Method of Sturm orbitals in calculations of physical characteristics of radiation from atoms and ions

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A technique for calculating spectroscopic constants of multielectron ions, such as energy levels, rates of radiative and collisional transitions, effectively taking into account the continuous spectrum and relativistic effects has been proposed. The technique uses a mixed basis composed of orbitals of discrete states derived from the Dirac equation and orbitals of the Sturm extension. The Sturm extension effectively covers a region of the phase space including an infinite set of real bound atomic wave functions as well as continuum states. The convergence of the technique in terms of the number of orbitals in the set has been investigated. The effect of the continuum on the kinetics of level population is important in calculations of the laser gain due to transitions from highly excited states of a multicharged ion. © *1996 American Institute of Physics.* [S1063-7761(96)01008-6]

1. INTRODUCTION

Calculation of emission spectra of plasma ions based on high-precision quantum-mechanical techniques is a practical tool which may be used instead of very expensive sophisticated experiments. Given systematic data about intensities of spectral lines and respective gains, one can establish basic rules of plasma motion in phase space. An important application of the theory of atomic spectra in plasma is determination of the optimum plasma excitation condition for lasing and discovery of new pumping approaches. Given an adequate theoretical model, one can, in principle, control plasma motion in phase space in order to optimize the lasing effect. An investigation of plasma motion over a large volume in phase space is necessary for detailed interpretation of experimental data on laser gain in plasma. In this connection, the relevant problem is to develop an efficient technique for calculating atomic constants and plasma kinetics taking into account regions of the phase space containing an infinite number of states. This technique is also important for the plasma diagnostics and understanding the evolution of the plasma by comparison between calculated and measured spectra.

Vinogradov *et al.*[1] proposed a radiative-collisional model of lasing on multicharged Ne-like ion transitions. This concept was later confirmed and developed in many laboratories.[2,3] In most of these studies, plasma was the lasing medium. Recently the capillary discharge has proved to be a convenient table-top source of far ultraviolet radiation.[4–6]

Dedicated investigations of capillary discharges demonstrated that electron velocities in them may be up to ultrarelativistic values under conditions of ultrashort discharge times (t_{dis}= 100–1000 ps). The atomic densities and energies of these discharges satisfy the conditions of lasing due to transitions in ions of the neon isoelectron series, including the krypton ion with electron structure similar to that of neon. Earlier[7] we proposed to use a two-step capillary discharge with the second ultrashort discharge in order to obtain a high laser gain. The basic idea was that the discharge conditions required to ionize an atom to the neon-like state and to excite the resulting ion should be essentially different. The first (preliminary) relatively long discharge converts the plasma to a quasi-uniform state whose dominant components are Na- and Ne-like ions. In the first stage, the electron temperature and degree of inversion are relatively low. The second ultrashort discharge generates beams of superhot electrons which transform the plasma to a highly-inverted state. Preliminary calculations of the gain due to flows of superhot electrons in pure plasma of iron,[8] selenium,[9] argon,[10] and krypton[11] demonstrated that the presence of superthermal electrons in plasma leads to a considerably higher degree of inversion and higher gains due to certain transitions in Ne-like ions. This inversion is short-lived (transient), and its duration is controlled by the rate of transitions from Ne-like ions to F-like ions with a higher ionization. Another characteristic time of this short stage is the time of electron thermalization (maxwellization). Both these processes directly determine the time of the second (ultrashort) discharge. In our previous publications[10,11] we demonstrated that under certain conditions it is possible to obtain essential gains (2–3 cm^{-1}) on the line of the $2s2p^6 3s[J=0]-2s^2 2p^5 3s[J=1]$ transition in a Ne-like ion, which is usually called the $2s-2p$ transition. Thus the laser wavelength can be shortened by tuning the discharge parameters. In our preliminary calculations, we did not take into account Rydberg and autoionizing states of Ne-like ions. In plasma processes involving high-energy electrons, however, these states largely determine the ratios of spectral line intensities. These states should be included in calculations of level collisional broadening, whereas line widths control laser gains in plasma. The importance of highly excited states in the kinetics was indicated by recent calculations of line intensities in the Ne-like selenium.[12] These calculations demonstrated that hundreds of electronic configurations should be included in the kinetics with due account of all elementary processes which change the ionization degree of a given ion by a unit. An important point is the consistency of calculations of popula-

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tion kinetics and decay rate coefficients of atomic states. In our previous paper[13] we proposed a radically new approach to the problem of inclusion of all elementary processes changing the ionization degree by a unit. We considered plasma containing Ne- and Na-like ions. The kinetic equations explicitly included 37 lower states of the Ne-like ions, and a set of Rydberg states (including those of the continuum) of the Na-like ion corresponded to each Ne-like state. The population as a function of the Rydberg electron energy was introduced for each set. All the elementary processes connecting these two ions, as well as those leading to transitions within each Rydberg set, were taken into account. This approach which effectively takes into account an infinite set of states of the ion of the previous ionization state allowed us to calculate precisely the ionization balance in plasma as a function of electron temperature and density.[10]

It is remarkable that practically all existing models of ion spectra in plasma yield results which are very different from experimental line intensities, even when the time-and-space resolution of measurements is fairly good. In our opinion, there are at least three important factors leading to disagreement between simulations of plasma spectra and experimental data: (1) inconsistency of atomic calculations, which leads to large errors in rates of elementary process in some cases; (2) uncertainty of the real electron-velocity distribution function and its time dependence in nonstationary plasma sources; (3) in calculations of the level population and atomic decay rates, one should, in principle, take into consideration an infinite set of states. Two aspects of the latter problem must be considered: (a) the effect of the continuum on the atomic decay rate; (b) the inclusion of Rydberg and autoionizing states of a given ion in the kinetic equations.

