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SPECTRUM OF ATOMIC ELECTRONS IONISED BY AN INTENSE FIELD

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Gauge problems arising in recent multiphoton ionisation computations are discussed. The recent results of Reiss [18],[19] based on his earlier formulae [20] are queried.

Nowadays the problem of multiphoton stripping of atoms including the above-threshold ionisation phenomena is intensely investigated both experimentally [1]–[8] and theoretically [9]–[16]. Some of the experiments [7] can be fairly well explained by the theory of Keldysh [17].

In two recent articles [18],[19] the spectrum of emitted atomic electrons originated from multiphoton ionisation of xenon was analysed and qualitative agreement was found with above-threshold ionisation experiments [6], [8]. Both articles [18], [19] are based on an earlier work of Reiss [20], where the effect of intense radiation field on weakly bound systems was discussed in a great detail using Volkov [21] solutions as final states but contrary to Keldysh [17], who used the electric-dipole (Göppert-Mayer, **xE**) gauge, Reiss [20] worked in the radiation (Coulomb, **pA**) gauge. It was claimed by Reiss [20] that the approximation used was equivalent to that of Keldysh [17]. However, differences in the formulae for the S matrix elements and transition probabilities per unit time appeared, although these must be invariant under gauge transformation. We think that these differences are the consequence of an incorrect use of the radiation gauge in Reiss's paper [20].

In order to discuss the situation we show that the S matrix element computed by Keldysh [17] in the electric-dipole gauge remains unchanged under transformation into the radiation gauge if one carefully carries out this transformation [22], [23], i.e. the S matrix element is gauge invariant as it is expected. Doing this we point out those steps of [20] which lead to formulae different from that of [17]. As the recent articles [18], [19] are based on the criticised formulae of [20] the results of these papers are also questioned.

The S matrix element, which describes the photodetachment, can be derived from formula (3) of [20]

$$(S-1)_{fi} = -\frac{i}{\hbar}\int dt \int d^3r\ \psi_f^{g*}(\mathbf{r},t)\ P^g(\mathbf{r},t)\ \psi_i^g(\mathbf{r},t), \tag{1}$$

where ψ_f^g is the final state of the free charged particle in the presence of the electromagnetic field and ψ_i^g is the initial, unperturbed bound state, which is well known.

$P^g(\mathbf{r},t)$ stands for that term which will be considered as perturbation in the time-dependent Schrödinger equation. The index g notes that the wave functions and the operator P depend on the choice of gauge.

Expression (1) is evaluated by Keldysh [17] in the electric-dipole gauge and by Reiss [20] in the radiation gauge. Therefore, first we briefly summarize those well known results which we need in order to treat the gauge transformation problem [22], [23].

The Schrödinger equation is written in the following form

$$\{\varepsilon^g(t) + F^g(t)\}\ \psi^g(\mathbf{r},t) = 0, \tag{2}$$

where $\varepsilon^g(t)$ is the time-dependent instantaneous energy operator of the system

$$\varepsilon^g(t) = \frac{1}{2m}\left(\mathbf{p} - \frac{e}{c}\,\mathbf{A}^g(\mathbf{r},t)\right)^2 + V(\mathbf{r}) \tag{3}$$

and $F^g(t)$ is defined as

$$F^g(t) = eU^g(\mathbf{r},t) - i\hbar\frac{\partial}{\partial t}. \tag{4}$$

Here $\mathbf{p}$ is the operator associated with the momentum of the particle, which has a gauge-independent form $\mathbf{p} = -i\hbar\nabla$, U_g and $\mathbf{A}^g$ are the scalar and vector potentials, respectively, describing the electromagnetic radiation and $V(\mathbf{r})$ stands for the atomic potential. If we transform the electromagnetic potentials from a gauge g to another one g' as usual, then the operators (e.g. operator O) and the state vector of a physical system are also transformed by a unitary transformation $T(\mathbf{r},t)$ as

