Validity and meaning of the momentum-translation approximation for bound states in a radiation field

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The conditions of validity for the momentum-translation approximation of Reiss are found much more stringent than was thought before: multiphotonic transitions induced by one strong field cannot be described by this method. First-order transitions between bound states dressed by a strong low-frequency field can be calculated, but the method gives nothing but a trivial result; only for the ionization of a dressed atom is the approximation found useful.

INTRODUCTION

Much interest arose when Reiss proposed[1,2] the "momentum-translation (MT) approximation" as a means of avoiding high-order perturbation theory in the calculation of multiphotonic transitons in a strong field. Many papers[3,4] using this approximation have been published since. However, doubts are now being raised[5] about the method itself; so it is necessary to reexamine the validity of the MT approximation and its significance.

I. TRANSITIONS IN ONE STRONG FIELD

A. Reiss approximation

With notations taken from Reiss,[1] the Schrödinger equation is ($\hbar = 1$)

$$i\partial_t \Psi = (H_0 + H')\Psi,$$

$$H_0 = \frac{\vec{p}^2}{2m} + V(\vec{x}),$$

$$H' = \frac{-2e\vec{A}\cdot\vec{p} + e^2\vec{A}^2}{2m},$$

where $\vec{A} = a\vec{\epsilon}\cos\omega t$. The dipole approximation for the field is thus made, as in all applications[1-4] of the method. Under the unitary transformation $\Psi = e^{ie\vec{A}\cdot\vec{x}}\overline{\Psi}$, the Schrödinger equation becomes

$$i\partial_t \overline{\Psi} = (H_0 + H_1)\overline{\Psi}, \quad H_I = e\frac{\partial \vec{A}}{\partial t}\cdot\vec{x}. \tag{1}$$

The solution of Eq. (1) can be written as an expansion in powers of H_I:

$$\overline{\Psi}(t) = \Phi(t) - i\int_{-\infty}^{t} dt'\, e^{-iH_0(t-t')} H_I(t')\overline{\Psi}(t'),$$

starting with the solution $\Phi = \phi e^{-iEt}$ of the unperturbed equation $i\partial_t\Phi = H_0\Phi$. The amplitude for transition from i to f is[1]

$$(S-1)_{fi} = -i\int_{-\infty}^{+\infty} dt\,\langle \Phi_f(t), H'(t)\Psi_i(t)\rangle, \tag{2}$$

$$(S-1)_{fi} = -i\int_{-\infty}^{+\infty} dt\,\langle \Phi_f(t), H'(t)e^{ie\vec{A}\cdot\vec{x}}\Phi_i(t)\rangle + (-i)^2\int_{-\infty}^{+\infty} dt\,\langle \Phi_f(t), H'(t)e^{ie\vec{A}\cdot\vec{x}} \times \int_{-\infty}^{t} dt'\, e^{-iH_0(t-t')} H_I(t')\Phi_i(t')\rangle + \cdots. \tag{3}$$

The first term in Eq. (3) is the Reiss approximation.

B. Higher-order corrections

Reiss considers[1] that the second term in (3) gives a contribution whose order of magnitude is $|H_I/E|$ times the contribution of the first term, from which he states the condition of validity (a_0 is the Bohr radius):

$$|eaa_0(\omega/E)| \ll 1. \tag{4}$$

This is an argument typical of a stationary problem (which does not mean that the conclusion is wrong). What is needed actually in such a nonstationary problem is the transition matrix element $T_{fi}^{(n)}$, which is the term proportional to $\exp[i(E_f - E_i + n\omega)t]$ in the integrand of (2). That is, of course, what is done by Reiss for the explicit calculation of transition probabilities, but it is also what should be done before a discussion of the validity of the approximation.

From the expansion (3) in powers of p of H_I, we can write

$$T_{fi}^{(n)} = \sum_{p=0}^{\infty}\langle \phi_f, U_p^{(n)}\phi_i\rangle, \tag{5}$$

where we want the zeroth-order operator $U_0^{(n)}$ to dominate over the remainder, for the MT approximation to be valid. That first term is easily calculated to give

$$U_0^{(n)} = -\frac{ea\vec{\epsilon}\cdot\vec{p}}{m} i^{n-1} J_n'(z) + \frac{e^2a^2}{2m} i^{n-2} J_n''(z)$$
$$= [H_0, i^n J_n(z)],$$
$$z = ea\vec{\epsilon}\cdot\vec{x}.$$

The second expression is used by Reiss and followers, but the first one serves our purpose better. Now, the first-order term is explicitly

$$U_1^{(n)} = [U_0^{(n-1)}(H_0 - E_i + \omega)^{-1} - U_0^{(n+1)}(H_0 - E_i - \omega)^{-1}](-\tfrac{1}{2}i\omega z). \quad (6)$$

