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SPECTROSCOPY OF AUTOIONIZATION STATES IN SPECTRA OF HELIUM, BARIUM AND LEAD ATOMS: NEW SPECTRAL DATA AND CHAOS EFFECT

We applied a generalized energy approach (Gell-Mann and Low S-matrix formalism) combined with the relativistic multi-quasiparticle (QP) perturbation theory (PT) with the Dirac-Kohn-Sham zeroth approximation to studying excites states and autoionization resonances (AR) in complex atoms and ions, in particular, energies and widths for the He and He-like ion, barium and lead atoms, with accounting for the exchange-correlation, relativistic corrections and the weak chaos effect.

1. Introduction

Traditionally an investigation of spectra, spectral, radiative and autoionization characteristics for heavy and superheavy elements atoms and multicharged ions is of a great interest for further development atomic and nuclear theories and different applications in the plasma chemistry, astrophysics, laser physics, etc. (look Refs. [1–10]). Theoretical methods of calculation of the spectroscopic characteristics for heavy atoms and ions may be divided into a few main groups [1-6]. First, the well known, classical multi-configuration Hartree-Fock method (as a rule, the relativistic effects are taken into account in the Pauli approximation or Breit hamiltonian etc.) allowed to get a great number of the useful spectral information about light and not heavy atomic systems, but in fact it provides only qualitative description of spectra of the heavy and superheavy ions. Second, the multi-configuration Dirac-Fock (MCDF) method is the most reliable version of calculation for multielectron systems with a large nuclear charge. In these calculations the one- and two-particle relativistic effects are taken into account practically precisely. In this essence it should be given special attention to two very general and important computer systems for relativistic and QED calculations of atomic and molecular properties developed in the Oxford group and known as GRASP (“GRASP”, “Dirac”; “BERTHA”, “QED”) (look [1-5] and refs. therein). In particular, the BERTHA program embodies a new formulation of relativistic molecular structure theory within the framework of relativistic QED. This leads to a simple and transparent formulation of Dirac-Hartree-Fock-Breit (DHFB) self-consistent field equations along with algorithms for molecular properties, electron correlation, and higher order QED effects. The DHFB equations are solved by a direct method based on a relativistic generalization of the McMurchie-Davidson algorithm [4]. In this paper we applied a new relativistic approach [11-15] to relativistic studying the excites states spectra and autoionization states for the He and He-like ion, barium and lead atoms. New approach in optics and spectroscopy of heavy atomic systems is the combined the generalized energy approach and the gauge-invariant QED many-QP PT with the Dirac-Kohn-Sham (DKS) “0” approximation (optimized 1QP representation) and an accurate accounting for relativistic, correlation, nuclear, radiative effects. [17-20].

2. Method

The generalized gauge-invariant version of the energy approach has been further developed in Refs. [12,13]. In relativistic case the Gell-Mann and Low formula expressed an energy shift DE through the QED scattering matrix including the interaction with as the photon vacuum field as the laser field. The first case is corresponding to definition of the traditional radiative and autoionization characteristics of multielectron atom. The wave function zeroth basis is found from the Dirac-Kohn-Sham equation with a potential, which includes the ab initio (the optimized model potential or DF potentials, electric and polarization potentials of a nucleus; the Gaussian or Fermi forms of the charge distribution in a nucleus are usually used) [5]. Generally speaking, the majority of complex atomic systems possess a dense energy spectrum of interacting states with essentially relativistic properties. Further one should realize a field procedure for calculating the energy shifts DE of degenerate states, which is connected with the secular matrix *M* diagonalization [8-12]. The secular matrix elements are already complex in the second order of the PT. Their imaginary parts are connected with a decay possibility. A total energy shift of the state is presented in the standard form:

$$\Delta \mathrm{E} = \mathrm{Re}\,\Delta E + i\,\mathrm{Im}\,\Delta E \qquad \mathrm{Im}\,\Delta E = -\Gamma/2\,, \tag{1}$$

where Γ is interpreted as the level width, and the decay possibility $\mathrm{P} = \Gamma$. The whole calculation of the energies and decay probabilities of a non-degenerate excited state is reduced to the calculation and diagonalization of the *M*. The *jj*-coupling scheme is usually used. The complex secular matrix *M* is represented in the form [9,10]:

$$M = M^{(0)} + M^{(1)} + M^{(2)} + M^{(3)}. \tag{2}$$

where $M^{(0)}$ is the contribution of the vacuum diagrams of all order of PT, and $M^{(1)}$, $M^{(2)}$, $M^{(3)}$ those of the one-, two- and three-QP diagrams respectively. $M^{(0)}$ is a real matrix, proportional to the unit matrix. It determines only the general level shift. We have assumed $M^{(0)} = 0$. The diagonal matrix $M^{(1)}$ can be presented as a sum of the independent 1QP contributions. For simple systems (such as alkali atoms and ions) the 1QP energies can be taken from the experiment. Substituting these quantities into (2) one could have summarized all the contributions of the 1QP diagrams of all orders of the formally exact QED PT. However, the necessary experimental quantities are not often available. So, the optimized 1-QP representation is the best one to determine the zeroth approximation. The correlation corrections of the PT high orders are taken into account within the Green functions method (with the use of the Feynman diagram's technique). All correlation corrections of the second order and dominated classes of the higher orders diagrams (electrons screening, polarization, particle-hole interaction, mass operator iterations) are taken into account [10-14]. In the second order, there are two important kinds of diagrams: polarization and ladder ones. Some of the ladder diagram contributions as well as some of the 3QP diagram contributions in all PT orders have the same angular symmetry as the 2QP diagram contributions of the first order [10-12]. These contributions have been summarized by a modification of the central potential, which must now include the screening (anti-screening) of the core potential of each particle by two others. The additional potential modifies the 1QP orbitals and energies. Then the secular matrix is : $M \simeq \tilde{M}^{(1)} + \tilde{M}^{(2)}$, where $\tilde{M}^{(1)}$ is the modified 1QP matrix (diagonal), and $\tilde{M}^{(2)}$ the modified 2QP one. $\tilde{M}^{(1)}$ is calculated by substituting the modified 1QP energies), and $\tilde{M}^{(2)}$ by means of the first PT order formulae for $M^{(2)}$, putting the modified radial functions of the 1QP states in the interaction radial integrals. Let us remind that in the QED theory, the photon propagator D(12) plays the role of this interaction. Naturally, an analytical form of D depends on the gauge, in which the electrodynamic potentials are written. In general, the results of all approximate calculations depended on the gauge. Naturally the correct result must be gauge invariant. The gauge dependence of the amplitudes of the

photoprocesses in the approximate calculations is a well known fact and is in details investigated by Grant, Armstrong, Aymar-Luc-Koenig, Glushkov-Ivanov [1,2,5,9]. Grant has investigated the gauge connection with the limiting non-relativistic form of the transition operator and has formulated the conditions for approximate functions of the states, in which the amplitudes are gauge invariant. These results remain true in an energy approach as the final formulae for the probabilities coincide in both approaches. In ref. [16] it has been developed a new version of the approach to conserve gauge invariance. Here we applied it to get the gauge-invariant procedure for generating the relativistic DKS orbital bases (abbreviator of our method: GIRPT). The autoionization width is defined by the square of interaction matrix element [9]:

$$V^{\omega}_{1234} = [(j_1)(j_2)(j_3)(j_4)]^{1/2} \sum_{\lambda\mu} (-1)^{\mu} \begin{pmatrix} j_1 j_3 & \lambda \\ m_1 - m_3 & \mu \end{pmatrix} \times \times \mathrm{Re}\, Q_{\lambda}(1234) \tag{3}$$

The real part of the interaction matrix element can be expanded in terms of Bessel functions [5,8]:

$$\frac{\cos|\omega| r_{12}}{r_{12}} = \frac{\pi}{2\sqrt{r_1 r_2}} \sum_{\lambda=0} (\lambda) J_{\lambda+1/2}(|\omega| r_<) J_{-\lambda-1/2}(|\omega| r_>) P_{\lambda}(\cos \mathbf{r}_1 \mathbf{r}_2) \tag{4}$$