The paper is dedicated to the fundamental problem of calculating the radiative decay rates of atomic states using the technique of Sturm orbitals. The aim of this study is to develop an efficient method taking into account the continuous spectrum and to investigate using numerical techniques the convergence of the method as the number of orbitals taken into account increases, i.e., as the number of electron configurations included in the energy matrix increases. The technique is illustrated by a calculation of energy levels, including high-energy states, and rates of radiative transitions in a Ne-like argon ion. This is a continuation of our previous research.[10,11,13–15]

Earlier[14] we demonstrated that many versions of multiconfigurational atomic calculations, including their relativistic generalizations, contradict the principles of the consistent quantum-electrodynamic (QED) theory. This may lead to enormous errors in calculations of radiative and collisional decay rates, and in this case the interpretation of lasing plasma spectra will be erroneous. Our technique for calculating atomic constants is based on the consistent QED theory[16–19] and is, in principle, free from many flaws of multiconfigurational techniques. Nevertheless, a correct calculation inevitably includes an investigation of the convergence with the number of orbitals taken into account. The difficulties of this investigation are self-evident; they derive primarily from the large number of components in the energy matrix. The major problem is to take into account the contribution of the continuous spectrum to the matrix elements.

A way of taking into account the effect of the continuum using only the states of the discrete spectrum was first indicated by Fock.[20] Later this approach was used in calculations of perturbation corrections to states of hydrogen-like atoms.[21–23] The Sturm extensions in the Hartree–Fock method were used in calculations of the Stark effect,[24] static and dynamic polarizabilities of atoms and ions.[25]

In this paper we suggest to apply the Sturm extension to conventional calculations explicitly taking into account many-body effects. Section 2 of the paper gives a systematic description of the energy approach to calculations of decay rates. In Sec. 3 we discuss the spectral representation of the Green's function in terms of configuration superposition. The method of generating the one-particle basis of the usual Dirac orbitals and its Sturm extension is described in Sec. 4. Section 5 gives a scheme of calculating the rates of radiative transitions between exited states of a Ne-like ion using the energy approach within the framework of the QED theory. Numerical results are discussed in Sec. 6, where the convergence of the results with the number of orbitals in different bases composed from orbitals of the usual (real) states and orbitals of the Sturm extension is investigated.

2. ENERGY APPROACH TO CALCULATIONS OF TRANSITION RATES

In terms of the energy approach to the consistent QED theory, the level width, irrespective of the underlying physical cause, is determined by the imaginary part of its energy. Strictly speaking, the Dirac equation is only applicable to a single-electron atom. Nonetheless, the QED approach which uses the Dirac equation with a model (bare) potential as a zeroth approximation of the multielectron atom[26,27] and is exact from the formal viewpoint is valid. The bare potential imitates the potential generated by the atomic nucleus and core electrons. In this model, the outer electrons and vacancies in the inner shells (quasi-particles) move in the field of the bare potential and interact with each other directly via electromagnetic field, via polarized inner shells, and also through the polarization of the electron-positron field vacuum. In studying a system of two or more quasi-particles by using the energy approach, one should calculate a complex secular matrix for a set of degenerate or nearly degenerate atomic states. In the lowest order of perturbation theory, the secular matrix is identical to the energy matrix.[28] In the case of an isolated atom, the imaginary part of the first order correction due to the electron-electron interaction (the second order of QED) directly yields the spontaneous radiative decay rate of the state.[29–31] The autoionization decay of the state can be manifested only in the second order of perturbation theory, in which the imaginary parts of the higher-order corrections contain terms due to multiphoton and interference effects.[29,30] We proposed the energy approach based on the consistent QED theory for calculating cross sections of electron-atomic collisions in our earlier publication.[18] Note that the procedure of the consistent QED calculation can be always compared to a nonrelativistic quantum-

mechanical or quasi-classical calculation, but in the latter two cases, the QED prescriptions should be followed closely.[14] On the base of these principles, we analyzed the processes of light-induced ionization, radiative recombination, two-electron and triple recombination in plasma containing Ne- and Na-like ions.[13]

Initially, the secular matrix is constructed in the representation of the jj-coupling of angular momenta. Excited two-quasi-particle $2lnl$-states of a Ne-like ion in the jj-representation have the form

$$\sum_{ie,iv} C^{JM}_{ie,iv} a^{+}_{ie} a_{iv} \varphi_0, \tag{1}$$

where a^+ and a are the creation and annihilation operators of a quasi-particle, $ie = n_{ie} l_{ie} j_{ie} m_{ie}$ is the set of quantum numbers of one electron over the core, $iv = n_{iv} l_{iv} j_{iv} m_{iv}$ is the set of quantum numbers of one vacancy in the inner shells, J and M are the total angular momentum of a two-quasi-particle system and its projection, φ_0 is the state of the core, and the factor $C^{J,M}_{ie,iv}$ contains the Clebsh–Gordan coefficient and a phase factor to account for tensor properties of the annihilation operator a_{iv}.[18] In order to transfer to the intermediate coupling scheme, one should diagonalize the secular matrix. Since the imaginary parts of the matrix elements are much smaller than their real parts, it is sufficient only to diagonalize the real matrix in order to determine the vectors of eigenstates, i.e., the matrix $B^I_{ie,iv}$ of the transition to the intermediate coupling scheme.[18] The latter is used to transform the imaginary part of the secular matrix. As a result of this transformation, we obtain a matrix whose diagonal elements are level widths in the intermediate coupling scheme. They are sums over the complete set of states in the one-quasi-particle representation. Then this sum can be transformed to the sum over two-quasi-particle states in the intermediate coupling scheme. The entire transformation procedure is independent of the physical nature of the level width because it only uses the unitary property of the matrix relating the one-quasi-particle representation ie,iv to the two-quasi-particle representation I.[18]

The final expression for the level width is interpreted as a sum of the contributions from specific transitions between states in the intermediate coupling scheme. In the lower orders of perturbation theory, contributions of radiative, autoionization, and collisional decays can be separated.[17–19,13]

3. SUPERPOSITION OF CONFIGURATIONS AND SPECTRAL REPRESENTATION OF GREEN'S FUNCTION

Perturbation theory corrections to the imaginary and real parts of the energy matrix can be expressed in terms of the Green's function $G(\mathbf{r}_1, \mathbf{r}_2)$ of the Dirac equation with a bare (model) potential and a convolution of two, three, etc. Green's functions.[28] The spectral expansion includes discrete and continuous components. Commonly, a reduced spectral expansion of the Green's function over a complete set of one-quasi-particle states, which are solutions of the Dirac equation, is used. In what follows, we will call them real states. In this case the reduced expansion means that nearly degenerate states of the secular matrix are omitted in the Green's function expansion. In reality, only a few terms of the spectral expansion can be explicitly included. One way to refine the calculation technique is to take more terms of the spectral expansion. It is well known, however, that the spectral expansion over one-particle discrete real states does not converge to the desired function because there is always a contribution from the continuum. Direct calculations indicate[32] that the continuum contribution to the excitation energy of lower levels for almost all atomic systems is at least several thousands of reciprocal centimeters. Moreover, this assertion is universal for any basis including functions of the continuum, i.e., functions of a scattered electron with asymptotic forms oscillating as $r \to \infty$.

