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Expansion of the Irregular Solution in the Theory of Stark Effect in Hydrogenic-Like Rydberg Atoms

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Abstract—We derive the expansion of the irregular physical solution over the spherical solutions at negative energies, which is necessary for obtaining the $S$ matrix of the process. The relation of this expansion to the theory developed by Giannakees et al., Phys. Rev. A 94, 013419 (2016), is analyzed. In particular, we show that the expansion of the irregular solution missing in Giannakees et al. theory can be derived from one of their main postulates. The expansion thus obtained turns out to be numerically equivalent to our expansion up to high angular momenta. Analytical expressions for the key matrix of both expansions are derived.

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1. INTRODUCTION

In order to quantitatively describe the Stark photoabsorption spectra of alkali-metal atoms, Harmin [1] used the local frame transformation (LFT) approach by Fano [2]. In frame of this approach, the “physical” solutions in parabolic coordinates, i.e. the ones that are bounded at infinity, are to be matched near the core with the spherical solutions that satisfy the boundary condition defined in the quantum-defect theory [3, 4]. According to this theory, the solutions outside the core have the form of a specific linear combination of the regular and irregular spherical Coulomb functions. In the presence of the electric field, this remains valid up to the distances $r$ where mixing of the states with different angular momenta $l$ by the field can be neglected. At larger distances, the parabolic Stark solutions are to be used. Therefore, in order to obtain the solution valid within the full range of radial coordinate $r$ outside the core, one has to match the physical parabolic solution to the spherical solutions with definite values of $l$. Such matching is possible only locally, within the range of intermediate distances, where both the effect of core structure and the influence of the external field are negligible in comparison with the Coulomb attraction to the core. The matching procedure is based on the mutual expansions between the regular and irregular parabolic and spherical solutions. These expansions form the base for calculation of the observable quantities. Harmin’s theory was successfully applied to calculations of the photoionization cross section of sodium atoms and to interpret the photoionization experiments. However, its application to calculations of the differential cross section in experiments on ionization microscopy and comparison with the results of highly accurate experiments (see e.g. [5]) turned out to be unsatisfactory. Moreover, it was found that the LFT of the irregular wave function defined in Harmin’s theory does not obey some necessary requirements [5–8].

Giannakees et al. have recently developed a generalized LFT (GLFT) [6] that employs the formal use of a single-particle potential and the operator algebra. The GLFT approach avoids the explicit use of the LFT of the irregular solution, which is nevertheless implicitly present in this theory and can be derived from one of its key postulates. In this paper, we first present the LFT of the irregular solution obtained by our approach and derive an explicit analytical expression for the LFT key matrix (Section 2). Second, we derive the LFT-transformation matrix for the irregular solution from Giannakees et al. GLFT theory and compare it with our respective matrix (Section 3). We found that the two matrices give numerically equivalent expansions of the irregular solution.
2. EXPANSION OF THE IRREGULAR SOLUTION

In the limit of small external field, $F \ll 1$ (atomic units are used), one can specify the core range of distances, $r \leq 1$, and an intermediate, Coulomb range, $1 \ll r \ll F^{-1/2}$, where the potential is Coulombic, $-1/r$, and the external field is weak, $F \ll 1/r$. Yet, mixing of the states with different angular momenta $l$ by the external field can take place within the Coulomb region at distances where the field energy is larger than the centrifugal energy difference between neighboring states $l$ and $l-1$, $Fr \gtrsim l/r^2$. Hence, the mixing can be neglected only within the “near-Coulomb” region next to the core, $1 \ll r \ll F^{-1/2}$, which is much narrower than the Coulomb one. At such small $r$, i.e. in the near-Coulomb region, the spherical and parabolic solutions coexist so that the LFT between them can be performed, whereas only the latter exists at large $r$, i.e. outside it. Because of the $l$-mixing within the Coulomb region, the matched spherical functions must unavoidably involve a linear combination of states with different momenta. The resulting matching equation is

