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RELATIVISTIC OPERATOR PERTURBATION THEORY IN SPECTROSCOPY OF MULTIELECTRON ATOM IN AN ELECTROMAGNETIC FIELD

We present the theoretical basis of a new relativistic operator perturbation theory (OPT) approach to multielectron atom in an electromagnetic field combined with a relativistic many-body perturbation theory (RMBPT) formalism for a free multielectron atom. As illustration of application of the presented formalism, the results of energy and spectral parameters for a number of atoms are presented. The relativistic OPT method is tested for the multielectron systems such as Fr and Tm. New approach is elaborated for an accurate, consistent treatment of a strong field Stark effect in multielectron atoms.

Keywords: multielectron atom in a dc electric field – modified operator perturbation theory – Rydberg autoionization resonances

1. Introduction

An investigation of spectra, optical and spectral, radiative and autoionization characteristics for the rare-earth elements (isotopes) and corresponding ions is traditionally of a great interest for further development quantum optics and atomic spectroscopy and different applications in the plasma chemistry, astrophysics, laser physics, quantum and nano-electronics etc. (see Refs. [1–42]).

The calculation difficulties in description of the multielectron atoms in electromagnetic (electric) field inherent to the standard quantum mechanical approach are well known. Here one should mention the well-known Dyson phenomenon for a Strong Filed AC, DC Stark effect. Besides, in contrast to the hydrogen atom, the non-relativistic Schrödinger and relativistic Dirac equations for an electron moving in the field of the atomic core in many-electron atom and a uniform external electric field does not allow separation of variables in the parabolic coordinates.

The Wentzel-Kramers-Brillouin (WKB) approximation overcomes these difficulties for the states lying far from the " new continuum" boundary. The detailed review of a modern states of art for spectroscopy of multielectron atoms in an electric (laser) field is presented in Refs. [8,16].

In this paper we present the theoretical basis of a new relativistic operator perturbation theory (OPT) approach to multielectron atom in an electromagnetic field combined with a relativistic many-body perturbation theory (RMBPT) formalism for a free multielectron atom. The relativistic OPT approach is tested for the multielectron systems such as francium Fr and thulium Tm.

The relativistic density-functional approximation with the Kohn-Sham potential is taken as the zeroth approximation in the RMBPT formalism. There have taken into account all exchange-correlation corrections of the second order and dominated classes of the higher orders diagrams (polarization interaction, quasiparticles screening, etc.). New form of the multi-electron polarization functional has been used.

As illustration of application of the presented formalism, new data on the energy and spectral parameters for two complex multielectron atoms in a electrioc (electromagnetic) field are presented.

2. Relativistic operator perturbation theory for multielectron atoms in an electromagnetic field

Here we present a new relativistic quantum approach to modeling the chaotic dynamics of atomic systems in a dc electric and ac electromagnetic fields, based on the theory of quasistationary quasienergy states, optimized operator perturbation theory, method of model-potential, a complex rotation coordinates algorithm method [16,43]. The universal chaos-geometric block will be used further to treat the chaotic ionization characteristics for a number of heavy atomic systems.

Let us remind that in the case of the electromagnetic field atomic Hamiltonian is usually as follows:

$$H = \frac{1}{2}p^2 + V_{at}(r) + zF_0\cos(\omega t) \quad (1)$$

The field is periodic, of course one should use the Floquet theorem; then the eigen Floquet states and quasienergies E_j are defined as the eigen functions and eigen values of the Floquet Hamiltonian ∂ . In the general form with using the method of complex coordinates the problem reduces to the solution of stationary Schrödinger equation, which is as follows in the model potential approximation:

$$(-1/2\cdot\nabla^2 + V_{at}(r) + \omega L_z + F_o z)\Psi_E(r) = E\Psi_E(r) \quad (2)$$

i.e. to the stationary eigen value and eigen vectors task for some matrix A (with the consideration of several Floquet zones): $(A - E_jB)|E_j\rangle=0$. As a decomposition basis, system of the Sturm functions of the operator perturbation theory basis is used.

