ФІЗИЧНІ, ХІМІЧНІ ТА ІНШІ ЯВИЩА, НА ОСНОВІ ЯКИХ МОЖУТЬ БУТИ СТВОРЕНІ СЕНСОРИ

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WANNIER-MOTT EXCITONS AND ATOMS IN A DC ELECTRIC FIELD: PHOTOIONIZATION, STARK EFFECT, RESONANCES IN THE IONIZATION CONTINUUM

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Abstract. A numerical calculation of the DC Stark effect for hydrogen and sodium atoms and Wannier-Mott excitons in an external uniform DC electric field is carried out within the operator perturbation theory method. New adequate model for description of the unique especialitites in the photoionization spectra of the hydrogenic atoms is proposed. It is found that the Stark shift for the n=2 state of excitons in the Cu_2O semiconductor (yellow series) at the electric field strength 600 V/cm results in — 3,1 10^{-4} eV which agrees well with experimental data of Gross et al. It is indicated also that the analogous unique especialitites may possibly take a place in the Wannier-Mott excitons spectra near the threshold boundary.

Keywords: atom, Wannier-Mott exciton, Stark effect, photoionization

ЕКСІТОНИ ВАН'Є-МОТТА І АТОМИ У ПОСТІЙНОМУ ЕЛЕКТРИЧНОМУ ПОЛІ: ФОТОІОНІЗАЦІЯ, ШТАРК ЕФЕКТ ТА РЕЗОНАНСИ У ІОНІЗАЦІЙНОМУ КОНТИНУУМІ

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Анотація. На підставі методу операторної теорії збурень виконано розрахунок Штарк-ефекту для атома водню, натрію та ексітонів Ван'є-Мотта (напівпровідник Cu_2O) у однорідному електричному полі. Незвичайні особливості у спектрі фотоіонізації воднеподібних систем описані на підставі адекватної квантово-механічної моделі. Енергії високо розташованих резонансів у іонізаційному континуумі добре узгоджуються з відомими експериментальними даними Gross et al. Розраховані штарківські зсуви для ряда станів ексітону у напівпровіднику Cu_2O (жовта серія) в електричному полі 600 В/см. Прогнозується можливість

проявлення незвичайних особливостей у спектрах екситонів Ван'є-Мотта поблизу границі іонізації останніх.

Ключові слова: атом, екситон Ван'є-Мотта, Штарк-ефект, фотоіонізація

ЭКСИТОНЫ ВАНЬЕ-МОТТА И АТОМЫ В ПОСТОЯННОМ ЭЛЕКТРИЧЕСКОМ ПОЛЕ: ФОТОИОНИЗАЦИЯ, ШТАРК ЭФФЕКТ И РЕЗОНАНСЫ В ИОНИЗАЦИОННОМ КОНТИНУУМЕ

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Аннотация. На основе метода операторной теории возмущений выполнен расчет Штарк-эффекта для атома водорода, натрия и экситонов Ванье-Мотта (полупроводник Cu_2O) в однородном электрическом поле. Необычные особенности в спектре фотоионизации водородоподобных систем описаны на основе адекватной квантовой модели. Энергии высоко лежащих резонансов в ионизационном континууме находятся в хорошем согласии с известными экспериментальными данными Gross et al. Рассчитанные штарковские сдвиги для ряда состояний экситона в полупроводнике Cu_2O (желтая серия) в электрическом поле 600 В/см. Предсказана возможность проявления необычных особенностей в спектрах экситонов Ванье-Мотта вблизи границы ионизации последних.