$$P_l^{(1)}(\cos \theta) G_1(r) + \sum_{l'=m} \gamma_{l,l'} P_{l'}^{(1)}(\cos \theta) F_{l'}(r) = \sum_{k=1} \gamma_{l,k} \psi_k(\xi, \eta),$$

where $P_l^{(1)}(\cos \theta)$ are the Legendre polynomials, $F_{l'}(r)$ and $G_1(r)$ are the regular and irregular (at $r = 0$) solutions of the radial Schrödinger equation in the pure Coulomb potential, and $\psi_k(\xi, \eta)$ are the physical, i.e. not increasing at infinity, irregular parabolic solutions in the pure Stark potential. The coefficients $\gamma_{l,l'}$ and $\gamma_{l,k}$ are uniquely defined by the matching conditions. The right-hand side of Eq. (1) represents the irregular physical solution at large $r$, i.e. everywhere outside the core region. Inside the near-Coulomb region, where the spherical solutions are simultaneously exist, it can be locally transformed to a linear combination of the spherical functions. The solutions in the parabolic and spherical frames must approximately coincide locally, i.e. within the near-Coulomb region, as is expressed by Eq. (1). The details of the derivation are given elsewhere [9].

In [9], matrix $\gamma_{l,l'}$ entering Eq. (1) could be calculated only numerically. In this section, we derive an analytical expression for this matrix. The radial spherical functions in the left-hand side of Eq. (1) are defined in [9] as

$$F_{l}(r) = \left[ \frac{2}{n} \right]^{l} e^{-r/n} \Phi(-n + l + 1, 2l + 2, 2r/n),$$

and

$$G_{l}(r) = \left[ \frac{2}{n} \right]^{l} e^{-r/n} \Psi(-n + l + 1, 2l + 2, 2r/n),$$

where $n = 1/\sqrt{2}E$, $E$ is the energy, $\Phi(a, b, x)$ and $\Psi(a, b, x)$ are Kummer functions $M(a, b, x)$ and $U(a, b, x)$, respectively [10]; functions $F_{l}(r)$ and $G_{l}(r)$ are not normalized. We consider the case of $n \gg 1$, which corresponds to highly excited Rydberg states.

In the right-hand side of Eq. (1), functions $\psi_{k}(\xi, \eta)$ are the irregular parabolic solutions of the pure Stark problem,

$$\psi_{k}(\xi, \eta) = \chi_{\psi_k}(\xi) \psi_{\mu_k}(\eta).$$

Here, $\chi_{\psi_k}(\xi)$ are the normalized-to-unity solutions of the eigenvalue problem for the finite motion along the parabolic coordinate $\xi$ and $\psi_{\mu_k}(\eta)$ are the irregular parabolic solutions of the Stark equation for the infinite motion along $\eta$. The quantum numbers $\nu_k = n \beta_k - (m + 1)/2$ and $\mu_k = n - v_k - m - 1$ are non-integers. They correspond to the discrete set of eigenvalues $\beta_k$ of the separation constant $\beta$ (partial charge) of the separable Schrödinger equation in the parabolic coordinates. At small $r$, i.e. in the near-Coulomb region, these functions approximately coincide with the Coulomb parabolic solutions,

$$\chi_{\psi_k}(\xi) \approx c_k f_{\nu_k}(\xi), \quad \psi_{\mu_k}(\eta) \approx g_{\mu_k}(\eta),$$

with $c_k$ being the normalization constants. The regular and irregular Coulomb parabolic functions are defined as

$$f_{\nu}(\xi) = \left[ \frac{\sqrt{\lambda}}{\eta} \right]^{m/2} e^{\epsilon \nu^2 \mu} \Phi(-\nu, m + 1, \xi/\eta),$$

where $\xi = \xi$ and $\nu = \eta$ (or, as required below in Eq. (10), $\xi = \eta$ and $\nu = \mu$) and

$$g_{\mu}(\eta) = \left[ \frac{n^{m/2}}{\eta} \right] e^{-\eta^2/\mu^2} \Psi(-\mu, m + 1, \eta/\mu),$$

respectively. The irregular spherical and parabolic functions (3) and (7) are chosen from the condition that the solutions are bounded at infinity.