The Coulomb part $Q^{\mathrm{Qul}}_{\lambda}$ is expressed in the radial integrals R_1, angular coefficients S_1 as follows:

$$\mathrm{Re}\, Q^{\mathrm{Qul}}_{\lambda} \sim \mathrm{Re}\{R_I(1243) S_{\lambda}(1243) + R_{\lambda}(\tilde{1}\,24\tilde{3}) S_{\lambda}(\tilde{1}\,24\tilde{3}) + R_{\lambda}(1\tilde{2}\tilde{4}3) S_{\lambda}(1\tilde{2}\tilde{4}3) + R_{\lambda}(\tilde{1}\,\tilde{2}\tilde{4}\,\tilde{3}) S_{\lambda}(\tilde{1}\,\tilde{2}\tilde{4}\,\tilde{3})\} \tag{5}$$

where $\mathrm{Re}Q_1(1243)$ is as follows:

$$\mathbf{Re}\, R_{\lambda}(1243) = \iint dr_1 r_1^2 r_2^2 f_1(r_1) f_3(r_1) f_2(r_2) f_4(r_2) Z^{(1)}_{\lambda}(r_<) Z^{(1)}_{\lambda}(r_>) \tag{6}$$

where f is the large component of radial part of the 1QP state Dirac function and function Z is :

$$Z^{(1)}_{\lambda} = [2/|\omega_{13}|\alpha Z]^{\lambda+1/2} J_{\lambda+1/2}(\alpha|\omega_{13}| r)/[r^{\lambda}\Gamma(\lambda + 3/2)]. \tag{7}$$

The angular coefficient is defined by standard way as above [3]. The calculation of radial integrals $\mathbf{Re}R_1(1243)$ is reduced to the solution of a system of differential equations:

$$\left.\begin{aligned} y_1' &= f_1 f_3 Z^{(1)}_{\lambda}(\alpha|\omega| r) r^{2+\lambda}, \\ y_2' &= f_2 f_4 Z^{(1)}_{\lambda}(\alpha|\omega| r) r^{2+\lambda}, \\ y_3' &= [y_1 f_2 f_4 + y_2 f_1 f_3] Z^{(2)}_{\lambda}(\alpha|\omega| r) r^{1-\lambda}. \end{aligned}\right\} \tag{8}$$

In addition, $y_3(\infty)=\mathbf{Re}R_{\lambda}(1243)$, $y_1(\infty)=X_{\lambda}(13)$. The system of differential equations includes also equations for functions $f/r^{|æ|-1}$, $g/r^{|æ|-1}$, $Z^{(1)}_{\lambda}$, $Z^{(2)}_{\lambda}$. The formulas for the autoionization (Auger) decay probability include the radial integrals $R_a(akgb)$, where one of the functions describes electron in the continuum state. When calculating this integral, the correct normalization of the function Ψ_k is a problem. The correctly normalized function should have the following asymptotic at $r \to 0$:

$$\left.\begin{matrix} f \\ g \end{matrix}\right\} \to (\lambda\omega)^{-1/2} \begin{cases} [\omega + (\alpha Z)^{-2}]^{-1/2} \sin(kr + \delta), \\ [\omega - (\alpha Z)^{-2}]^{-1/2} \cos(kr + \delta). \end{cases} \tag{9}$$

When integrating the master system, the function is calculated simultaneously:

$$N(r) = \{\pi\omega_k [f_k^2 [\omega_k + (\alpha Z)^{-2}] + g_k^2 [\omega_k + (\alpha Z^{-2})]]\}^{-1/2} \tag{10}$$

It can be shown that at r®¥, $N(r) \to N_k$, where N_k is the normalization of functions f_k, g_k of continuous spectrum satisfying the condition (9). Other details can be found in refs.[10-13,16-20].

3. Results and conclusions

In Table 1 we present the results of our calculating energies of levels in Pb: experimental data and the results of the calculation method in the framework of the relativistic Hartree-Fock (RHF) (see [21] and references therein) and our theory. Our data are in quite good agreement with the experimental data.