Ключевые слова: атом, экситон Ваньє-Мотта, Штарк-эффект, фотоионизация

Introduction

Observation of the Stark effect in a constant (DC) electric field near threshold in hydrogen and alkali atoms led to the discovery of resonances extending into the ionization continuum by Glab et al and Freeman et al (c.f.[1-5]). Though the known semi-empirical approach of Harmin [5] (c.f.[3]) is effective enough, a full adequate consistent theory of this phenomena is absent hitherto. Calculation of the atomic characteristics in a strong electric DC field remains very important problem of modern atomic physics and also of the physics of semiconductors [6-30]. It is well known [28] that the availability of excitons in semiconductors resulted experimentally in the special form of the main absorption band edge and appearance of discrete levels structure (f.e. hydrogen-like spectrum in Cu_2O). Beginning from known papers of Gross-Zaharchenya, Thomas and Hopfield et al (c.f.[28-30]), a calculation procedure of the Stark effect for exciton spectrum attracts a deep interest permanently. As it is well known [11], external electric field shifts and broadens the bound state atomic levels. The standard quantum-mechanical approach mutually relates complex eigen-energies (EE) $E = E_r + 0{,}5iG$ and complex eigen-functions (EF) to the resonances' shape. The calculation difficulties in the standard quantum mechanical approach are well known [3]. The WKB approximation overcomes these difficulties for the states, lying far from "new continuum "boundary and, as a rule, is applicable in the case of a relatively weak electric field. The same could be regarded to the widespread asymptotic phase method (c.f.[1-4], based on the Breit-Wigner parameterisation for the phase shift dependence on scattering energy. Some modifications of the WKB method were introduced by Popov et al. and Ostrovsky et al.(c.f. [3]). Quite another calculation procedures are used in the Borel summation of the divergent perturbation theory (PT) series and in the numerical solution of the difference equations following from expansion of the wave-function over finite basis. In refs. [10-13] a principally new consistent uniform quantum — mechanical approach to the non-stationary state problems solution had been developed including the Stark effect and also the scattering problems. The essence of the method is the inclusion of the well known method of "distorted waves approximation" in the frame of the formally exact PT [11]. The zeroth order Hamiltonian H_0 of this PT possesses only stationary bound and scattering states. In order to overcome the formal difficulties, the zeroth order Hamiltonian was defined using the set of the orthogonal EF and EE without specifying the explicit form of the corresponding zeroth order potential. In the case of the optimal zeroth order spectrum , the PT smallness parameter is of the order of G/E, where G and E are the field width and bound energy of the state level.

One could see that $G/E \leq 1/n$ even in the vicinity of the "new continuum" boundary (where n is the principal quantum number). This method is called the operator PT (OPT) approach [11,12]. It is very important to note that the hamiltonian H_0 is defined so that it coincides with the general Hamiltonian H at $\varepsilon \Rightarrow 0$. ($\varepsilon$ is the electric field strength). Let us note that perturbation in OPT does not coincide with the electric field potential though they disappear simultaneously. An influence of the corresponding electric potential model function choice on the values of the Stark resonances energies and bandwidths does not significantly change the final results for the resonances shifts and widths [11,12]. All said above regards the Wannier-Mott exciton characteristics in semiconductors as well.

In ref. [13-17] the OPT approach have been used for solution of the isotopes separation problem and an account of the non-hydrogenic effects was done as well as the improvement of the convergence procedure. In ref. [18-24] the OPT approach have been successfully used for studying new laser-electron nuclear spectral effects in thermalized plasma (speech is about new cooperative laser-electron-nuclear processes), new laser-electron-nuclear effects in atoms, ions and diatomic molecules. There is very effective application of the OPT approach in conjuction with S-matrix Gell-Mann and low formalism to studying the resonance states of compound super-heavy nucleus and electron-positron pair production in heavy nucleus and ioons collisions and under availibuility of the external superintense electromagnetiv filed, when the EPPP chanell is opened [22-24].

In this paper we have used the OPT method [11,12] for studying and exact calculation of the DC Stark effect for hydrogen, sodium atoms and Wannier-Mott excitons in an external uniform DC electric field and the corresponding photoionization spectra. New adequate model for description of the unique especialitites in the photoionization spectra of the atoms is proposed. It is found that the Stark shift for the n=2 state of excitons in the Cu_2O semiconductor (yellow series) at the electric field strength 600 V/cm results in — 3,1 10^{-4} eV which agrees well with experimental data of Gross et al. It is indicated also that the analogous unique especialitites may possibly take a place in the Wannier-Mott excitons spectra near the threshold boundary.

