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SYMMETRY-BASED STUDY OF QUASI-ONE-DIMENSIONAL SYSTEMS RELEVANT TO SOLAR CELL APPLICATIONS

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Abstract

The importance of nanorods and nanotubes are presented in the field of solar cell developments. The investigations of nanostructured related efficiency-increasing processes in external fields are more invited. A group theoretical method for the study of electron states of semiconductors can be relevant to solar energy too is proposed. For further applications a possible extension is raised up.

Keywords

carrier multiplication, vertically aligned nanowire array, line groups, projective representations, magnetic field

1. Introduction

Quasi –one-dimensional systems (Q1D), namely carbon nanotubes (CNT), ZnO nanowires and nanorods and conjugated polymers (CP) have great interests on the field of study of solar cell applications nowadays thanks to the results of Nanotechnology. One of the main ones is the detectability of the so-called Carrier Multiplication process (CM) (or Multiply Exiton Generation one) in some CNT structures, for example in photodiodes [1], or in (6,5) chirality tubes [2], in which a single falling photon can create two or more electron-hole pairs (excitons). With the help of this process the so-called and referencia-considered Shockley-Quisser (SQ) theoretical limit [3] can be overcome. However the CM has been observed in bulk semiconductors since 1950s [4] the contribution of CM for efficiency-increasing is significant in the solar spectrum in quantum-confined semiconductor nanocrystals (in bulk semiconductors this contribution is insignificant) namely in quantum dots – the possible applicability in solar energetics can be originated from Nozik [5] – furthermore in the other type of Q1D nanorods (for example PbSe) [4, 6] and graphene [2, 7-8]. The CM effect in CNTs - due to the fact that they have no direct bulk counterparts- is also important in the understanding the fundamental processes responsible for the effect itself in the case of those nanocrystals at which the effect is enhanced related to their bulk counterparts. This is an open question yet [2]. Another aspect of the importance of the usage of Q1D materials in solar cells is in their technology known as vertically aligned nanowire array. With the help of its oriented geometry high carrier collection efficiency can be reached [9]. Xu et al. [10] gave a

generalized theoretical limit-for efficiency - maintaining the same detailed balance principle on which stand the Shockley and Quisser's results- for nanostructured solar cells and proved its equality to the case of the usage of optical concentrators in Shockley and Quisser's article (42%). Authors also gave examples for published efficiencies of the vertically aligned nanowire-based PV cells (in Figure 1b in their paper). For CNTs array (Figure 1.) as absorbent Bierman et al. [11] reported a solar thermophotovoltaic device and discussed the implications of surpassing the SQ limit. For ZnO nanowire arrays an important example is given by Bi et al. [12] for MEH-PPV/ZnO hybrid solar cell. Further solar cell applications are presented in Peng and Qin's [13] review (Figure 2).

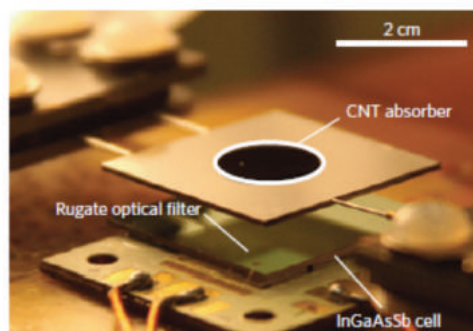


Figure 1. Array of CNTs as solar absorbent in a solar thermophotovoltaic device [11]

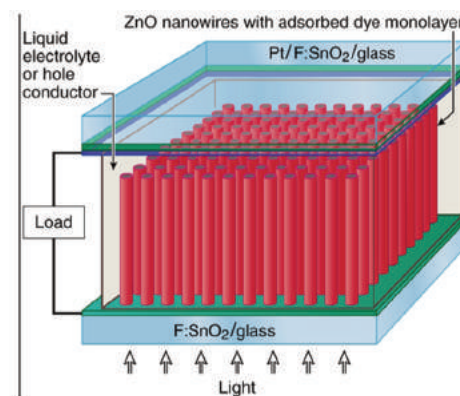


Figure 2. Schematic diagram of the nanowire-based dye-sensitized solar cell [13]

However the increasing of power efficiency is not the only reason for developments, this is the case when solar cells are needed to be installed onto uneven surfaces. For this installations the CP based solar cells are suitable against to the traditional Si-based ones. However the efficiency of polymer solar cells are much lower than Si-solar cells there are some cases when this type of solar cells gives the only result for using solar cells for example installation onto top of tents or onto the surfaces of rucksacks or onto the surfaces of human body thank to their flexibility.