Table 2 shows our calculated result for transition probabilities corresponding to the line 1278.9 nm in the spectrum of Pb, together with the results of alternative theories, in particular, the data obtained on the basis of the calculation in the many approximation DF using, respectively, the theoretical and experimental values of the DF

transition energy (MCDF: theor.E; exp.E), in the many relativistic random phase approximation (MCRRPA), in the relativistic Hartree-Fock (RHF), in the intermediate-coupling approximation (ICC) and finally, the method of empirical MP (EMP) [22].

Table.1.

The energies of the levels of the np2, npn's (n = 6, n '= 7) lead (see text)

		Exp.	RHF	RPT
np^2	$^3P_0$	59821	59960	59862
	$^3P_1$	52002	51996	52005
	$^3P_2$	49171	49174	49172
	$^1D_2$	38363	38301	38352
	$^1S_0$	30354	30282	30332
npn's	$^3P_0$	24861	25163	24924
	$^3P_1$	24534	25721	24598

Table 2.

Probability (s^{-1}) of transition corresponding to the line 1278.9 nm in Pb

RPT-EA (our data)	6.95
MCDF (exp.E); Horodecki et al	7.27
MCDF (theor.E); Horodecki et al	4.29
EMP; Horodecki et al	7.50
RHF (Biemont-Quinat)	7.85
MCRRPA (Chou-Huang)	7.00
RHF (Dzuba et al)	7.08
ICC (Garstang)	7.14
Laser absorption	6.1±1.5

In the next table 3 we present the measured and calculated autoionization width (cm^{-1}) for 5d5g levels of the Ba atom with angular momentum J <6: (3) Experiment (4) Theory: HF method with a partial view of the correlations (Van Leeuwen et al; (5) R-matrix method without into account the effect of the polarization shells basis: 6snl +5 dnl (Luc-Koenig et al); (6) R-matrix method taking into account the polarization of the c shell and without dielectronic polarization interaction (Luc-Koenig et al); (7) R-matrix method taking into account the effect of the polarization of shells and dielectronic polarization interaction (Luc-Koenig et al); (8) - our theory, taking full account of correlations. As indicated in Ref. [23]. The experimental studying of the spectrum considered had been performed in the remarkable work [24].

Analyzing shown in Table 3 the data on measured and calculated widths for 5d5g levels of the Ba atom with momentum J = 6, we note that to reach a physically reasonable agreement between theory and experiment it is necessary to carry put a precise account of the polarization and dielectronic interaction effects. This has been accurately done in the framework of our theory and the R-matrix method Luc-Koenig et al [24]. In the last paper the known polarization potentials have used with such a parameter as static dipole polarizability. In our theory we have used the multiparticle polarization functionals [12], which have been obtained by summation of the PT polarization diagrams sequence in the Thoams –Fermi approximation.

Another interesting feature of the auto resonances spectra in barium is connected with elements of the weak chaos. In ref. [25] we have treated a problem of a chaos manifestation in dynamical systems from the geometrical and spectral points point of view. There3 we have presented the theoretical basis's of a consistent chaos-geometrical approach to treating of chaotic dynamical systems which combines together the non-linear analysis methods to dynamics, such as the wavelet analysis, multi-fractal formalism, mutual information approach, correlation integral analysis, false nearest neighbour algorithm, Lyapunov exponent's analysis, surrogate data method etc. Application of this approach to studying dense and very complicated spectrum of barium (there are taken into account more 400 levels and resonances, including the Rydberg, twice excited Autoionization resonances etc)) has shown that one could find the elements of the weak chaos (the first two spectral Lyapunov's exponents have positive and a little negative values).

The next system to be studied is the systems of the autoionization resonances in helium, which includes states with double excited electrons. Table 4 shows the data of our theory for the energy and width of the Autoionization resonance 3s3p

$^1P_0$, along with those of other theories (see details in [27] and references therein). In the next table 5 we present some of the theoretical and experimental data on the energy and the width of the resonance 3s3p $^1P_0$, (theoretical): our theory, the method of complex rotation method diagonalization MHF R-matrix method; Experiment: NIST (NBS 2SO-MeV electron synchrotron storage ring (SURF-II)), Wisconsin laboratory (Wisconsin Tantalus storage ring), Stanford Synchrotron Radiation Laboratory (SSRL), Berlin electron storage ring (BESSY), Daresbury Synchrotron Radiation Source (DSRS) [27]. With regard to the data of different methods it should be noted that these data are different theories degree of agreement with the experimental data. Surely, the value of of any theory is determined by not only the studying the helium atom, but also of other atoms, as well as the level of complexity of the computational procedure. In this sense, the method of complex rotation stand, MHF, R-matrix approach is now actively used in different tasks.