An alternative method of refining the calculation technique is to increase the matrix dimension by including additional states, i.e., by taking a superposition of configurations, the order of the perturbation theory for each matrix element remaining unchanged. The additional states should be excluded from the spectral representation of Green's function because these states are explicitly included in all orders of the perturbation theory, similarly to multiconfigurational versions of the self-consistent field technique. The convergence of this calculation procedure with respect to the number of states (configurations) explicitly included in the energy matrix is determined by the convergence of the Green's function expansion over real states. The convergence of the latter has been discussed previously. The central problem is that in both approaches, the complete functional space of atomic states is spanned by the one-quasi-particle basis of real states, which is not optimal for our purpose.

Contemporary atomic calculations widely use the technique of optimizing the real energy matrix in the first order of perturbation theory by introducing variational parameters either directly into the functions of real states or into the Hamiltonian generating these states. We have in mind various versions of the Dirac–Fock and Hartree–Fock multiconfigurational theories. This technique, of course, allows one to attain satisfactory accuracy in calculations of energy levels using rather limited sets of superposed configurations, the accuracy always being better than that deriving from the approximate nature of the theory on which the calculation is based. At least, the ignored contributions of Green's functions are usually larger than errors in the energy calculations. It is clear that this "superaccuracy" of the varied energies leads to errors in matrix elements of other operators calculated in the basis of the same real states and also to errors in the energy terms of higher orders of the perturbation theory.

There is another essential flaw inherent in many contemporary versions of multiconfigurational atomic programs. It is manifested in calculations of rates of spontaneous radiative transitions between groups of degenerate or nearly degenerate states. We discussed this problem in our earlier publication[14] taking as an example the 3–3 radiative transitions in Ne-like iron and argon. In traditional techniques, the calculation is performed in three stages: (1) construction of the transition-operator matrix in LS- or jj-representation; (2) transformation of this matrix to the intermediate momentum-coupling scheme; (3) multiplication of squared matrix elements of the transformed radiative transition ma-

trix by the energy factor E_{tr}^3 (in the case of electric dipole transition), where E_{tr} is the transition energy determined with the highest possible accuracy. Earlier[14] we demonstrated that this scheme contradicts the principles of QED theory. The scheme has two basic flaws: (1) inconsistency of the approximation for transition matrix elements with that for the transition energy; (2) an incorrect sequence of operations, namely, first the transformation to the intermediate coupling scheme, then multiplication by the energy multiplier. As a consequence, correlation effects are not correctly taken into account, and terms of different orders of perturbation theory in final results may not cancel out. Note that such a cancellation is an important feature of the atomic perturbation theory and demands that formal conditions of the theory should be followed closely.[28,16] In some cases, which are rather rare but apply to practically important systems, deviations from the formal conditions can lead to enormous errors. In this context, the case of two lasing transitions in Ne-like ions, $1s^22s^22p^53p[J=0]\rightarrow 1s^22s^22p^53s[J=1]$ and $1s^22s^22p^53p[J=2]\rightarrow 1s^22s^22p^53s[J=1]$ is very instructive. The anomalously large contribution of electron correlations to the higher state $2p3p[J=0]$ results in a high susceptibility of its parameters to the calculation procedure, namely the available Dirac–Fock calculations of the radiative 0–1 transition differ by a factor of several units from calculations obtained using the same basis, but the correct sequence of operations.[14]

It is well known that the complete basis of atomic functions can be expressed in terms of the set of Sturm orbitals, which is discrete and countable. This allows one to ignore the continuum in calculations in the framework of a formally exact theory.[20,21] In calculations of atomic parameters, this standardizes the procedure and, naturally, makes it easier. In the case of kinetic equations for level populations, the method allows one to bypass the problem of taking into account an infinite set of intermediate states, including those of the continuum.

A set of Sturm orbitals can be introduced with a predetermined asymptotic form, which is fundamentally important for the convergence of the spectral expansion. In principle, the transition to the Sturm basis resolves the convergence problem for the Green's function spectral expansion. It is clear that the optimum basis for an expansion of an arbitrary function in the form of a discrete set is that in which all the functions have the right asymptotic limit. In this context, the flaws of the real-state basis composed of an infinite set of discrete states and those of the continuum with undesired asymptotics are self-evident. Sometimes one can overcome this difficulty by replacing a sum over "Rydberg" states and adjacent continuum states with an integral of a quasiclassical continuous function approximating the spectral distribution of the desired function, which is convenient for some specific problems. In this paper, however, we are trying to formulate a universal approach to the kinetic problem of atomic transitions, which could be easily refined using a formally exact procedure.

In specific calculations, we will use a mixed basis composed of several functions of mixed states supplemented with Sturm functions. The functions of the mixed basis are not orthogonal to one another, which is taken into account in constructing the secular matrix. Two-quasi-particle states composed only of orbitals of real states are real two-quasi-particles states (states of a Ne-like ion in this work). States including a $2l$ vacancy and an electron Sturm function correspond to fictitious (virtual) states. Diagonalization of the secular matrix yields a set of orthonormal two-quasi-particle states. The mixing of real and virtual states effectively takes into account superposition of an infinite set of real states with one excited electron, including scattering states, i.e., those in which the ionization degree of the atom is a unit larger.