Coefficients $\gamma_{l,l'}$ and $\gamma_{l,k}$ are uniquely defined by the matching of the physical, i.e. not increasing at infinity parabolic solution with the spherical solutions in the near-Coulomb region and by the choice of the functions in the form of Eqs. (3) and (7). The transformation matrix $\gamma_{l,k}$ has the form [9]

$$\gamma_{l,k} = \frac{W_{l}}{m! N_m} A_{l,m} c_k \Gamma(-\mu_k),$$

where
is the LFT matrix for the regular solutions. The regular solutions are given by the product of two functions $f_n(\xi)$ defined in Eq. (6), and the LFT for them is given by (see [1] and Appendix A in [9])

$$f_n(\xi) = \sum_{l=0}^{\infty} A_{nl} P_l^m(\cos \theta) F_l(r).$$

In Eq. (8), $W_l$ and $N_{lm}$ are the Wronskian and the normalization constants for the spherical Coulomb functions,

$$W_l = \frac{n(2l + 1)!}{2^{2l+1} l! (l - n)!}, \quad N_{lm} = \frac{2l + 1 (l - m)!}{2 (l + m)!}.$$  

Our method to derive Eq. (1) is based on the exact expansion of the irregular spherical functions over the irregular parabolic Coulomb solutions,

$$\gamma_l(r, \theta) = P_l^m(\cos \theta) G_l(r)$$

$$= \sum_{n=0}^{\infty} B_{nl} f_n(\xi) g_n(n),$$

where the transformation matrix $B_{nl}$ is given by [9]

$$B_{nl} = \frac{W_l}{m! N_{lm}} A_{nl} N_{ll}^2 \Gamma(-n_2)$$

and $n_2 = n - n_1 - m - 1$. Parameter $N_{nl}$ is the normalization constant for the regular parabolic function $f_n(\xi)$,

$$N_{nl} = \frac{1}{m! n!} \Gamma(n + 1).$$

Expansion (12) was derived in [9] for arbitrary integer and non-integer $n$ and for a special, unique choice of the radial spherical function $G_l(r)$, Eq. (3), such that it exponentially decreased at infinity. Owing to this choice, the spherical function on the left of Eq. (12) at any fixed $\mu = m$ can be expanded over the quantized basis of $f_n(\xi)$. The coefficients of this expansion are proportional to the bounded at infinity parabolic solution (7) with $\mu = n_2$. Note that any irregular radial function other than $G_l(r)$ increases exponentially and the corresponding spherical function cannot be expanded over the parabolic solutions.

The physical irregular Stark wavefunction, which is given by the sum in the right-hand side of Eq. (1) with coefficients $Y_{l,k}$ determined by Eq. (8), at any finite $r$ converges to a regular function of $\theta$, while the singular (at $r = 0$) part of this function coincides with the singular part of $\gamma_l(r, \theta)$ (see [9]). Therefore, the difference between these two functions is a regular function of $r$ and $\theta$, which can be expanded in a series over the regular spherical functions,

$$\Psi_{l, reg}(\xi, \eta) = \sum_{k=1}^{\infty} Y_{l,k} X_{\nu_k}(\xi) \psi_{\mu_k}(\eta) - \gamma_l(r, \theta)$$

$$= \sum_{l'=m}^{\infty} \gamma_{l',l} P_{l'}^m(\cos \theta) F_{l'}(r)$$

which leads to Eq. (1).

In [9], matrix $\gamma_{l,l'}$ was found numerically. Here, we will find its analytical representation. To begin with, we note that the main difficulty in calculating the matrix elements $\gamma_{l,l'}$ is related to the nonuniform convergence of the sum on the left-hand side of Eq. (15), see [9]. The term by term projection of this sum onto the Legendre polynomials is impossible because it leads to a divergent series. The truncated sum has a singularity at $\eta = 0$ and the contribution of this singularity to the integral does not disappear when the truncation limit tends to infinity.