2. Symmetry-based study of 01d solar cell materials

The above mentioned Q1D materials for solar cell applications have special space symmetry described by the so-called line group. Line groups are algebraical groups of Euclidean symmetries leaving invariant a straight line conventionally the z-axis. For the concreteness let us see polymers: they are built up from monomers. Monomers have own symmetry: rotations, mirrors and their combinations leaving the z-axis invariant. This is the one of the 7 possible point groups, namely: "... C_n (successive rotations C_n by $2\pi/n$ around z-axis), S_{2n} , C_{nh} , C_{nv} , D_n (which combine C_n with rotation by π/n followed by horizontal mirror reflection, horizontal and vertical mirror plane and rotation U by π around x-axis, respectively) and D_{nd} and D_{nh} (vertical mirror reflection combined with S_{2n} and C_{nh})..." [14]. But (identical) monomers are regularly repeated along the z-axis (regularly arrangement) and a monomer can achieved from the previous one not by pure translation but by screw axis or glide plane (generalized translations). Every line group can be set up a factorized form: $L=ZP$, where Z the set of generalized transformations and P the point group of the monomer. The generalized translations are needed to be combined with point group operations for getting an algebraical group at the end of procedure. There are infinitely many line groups gathered into 13 families. For example for polyacetylen- it was the one of the first CP for solar cells and it is under theoretical studies nowadays too [15] - the line group is $L2_1/mcm$ and this belongs to the 13th family. The capital L refer to the line group structure, 2_1 means that the generalized translation in this case a rotation by π around z-axis followed by a translation $a/2$, the half of the translation unit along z-axis (the symmetry element of it is a glide plane) the point symmetry group consists of a σ_h horizontal mirror (in the $z=0$ plane) and a σ_v vertical one (in the plan containing the polymer), their products and powers. This point group is D_{2h} . [16]. For getting the line group structure of CNTs we need to follow the so-called rolling up procedure of graphene sheet with identifying carbon atoms by the so – called chiral vector (Figure 3-6) [17].

We wish to study these materials in the presence of external uniform magnetic field. The reasons of our choice are the following: solar cells are under investigations in the presence of external magnetic field both experimenetally and theoretically nowadays [18-22]. However, there are no symmetry-based considerations of solar cells using Q1D materials in external magnetic field to our knowledge. In the field of study of quantum confined nanostructures, namely quantum dots (QD), quantum wells (QW) [23], superlattices (SL) [24] for solar cell applications have great importance but the investigation of CM processes in the presence of external magnetic field has no already received enough attention yet. Beside these facts some important research work have been already published about Q1D materials in constant external magnetic field. For example Trencsényi et al. [25] analyzed the bond connected hexagon chain structures of polyphenylene type of materials placed in perpendicular field to the surface containing the system. This structure is of some CP like PPV whose derivates (for example MEH-PPV as above,

MDMO-PPV) are the one of the most frequently applied materials at polymer based solar cells. For the future applications the Dzyaloshinsky and Kats's [26] result about magnetic field induced order (superconductivity) in Q1D materials might be decisive.

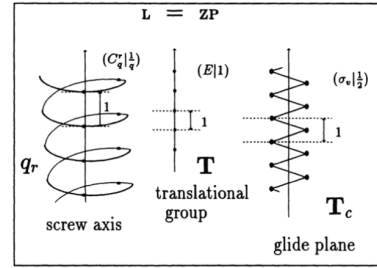


Figure 3. The types of generalized translations of line groups [16]

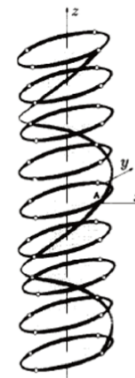


Figure 4. The so-called orbits of the line groups. [16]

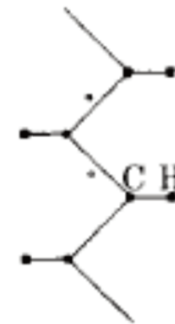


Figure 5. The structure of trans-polyacetylene [16]

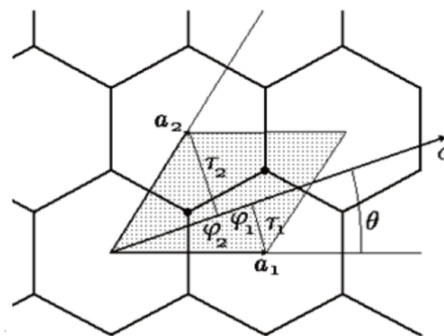


Figure 6. The elementary cell (shaded) and the chiral vector c and angle θ of graphene [17]

