

Removal of the intermediate states in an all-order theory of multiphoton processes

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The implicit summation technique used to calculate the $N - 1$ sums over the intermediate states appearing in the expression of the N -photon transition amplitude provided by the lowest-order perturbation theory is generalized to the calculation of the transition amplitude resulting from the resummation of the whole perturbation series. The method is illustrated by two-photon ionization.

The accurate calculation of the $N - 1$ sums running over the complete set of atomic states appearing in the N th-order transition matrix element describing N -photon absorption is a considerable task. This problem has been tackled by generalizing to arbitrary order¹ an implicit summation technique²⁻⁴ within the framework of the lowest-order perturbation theory (LOPT), i.e., when only the lowest-order nonvanishing term of the perturbation series is retained. We know that LOPT is not able to give satisfactory predictions regarding the behavior of multiphoton processes in which higher-order contributions were expected to play an important role, i.e., resonant absorption.^{5,6} A higher-order perturbation theory (HOPT) has been proposed.⁷ As a result of the resummation of the perturbation series, the transition amplitudes were expressed in terms of continued fractions of the absorption and emission operators of a photon. In the basis of atomic states, the calculation of the probability still involves infinite summations over the complete set of atomic states. Up to now, most of the calculations have proceeded via inversion of matrices of finite sizes.⁸ Such a technique, in which one considers only a limited number of states, gives satisfactory results when few levels contribute to the probability (resonant transitions). It cannot be used when the whole spectrum of the atom must be taken into account. Typical examples of these problems are encountered in the HOPT approach of resonant multiphoton ionization and in multiphoton absorption processes above the ionization threshold (ATI).⁹

The aim of this paper is to show how the implicit summation technique, extensively involved in LOPT, can be extended to the calculation of the resummed expressions of the probability amplitudes provided by HOPT. Our approach, whose general principles are enunciated elsewhere,¹⁰ is

called GIST (generalized implicit summation technique). We show below how it enables to complete the exact calculation of nonperturbative expressions of any transition amplitude. As an example, we consider the case of two-photon ionization. Within the resolvent formalism¹¹ we have to calculate the matrix $G_{ba}^{(2)} = \langle b | G^{(2)} | a \rangle$, where $|a\rangle = |g\rangle \otimes |n\rangle$ and $|b\rangle = |f\rangle \otimes |n-2\rangle$; $|g\rangle$ and $|f\rangle$ being the initial and the final atomic states, respectively, and $|n\rangle$ and $|n-2\rangle$ the corresponding field states.

As a result of the resummation of the perturbation series, $G^{(2)}$ is given by⁷

$$G^{(2)} = \frac{1}{z - H^0 - R^+ - R^-} \times V^- \frac{1}{z - H^0 - R^-} V^- \frac{1}{z - H^0 - R^-}, \quad (1a)$$

where

$$R^\pm = V^\pm G^\pm V^\mp \quad (1b)$$

and

$$G^\pm = \frac{1}{z - H^0 - R^\pm}. \quad (1c)$$

In Eqs. (1), H^0 is the free Hamiltonian of the system atom-plus field, z is a complex variable, $V^- = Da$, and $V^+ = Da^\dagger$; D is the atomic dipole operator and a and a^\dagger are the destruction and the creation operators of a photon, respectively.

It is convenient to eliminate the field operators in Eqs. (1). To this end we define any averaged operator over the field states by ${}^{(n)}O = \langle n | O | n \rangle$. In applying successively the operators $z - H^0 - R^+ - R^-$ and $z - H^0 - R^-$ to the left of $G^{(2)} | a \rangle$, we obtain the following set of hierarchical equations:

$$(z - {}^{(n-2)}H^0) | V_g^{(n-2)} \rangle - {}^{(n-2)}R | V_g^{(n-2)} \rangle = D | V_g^{(n-1)} \rangle, \quad (2a)$$

$$(z - {}^{(n-1)}H^0) | V_g^{(n-1)} \rangle - {}^{(n-1)}R | V_g^{(n-1)} \rangle = D | V_g^{(n)} \rangle, \quad (2b)$$

$$(z - {}^{(n)}H^0) | V_g^{(n)} \rangle - {}^{(n)}R | V_g^{(n)} \rangle = | g \rangle. \quad (2c)$$