In this paper, we also effectively take into account superposition of doubly excited states (corresponding to excitation of two core electrons). To this end, the matrix element of the polarization interaction between two quasi-particles (an electron and a vacancy) via the polarizable core of the filled shells is calculated similar to Refs. 16 and 17. Polarization corrections are calculated for both real and imaginary parts of the secular matrix elements.

4. ONE-PARTICLE BASIS OF REAL STATES AND STURM EXTENSION

In what follows we will only discuss radial Green's functions and versions of mixed bases for specific calculations. All the one-quasi-particle orbitals are generated by solving the one-electron Dirac equation

$$[K(r)+V_n(r)+\delta_i V_{el}(r|\beta_i)-\varepsilon_i]\varphi_i=0, \qquad (2)$$

where K is the kinetic energy, $V_n(r)$ is the electron-nucleus interaction, $V_{el}(r|\beta)$ is the bare potential due to the core electrons (its construction was described in Refs. 16–18), and β_i is the parameter of the bare potential for the ith orbital. In order to generate the Sturm functions, we have introduced a second parameter δ_i, which equals unity for all functions of real states and is a quantization parameter for the additional Sturm functions. In this paper the two parameters β_i and δ_i correspond to each (ith) orbital of a real or Sturm state. In different stages of the calculation, the Dirac equation (2) is treated as an eigenvalue problem for the state φ_i and one of the three quantization parameters β_i, ε_i, or δ_i, the other two parameters being fixed.

The smallest basis discussed in this paper includes eight one-quasi-particle orbitals of real states. Three of them are states with vacancies in the core, $2s_{1/2}$, $2p_{1/2}$, and $2p_{3/2}$, and five have electrons in the outer shells, $3s_{1/2}$, $3p_{1/2}$, $3p_{3/2}$, $3d_{3/2}$, and $3d_{5/2}$. The exact (experimental) one-quasi-particle energies ε_{2lj} and ε_{3lj} are substituted into Eq. (2) to determine the respective parameters β_{2lj} and β_{3lj} of the bare potential, which are quantization parameters at this stage. We recall that $\delta_i=1$ holds for real-states orbitals. In what follows, this smallest basis is denoted $\{2;3\}$. In this simplest scheme, the secular matrix is calculated on the basis of 36 functions of two-quasi-particle states $2lj$; $3lj$ constructed from eight orbitals of real states of the $\{2;3\}$ basis. The basis $\{2;3,4\}$ includes five additional $4lj$ ($l=0,1,2$) one-quasi-particle real states derived from the Dirac equation (2) with "exact" (experimental) one-electron energies ε_{4lj} and $\delta_i=1$. Then by adding five $5lj$ orbitals of real states, we

obtain the one-quasi-particle {2;3,4,5} basis. Since the "exact" energies ε_{5lj} are not available, we take $\beta_{5lj}=\beta_{4lj}$. The respective energies ε_{5lj} are derived from the Dirac equation (2) by solving the eigenvalue problem for φ_{5lj} and ε_{5lj}. The secular matrix in the {2;3,4} and {2;3,4,5} bases contains 72 and 108 states, respectively. For each of the above bases of real-state orbitals, the Sturm extension is generated. In the case of the {2;3} basis the orbitals $\varphi_{S,nlj}$ ($n>3$) of the Sturm extension satisfy the Dirac equation (2), which is solved in this case as an eigenvalue problem for the parameter $\delta_{S,nlj}$ with given energy $\varepsilon_{S,nlj}=\varepsilon_{3lj}$ and the bare-potential parameter $\beta_{S,nlj}=\beta_{3lj}$, i.e., the energy and bare-potential parameter coincide with the respective parameters of real-state orbitals of the same symmetry. The spectrum of the Sturm extension may contain an infinite but countable set of solutions. The eigenvalue $\delta_{3lj}=1$ corresponds to the orbital φ_{3lj} of the real state, the other values $\delta<1$ correspond to the Sturm-extension orbitals. All the orbitals of the Sturm extension for Eq. (2) have an exponential asymptotic form at $r\to\infty$, which coincides with the asymptotic form of the last real state in the respective basis. The principal quantum number n of the extension function is uniquely related to the number of nodes in its radial part.

The Sturm extensions for the {2;3,4} and {2;3,4,5} bases are generated similarly, but with different pairs of fixed parameters ε and β: namely, for each orbital of the Sturm extension they coincide with the corresponding parameters of the real-state orbital with the largest principal quantum number n and the same angular symmetry. In this paper, we present calculations using the following bases: {2;3}, {2;3,*S*4}, {2;3,4,*S*5}, and {2;3,4,5,*S*6}. The latter three bases contain the Sturm orbitals $4lj$, $5lj$, or $6lj$ with $l=0,1,2$, i.e, each set of orbitals is only supplemented with one Sturm orbital of the same symmetry and generating 36 additional virtual states of a Ne-like ion. For simplicity f-orbitals are not included in the scheme because their contribution to the calculated parameters is negligible.

In each case, the functions of explicitly included states constitute a reduced spectral representation of the Green's function $G(\mathbf{r}_1,\mathbf{r}_2)$. The residual part drops proportionally to $\exp[-r\sqrt{(-2\varepsilon)}]$ as $r\to\infty$ ($r=r_1,r_2$ and ε is the eigenenergy of the last explicitly included real state). All the orbitals of the Sturm extension have the same asymptotic form. As was noted above, this is essential for the convergence of the technique. In specific calculations, we retained only one function of the Sturm extension in each basis. This is justified because, in addition to the asymptotic form, the residual part of the complete Green's function has the same number of nodes on both the r_1 and r_2 axes as the first function of the Sturm extension. The number of explicitly included functions of real states is determined, as usual, by studying the convergence of the calculated parameters using numerical techniques.