We shall replace the "Green operators" in Eq. (6) by a mean energy difference ΔE. This trick is of common use in stationary perturbation computations, and retains some validity in nonstationary problems, as exemplified by the work of Bebb and Gold[6]: for the multiphoton ionization of a ground-state hydrogen atom, they show that ΔE can be chosen (away from resonances) as the first excitation energy $E_2 - E_1$; the mean energy approximation should be still more reliable for bound-bound multiphoton transitions such as $1s$-$2s$ excitation of atomic hydrogen, where there are no resonances. Within this approximation, the general perturbation term can be written

$$U_p^{(n)} \simeq \left[-\frac{ea\vec{\epsilon}\cdot\vec{p}}{m} i^{n-1} \sum_q \binom{p}{q} J'_{n-p+2q}(z) + \frac{e^2a^2}{2m} i^{n-2} \sum_q \binom{p}{q} J''_{n-p+2q}(z)\right] \left(-\frac{\omega z}{2\Delta E}\right)^p.$$

(For convenience of notation, ΔE has been put constant in all terms.)

C. Low-field limit

For not too strong fields ($z \ll 2|n|^{1/2}$), we can use the expansion of Bessel functions in powers of the argument:

$$J_n(z) = (\tfrac{1}{2}z)^n \sum_{k=0}^{\infty} \frac{(-\frac{1}{4}z^2)^k}{k!(n+k)!},$$

to get, for $p < |n| - 1$,

$$U_p^{(n)} \simeq \left[-\frac{ea\vec{\epsilon}\cdot\vec{p}}{m} \frac{(\frac{1}{2}iz)^{|n|-1}}{2(|n|-p-1)!} + \frac{e^2a^2}{2m} \frac{(\frac{1}{2}iz)^{|n|-2}}{4(|n|-p-2)!}\right] \left(\frac{-\omega}{\Delta E}\right)^p \left(\frac{n}{|n|}\right)^p.$$

For $p \geq |n| - 1$ the discussion is more tedious (but less important), and one can find the order-of-magnitude equation

$$U_{p+2}^{(n)} \sim U_p^{(n)} (2\omega z/p\Delta E)^2.$$

We conclude that for $|n| \geq 2$, the first term in Eq. (5) dominates if

$$|n\omega/\Delta E| \ll 1. \quad (7)$$

The discussion is different for $|n| = 0$ or 1, and the condition of validity is found to be

$$|z\omega/2\Delta E| \ll 1. \quad (8)$$

Condition (7) is far more drastic than Reiss's condition (4), which is just (8), and $|\omega/E| \ll 1$.[1] Condition (8) for $|n| = 0$ or 1 means simply that the Reiss approximation is at least as good as first-order perturbation theory, which we knew.[1] On the contrary, condition (7) means that the MT approximation cannot be used for the calculation of multiphoton transitions induced by the field itself, since then $|n\omega| = |E_f - E_i|$ cannot be much less than $|\Delta E|$; $|n\omega/\Delta E|$ is more likely to be of order unity, which is the conclusion reached independently by Cohen-Tannoudji *et al.*[7]

D. Strong-field limit

Let us now consider the strong-field limit $z \gg 1$. We can use the Hankel asymptotic expansion of Bessel functions[8] to show that

$$U_1^{(n)} \simeq U_0^{(n)}(-n\omega/\Delta E).$$

The condition of validity (7) is thus still necessary. Further study could lead to a condition for the first term in (5) to dominate over the whole remainder: This is a difficult task. I have made the calculation for $|n| \gg 1$, and the resulting condition was

$$|z^{1/2}\omega/\Delta E| \ll 1, \quad (9)$$

which is not very different from Reiss's condition (4).

E. Another discussion is needed for two-field cases

Considering condition (7), it may be thought that the MT approximation could keep its validity in the case where the greatest part of the transition energy is brought in by a weak field $\vec{\alpha}$ (treated in first-order perturbation theory), while the strong field $\vec{A}$ supplies a little remainder $|n\omega| \ll \Delta E|$. The method used by Reiss and others for two-field problems[1-3] is not valid under condition (7), so we shall give a new one in Sec. II. The analysis has been carried on for a monochromatic linearly polarized field. Generalizations are straightforward, but tedious.

II. MEANING OF APPROXIMATION AND VALIDITY FOR TRANSITIONS IN TWO FIELDS

A. Link with perturbation theory

In this section, I want to show directly which processes are described by the MT approximation.