In our new theory we start from the Dirac Hamiltonian (in relativistic units):

$$H = \alpha p + \beta - \alpha Z/r_i + \sqrt{\alpha}Fz\,, \quad (3)$$

Here a field strength intensity is expressed in the relativistic units (F_{rel}= $a^{5/2}F_{at.un.}$; a is the fine structure constant). One could see that a relativistic wave function in the Hilbert space is a bi-spinor. Using the formal transformation of co-ordinates $r \to r\exp(i\theta$ in the Hamiltonian (11), one could get:

$$H(\theta) = (\alpha cp - Z/r)\exp(-i\theta) + \beta - \sqrt{\alpha}Fz\exp(i\theta) \quad (4)$$

In comparison with an analogous non-relativistic theory, here there is arisen a technical problem. In formulae (11) there is term b, which can not be simply transformed. One of the solving receptions os a limitation of a sub-space of the Hamiltonian eigen-functions by states of the definite symmetry (momentum J and parity P). Thes states can be described by the following functions:

$$\Psi_{PJ}^M = 1/r\begin{pmatrix} f(r)\Upsilon_{lJ}^M(n,\sigma) \\ g(r)\Upsilon_{l'J}^M(n,\sigma)\end{pmatrix} \quad (5)$$

Here l (l') and spin ½ in the coupling scheme give a state with the total momentum J and its projection M_J=M. Action of the Hamiltonian (11) on the functions (13) with definite J results in:

$$\hat{H}(\theta)\Psi_{PJ}^M = \alpha_r(\hat{p}_r - \frac{i\omega(J+1/2)}{r}\beta)\exp(-i\theta)\Psi_{PJ}^M + \\ +(\beta - \frac{\alpha Z}{r}\exp(-i\theta) - \sqrt{\alpha}Fz\exp(-i\theta))\Psi_{PJ}^M \quad (6)$$

$p_r = -i(1/r)(d/dr)r$, $\vec{n} = \vec{r}/r$, σ – the Pauli matrices; parameter w=-1, if l=J-1/2 and w=1, if l=J+1/2.

In order to further diagonalize the Hamiltonian (6), we need to choose the correct basis of functions in the subspace (5), in particular, by choosing the following functions (the sitter or water-like type):

$$\Psi_{PJ}^{a,M} = 1/r\begin{pmatrix} F(r)\Upsilon_{lJ}^M(n,\sigma) \\ 0\end{pmatrix} \quad (7)$$

$$\Psi_{PJ}^{b,M} = 1/r\begin{pmatrix} 0 \\ iG(r)\Upsilon_{l'J}^M(n,\sigma)\end{pmatrix} \quad (8)$$

It is easy to see that the matrix elements (6) will be no-zeroth only between the states with the same M_J. In fact this moment is a single limitation of the whole approach.

Transformation of co-ordinates in the Pauli Hamiltonian (in comparison with the Schrodinger equation Hamiltonian it contents additional potential term of a magnetic dipole in an external field) can be performed by the analogous way. However, procedure in this case is significantly simplified. They can be expressed through the set of one-dimensional integrals, described in details in Refs. [8,14,47].

In contrast to the hydrogen atom, the non-relativistic Schrödinger equation for an electron moving in the field of the atomic core in many-electron atom (in particular, an alkali element) and a uniform external electric field does not allow separation of variables in the parabolic coordinates x, h, j [14].One of the ways this problem could be related to the use of effective potentials, chosen in such a way (for example, in the Miller-Green approximation (see [1,2]) that to achieve the separation of variables in the Schrödinger equation. Here the model potential approach or the quantum defect approximation can be used. One may introduce the ion core charge for the multielectron atom. According to standard quantum defect theory, the relation between quantum defect value , electron energy E and principal quantum number n is: $\mu_1 = \grave{o} - z^*(-2E)^{-1/2}$. The quantum defect in the parabolic coordinates $\delta(n_1 n_2 m)$ is connected to the quantum defect value of the free $(\varepsilon = 0)$ atom by the following relation [43]:

$$\delta(n\ n\ m) \quad (1/n)\sum_{l\ m}^{n}(2l \quad 1)(\ C^{M}_{J\ M\ \ mlm} \quad \mu_l \tag{9}$$

Such a scheme provides a general receipt to combine the OPT method with the RMBPT in spherical coordinates for a free atom. The details of the used method can be found in the references [8,16,43].

