by decrease in the spectral density of the incident light. Emin corresponds to the bottom of the conduction band.

phonons (with the frequency linear in the momentum) would lead to high mobilities of the order of $10^3 \, \mathrm{cm}^2 \, \mathrm{V}^{-1} \, \mathrm{s}^{-1}$ and, therefore, this coupling is not the limiting factor for the observed low mobilities. Therefore, we concentrate on the relevant coupling to optical phonons. The coupling is due to the asymmetry of the field and change of the hopping integrals due to change in the interatomic distances. The value of the deformation potential constant is attributed to two main effects. First effect is the change in the site energy, corresponding to atomic displacement in the crystal field formed by its interaction with surrounding ions. Second effect is the changing in the overlap transfer integrals between the iodine and the lead orbitals, contributing to the electron energy as well.

The energy relaxation of the photoexcited electrons due to electron–phonon coupling with optical phonons, is relatively fast and occurs on the time scale of the order of 10 ps. This fast relaxation demonstrates that a thermalized room-temperature energy distribution is quickly produced. As a result, the performance of the photovoltaic elements with typical involved time scales of the order of 1–10 ns, is determined by the thermalized distributions, where the static local defects, either charged or not, structural disorder, and low-frequency optical phonons can play a role for the kinetics of the carriers distributions. The relation of these energy relaxation processes to the photovoltaic performance of real solar cells needs experimental studies and remains to be investigated.

The light absorption is efficient due to the band structure of perovskite materials having a direct bandgap close to 1.5 eV in the vertex point of the Brillouin zone. As a result, almost the entire sunlight spectrum can be absorbed. The efficiency of the absorption, in addition, is enhanced by relatively large momentum matrix elements between the group-IV heavy metal and halogen atoms resulting from their spatial overlap what makes perovskite material promising material for III generation of photovoltaic.

3. Quantum dots for photovoltaics

Quantum dots are one of the most interesting objects in modern fundamental and applied solid state physics, including applications for photovoltaics systems [10]. Typical sizes of the quantum dots of the order of 10 nanometers determine the majority of their physical properties, including the spectra of light

absorption and properties of light-injected carries. These spectra determine the application of quantum dots in photogalvanics and photovoltaics systems. In contrast to bulk materials, where free electron–hole pairs can be produced optically, a strong confinement of carriers in quantum dots and resulting interaction between them leads to formation of exciton-like states. This effect qualitatively modifies optical properties of quantum dots and can make them useful for solar cell elements applications [10].

Various types of quantum dots can be used in photovoltaics: semiconductor polycrystalline and granular materials, quantum dots obtained by epitaxial methods or from colloidal solutions, nanoparticles of organic dyes. There are also a number of possibilities for the architecture of photovoltaic cells. Their common feature is that the phenomenon of multiple exciton excitation in dots a is used the energized charges (electrons and holes) are conducted to the electrodes in various ways, ensuring, however, their spatial separation. One possibility is to use scatter dots.

in a conductive material (e.g. in organic polymers). With the appropriate concentration of the dots, the discharge of the charge from the dots to the electrodes can be accomplished by coupling between the quantum dots. For regular networks of dots (single, double, or three-dimensional), discrete states of dots are transformed into mini-electron bands, ensuring charge transport. Photovoltaic cells with the use of regular quantum dot networks and their electronic mini-band structure (also called intermediate bands) have become one of the important directions in the development of photovoltaics. The essence of this type of solution is the fact that in the area between the electrodes in the p-n junction there is a layer containing dots quantum between which the distance is so small that an intermediate band is created in this area during the energy gap. This allows the use of low-energy photons (with energy lower than the width of the output semiconductor gap) to generate electrons in the conduction band and holes in the valence band. This is due to optical transitions from the valence band to the intermediate band and from the intermediate band to the conduction band. An important element is also that the recombination processes are in the case of the intermediate band much less likely than in the case of isolated quantum dots. In this case, it is enough for the wave functions of the dots to be quite delocalized. This can be achieved in systems with complexes quantum suppositories instead of regular networks.