5. ENERGY APPROACH WITHIN THE QED THEORY TO CALCULATIONS OF RATES OF RADIATIVE TRANSITIONS BETWEEN EXCITED STATES OF A NE-LIKE ION

Radiative transition rates are contained in imaginary components of the secular matrix. They are determined by the delay factor of the interaction among the electrons. The relativistic radiative decay of the one- and two-quasi-particle states was previously studied by one of the present authors.[29,30] Later[17] this technique was applied to calculations of dipole, quadrupole, and octupole radiative transitions to ground states of Ne-like ions. As in the previous publication,[18] here we quote the contribution of the four-vertex $iklm$ Feynman diagram corresponding to the rate of radiative transition between excited states of a Ne-like ion:

$$\mathbf{P}(I\to F)=\sum_{ie,iv}\sum_{le,lv}\sum_{ke,kv}\sum_{me,mv}B^{I}_{ie,iv}B^{F}_{ke,kv}(2J_F+1)\times\langle ie,iv,J_I;ke,kv,J_F|\mathbf{A}|le,lv,J_{I'};me,mv,J_{F'}\rangle\times B^{F}_{me,mv}B^{I}_{le,lv}. \quad (3)$$

The indices i and l correspond to higher (J_I) states, and k and m to lower (J_F) states. In the case when the electron is an active particle (the vacancy in this case is a "spectator"),

$$\langle\mathbf{A}\rangle=\delta_{lv,mv}\delta_{iv,kv}\sqrt{(2j_{ie}+1)(2j_{ke}+1)(2j_{le}+1)(2j_{me}+1)}\times\begin{Bmatrix}A & J_F & J_I\\ j_{iv} & j_{ie} & j_{ke}\end{Bmatrix}\begin{Bmatrix}A & J_{F'} & J_{I'}\\ j_{lv} & j_{le} & j_{me}\end{Bmatrix}\times(-1)^{(j_{ie}+j_{le}+j_{ke}+j_{me}+j_{kv}+j_{mv})}Q_A(ieme;le\,ke). \quad (4)$$

If the active particle is a vacancy, then

$$\langle\mathbf{A}\rangle=\delta_{le,me}\delta_{ie,ke}\sqrt{(2j_{iv}+1)(2j_{kv}+1)(2j_{lv}+1)(2j_{mv}+1)}\times\begin{Bmatrix}A & J_F & J_I\\ j_{ie} & j_{iv} & j_{kv}\end{Bmatrix}\begin{Bmatrix}A & J_{F'} & J_{I'}\\ j_{le} & j_{lv} & j_{mv}\end{Bmatrix}\times(-1)^{(j_{ie}+j_{le})}Q_A(ivmv;lv\,kv), \quad (5)$$

where j_{ie} and j_{iv} is the one-particle total momentum of the electron and vacancy, respectively, J_i is the total momentum of the ith state of two-quasi-particle system, and A is the transition multiplicity. The expression for the integral $Q_A(1\ 2;4\ 3)$ is given in Refs. 18 and 19. Equations (3)–(5) were used in our previous publication[14] and in calculating the data for Table 4 of this paper (see below). The transition rate is expressed in Coulomb time units. The Coulomb time units are converted to seconds using the formula $t_s=0.827\cdot 10^{17}Zt_{\text{C.u.}}$, where Z is the nucleus charge.

The imaginary part of the radial integral $Q_A(\ldots)$ depends on the calibration of the photon propagator D in the matrix element of interelectron interaction. The family of propagators

$$D=D_T+CD_L,\quad D_T=\delta_{\mu\nu}/(k_0^2-k^2),\quad D_L=-k_\mu k_\nu/(k_0^2-k_2^2) \quad (6)$$

was studied numerically in Ref. 33 [the notations in Eq. (6) are the same as in Ref. 31]. D_T describes the interelectron interaction via transverse photon exchange, D_L describes the interaction via the exchange of longitudinal photons. At $C=0$ and $C=1$ the propagator D is appropriate to the Lorentz and Landau gauge, respectively. In the nonrelativistic limit different calibrations lead to different forms of the radiative-transition operator, namely, in the forms of length, velocity, and acceleration. The final calculations should be

TABLE I. Calculated positions of $2l3l$ levels with $J=0,1$ of Ne-like argon with respect to the ground state $1s^22s^22p^6$ in units of 100 cm^{-1}. The calculations in four bases are compared to data from Refs. 37 and 35 obtained by fitting calculations to measured energy levels. The states are numbered corresponding to their positions.

Number	State	J	Calculations of this work in four bases				Theory Ref. 18	Fitting Ref. 37
			{2;3}	{2;3,*S*4}	{2;3,4,*S*5}	{2;3,4,5,*S*6}		
					$2p3s$			
3	$2p_{3/2}3s_{1/2}$	1	20361	20324	20332	20336	20333	20331
4	$2p_{1/2}3s_{1/2}$	0	20454	20411	20421	20425	20444	20445
5	$2p_{1/2}3s_{1/2}$	1	20529	20499	20505	20508	20523	20518
					$2p3p$			
6	$2p_{3/2}3p_{3/2}$	1	21589	21464	21484	21491	21498	21493
9	$2p_{3/2}3p_{1/2}$	1	21833	21758	21767	21771	21763	21767
11	$2p_{1/2}3p_{1/2}$	1	21947	21874	21881	21885	21889	21892
12	$2p_{1/2}3p_{1/2}$	0	21991	21928	21935	21938	21916	21925
14	$2p_{1/2}3p_{3/2}$	1	22002	21934	21945	21949	21955	21961
15	$2p_{3/2}3p_{3/2}$	0	22705	22696	22697	22698	22754	22639
					$2p3d$			
16	$2p_{3/2}3d_{3/2}$	0	23631	23482	23496	23503	23486	23494
17	$2p_{3/2}3d_{3/2}$	1	23651	23501	23517	23524	23507	23513
23	$2p_{3/2}3d_{5/2}$	1	23917	23821	23828	23832	23815	23810
27	$2p_{1/2}3d_{3/2}$	1	24188	24169	24163	24162	24187	24110
					$2s3s$			
28	$2s_{1/2}3s_{1/2}$	1	26397	26342	26350	26355	26397	26419
29	$2s_{1/2}3s_{1/2}$	0	26673	26642	26645	26647	26688	26697
					$2s3p$			
30	$2s_{1/2}3p_{1/2}$	0	27853	27690	27799	27802	27802	27851
31	$2s_{1/2}3p_{1/2}$	1	27859	27660	27805	27808	27810	27859
33	$2s_{1/2}3p_{3/2}$	1	27958	27901	27904	27907	27927	27974
					$2s3d$			
34	$2s_{1/2}3d_{3/2}$	1	29779	29668	29667	29673	29674	29723