3. Method of relativistic many-body perturbation theory

Generally speaking, the energy spectra for the majority of complex atomic systems (naturally including the rare-earth elements) are characterized by a great density. Moreover, these spectra have essentially relativistic properties. So, correct theoretical method of their studying can be based on the convenient field procedure, which includes computing the energy shifts DE of the degenerate electron states. More exactly, speech is about constructing secular matrix M *(*with using the Gell-Mann and Low adiabatic formula for DE), which is already complex in the relativistic theory, and its further diagonalization [26-32]. In result one could compute the energies and decay probabilities of a non-degenerate excited state for a complex atomic system [26]. The secular matrix elements can be further expanded into a PT series on the inter-electron interaction. Here the standard Feynman diagrammatic technique is usually used.

Generally speaking, the secular matrix M can be represented as follows:

$$M = M \quad + M \quad + M \quad + M \quad + \ldots + M^{k} \tag{10}$$

where $M^{(0)}$ is the contribution of the vacuum diagrams of all PT orders (this contribution determines only the general levels spectrum shift); $M^{(1)}$, $M^{(2)}, M^{(3)}$ are contributions of the 1-, 2- and 3- quasiparticle (QP) diagrams respectively. The matrix $M^{(1)}$ can be presented as a sum of the independent one-QP contributions. Substituting these quantities into (1) one could have summarized all the one-QP diagrams contributions. In the empirical methods here one could use the experimental values of one-electron energies, however, the necessary experimental quantities (especially for the rare-earth and other elements) are not often available. The detailed procedure for computing $\mathrm{Re}M^{(2)}$ is presented, for example, in Ref. [3].

We will describe an atomic multielectron system by the relativistic Dirac Hamiltonian (the atomic units are used) as follows [41-43]:

$$H=\sum_{i}\{\alpha c p_i-\beta c^2-Z/r_i\}+\sum_{i>j}\exp(i\,|\,\omega\,|\,r_{ij})(1-\alpha_i\alpha_j)/r_{ij} \quad (11)$$

where Z is a charge of nucleus, a_i, a_j are the Dirac matrices, w_{ij} is the transition frequency, c – the velocity of light. The interelectron interaction potential (second term in (3)) takes into account the retarding effect and magnetic interaction in the lowest order on parameter of the fine structure constant. In the PT zeroth approximation it is used ab initio mean-field potential:

$$V^{DKS}(r)=[V^{D}_{Coul}(r)+V_X(r)+V_C(r\,|\,a)], \quad (12)$$

with the standard Coulomb, exchange Kohn-Sham V_X and correlation Lundqvist-Gunnarsson Vc potentials (look details in Refs. [46-49]). An effective approach to accounting the multi-electron polarization contributions is described earlier and based on using the effective two-QP polarizable operator, which is included into the PT first order matrix elements.

In order to calculate the radiation decay probabilities and autoionization energies and widths a gauge invariant relativistic energy approach (version [43]) is used. In particular, a width of the state, connected with an autoionization decay, is determined by a coupling with the continuum states and calculated as square of the matrix element [43]:

$$V_{\beta_1\beta_2;\beta_4\beta_3}=\sqrt{(2j_1+1)(2j_2+1)(2j_3+1)(2j_4+1)}$$

$$\text{x}\sum_{a\mu}(-1)^{\mu}\begin{pmatrix} j_1 & j_3 & a \\ m_1-m_3 & & \mu\end{pmatrix}\begin{pmatrix} j_2 & j_4 & a \\ m_2-m_4 & & \mu\end{pmatrix}\times$$

$$\text{x}Q_a\left(n_1l_1j_1n_2l_2j_2;n_4l_4j_4n_3l_3j_3\right) \quad (13)$$

Here $=Q_a^{\mathrm{Qul}}+Q_a^{\mathrm{B}}$, where Q_a^{Qul}, Q_a^{B} correspond to the Coulomb and Breit parts of the relativistic interelectron potential in (3) and express through Slater-like radial integrals and standard angle coefficients. Other details can be found in Refs. [44-57].