The results of our theory is physically reasonable, fairly good agreement with the experimental data obtained in a number of well-known laboratories.

Table 3. The measured and calculated autoionization width (cm^{-1}) for 5d5g levels of the Ba atom with angular momentum J <6:
(3) Experiment (4) Theory: HF method (5) R-matrix method without account of polarization; (6) R-matrix method with account polarization and without dielectronic interaction; (7) R-matrix method with account of polarization of and dielectronic interaction; (8) - our theory

1:j	K	J	2: εl	(3)	(4)	(5)
$5d_{3/2}$	5/2	2	εd	0.062(4)	0.0074	0.077
$5d_{3/2}$	5/2	3	εd	0.056(5)	0.0074	0.048
$5d_{3/2}$	7/2	3	εg	0.34(3)	1.96	2.34
$5d_{3/2}$	7/2	4	εg	0.34(3)	1.96	2.41
$5d_{3/2}$	9/2	4	εg	0.18(2)	1.00	1.11
$5d_{3/2}$	9/2	5	εg	0.18(2)	1.00	1.19
$5d_{3/2}$	11/2	5	εi	-	2.90	3.19
$5d_{3/2}$	11/2	6	εi	-	2.90	3.20
$5d_{5/2}$	3/2	1	εd	0.046(3)	0.012	0.038
$5d_{5/2}$	3/2	2	εd	0.057(4)	0.012	0.077
$5d_{5/2}$	5/2	2	εd	0.0107(15)	0.0041	0.025
$5d_{5/2}$	5/2	3	εd	0.0174(23)	0.0041	0.017
$5d_{5/2}$	7/2	3	εg	0.29(3)	1.43	1.61
$5d_{5/2}$	7/2	4	εg	0.29(3)	1.43	1.73
$5d_{5/2}$	9/2	4	εg	0.43(4)	2.29	2.57
$5d_{5/2}$	9/2	5	εg	0.43(4)	2.29	2.71
$5d_{5/2}$	11/2	5	εi	0.28(3)	0.45	0.56
$5d_{5/2}$	11/2	6	εi	0.32(3)	0.45	0.55
$5d_{5/2}$	13/2	6	εi	-	3.37	3.95
1:j	K	J	2: εl	(6)	(7)	(8)
5d3/2	5/2	2	εd	0.072	0.018	0.058
5d3/2	5/2	3	εd	0.046	0.025	0.055
5d3/2	7/2	3	εg	0.12	0.34	0.34
5d3/2	7/2	4	εg	0.12	0.33	0.33
5d3/2	9/2	4	εg	0.11	0.23	0.20
5d3/2	9/2	5	εg	0.11	0.23	0.19
5d3/2	11/2	5	εi	1.44	1.74	1.52
5d3/2	11/2	6	εi	1.44	1.73	1.51
5d5/2	3/2	1	εd	0.066	0.041	0.044
5d5/2	3/2	2	εd	0.061	0.025	0.053
5d5/2	5/2	2	εd	0.023	0.008	0.009
5d5/2	5/2	3	εd	0.021	0.013	0.015
5d5/2	7/2	3	εg	0.13	0.29	0.28
5d5/2	7/2	4	εg	0.13	0.31	0.30
5d5/2	9/2	4	εg	0.22	0.48	0.45
5d5/2	9/2	5	εg	0.22	0.48	0.46
5d5/2	11/2	5	εi	0.25	0.30	0.28
5d5/2	11/2	6	εi	0.25	0.30	0.29
5d5/2	13/2	6	εi	1.83	2.15	2.17

Table 4. Energy and width of the resonance 3s3p $^1P_0$ (theoretical): our theory, the method of complex rotation (MCR), the algebraic close coupling (ACC), a method of diagonalization (DM), multiconf. HF, R-matrix method, the adiabatic potential curves method (APC), L^2 technique with using the Sturmian expansions, Feshbach method (FM), K -matrix (KM)