To find matrix $\gamma_{l,l'}$, we transform function $\Psi_{l, reg}$ using Eqs. (8), (12), and (13),

$$\Psi_{l, reg}(\xi, \eta) = \sum_{k=1}^{\infty} Y_{l,k} X_{\nu_k}(\xi) \psi_{\mu_k}(\eta) - \sum_{n=0}^{\infty} B_{nl} f_n(\xi) g_n(n)$$

$$= \sum_{k=1}^{\infty} \gamma_{l',l} P_{l',m}^m(\cos \theta) F_{l'}(r) + \sum_{n=0}^{\infty} A_{nl} c_{n}^2 \Gamma(-n_2) f_n(\xi) g_n(n).$$

Both sums in the right-hand side of Eq. (16) converge non-uniformly at $\eta = 0$, and when the truncation limits are chosen arbitrarily, the difference between these sums turns out to be a singular function that cannot be expanded over the spherical harmonics. However, this singularity can be eliminated if specially selected cutoff functions are introduced into the sums.

At asymptotically large values of $k$ and $n_1$ coefficients $A_{l, kj}$ and $A_{l, n_1, j}$ are smooth functions of indices $A_{l, kj} = (-1)^j \sqrt{j^{-m}} \frac{2^j (m!)^j}{(l!)^2 (l+m)!}$.

Product $\Gamma(-\mu) g_{l, n_1}(\eta)$ at large negative $\mu$ is also a smooth function of $\mu$. Due to a smooth dependence of the terms in the sums, summations for large values of indices can be replaced by integrations,
\[ \sum_{n=0}^{\infty} F_{\text{coul}}(n_{1}) A_{n_{1}j,l} N_{l}^{2} (-\mu_{k}) f_{n_{1}}(\xi_{k}) g_{n_{1}}(\eta) \]
\[ = \int F_{\text{coul}}(n_{1}) A_{n_{1}j,l} N_{l}^{2} (-\mu_{k}) f_{n_{1}}(\xi_{k}) g_{n_{1}}(\eta) dn_{1}, \quad (18) \]

and
\[ \sum_{k=0}^{\infty} F_{\text{st}}(v_{k}) A_{v_{k}j,l} N_{l}^{2} (-\mu_{k}) f_{v_{k}}(\xi_{k}) g_{v_{k}}(\eta) \]
\[ = \int F_{\text{st}}(v) A_{v_{k}j,l} N_{l}^{2} (-\mu_{k}) f_{v_{k}}(\xi_{k}) g_{v_{k}}(\eta) \frac{dk}{dv}. \quad (19) \]

Here \( F_{\text{st}}(v_{k}) \) and \( F_{\text{coul}}(n_{1}) \) are the cutoff functions for the Stark and Coulomb sums. At large values of \( k \) and \( n_{1} \) the derivative \( dk/dv \) is calculated as
\[ \frac{dk}{dv} = \frac{N_{l}^{2}}{c_{k}^{2}}, \quad (20) \]

and
\[ \Psi(\mu, s) = \psi(-\mu + s) - \psi(1 + m + s) - \psi(1 + s) \quad (23) \]
(\( \psi \) is the digamma function).

Note that convergence of the sums in Eq. (21) is provided by the cutoff functions, which have the same functional form in both sums. Note also that the true matching of the Stark wave function to the spherical Coulomb solutions is realized only in the asymptotic limit of \( n \rightarrow \infty \). At large yet finite \( n \), the exact cancelation of residuals of sums in Eq. (16) is impossible. This circumstance reflects the approximate nature of the matching, which fully neglects the external field in the near-Coulomb region. The accuracy of matching is also influenced by an important physical parameter
\[ \delta = 16 F_{n}^{4}, \quad (24) \]
which determines the height of the potential barrier for ionization: \( \delta = 1 \) corresponds to the classical ionization threshold. In practice, when \( \delta \) is on the order of unity, a high accuracy of the matching can be achieved already for \( n > 10 \) by a reasonable choice of the cutoff function.