Our considerations are based on Tronc and Smirnov's [27] symmetry methods worked out for various semiconductor types

in the presence of external uniform magnetic field. The usages of symmetry methods mean a powerful tool for generating the so-called (optical) selection rules for transitions between electron states allowed by the symmetry of the systems. The symmetry method name means that we have to deal with the symmetry group of the Hamilton operator (Hamiltonian) of the system, with a group of operators representing some coordinate transformation which commute with Hamiltonian. The main problem in the field of study of an electron being in a crystal potential in the presence of external magnetic field that however a uniform field does not destroy the translational invariance of the system physically the operators of pure geometrical translations did not commute with Hamiltonian and these operators do not belong to the symmetry group of Hamiltonian. Brown [28] showed that multiplying with Peierls's phase factor they will have already been commute with it. In this case the representation of the usual translation group will become projective and the symmetry group form a so-called ray group. In the other way Zak [29, 30] defined an operator group (not a ray group) which is homomorphic to the usual translation group and commute with Hamiltonian. of the electron immersed to a crystal in magnetic field. This two constructions will be the same in the finite case and called by Magnetic Translation Group (MTG). But this method do not treat with point symmetry operations and its application for concrete semiconductor structure can be problematic. For this difficulty Tronc and Smirnov [27] gave a method. (A group-theoretical extension for the case of all the admissible rotations, reflections and time reversal is given by Tam [31] too.) Let us see the „General Considerations” of Tronc and Smirnov [27] (sec 2.) in more detail:

The Schrödinger equation for an electron in the crystal potential $V(r)$ placed in external magnetic field:

$$H\Psi_{j,n}(\mathbf{r}) = E_j\Psi_{j,n}(\mathbf{r}), \quad (1)$$

$$H = \frac{1}{2m} \left(\mathbf{p} + \frac{e}{c} \mathbf{A} \right)^2 + V(\mathbf{r}) + \frac{e}{mc} \hat{\mathbf{S}} \mathbf{B}, \quad (2)$$

where H is the Hamiltonian, \mathbf{A} is the vector potential of the magnetic field, \mathbf{p} is the momentum operator ($=i\hbar \text{grad}$), $\hat{\mathbf{S}}$ the spin operator, j the eigenvalue index and n enumerates the eigenstates of the same energy.

Let us choose the form of vector potential in the symmetric gauge:

$$\mathbf{A} = \frac{1}{2} [\mathbf{B} \times \mathbf{r}]. \quad (3)$$

Let be a general element of the symmetry group of the crystal $g = (R|a)$, which labels the rotation R as the coordinate transformation followed by the translation a . The „symmetry element” expression means that $V(r)$ crystal potential do not changes under this operation. This is the same case at the last term of the Hamiltonian since does not depend on r . The action of the symmetry element of the crystal on the geometrical space vectors can be written in the form: $r \rightarrow g^{-1} r = R^{-1}(r-a)$. From the fact that the scalar product of two vectors does not change under orthogonal transformations and after the acting of operation g the transformation R^{-1} „can be raised up” from the expressions for p and \mathbf{A} , the change in the expression of the Hamiltonian is solely:

$$\mathbf{A} \rightarrow \mathbf{A}(\mathbf{r}-\mathbf{a}). \quad (4)$$

At this time the Hamilton operator in the Schrödinger equation acts on $\Psi_{j,n}(g^{-1} r)$. But this change can be reached in the starting Schrödinger equation with a gauge transformation too. Since $\mathbf{B} = \text{rot} \mathbf{A} = \text{rot}(\mathbf{A} + \text{grad} f(r))$, where $f(r)$ an arbitrary function of coordinates if add to \mathbf{A} the gradient of:

$$f(\mathbf{r}) = \frac{1}{2} (\mathbf{B} \times \mathbf{a}) \cdot \mathbf{r}, \quad (5)$$

$$\mathbf{A} + \text{grad} f(\mathbf{r}) = \mathbf{A}(\mathbf{r}-\mathbf{a}). \quad (6)$$

According to the teaching of quantum mechanics the Schrödinger equation remain unchanged if we carry out at the same time a transformation on the wave function:

$$\Psi_{j,n}(g^{-1} \mathbf{r}) = \exp(-i \frac{e}{2\hbar c} [\mathbf{B} \times \mathbf{a}] \cdot \mathbf{r}) \Psi_{j,m}(\mathbf{r}) \quad (8)$$