gauge invariant, but in lower orders of perturbation theory, there is, of course, a component of imaginary energy which is not guage invariant. The study described in Ref. 33 was dedicated to constructing a bare potential generating a one-electron basis to minimize the gauge noninvariant component in the lowest, second order of perturbation theory (fourth order of the QED perturbation theory). The rates of strong (dipole) transitions for the gauges mentioned above differ by less than 0.16%. The representation constructed in Ref. 33 has been used in the present work.

6. DISCUSSION

Table I lists energies of excited $2l3l$ states of a Ne-like argon ion. For brevity only the states with $J=0,1$ are given (some of these states are interesting from the standpoint of applications). Table I demonstrates good convergence of the numerical results as the basis of the one-quasi-particle orbitals is enhanced. We have also calculated energies using the {2;3,4,*S*5,6} basis, which includes the {2;3,4} basis of real states. Each function of this basis is supplemented with two Sturm orbitals of the same symmetry with $n=5,6$. The calculations using this basis are practically identical to those with the {2;3,4,5,*S*6} basis. Note that the inclusion of f orbitals leads to small shifts of the states $2l3l$, i.e., these shifts are smaller than the difference between calculations using the two longest bases, {2;3,4,*S*5} and {2;3,4,5,*S*6}. This leads us to a conclusion that all singly excited states $2lnl$ are fairly adequately accounted for in the calculation using the latter "best" basis. At least, the superposition of these states is not the main source of uncertainties in the calculation. The energy of the $2l3l$ level calculated using the {2;3,4} basis usually ranges between the calculations using the {2;3} and {2;3,*S*4} bases (closer to the former figure). This indicates that the level energy calculated using a mixed basis of real states and additional Sturm orbitals converges faster than in the case of a discrete basis of only real states.

Note that the energy gap between the fifteenth level $2p3p[J=0]$ and lower levels of this configuration is anomalously large. This can be accounted for in terms of large correlation effects in this state. Other manifestations of this effect will be discussed below.

It is noteworthy that the feature distinguishing this calculation from the previous ones[16–18,34] is the absence of any variational procedure. The variational procedure was used previously[16–18,34] to determine the screening parameter of the electron-vacancy interaction, which was introduced to the bare potential in order to improve the efficiency of inclusion of higher-order perturbation corrections. The approach employed in this work is more consistent. On an average, the "variational" calculation of Ref. 16 is more accurate than the best version of this work, namely the calculation in the {2;3,4,5,*S*6} basis, except the two usually noted anomalies: level 15 of the state $2p3p[J=0]$ and level 27 of the state $2p3d[J=1]$. It is well known that these levels cause additional difficulties in all theoretical approaches. In the approximation used in this work, the calculation error of these two levels is significantly smaller, whereas these levels are quite important for applications. We also assume that the

TABLE II. Calculated positions of the $2l4l$ levels with $J=0,1$ of Ne-like argon with respect to the ground state $1s^22s^22p^6$ in units of 100 cm^{-1}. The calculations in three bases are compared to data from Ref. 37 obtained by fitting calculations to experimental data. The states are numbered according to their positions.

Number	State	J	Calculations of this work in three bases {2;3,4}	{2;3,4,*S*5}	{2;3,4,5,*S*6}	Fitting Ref. 37
				$2p4s$		
39	$2p_{3/2}4s_{1/2}$	1	27169	27109	27117	27040
40	$2p_{1/2}4s_{1/2}$	0	27319	27249	27258	27186
41	$2p_{1/2}4s_{1/2}$	1	27334	27271	27278	27208
				$2p4p$		
42	$2p_{3/2}4p_{1/2}$	1	27682	27588	27600	27552
45	$2p_{3/2}4p_{1/2}$	1	27724	27646	27654	27610
47	$2p_{3/2}4p_{3/2}$	0	27807	27743	27750	27729
48	$2p_{1/2}4p_{1/2}$	1	27881	27791	27800	27769
50	$2p_{1/2}4p_{3/2}$	1	27886	27808	27819	27794
51	$2p_{1/2}4p_{1/2}$	0	28033	28012	28012	28029
				$2p4d$		
52	$2p_{3/2}4d_{3/2}$	0	28398	28255	28269	28220
53	$2p_{3/2}4d_{3/2}$	1	28403	28264	28280	28231
59	$2p_{3/2}4d_{5/2}$	1	28492	28400	28409	28361
63	$2p_{1/2}4d_{3/2}$	1	28665	28599	28600	28552

wave functions derived by the present method are more accurate for calculations of other parameters since the energies have been calculated without using any fitting parameters and variational procedures. The uncertainty in the level energies is mainly due to the approximation used to account for the corrections due to the polarization interaction between quasiparticles—an electron and a vacancy. A detailed description of how the polarization interaction operator is constructed and its calculation were given in Ref. 16.

One may come to similar qualitative conclusions considering the group of $2l4l$ levels with $J=0,1$ listed in Table II. The states $2p4l$ are strongly mixed with $2s3l$ states, thus both groups of these states should be calculated together and the same theoretical accuracy in both groups should be provided.

Table III lists the rates of most strong (resonant) radiative transitions to the ground state. The calculations have been performed using three bases. One can see fast convergence of the energies of these transition with respect to the basis dimension; the convergence of other transition probabilities is also fairly fast. Besides, the results are compared to our previous calculations,[14] which were performed using a variational parameter. The last column of this table is given to compare our results to calculations of Ref. 35, which used the traditional approach to the calculation of transition rates, i.e., squared "exact" matrix elements of the nonrelativistic transition operators in the intermediate scheme were multiplied by "exact" transition energies E_{tr}^3. The electronic basis in Ref. 35 included all Slater orbitals (including the $5f$-orbital), each orbital being optimized separately, i.e., the energy matrix was fitted to an experimentally measured spectrum. Thereafter oscillator strengths were calculated.