The new features of our LFT expansion, Eq. (1), as compared to Harmin’s LFT expansion [1] are that the matching of the spherical and parabolic solutions is performed within the near-Coulomb region, which is narrower than the full Coulomb one, and that the sum over the regular spherical solutions in the Coulomb potential is added to the left-hand side of the matching Eq. (1). Outside the near-Coulomb region, strong \( l \)-mixing takes place, therefore the physical solution extended to the region of spherical symmetry must involve the states with various \( l \). This drastically differs from Harmin’s matching equation where the physical solution is matched to a spherical function with a definite \( l \) within the full Coulomb region.

The advantage of our expansion over the respective Harmin’s expansion was demonstrated numerically for \( l = 1, 3, 5 \), \( r = 10–80 \), and \(-1 < \cos \theta < 1\) by comparison with the exact solution [9].

Using the expansion of the irregular solution (1) and a similar expansion of the regular solution [1, 6, 9], we derived the \( S \) matrix [9] suitable for calculating the observed quantities such as photoionization cross-sections. A different GLFT approach developed by Giannakes et al. [6] avoids using the irregular solution. Nevertheless, the explicit expansion of the irregular solution can be derived from the basic relations of the GLFT theory, which can be compared with our result. This is performed in the next section.

\section{3. COMPARISON WITH THE GLFT THEORY}

In this section, notations of [6] are used, with prefix “G” added to the equation numbers. One of the basic strong statements of the GLFT theory is the
equivalence of two Green functions at small distances (see Eqs. (G18)–(G21) and subsequent text in [6]),
\[ G_{\text{smooth}}^{C-S}(\mathbf{r}, \mathbf{r}') = G_{\text{smooth}}^{C-S}(\mathbf{r}, \mathbf{r}') . \] (25)

Both functions are defined as divergent sums over the separation constant \( \beta \) (the partial charge) running an infinite set of discrete values, but the actual summations are performed up to a common maximum value of \( \beta \). Substituting the explicit definitions of the relevant functions given in Eqs. (G13), (G15), (G18), (G19), and (G22) and expanding all the regular parabolic functions over the basis of the spherical harmonics (Eq. (G10)), we obtain the expansion of the irregular function in the form
\[ g_{\text{irr}}(\mathbf{r}) = \sum_{\beta'} [U^T(\mathbf{e})]_{\beta' \mathbf{m} e} \nu_{\beta' \mathbf{m} e}(\mathbf{r}) - \sum_{l} \tilde{\gamma}_{l l'} f_{l l'}(\mathbf{r}) , \] (26)

where
\[ \tilde{\gamma}_{l l'} = J_{l l'} + \cot(\pi n) \] (27)
and \( J_{l l'} \) is given by Eq. (G22). Rewriting Eq. (26) in terms of our functions and notations, we finally obtain it in the form of Eq. (1) with
\[ \gamma_{l l'} = \frac{W_l}{N_{lm}(m!)^2} \left[ \sum_{n>0} A_{\mathbf{n} \mathbf{n}' l} N_n^2 \Omega(n) A_{\mathbf{n} \mathbf{n}' l} - \sum_{k=1}^{\infty} A_{\mathbf{v} \mathbf{u} \mathbf{n} \mathbf{n}' l} c_k^2 \Omega(\mu) A_{\mathbf{v} \mathbf{u} \mathbf{n} \mathbf{n}' l} \right] . \] (28)

where
\[ \Omega(\mu) = \frac{\Gamma(1 + m + \mu)}{\Gamma(1 + \mu)} \left[ \psi(1 + m + \mu) + \psi(1 + \mu) - 2 \ln n + \pi \cot(\pi \mu) \right] . \] (29)

**Fig. 1.** Comparison of two matched irregular spherical solutions with the exact solution at \( n = 10.5, m = 1, l = 3, \) and \( \delta = 1.3. \) Full line and symbols at the upper panels (a) and (b), the exact and matched solutions, respectively. Since the matched solutions are indistinguishable at this scale, their differences with the exact solution are shown at the lower panels (c) and (d) by full and dashed lines for matrices (21) and (28), respectively. All functions are divided by the value of \( g_{l l'}(r = 30, \cos \theta = 0). \)
Both sums in Eq. (28) are divergent, therefore the actual summation is performed up to a common maximum value of $\beta$ using a suitable cutoff function.