Semiconductor quantum dots are usually produced of two types of binary materials. First type is usually referred to as III-V semiconductors, where one of the elements belongs to the III group of the periodic table of elements and the other belongs to the V group such as GaAs, InAs, InSb, and similar ones. The other group, named in the same way, is the II-VI semiconductors such as ZnS, ZnSe, CdS, and similar ones. In addition, coated quantum dots, where the core and the coating layer are made of different materials of the same (usually II-VI) type can be produced and used for different applications. Conventionally produced quantum dots show a variety of sizes and shapes. On one hand, this variety of quantum dot geometries extends the ability to use them for various applications, including photovoltaics. On the other hand, this variety hampers controllability of their applications. This circumstance should be taken into account in the analysis of all applications quantum dots.

Direct calculations of properties of quantum dots are very difficult, simple, but still highly efficient, approach relies on employing of the effective mass approximation with the Hamiltonian

$$H_0 = \frac{\hbar^2 k^2}{2m^*} + U(r)$$

where $\hbar^2 k^2 / 2\,m^*$ is the kinetic energy, k is the electron momentum, m_ is the electron effective mass, and U (r) is the effective confining potential of the quantum dot. For model calculations the anisotropic oscillator form of the confinement

$$U(R) = \frac{m^*}{2}\left(\Omega_x^2 x^2 + \Omega_y^2 y^2 + \Omega_z^2 z^2\right)$$

where Ω i are the corresponding frequencies, is assumed. One-dimensional representation of this potential and corresponding wave functions are shown in **Figure 5**. This form, being useful for basic understanding, especially of the ground state of the system, has strong limitations for the analysis of applications of quantum dots in photovoltaics and photogalvanics, where highly excited states are involved.

Another form of the potential is given by:

$$U(r) = 0 \text{ for r inside the quantum dot}$$

$$U(r) = \infty \text{ for r outside the quantum dot}$$

and determines the quantum dot shape. Usual model shapes of quantum dot are ellipsoidal (in simple realizations, spherical) or coin-like cylindrical with the radius much larger than the width, as shown in **Figure 6**.

It is well-known that in semiconductors, although the band electron velocities, being of the order of v ∼ 10^8 cm/s, are much smaller than the speed of light c = 3 × 10^{10} cm/s, the relativistic effects, dependent on the v/c ratio, should be taken into account. In both types of semiconductors these relativistic effects lead to a coupling between electron momentum and electron spin, which appears due to the effect of the electric field in the unit cell of a binary semiconductor without inversion symmetry.

The Hamiltonian describing the spin-orbit coupling in bulk III-V materials has the form

$$H_{SO} = \alpha_D\left(\sigma\kappa\right)$$

presenting the Dresselhaus realization of the spin-orbit coupling [11]. In this Hamiltonian σ is the vector of Pauli matrices, and κ is defined by.

$$\kappa_x = k_x(k_y^2 - k_z^2).$$

with the cyclic permutation of the indices defining the other two components. The coupling constant α_D being of the order of 10 eVA^3 leads to spin splitting of the band electron states of the order of 1 meV at electron concentrations of the order of 10^{18} cm^{-3}.

For II-VI compounds the bulk Hamiltonian has the form

$$H_{SO} = \alpha\left(\sigma_y k_x - \sigma_x k_y\right),$$

usually referred to as the" Rashba Hamiltonian".

The same form of the Hamiltonian describes the spin-orbit coupling in two-dimensional systems with a structural inversion asymmetry. Nonzero values of acan be achieved, in addition, by applying an electric field across the two-dimensional structure or a quantum dot. Usually both (Rashba and Dresselhaus) terms are present in two-dimensional electron systems and quantum dots.

The corresponding spectrum of the Rashba Hamiltonian is given by:

$$E(k) = \frac{\hbar^2 k^2}{2m^*} \pm \alpha k$$

as shown in **Figure 7**.