The convergence of our technique applied to the rates of transitions with $\Delta n=0$ is illustrated by Table 4 taking as an example the four transitions that attract considerable attention of researchers of lasing effects in plasma with Ne-like ions. As was mentioned above, there is a considerable discrepancy between the various approaches to these transi-

TABLE III. Rates of spontaneous 1–0 resonant radiative transitions to the ground state measured in s^{-1}. Numbering corresponds to that of Tables I and II.

Number	Calculations of this work in different bases {2;3,4}	{2;3,4,*S*5}	{2;3,4,5,*S*6}	Theory Ref. 14	Theory Ref. 35
3	6.63+10	6.66+10	6.68+10	5.06+10	6.69+10
5	1.49+11	1.61+11	1.53+11	1.38+11	1.62+11
17	2.10+10	1.32+10	1.40+10	5.85+09	6.63+09
23	6.49+10	7.20+10	7.27+10	1.44+11	1.87+11
27	1.97+12	2.07+12	2.05+12	3.17+12	2.37+12
31	3.20+08	5.71+09	6.09+09	8.21+09	8.48+09
33	5.40+11	6.74+11	6.44+11	4.43+11	4.85+11
39	3.75+09	5.35+09	3.93+09		
41	1.12+10	2.04+10	1.62+10		
53	5.99+09	4.89+09	3.79+09		
59	1.71+11	1.32+11	1.33+11		
63	4.36+11	8.02+11	7.41+11		

TABLE IV. Rates of four radiative transitions: $2p3p[J=0]-2p3s[J=1]$ in units of s^{-1}. Numbering corresponds to that of Table I.

Transition	Calculations of this work in different bases				Theory Ref. 14	Theory Ref. 38	Theory Ref. 35
	{2;3}	{2;3,S4}	{2;3,4,S5}	{2;3,4,5,S6}			
12–3	2.3+09	1.1+09	9.6+08	1.0+09	1.7+09	2.5+09	2.5+09
12–5	5.5+08	1.5+08	1.3+08	1.6+08	3.9+08	4.4+08	4.4+08
15–3	3.8+08	5.0+08	4.8+08	4.6+08	4.6+08	3.3+09	2.4+09
15–5	2.5+09	2.9+09	2.7+09	2.7+09	1.7+09	9.5+09	6.8+09

tions because the correlation effects on level 15 ($2_{3/2}3p_{3/2}[J=0]$) are anomalously strong and this state is strongly admixed to that of level 12 ($2p_{1/2}3p_{1/2}[J=0]$). The fundamental difference between the consistent QED approach and the widely used traditional procedure based on the multiconfigurational method was discussed in detail in Ref. 14, where we demonstrated how the violation of QED principles in traditional schemes leads to enormous errors in calculations of radiative and collisional transition rates.

The main result of this work is the fast convergence of the calculations with respect to the dimension of the basis composed of real states and additional Sturm orbitals. It is also important that, although our previous calculation using the {2;3} basis[14] and variational parameter yielded more accurate values of energies, the accuracy of transition rates obtained by that method is not equally good.

Sampson *et al.*[36] analyzed six versions of the multiconfigurational approach taking as an example the resonant 2–3 transitions in Ne-like ions. The comparison demonstrated that the calculated oscillator strengths of some transitions differ by a factor of three to four, although the approximations used in these six versions were quite similar. This large disagreement among the calculations was ascribed to the difference in how the correlation effects were included in the different versions of the multiconfigurational techniques. We noted in Ref. 14 that there are other factors leading to disagreement among transition rates calculated in different approximations. They include, for example, the relativistic "contraction" of orbitals. The result may also be affected by the form of the transition operator, by whether it is relativistic or nonrelativistic, whether its polarization component has been taken into account, etc. Possible uncertainties due to the gauge of the photon propagator were also analyzed in Ref. 33. In this connection, the issue of testing the calculations experimentally is quite urgent.

7. CONCLUSIONS

We have analyzed the possibilities of contemporary techniques in calculating fundamental spectroscopic constants which determine emission spectra of ions in plasma. Our numerical results clearly indicate the importance of the convergence of calculations with respect to the number of orbitals included in the basis, as well as the reproducibility of the calculations performed in different approximations. This statement is more important when theoretical transition rates are employed to predict gains on lasing lines in plasma (Table IV). The reported study allows one to select an adequate basis for calculating populations of levels of a Ne-like ion. By comparing our calculations of radiative transition rates to the respective data by Hibbert *et al.*[35], we have discovered the cause of disagreement among these constants calculated using different approaches.[14] Our analysis indicates that we can, in principle, mimic various multiconfigurational calculations using the same theoretical scheme. The inevitable question is which data are more reliable. The traditional technique yielded fairly accurate results in some cases. A situation may occur, however, when the oversimplifications and inconsistency of traditional techniques lead to enormous errors in decay rates of atomic states. Therefore the topical problem is testing calculations of fundamental constants. For example, spectra of Ne-like ions measured at the EBIT (electron-beam ion trap) facility at an electron density of 10^{12}–10^{15} cm^{-3} can be used in testing calculations of radiative transition rates in this ion. At all electron densities the collisional mixing of states can be ignored, and the kinetics is largely controlled by collisional excitations from the ground state. The efficiency of electron-ion collisions from the ground state to an excited state are approximately equal for strong transitions in all approximations.[18] Unlike other plasma sources, the energy and density of electrons in the EBIT facility are well known. Kinetic equations should take into account all important elementary processes in a system with an infinite number of levels.[13] Thus, given spectral measurements of transitions with $\Delta n=0$ and an approximately adequate kinetic model, one can test calculations of radiative transition rates in Ne-like ions. No less important information for testing the theory can be derived from spectra of resonant 2–3 transitions and their satellites.