Method		E_r (Ry)	Γ/2 (Ry)
PT-EA	This	-0.668802	0.006814
MCR	Ho	-0.671252	0.007024
ACC	Wakid etaa	-0.670	0.00695
MD	Senashenko	-0.6685	0.00548
MHF	Nicolaides	-0.671388	-
R-matr	Hayes-Scott	-0.6707	0.00660
APC	Koyoma et al	-0.6758	-
APC	Sadeghpour	-0.67558	-
L^2 tech.	Gershacher	-0.67114	0.00704
FM	Wu et al	-0.669 27	0.00420
KM	Moccia et al	-0.670 766	0.00676

Table 5. Theoretical and experimental data on the energy and the width of the resonance 3s3p $^1P_0$, (theoretical): our theory, the method of complex rotation method diagonalization MHF R-matrix method; Experiment: NIST (NBS), Wisconsin (Wisconsin storage ring), Stanford - SSRL; Berlin-BESSY; Daresbury-DSRS.

	E_r (eV)	Γ/2 (eV)
This	69.9055	0.1854
MCR	69.8722	0.1911
MHF	69.8703	-
R-matr.	69.8797	0.1796
NBS	69.919±0.007	0.132±0.014
Wisconsin	69.917±0.012	0.178±0.012
SSRL	69.917±0.012	0.178±0.012
BESSY	69.914±0.015	0.200±0.020
DSRS	69.880±0.022	0.180±0.015

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SPECTROSCOPY OF AUTOIONIZATION RESONANCES IN SPECTRA OF HE-LIKE IONS AND ALKALI-EARTH ATOMS: NEW SPECTRAL DATA AND CHAOS EFFECT

Abstract

We applied a generalized energy approach (Gell-Mann and Low S-matrix formalism) combined with the relativistic multi-quasiparticle (QP) perturbation theory (PT) with the Dirac-Kohn-Sham zeroth approximation to studying excites states and autoionization resonances (AR) in complex atoms and ions, in particular, energies and widths for the He and He-like ion, barium and lead atoms, with accounting for the exchange-correlation, relativistic corrections and a chaos effect.

Key words: spectroscopy of autoionization resonances, relativistic energy approach

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СПЕКТРОСКОПИЯ АВТОИОНИЗАЦИОННЫХ РЕЗОНАНСОВ В СПЕКТРАХ He-ПОДОБНЫХ ИОНОВ И ЩЕЛОЧНО-ЗЕМЕЛЬНЫХ АТОМОВ: НОВЫЕ СПЕКТРАЛЬНЫЕ ДАННЫЕ И ЭФФЕКТ ХАОСА

Резюме

Обобщенный энергетический подход (S-матричный формализм Гелл-Мана и Лоу) и релятивистская теория возмущений с дирак-кон-шэмовским нулевым приближением применены к изучению спектра возбужденных состояний и автоионизационных резонансов в сложных атомах, в частности, энергий и ширин автоионизационных резонансов в гелии, барии и свинце, с учетом обменно-корреляционных и релятивистских поправок и эффекта хаоса.

Ключевые слова: спектроскопия автоионизационных резонансов, релятивистский энергетический подход

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СПЕКТРОСКОПІЯ АВТОІОНІЗАЦІЙНИХ РЕЗОНАНСІВ В СПЕКТРАХ He- ПОДІБНИХ ІОНІВ ТА ЛУЖНО-ЗЕМЕЛЬНИХ АТОМІВ: НОВІ СПЕКТРАЛЬНІ ДАНІ ТА ЕФЕКТ ХАОСУ

Резюме

Узагальнений енергетичний підхід (S-матричний формалізм Гелл-Мана та Лоу) и релятивістська теорія збурень з дірак-кон-шемівським нульовим наближенням застосовані до вивчення збуджених спектрів збуджених станів та автоіонізаційних резонансів у складних атомах, зокрема, енергій та ширин автоіонізаційних резонансів у гелію, барії та свинцу з урахуванням обмінно-кореляційних і релятивістських поправок та ефекту хаосу.

Ключові слова: спектроскопія автоіонізаційних резонансів, релятивістський енергетичний