The exact wave function $\Psi_i(t)$ is given by the usual time-dependent perturbation theory from the development in powers of H' and the time integrations of

$$\Psi_i(t) = \Phi_i(t) - i \int_{-\infty}^{t} dt' e^{-iH_0(t-t')} H'(t') e^{\eta t'} \Psi_i(t'),$$

where $\eta \to 0_+$ for an adiabatic switching of the perturbation. The result includes a number of sums on intermediate states j with denominators $(E_i - E_j + p\omega + i\eta)^{-1}$. Condition (7) of Sec. I suggests that the MT approximation could be obtained by neglecting $|p\omega| \ll |E_i - E_{j\neq i}|$ everywhere. There remains the questions of intermediate i states; fortunately, we can write the exact $\Psi_i(t)$ in the form[9]

$$\Psi_i(t) = \varphi_i(t) \langle \varphi_i(t) | \varphi_i(t) \rangle^{-1/2} \exp\left[-i \left(E_i t + \int_{-\infty}^{t} dt' \, \mathrm{Re} \langle \phi_i | H'(i') e^{\eta t'} | \varphi_i(t') \rangle \right) \right],$$

where $\langle \phi_i | \varphi_i(t) \rangle = 1$ and the perturbation development of $\psi_i(t)$ contains only[10] denominators $(E_i - E_j + p\omega + i\eta)^{-1}$ with $j \neq i$. If we neglect the $p\omega$ terms everywhere (an approximation to be discussed in Sec. IIB), $\varphi_i(t)$ becomes the corresponding wave function of the *static* perturbation theory,[11] as does $\Psi_i(t)$; the normalized wave function of the static perturbation theory (i.e., as if $\vec{A}$ were time independent) is exactly $e^{ie\vec{A}\cdot\vec{x}}\Phi_i(t)$, and the approximation is thus

$$\Psi_i(t) \simeq e^{ie\vec{A}\cdot\vec{x}} \Phi_i(t).$$

The first-order transition amplitude in a weak field of circular frequency Ω can now be easily calculated as the term proportional to $\exp i(E_f - E_i - \Omega - n\omega)t$ in

$$\langle \Psi_f(t) | \left(\frac{-e\vec{\alpha}\cdot\vec{p}}{m} + \frac{e^2\vec{\alpha}\cdot\vec{A}}{m} \right) | \Psi_i(t) \rangle \simeq \langle \Phi_f(t) | e^{-ie\vec{A}\cdot\vec{x}} \left(\frac{-e\vec{\alpha}\cdot\vec{p}}{m} + \frac{e^2\vec{\alpha}\cdot\vec{A}}{m} \right) e^{ie\vec{A}\cdot\vec{x}} | \Phi_i(t) \rangle = \langle \Phi_f(t) | \left(\frac{-e\vec{\alpha}\cdot\vec{p}}{m} \right) | \Phi_i(t) \rangle, \tag{10}$$

i.e., exactly the same result as if the dressing field $\vec{A}$ were not present.

B. Discussion

If the condition

$$\forall \; j \neq i, \quad |N\omega| \ll |E_i - E_j| \tag{11}$$

is fulfilled for some N, we can consistently neglect the $p\omega$ terms in the perturbation development of $\varphi_i(t)$ up to the Nth order in $|\vec{A}|$. The MT approximation can thus be used for the calculation of a transition involving $n \leq N$ quanta ω (and one quantum Ω) if the dressing field $\vec{A}$ is not too strong: for there is certainly a limitation on the strength of the field, since in very strong fields $|p\omega|$ may be much greater than $n\omega$ in some intermediate states of some amplitudes that contribute significantly to the transition. This limitation can be determined approximately from arguments similar to those of Sec. I, leading to condition (4) [or perhaps (9)]; conditions like (7) or (8) could also be obtained, but they are weaker than (4) and (11).

The first-order transition amplitude of Eq. (10) could have been calculated equivalently in the representation where the wave equation is (1) and the perturbation is H_I. The MT approximation is therefore the zeroth-order approximation, and the trivial result obtained in (10) is now evident. What we have gained is the knowledge that the same trivial result holds in perturbation theory to the Nth order—not only to the zeroth order—provided conditions (4) and (11) are satisfied.

C. Conclusion

The MT approximation cannot be used to calculate multiphotonic transitions in a strong field. It can be used under conditions (4) and (11) to compute first-order transitions between bound states dressed by a strong field, but it gives nothing but trivial results. The only open way I could find is the calculation of bound-free transitions in a dressed atom, the bound state being treated in the MT approximation, while another approximation is used for the free state, e.g., the transformation of Schwinger[12] and Henneberger,[13] plus the Born approximation for high energies:

$$\Psi_f(t) \simeq \exp\left(-i \int_{-\infty}^{t} dt' \, H'(t') e^{\eta t'} \right) e^{-iE_f t} e^{i\vec{k}\cdot\vec{x}}.$$

This could lead to nontrivial results.

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