Operator perturbation theory approach [10-13]

As usually, the Schrodinger equation for the electronic eigen-function taking into account the uniform DC electric field and the field of the nucleus (Coulomb units are used: a unit is $h^2/Ze^2 m$ and a unit of mZ^2e^4/h^2 for energy) looks like:

$$[-(1 - N/Z)/r + \varepsilon z - 0{,}5\Delta - E]\psi = 0, \quad (1)$$

where E is the electronic energy, Z — charge of nucleus, N — the number of electrons in atomic core. Our approach allow to use more adequate forms for the core potential (c.f.[25-27]), including the most consistent quantum electrodynamics procedure for construction of the optimized one-quasi-electron representation and *ab initio* core potential , providing a needed spectroscopic accuracy. For multielectron atom one may introduce the ion core charge z^*. According to standard quantum defect theory (c.f.[26]), relation between quantum defect value μ_l, electron energy E and principal quantum number n is: $\mu_l = n - z^*(-2E)^{-1/2}$. As it is known, in an electric field all the electron states can be classified due to quantum numbers: n, n_1, n_2, m (principal, parabolic, azimuthal: $n = n_1 + n_2 + m + 1$). Then the quantum defect in the parabolic co-ordinates $\delta(n_1 n_2 m)$ is connected with the quantum defect value of the free ($\varepsilon = 0$) atom by the following relation [14]:

$$\delta(n_1 n_2 m) = (1/n)\sum_{l=m}^{n-1}(2l+1)(C^{JM}_{J,M-m;lm})^2\mu_l\,,\ J = (n-1)/2,\ M = (n_1 - n_2 + m)/2.$$

Naturally, it is possible to use more complicated forms for the ion core potential (c.f.[3,17]). After separation of variables, equation (1) in parabolic co-ordinates could be transformed to the system of two equations for the functions f *and* g:

$$f' + \frac{|m|+1}{t} f' + [0{,}5E + (\beta_1 - N/Z)/t - 0{,}25\,\varepsilon(t)\,t]f = 0; \quad (2)$$

$$g'' + \frac{|m|+1}{t} g' + [0{,}5E + \beta_2/t + 0{,}25\,\varepsilon(t)\,t]g = 0, \quad (3)$$

coupled through the constraint on the separation constants:

$$\beta_1 + \beta_2 = 1.$$

For the uniform electric field $\varepsilon(t) = \varepsilon$. In ref. [11], the uniform electric field ε in (3) and (4) was substituted by model function $\varepsilon(t)$ with param-

eter τ (τ = 1.5 t_2) . Here we use similar function, which satisfies to necessary asymptotic conditions (c.f.[11,12]) :

$$\varepsilon(t) = \frac{1}{t}\,\varepsilon\left[(t-\tau)\frac{\tau^2}{\tau^2+t^2}+\tau\right]. \qquad (4)$$

Potential energy in equation (4) has the barrier. Two turning points for the classical motion along the η axis, t_1 and t_2 , at a given energy E are the solutions of the quadratic equation (β = β_1, $E = E_{0)}$. It should be mentioned that the final results do not depend on the parameter τ . It is necessary to know two zeroth order EF of H_0: bound state function Ψ_{Eb} (ε, ν, φ) and scattering state function Ψ_{Es} (ε, η, φ) with the same EE in order to calculate the width G of the concrete quasi-stationary state in the lowest PT order. Firstly, one would have to define the EE of the expected bound state. It is the well known problem of states quantification in the case of the penetrable barrier. We solve the (2, 3) system here with the total Hamiltonian H using the conditions [11]:

$$f(t)\to 0 \text{ at } t \Rightarrow \infty,$$

$$\partial x(\beta, E) / \partial E = 0, \qquad (5)$$

with

$$x(\beta, E) = \lim_{t\Rightarrow\infty} [g^2(t) + \{g'(t) / k\}^2]\, t^{|m|+1}.$$