In the absence of an external magnetic field the presented states are double-degenerate as dictated by the time-reversal symmetry of the spin-orbit coupled Hamiltonian. In the presence of such a field, the spin-orbit coupling terms should be augmented by the Zeeman coupling for the interaction between electron spin and external magnetic field in the form:

$$Hz = \frac{g}{2}(\sigma B)$$

Here B is the magnetic field applied to the quantum dot, and g is the g-(Lande) factor.
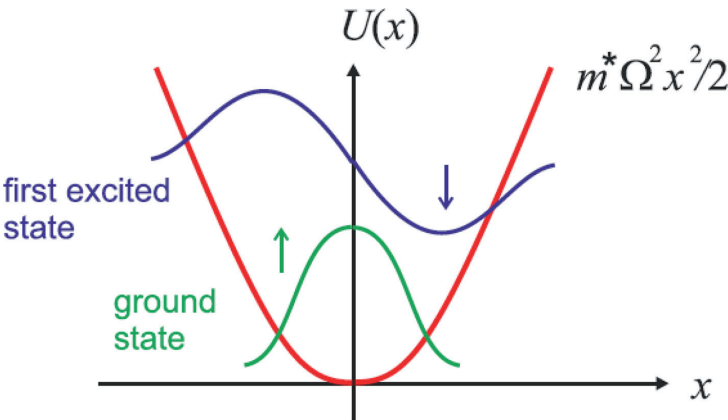
Figure 5.
One-dimensional parabolic potential modeling a simple quantum dot. Schematic plots of the ground and first excited states with opposite spins (marked by up- and down-arrows) are presented. Spin-orbit coupling couples these two spatial states with opposite spins and, as result, leads to the spin-position entanglement and modification of the charge densities.
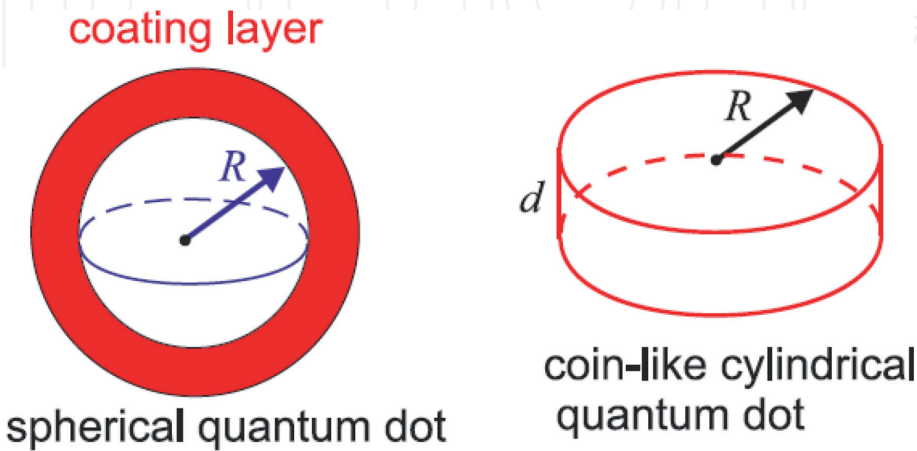
Figure 6.
(a) Typical spherical quantum dot with a coating layer. Typical value of the radius R is of the order of 10 nm.
(b) Typical coin-like cylindrical model of a quantum dot. The width d is of the order or less than 10 nm.
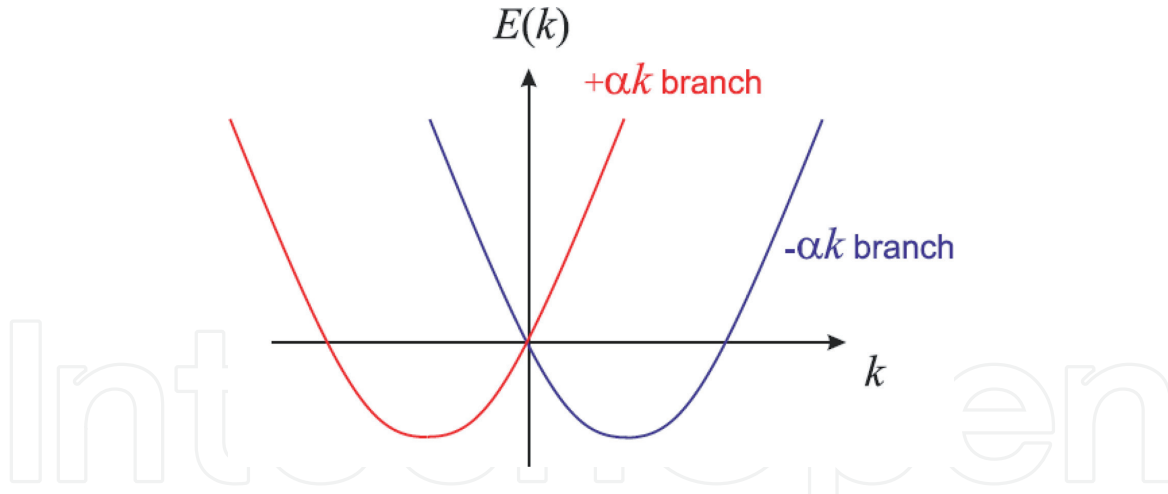
Figure 7.
Two branches of spectrum of two-dimensional carriers in the presence of the Rashba spin-orbit coupling with
$E(k) = \dfrac{\hbar^2 k^2}{2m^*} \pm \alpha k.$ *This shape of the spectrum corresponds to the anomalous velocity in the presence of the spin-orbit coupling: The zeromomentum states have nonzero velocities while finite-momentum states at*
$k = \dfrac{m^* \alpha}{\hbar}$ *have zero velocities.*