Despite the apparent difference between expressions (21) and (28) for $\gamma$, both matrices turn out to be numerically equivalent up to high angular momenta. In order to compare $\gamma_{l}$ from Eqs. (21) and (28), we used the cutoff function of [6]. Two matrices in Eqs. (21) and (28) determine two irregular spherical solutions $\gamma_{l, \text{matched}}(r, \theta)$ matched to the physical irregular parabolic solution by two different methods. The upper panels on Figs. 1 and 2 compare them with the exact function $\gamma_{l}(r, \theta)$ whereas the lower ones show the respective differences. It is seen in Fig. 1 that the matched functions ideally coincide with the exact function at the selected parameters, their difference being only on the order of $10^{-3}$ over the most of the variables intervals.

Since the matching is correct only asymptotically at $n \to \infty$ (i.e. $F \to 0$ at fixed $\delta$ of Eq. (24)), Fig. 2 demonstrates the rate of convergence when $n$ increases from 10.5 to 28.5. The difference of both matched functions with the exact function drops down to $10^{-4}$.

Note that the discrepancy between the exact and matched functions in the figures increases at small $\eta$ (cos$\theta \approx +1$). This discrepancy is not a consequence of the matching error as such, rather it is determined by using a finite basis when calculating the sum in the right-hand side of Eq. (1) and by the non-uniform convergence of this sum at $\eta = 0$.

4. DISCUSSION

The major source of inaccuracies of the Harmin LFT theory is the approximation that enables matching, at short $r$, of the irregular physical Stark wavefunction to the irregular spherical solution with a definite value of the orbital angular momentum $l$. However, such a matching is, strictly speaking, physically impossible because essential $l$-mixing takes place at short $r$ despite the fact that the external field is weak as...
compared with the Coulomb one. Giannakees et al. derived a general expression for the $K$ matrix (i.e. the real scattering matrix which couples the standing waves at infinity, see Eq. (G6) in [6]) without using the explicit matching of the irregular solution. They introduced a one-particle potential modeling the quantum-defect boundary condition near the core and invoked the Lippmann–Schwinger formalism. In fact, as was demonstrated in the present paper, the matching of the irregular Stark and Coulomb solutions had been implicitly present in Giannakees et al. theory, and here we have shown how it can be deduced from one of its basic postulates, Eq. (G22), with the matching being governed by Eqs. (26) and (27). Using these equations together with the expansion of the regular solutions, Eq. (G10), one can obtain exactly the same $K$ matrix as in [6] without invoking any one-particle potential. The key matrix $\gamma$ in our notations has the form of Eq. (28). Our alternative approach is based on the exact expansion of the irregular spherical Coulomb function over the irregular parabolic Coulomb solutions, Eq. (12), derived in [9]. The resulting Eq. (21) for $\gamma$ seemingly differs from Eq. (28). Yet, the numerical tests showed that the two expressions have similar accuracy and the identical ranges of validity.

5. CONCLUSIONS

We performed an analytical matching, in the region of spherical symmetry, of the physical irregular solution of the Stark problem in the hydrogen-like Rydberg atoms to a linear combination of the irregular and regular spherical solutions in the pure Coulomb field. We took into account the fact earlier ignored by researchers in the field that mixing of the states with different momenta occurs within the Coulomb region where the external field is weak as compared to the Coulomb one. With the well-known similar matching for the regular solution, the $S$ matrix can be constructed using standard procedures [9].

This paper is dedicated to the centenary anniversary of the outstanding scientist, the Founder and long-term Director of the L.D. Landau Institute for Theoretical Physics, and merely charming person Academician Isaak Markovich Khalatnikov, “Khalat” among the colleagues and students. One of us (E.S. Medvedev) preserves warm memories of 1957–1963 studies at Moscow Physical–Technical Institute and P.L. Kapitza Institute of Physical problems where Khalat was supervisor of the 722 group.

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