These two conditions quantify the bounding energy E, with separation constant β_1 . The further procedure for this two-dimensional eigenvalue problem results in solving of the system of the ordinary differential equations(2, 3) with probe pairs of E, β_1. The bound state EE, eigenvalue β_1 and EF for the zero order Hamiltonian H_0 coincide with those for the total Hamiltonian H at ε ⇒ 0, where all the states can be classified due to quantum numbers: n, n_1, l , m (principal, parabolic, azimuthal) that are connected with E, β_1, m by the well known expressions. We preserve the n, n_1 ,m states-classification in the ε≠0 case . The scattering states' functions must be orthogonal to the above defined bound state functions and to each other. According to the OPT ideology [11,12], the following form of $g_{E's}$:is possible:

$$g_{E's}(t) = g_1(t) - z_2' g_2(t), \qquad (6)$$

with $f_{E's}$, and $g_1(t)$ satisfying the differential equations (2) and (3). The function g_2(t) satisfies the non-homogeneous differential equation, which differs from (3) only by the right hand term, disappearing at $t \Rightarrow \infty$. The coefficient z_2' ensures the orthogonality condition and could be defined as [11]:

$$z_2' = \{\iint d\zeta d\eta\, (\zeta+\eta) f^2_{Eb}(\zeta) g_{Eb}(\eta) g_1(\eta)\} /$$

$$/ \{\iint d\zeta d\eta\, (\zeta+\eta) f^2_{Eb}(\zeta) g_{Eb}(\eta) g_2(\eta)\}.$$

The imaginary part of state energy in the lowest PT order is:

$$Im\, E = G/2 = \pi < \Psi_{Eb} |H| \Psi_{Es} >^2, \qquad (7)$$

with the general Hamiltonian H (G- resonance width). The state functions Ψ_{Eb} and Ψ_{Es} are assumed to be normalized to unity and by the δ(k -k')-condition, accordingly.. The photoionization cross section could be defined as follows:

$$\sigma^F = 4\pi^2 \hbar\omega / 137 \times$$

$$\times \sum_{l,l'} < 0 | r_m | \Psi_{l'} > [< \Psi' | \Psi >^{-1}]_{ll'} < \Psi_l | r_m | 0 >, \qquad (8)$$

where |0> is the initial state of the atom, $r_m = z$ for π-polarized light and $r_m = (1/\sqrt{2})(x \pm iy)$ for σ-polarization; $< \Psi_{l'}' | \Psi_l >$- the overlap matrix of the set {Ψ} (see details of its definition in [3] and cited ref. therein). Note then that the whole calculation procedure at known resonance energy E and separation parameter β has been reduced to the solution of one system of the ordinary differential equations. For its solution we use our numeral atomic code ("Superatom" package [3, 10-16,24-27]).

Stark resonances energies and widths calculation results

The calculation results for Stark resonances energies and bandwidths for some states of H atom are presented in Tables 1. For comparison we have indicated the data, obtained within another approach — complex eigen-values and numerical calculation (c.f.[1,3,11]. In table 2 we present the calculation results for Stark resonances energies for some Rydberg states of *Na* atom in an electric field 3,59 kV/cm. For comparison we have also presented the experimental data [5], the results of calculation within the *1/n*-expansion method by Popov etal , semi-empirical approach of Harmin (c.f. [5,14]).

For the most long-living Stark resonances with quantum numbers $n_2 = 0$, m = 0, a width of energy level is significantly less than a distance between them. These states are mostly effectively populated by π-polarized light under transitions from states with (n_1-n_2) = max, m = 0. As a result, the sharp

isolated resonances (their positions under E>0 are determined by energies of quasi-stationary states with n_2=0 , m=0) are appeared under photo ionization from these states in a case of π-polarization. In particular, calculated values of photo ionization cross-sections (ε=*6,5 kV/cm.*): (24, 23, 1, 0) σ^F= 0,63 (atomic units), (24, 23, 0, 0) σ^F= 1,7 (atomic units) [14]. In general, the agreement between theory and experiment is good. Let us note that our results are obtained in the first PT order, i.e. already the first PT order provides the physically reasonable results.