The most complicated problem of the relativistic PT computing the complex multielectron lements spectra is in an accurate, precise accounting for the multi-electron exchange-correlation effects (including polarization and screening effects, a continuum pressure etc), which can be treated as the effects of the PT second and higher orders . Using the standard Feynman diagrammatic technique one should consider two kinds of diagrams (the polarization and ladder ones), which describe the polarization and screening exchange-correlation effects. The detailed description of the polarization diagrams and the corresponding analytical expressions for matrix elements of the polarization QPs interaction (through the polarizable core) potential is presented in Refs. [34-36]. An effective approach to accounting of the polarization diagrams contributions is in adding the effective two-QP polarizable operator into the PT first order matrix elements. In Ref. [27] the corresponding non-relativistic polarization functional has been derived. More correct relativistic expression has been presented in the Refs. [2] and used in our computing. The contribution of the ladder diagrams (these diagrams describe the immediate QPs interaction) is summarized by a modification of the PT zeroth approximation mean-field central potential (look below), which include the screening (anti-screening) of the core potential of each particle by the two others. The details of this contribution can be found in Refs. [44-57].

4. Results and Conclusions

In the framework of the development of spectroscopy of the AS of heavy atoms in the external field, a quantitative study of the effects of the non-conductive electric field on the parameters of the AS in the spectra of the lanthanide atoms was performed. Based on our theory, for the first time, the widths of the autoionization states for the Tm $4f^{13}{}_{7/2,5/2}\,6s_{1/2}$ (3,2) ns,np i $4f^{13}{}_{5/2}6s_{1/2}(2)nsp_{1/2}[3/2]$ (n=26,30) i Yb $4f^{13}$ $[^2F_{7/2}]$ $6s^2np[5/2]_2$ $4f^{13}$ $[^2F_{7/2}]$ $6s^2nf[5/2]_2$. In Table 1 we list our data on the widths of the $4f^{13}{}_{7/2,5/2}\,6s_{1/2}$ (3,2) ns,np states, which are mixed with the resonances of the opposite parity in a rather weak DC electric field.

Table 1.

The widths Γ (cm^{-1}) of autoionization states of the Tm $4f^{-13}{}_{7/2}6s_{1/2}$(3)ns,np, which are mixed with resonances of opposite parity for different DC electric fields

F(V/cm)		$4f^{13}{}_{7/2}6s_{1/2}(3)ns[5/2]$ n=26 n=30
Γ	F=0	1.13D-5 6.12D-6
Γ	F =50	1.11D-04 5.88D-5
Γ	F =100	4.05D-04 2.15D-4
Γ	F =150	8.15D-04 4.13D-4
F(V/cm)		$4f^{13}{}_{7/2}6s_{1/2}(3)np_{3/2}[3/2]$ n=26 n=30
Γ	F=0	4.22D-5 2.42D-5
Γ	F =50	4.07D-4 2.36D-4
Γ	F =100	1.56D-3 8.88D-4
Γ	F =150	3.08D-3 1.76D-3
F(V/cm)		$4f^{13}{}_{5/2}6s_{1/2}(2)np_{1/2}[3/2]$ n=26 n=30
Γ	F=0	2.36D-5 1.27D-5
Γ	F =50	2.23D-4 1.22D-4
Γ	F =100	8.37D-3 4.28D-4
Γ	F =150	1.64D-3 8.63D-3

Note: 1.13D-5=1.13×10^{-5};

From these data one could see that in this case there is the effect of a giant broadening of the resonance widths. For the first time, for Tm, the possibility of such an effect was foreseen in the papers by Glushkov-Ivanov-Letokhov, which was later confirmed in the known ISAN experiments by V.S. Letokhov etal (look details in Refs. [3,8]). Similar data are obtained for Yb, for which we first detected the effect of strong amplification of the AU .

We also present our results of numerical modelling ionization dynamics for Rydberg atoms Rb, Cs, Fr (Rb: *n*=50-80; Cs, Fr: *n*=60-80) in a microwave field (F=$(1.2\text{-}3.2)\times10^{-9}$ a.u.; w/2p=8.87, 36 HGz). The preliminary estimate a dependence of the Rb ionization probability P upon the F, interaction time "atom-field" and comparison with available data by Krug-Buchleitner [19] and Glushkov-Ternovsky etal [49] shows that all listed data are in a reasonable agreement with experiment, however, the best accuracy is provided by relativistic theory. In Table 2 we firstly present new data on dependence of the Fr ionization probability upon the F value, interaction time "atom-field". Unfortunately, here there are no any alternative theoretical or experimental data.