The matrix element $G_{ba}^{(2)}$ is given in terms of $| V_g^{(n-2)} \rangle$ by

$$G_{ba}^{(2)} = \sqrt{n(n-1)} \langle f | V_g^{(n-2)} \rangle.$$

The kets appearing in Eqs. (2) are given by $| V_g^{(n-2)} \rangle = {}^{(n-2)}G^{-D} | V_g^{(n-1)} \rangle$, $| V_g^{(n-1)} \rangle = {}^{(n-1)}G^{-D} | V_g^{(n)} \rangle$, and $| V_g^{(n)} \rangle = {}^{(n)}G^{-} | g \rangle$. For shortness we have set $R = R^+ + R^-$. In the polar coordinate representation, the H^0 's contain the radial (differential) Schrödinger operator but the corresponding equations cannot be handled like ordinary differential equations of a single variable.

One of the reasons lies in the presence of the operators R which come from the resummation of the perturbation series (we note that within the lowest-order theory these operators are lacking). The other reason is that each H^0 includes the orbital momentum operator L^2 . In what follows, we describe a method which enables to put Eqs. (2) in a fully tractable form involving only radial differential operators. We note that in Eqs. (2) each unknown ket is the solution of an inhomogeneous equation, the ket in the inhomogeneous term being given by the next equation, etc. Thus each equation can be discussed separately.

Let us consider Eq. (2c) with R iterated once according to Eq. (1b). One has

$$(z - {}^{(n)}H^0) | V_g^{(n)} \rangle - (n+1)D | W_g^{(n+1)} \rangle = | g \rangle, \quad (3a)$$

$$(z - {}^{(n+1)}H^0) | W_g^{(n+1)} \rangle - D | V_g^{(n)} \rangle - {}^{(n+1)}R {}^{(n+1)}G^{-D} | V_g^{(n)} \rangle = 0, \quad (3b)$$

where

$$| W_g^{(n+1)} \rangle = {}^{(n+1)}G^{-D} | V_g^{(n)} \rangle.$$

By iterating many times the operator R , each equation of (2) is replaced by a set of coupled equations which are infinite in number. However, this set can be truncated. The truncation is governed by

the contribution of the expectation values of the operators $R G^{-D} | V_g \rangle$ which are calculated for each iteration. For example, to solve the set of Eqs. (3) one neglects the last term in the left-hand side of Eq. (3b).

To eliminate the angular momentum operator L^2 in Eqs. (3), we express all the operators in the basis of the eigenstates of L^2 . In this representation, the operators H^0 and D are expressed by matrices of finite size, each matrix element being an operator of radial coordinates only. For clarity, we consider two values of the orbital quantum number, i.e., $l=0,1$. By taking into account the selection rules, we have to solve the following four coupled differential equations:

$$\begin{aligned} [z - {}^{(n)}H_0(r)] | V_0 \rangle - (n+1)D | W_1 \rangle &= | g \rangle, \\ [z - {}^{(n)}H_1(r)] | V_1 \rangle - (n+1)D | W_0 \rangle &= 0, \\ [z - {}^{(n+1)}H_0(r)] | W_0 \rangle - D | V_1 \rangle &= 0, \\ [z - {}^{(n+1)}H_1(r)] | W_1 \rangle - D | V_0 \rangle &= 0, \end{aligned} \quad (4)$$

to get the components $| V_0 \rangle$ and $| V_1 \rangle$ of $| V_g^{(n)} \rangle$. In this simplified notation, $| W_0 \rangle$ and $| W_1 \rangle$ are the components of $| W_g^{(n+1)} \rangle$, $| g \rangle$ is any atomic S state and

$${}^{(k)}H_l = -\frac{d^2}{dr^2} - \frac{2}{r} + \frac{l(l+1)}{r^2} + kE_p,$$

E_p being the photon energy.

Since the same discussion holds for Eqs. (2a) and (2b), we observe that our method enables one to calculate the components of $| V_g^{(n-2)} \rangle$ by solving a set of coupled second-order differential equations of a single variable r . Thus, the problem of the double summation has been formulated in a fully tractable form. The number of equations to be solved depends on the order of the process, on the number of iterations for R , and on the range of variation of l . For the sake of brevity, only the case of two-photon ionization has been considered but the method can be generalized to arbitrary order. Such a generalization as well as a quantitative discussion about the merit of this method will be presented in a more expanded account.

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¹¹See, for example, P. Lambropoulos, in *Advances in Atomic and Molecular Physics* (Academic, New York, 1976), Vol. 12, p. 87, and references quoted therein.