Table 1
The energies E_r (at.units) and widths G (at.units)of Stark resonances of the hydrogen atom ina DC electric field with strength: ε=*6,5 kV/cm.*

(n n_1 n_2 m)	*E*, Ref. [5]	G , Ref. [5]	*E* , Present paper	G , Present paper
24,23,0,0	0,1192	0,2752	0,1194	0,2754
25,23,1,0	0,2748	1,0868	0,2749	1,0871
25,23,0,1	0,8298	0,7484	0,8301	0,7487
25,24,0,0	1,4329	0,4175	1,4331	0,4177

Table 2
The energies (cm^{-1})of the Stark resonances for the Na atom (ε=*3,59 kV/cm)*:
A-experimental data ; B- Popov et al; C- semi-empirical approach of Harmin;
D- OPT approach.

State:$n_1 n_2 m$	δ	A	B	C	D
26,0,0	0,140	15,5	15,5	15,5	15,5
25,0,1	0,007	21,1	21,2	21,2	21,1
25,0,0	0,145	35,5	35,6	35,7	35,5
24,0,1	0,008	41,1	40,4	40,5	41,0
24,1,0	0,130	50,5	50,3	50,4	50,5
24,0,0	0,151	56,5	57,0	57,2	56,5
23,0,1	0,008	61,2	60,7	60,8	61,1
23,0,0	0,157	79,3	80,3	80,6	79,4
22,0,1	0,009	84,1	83,1	83,5	83,9
22,1,1	0,016	75,0	74,8	74,9	75,1

Wannier-Mott Excitons in a DC electric field

The analogous method can is formulated for description of the Stark effect in the Wannier-Mott excitons in semiconductors (CdS, Cu_2O). The Schrodinger equation for the Wannier-Mott exciton has a standard form:

$$[-\hbar^2\nabla_e^2/2m_e^* - \hbar^2\nabla_h^2/2m_h^* - -e^2/\varepsilon r_{eh} - eEr_e - eEr_h]\Psi = E\Psi \ . \qquad (9)$$

Here all notations are standard. A vector potential is as follows: *A(r)=1/2 [Hr].* Under transition to system of exciton masses centre by means of introducing the relative coordinates: $r = r_e - r_h$

$$\rho = (m_e^* r_e + m_h^* r_h)/(m_e^* + m_h^*)' ,$$

one could rewrite (9) as:

$$[-\hbar^2\nabla^2/2\mu - e^2/\varepsilon r - \hbar/2\times$$
$$\times(1/m_h^* - 1/m_e^*)K\cdot p - eEr]F = [E - \hbar^2K^2/8\mu]F \ .$$

This equation then could be solved by the method, described above. Preliminary estimates show that this approach, in a case of electric DC field, gives the results for Stark states in a reasonable agreement with known results of Thomas and Hopfield (TH) [28]. According to our preliminary estimate, the Stark shift for the n=2 state of excitons in the Cu_2O semiconductor (yellow series) at the electric field strength 600 V/cm results in — 3,1 10^{-4} eV. This value agrees well with experimental data of Gross et al.[28]. Ionization of the exciton in an electric DC field occurs if a change of potential on a small enough distance (the orbits diameter) is comparable with a bonding energy of particle on this orbit. According to data of Gross et al., the corresponding electric field is ~ 9 10^3 V/cm. Our calculation agrees with this value. Near ionization boundary, a hydrogen atom demonstrates a behaviour of quantum chaotic system, including the diffusion mechanism of ionization. Besides, for non-hydrogen atoms, there are unique especialitites in the photoionization spectra (alkali atoms) [5,14]. Probably, the analogous unique especialitites may take a place in the Wannier-Mott excitons spectra in semiconductors (of Cu_2O type) near the threshold boundary. One could suppose very interesting mechanism of the exciton ionization under different values of the electric field strength with possible positive energy resonances in spectra.

Conclusions

We have applied earlier developed the operator perturbation theory method [10-13] in numerical calculation of the DC Stark effect for hydrogen-like atoms and Wannier-Mott excitons in an external uniform DC electric field. We have demonstrated the existence of common features in behaviour of such a different physical objects as hydrogen atom, non-hydrogen (alkali) atoms and Wannier-Mott exciton under Stark effect caused by a DC electric

field.. The latter is to be the generator of the states mixture leading to the resonance states ionization threshold lying in the continuum, which can not be correctly described by standard available quantum-mechanical methods [4] as specch is about sufficiently strong fileds.

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