Similar analysis is necessary in studies of collisional widths of levels. These parameters are of primary importance in calculations of laser gain. Preliminary estimates[14] indicate that data by different authors on ratios of collisional widths of levels in Ne-like ions contradict one another.

[a]Deceased.

[1] A. V. Vinogradov, I. I. Sobel'man, and E. A. Yukov, Kvant. Élektron. **2**, 105 (1975) [Sov. J. Quantum Electron. **5**, 59 (1975)].
[2] J. A. Koch, B. J. MacGowan, L. B. Da Silva, D. L. Matthews *et al.*, Phys. Rev. A **50**, 1877 (1994).
[3] R. C. Elton, *X-Ray Lasers*, Academic Press, San Diego (1990).
[4] S. Glenzer and H.-J. Kunze, Phys. Rev. E **49**, 1586 (1994).
[5] H.-J. Shin, D.-E. Kim, and T.-N. Lee, Phys. Rev. E **50**, 1376 (1994).
[6] J. J. Rocca, F. G. Tomasel, M. C. Marconi *et al.*, Phys. Plasmas **6**, 2547 (1995).
[7] E. P. Ivanova, L. N. Ivanov, L. V. Knight, and B. G. Peterson, *Proceedings of the International Conference on Laser 93*, Lake Tahoe, NV, December 1993, ed. by V. J. Corcoran, T. A. Goldman, STS Press McLean, VA (1994), p. 704.

[8] J. P. Apruzese and J. Davis, Phys. Rev. A **33**, 3686 (1983).
[9] K. J. Whitney, A. Dasgupta, and P. E. Pulsifer, Phys. Rev. E **50**, 468 (1994).
[10] E. P. Ivanova, L. V. Knight, and B. G. Peterson, in *X-ray Lasers 1994*, ed. by D. C. Eder and D. L. Matthews, AIP Conference Proceedings 332, AIP PRESS, New York (1995), p. 393.
[11] E. P. Ivanova, L. V. Knight, and A. Reyes-Mena, *Proceedings of the International Conference on Laser 94*, December 1994, Quebec, Canada, STS Press * Mc Lean, VA (1995), p. 123.
[12] J. Abdallah, Jr., R. E. Clark, and J. M. Peek, Phys. Rev. A **45**, 3980 (1992).
[13] L. N. Ivanov, E. P. Ivanova, L. V. Knight, and A. G. Molchanov, Physica Scripta **53**, 653 (1996).
[14] L. N. Ivanov, E. P. Ivanova, and L. V. Knight, Phys. Lett. A **206**, 89 (1995).
[15] L. N. Ivanov and L. V. Knight, in *Proceedings of the International Conference on Lasers 94*, December 1994, Quebec, Canada, STS PRESS * McLean (1995), p. 127.
[16] E. P. Ivanova, L. N. Ivanov, A. V. Glushkov, and A. E. Kramida, Phys. Scripta **32**, 513 (1985).
[17] E. P. Ivanova and A. V. Gulov, Atom. Data Nucl. Data Tables **49**, 1 (1991).
[18] L. N. Ivanov, E. P. Ivanova, and L. V. Knight, Phys. Rev. A **48**, 4365 (1993).
[19] L. N. Ivanov, E. P. Ivanova, and E. V. Aglitskii, Phys. Reports **164**, 315 (1988).
[20] V. A. Fok, *Principles of Quantum Mechanics* [in Russian], Nauka, Moscow (1976), p. 194 [Transl. Mir., Moscow (1978)].
[21] M. G. Veselov and L. N. Labzovskii, *Theory of Atomic Structure of Electron Shells* [in Russian], Nauka, Moscow (1986), p. 29.
[22] C. V. Khristenko and S. I. Vetchinkin, Opt. Spektrosk. **31**, 503 (1971) [Opt. Spectrosc. **31**, 269 (1971)].
[23] A. I. Sherstyuk, Zh. Éksp. Teor. Fiz. **62**, 1238 (1972) [Sov. Phys. JETP **35**, 655 (1972)].
[24] A. I. Sherstyuk and P. F. Gruzdev, Opt. Spektrosk. **42**, 1198 (1977) [Opt. Spectrosc. **42**, 690 (1977)].
[25] P. F. Gruzdev, G. S. Solov'eva, and A. I. Sherstyuk, Izvestiya Akad. Nauk SSSR (Ser. Fiz.) **50**, 1356 (1986).
[26] A. P. Martinenko and R. N. Faustov, Teor. Mat. Fiz. **66**, 399 (1986).
[27] M. A. Brown, Teor. Mat. Fiz. **39**, 388 (1984).
[28] V. V. Tolmachev, Adv. Chem. Phys. **14**, 421, 471 (1969).
[29] L. N. Ivanov, M. N. Driker, Opt. Spektrosk. **49**, 113 (1980) [Opt. Spectrosc. **49**, 113 (1980)].
[30] E. P. Vidolova-Angelova and L. N. Ivanov, J. Phys. D: At. Mol. Opt. Phys. **24**, 4147 (1991).
[31] A. I. Akhiezer and V. B. Berestetskiĭ, *Quantum Electrodynamics* [in Russian], Nauka, Moscow (1969) [Transl. of earlier edition, Wiley, New York].
[32] E. P. Ivanova and U. I. Safronova, J. Phys. B: At. Mol. Phys. **8**, 1591 (1975).
[33] L. N. Ivanov and A. V. Glushkov, Physics Lett. A **170**, 33 (1992).
[34] A. L. Gogava and E. P. Ivanova, Opt. Spektrosk. **66**, 980 (1989) [Opt. Spectrosc. **66**, 572 (1989)].
[35] A. Hibbert, M. Le Dourneuf, and M. Mohan, At. Data and Nucl. Data Tables **53**, 23 (1993).
[36] D. H. Sampson, H. L. Zhang, A. K. Mohanty, and R. E. H. Clark, Phys. Rev. A **40**, 604 (1989).
[37] B. C. Fawcett, Physica Scripta **30**, 326 (1984).
[38] A. K. Bhatia, U. Feldman, and J. F. Seely, At. Data and Nucl. Data Tables **32**, 435 (1985).

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