$$\sum_m C_{mn}(g) \Psi_{j,m}(\mathbf{r}).$$

Thus to every g symmetry element of the crystal belongs a $C(g)$ matrix which represents it. From this procedure one can see the structure of the elements of the symmetry group of the Hamiltonian: after the geometrical transformation $r \rightarrow g^{-1} r$, where $g = (R|a)$ and $R\mathbf{B} = \mathbf{B}$, we have to carry out a gauge transformation with $f(r) = -f(r)$, since $\mathbf{A}(\mathbf{r}-\mathbf{a}+\mathbf{a}) = \mathbf{A}(\mathbf{r})$ remain unchanged and we returned to the starting Hamiltonian. The elements of this group labeled by g^* . the group itself labelled by G^* . G^* contains magnetic translations as subgroup. Tronc and Smirnov [27] pointed out that for the g^* . elements the representation of matrices are projective

$$g_2^* g_1^* = \omega(g_2, g_1) g_3^*, \quad (9)$$

$$C(g_2)C(g_1) = \omega(g_2, g_1)C(g_2g_1), \quad (10)$$

and

$$\omega(g_3, g_2) \omega(g_3g_2, g_1) = \omega(g_3, g_2g_1) \omega(g_2, g_1). \quad (11)$$

This method has other solutions for obtaining the so-called irreducible representations (IR) for the symmetry group of the Hamiltonian of bulk materials than MTG theory's ones. For example in the case of two symmetry operations with lattice translations are in (a_1, a_3) , where a_3 is in the direction of magnetic field do not commute in contrast with the theory of MTG.

The authors applied their results of their model namely the symmetry groups of bulk semiconductors with the wurtzite and zinc blende structure and for the related nanostructures as well such as SLs, QWs and QDs under magnetic field directed parallel or perpendicular to some symmetry elements (symmetry axis, mirror plane).

From the viewpoint of our research the most interesting results are about Q1D rods and tubes related to bulk materials with wurtzite structure specially for the ZnO nanorods and nanotubes [32]. Their space symmetry are described by the so-called rod groups a subperiodic (1-periodic) subgroups of 3-periodic 3-dimensional space groups [33]. We can get a rod group from the related space group by keeping translations only in the direction of z -axis. The order of rotations or screw axis can only be 1, 2, 3, 4 and 6. The point symmetry of them is the same as ones of their space groups [34]. For generating optical selection rules conventional group theoretical methods can be used just by referring to the corresponding space groups – for example the so-called little group method known as subgroup one [27, 32]. But this method for CNTs whose space symmetry described by general line groups – rod groups are considered as special line groups- due to the lack of bulk counterparts were mentioned at the beginning of this paper cannot be used, there are no „global” space groups related to their space symmetries. It seems to us that

this is an essential open problem yet. At this point can become relevant the mathematical study of projective representations of line groups which one is almost completely missing in the literature. (This idea is suggested by the second author of this paper.) A short description are in the Evarestov's book [34].

3. A possible extension of Tronc and Smirnov's method

Since the Tronc and Smirnov [27] method contains the theory of MTGs- but came to another result for generating IR of symmetry group of Hamiltonian- can be raised naturally the extension of it to the case when external electric field is present too. This type of generalization of the theory of MTG is given by Ashby and Miller [35], in which the Hamiltonian contains space and time periodic potentials. too (However Tronc and co-workers discussed the power of the electric field on the symmetry of ZnO nanorods and tubes and referred to Ashby and Miller [35] too did not considered the cases of time-varying fields). For the solar cell applications would be essential to study the case of the light wave falling onto the semiconductor. If so a perturbing term of this form:

$$\mathcal{K} = \frac{eE}{m} \left(\mathbf{p} + \frac{eA}{c} \right) \frac{1}{2i\omega} [\boldsymbol{\varepsilon} \exp(i\omega t) - \boldsymbol{\varepsilon}^* \exp(-i\omega t)], \quad (12)$$

where „m” is the free electronic mass, ω is the frequency of the radiation, E is the magnitude of the electric field, and $\boldsymbol{\varepsilon}$ is the unit vector in the direction of the field will appear in the Hamiltonian [36].

Our task is to find translation-type operators commuting with the perturbed Hamiltonian which translations are in not purely in the space but in the time too according to Ashby and Miller's [35] ideas. Arising from these ones the time-periodicity leads to the restrictions for possible magnitude of the electric field and because of this fact we have to proceed carefully.

4. Conclusions

However the Q1D materials have a great importance in the efficiency-increasing developments of solar cells but the added external magnetic field investigations of them have no enough attention. The investigations of CM processes are hoped to extend for the case of the presence of external magnetic field. The Tronc and Smirnov's symmetry method is a usefool tool for studying solar cells using Q1D materials under external uniform magnetic field. For more general cases as of the CNTs the research of the mathematical theory of projective representations of line groups would be fruitful. For further solar cell applications the extension for the case of falling electromagnetic wave onto the semiconductors would be a possible way.

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