Being a relativistic effect, the g − factor is strongly related to the spin-orbit coupling and the Zeeman term plays an important role in the physics of the quantum dot. The values of g- factor can vary by two orders of magnitude, e.g. between −0.45 for GaAs and approximately −50 in InSb. The corresponding spin splitting can reach the values of the order of 10 meV at realistic fields of the order of 10 T. We mention here that while the spin-orbit coupling for a given quantum dot is fixed by its material and shape [12], the Zeeman coupling can easily be modified by applying a magnetic field. Thus, the absorption spectra and optics-related properties of the quantum dot can be modified as well.

The geometry of a quantum dot plays the crucial role for its spectrum and spin-orbit coupling, and, therefore in the light absorption and photovoltaics effects. A qualitative effect of the spin-orbit coupling in quantum dots is the quantum entanglement of particle spin and its position, where the particle wavefunction cannot be presented as a product of the spin and coordinate states. This entanglement determines several processes in quantum dots.

Since in the optical absorption electrons and holes are produced, similar spin-orbit coupling and Zeeman Hamiltonians should be defined for the holes as well. The Coulomb interaction between electrons and holes plays the crucial role in the light absorption in quantum dots. The spectrum of holes is described by 4 × 4 matrices due to degeneracy of the spectrum consisting of "light" and "heavy" hole branches. Dependent on the material and shape of the quantum dot, spin-orbit coupling and the Zeeman coupling for holes can be much stronger than that for electrons. In the presence of spin-orbit coupling, spin-defined states are prone to relaxation and loosing well-defined spin values. In bulk systems, where electron momentum is a well-defined quantum number, spins relax mainly due to the celebrated Dyakonov-Perel mechanism [13] with the spin relaxation rate of the order of $\left(\dfrac{\alpha k}{\hbar} \right)^2 \tau_k$, where τ_k is the momentum relaxation time. In quantum dots the situation is qualitatively different since the localization suppresses the free motion of carriers. Here the relaxation is mainly due to the so-called" admixture mechanism" caused by the spin-orbit coupling between different orbital states with opposite spins and interaction of these orbital states with lattice vibrations. This mechanism strongly depends on the applied magnetic field. In relatively week magnetic fields, the spin relaxation rate is small, and spin states are conserved for

long times of the order of 1 ms or more. If the spin-orbit coupling is sufficient, the resulting spin-position entanglement influences the spin states, and, therefore, the orbital wave functions and the exchange interaction between the injected and original carriers in the quantum dots. In addition, spin states of the injected carriers can be controlled by polarization of the incident light [14]. These interactions should be clearly seen in the absorption spectra and, as a result, influence the photovoltaics and photogalvanic application.

We mention here that a new very large class of very promising for the photovoltaics materials such as perovskites or quantum dots show the spin-orbit coupling in the general form of the Rashba and Dresselhaus terms. Therefore, the spin-orbit coupling-related aspects of their applications can be treated in general terms similarly to semiconductors. This coupling can increase the carrier's lifetime - the quantity important for solar cell applications. However, these rather complex compounds, demonstrating a great variety of different properties, are not yet reliably functionalized in the form of quantum dots. At the same time, whether the combined effects of the spin-orbit coupling lead to an increase or to a decrease in the efficiency of the light-to-voltage conversion in solar cells, is not yet clear even on the qualitative scale.

Acknowledgements

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