Table 2.

Our data for ionization probability P for Fr (l_0=0, m_0=0, n_0=76-80) in dependence on n_0 F (at.units; field parameters: t = 327× 2p/w; frequency w_c=w/2p=36 GHz, 8.87 GHz)

n_0 ↓	Our data	Our data	Our data	Our data
F= ω_c=	2.8×10^{-9} 36GHz	3.1×10^{-9} 36GHz	2.8×10^{-9} 8.87GHz	3.1×10^{-9} 8.87GHz
77	0.47	0.50	0.43	0.46
80	0.58	0.61	0.54	0.56
83*	0.56	0.60	0.51	0.53
86	0.67	0.69	0.62	0.66

In whole, our modeling relativistic dynamics of ionization Rb, Cs, Fr Rydberg states in the electromagnetic field shows that there are the local violations of probability smooth growth associated with the complex Floquet spectrum, link between the quasi-stationary states and a continuum, the growing influence of multiphoton resonances. The picture becomes by more complicated due to the single-photon near-resonance transitions with quasi-random detuning from resonance and quantum phase shift due to scattering Rydberg electron on the atomic core.

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RELATIVISTIC OPERATOR PERTURBATION THEORY IN SPECTROSCOPY OF MULTIELECTRON ATOM IN AN ELECTROMAGNETIC FIELD

Summary

We present the theoretical basis of a new relativistic operator perturbation theory (OPT) approach to multielectron atom in an electromagnetic field combined with a relativistic many-body perturbation theory (RMBPT) formalism for a free multielectron atom. As illustration of application of the presented formalism, the results of energy and spectral parameters for a number of atoms are presented. The relativistic OPT method is tested for the multielectron systems such as Fr and Tm. New approach is elaborated for an accurate, consistent treatment of a strong field Stark effect in multielectron atoms.

Keywords: multielectron atom in a dc electric field – modified operator perturbation theory – Stark resonances

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РЕЛЯТИВИСТСКАЯ ОПЕРАТОРНАЯ ТЕОРИЯ ВОЗМУЩЕНИЙ В СПЕКТРОСКОПИИ МНОГОЭЛЕКТРОННОГО АТОМА В ЭЛЕКТРОМАГНИТНОМ ПОЛЕ

Резюме

Изложены теоретические основы нового аппарата релятивистской операторной теории возмущений (ОТВ) в спектроскопии многоэлектронного атома в электромагнитном поле, объединенного с формализмом релятивистской многочастичной теории возмущений для свободного многоэлектронного атома. В качестве иллюстрации тестирования представленного подхода представлены результаты оценки энергетических и спектральных параметров для ряда атомов. Релятивистский метод ОРТ тестируется для таких многоэлектронных систем как Fr и Tm. Новый подход разработан для последовательного описания эффекта Штарка в многоэлектронных атомах в сильном внешнем электромагнитном поле.

Ключевые слова: Многоэлектронный атом в электрическом поле - модифицированная операторная теория возмущений – штарковские резонансы

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РЕЛЯТИВІСТСЬКА ОПЕРАТОРНА ТЕОРІЯ ЗБУРЕНЬ В СПЕКТРОСКОПІЇ БАГАТОЕЛЕКТРОННОГО АТОМА В ЕЛЕКТРОМАГНІТНОМУ ПОЛІ

Резюме

Викладені теоретичні основи нового апарату релятивістської операторної теорії збурень (ОТЗ) в спектроскопії багатоелектронного атома в електромагнітному полі, об’єднаного з формалізмом релятивістської багаточастинкової теорії збурень для вільного багатоелектронного атома. В якості ілюстрації можливостей представленого підходу представлені результати оцінки деяких енергетичних і спектральних параметрів для ряду атомів. Релятивістський метод ОРЗ тестується для таких багатоелектронних систем як Fr і Tm. Новий підхід розроблений для послідовного опису ефекту Штарка в багатоелектронних атомах в сильному зовнішньому електромагнітному полі.

Ключові слова: багатоелектронний атом у електричному полі - модифікована операторна теорія збурень – штарківські резонанси