$$O^{g'} = T(\mathbf{r},t)\ O^g\ T^+(\mathbf{r},t), \tag{5a}$$

$$\psi^{g'}(\mathbf{r},t) = T(\mathbf{r},t)\ \psi^g(\mathbf{r},t), \tag{5b}$$

with

$$T(\mathbf{r},t) = \exp\ (ie\ \chi(\mathbf{r},t)/\hbar c). \tag{6}$$

Here $\chi(r,t)$ is an arbitrary function, which governs gauge transformation. The function

$$\chi_{ER}(\mathbf{r},t) = \mathbf{A}(t).r \tag{7}$$

corresponds to that gauge transformation which transforms from the electric-dipole gauge into the radiation gauge. Here and throughout the paper the long wavelength approximation (LWA) is used.

Only those operators can correspond to physical quantities which have gauge invariant eigenvalues. The instantaneous energy operator $\varepsilon^g(t)$, the operator $F^g(t)$ and the mechanical momentum $\pi^g = \mathbf{p} - e/c\ \mathbf{A}^g$ all represent physical quantities. In the LWA the eigenvalues of ε^g are time-independent and in the electric-dipole gauge $\varepsilon^E = H_0$ with $H_0 = \mathbf{p}^2/2m + V(\mathbf{r})$, which is the unperturbed Hamiltonian and represents a nonphysical quantity. Thus the eigenvalues of ε^g in all gauges are equal to the eigenvalues of H_0 which are usually known. Furthermore, the eigenstates of the two operators are also identical in the electric-dipole gauge.

Now in order to reconstruct Keldysh's calculation we consider the Schrödinger equation in the electric dipole gauge. In this case $\mathbf{A}^E = 0$ and $U^E = -\mathbf{r}.\mathbf{E}$ so (2) reads

$$\{H_0 - i\hbar\ \partial/\partial\ t - e\mathbf{r}.\mathbf{E}(t)\}\ \psi^E(\mathbf{r},t) = 0, \tag{8}$$

where we used the $\varepsilon^E = H_0$ and the $F^E = -i\hbar\ \partial/\partial\ t - e\mathbf{r}.\mathbf{E}(t)$ equalities. The index E refers to the electric-dipole gauge. If we start the solution of (8) with the time dependent wavefunction

$$\psi_n^E(\mathbf{r},t) = \exp\ (-iE_nt/\hbar)\ \Phi_n(\mathbf{r}), \tag{9}$$

which satisfies the $[H_0 - i\hbar\ \partial/\partial\ t]\ \psi^E(\mathbf{r},t) = 0$ equation, then the $-e\mathbf{r}.\mathbf{E}(t)$ term can be considered as perturbation in (1), i.e.

$$P^E(\mathbf{r},t) = -e\mathbf{r}.\mathbf{E}(t). \tag{10}$$

The wave function $\Phi_n(\mathbf{r})$ is that eigenstate of H_0 which has an eigenvalue E_n. Thus $\psi_n^E(\mathbf{r},t)$ of the form (9) corresponds to the initial state in (1). Before obtaining the final state we restrict ourself to the linearly polarized case investigated by Keldysh [17]. Then the electric field strength is given as $\mathbf{E}(t) = \mathbf{E}\ \cos(\omega t)$. As final state we use the wavefunction of a free electron in this field ([17], formula (6))

$$\psi_f^E(\mathbf{r},t) = \exp\frac{i}{\hbar}\left\{\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\ \sin(\omega t)\right)\cdot\mathbf{r} - \right.$$
$$\left. - \int_0^t \frac{1}{2m}\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\ \sin(\omega\tau)\right)^2 d\tau\right\}. \tag{11}$$

If we use in (1) the initial, final states and the perturbation P^E given above (formulas (9), (11) and (10), respectively), then we get

$$(S-1)_{fi} = -\frac{i}{\hbar}\int dt \int d^3r\ \psi_f^{E*}(\mathbf{r},t)\ P^E(\mathbf{r},t)\ \psi_n^E(\mathbf{r},t) \tag{12}$$

and from (12) with the usual procedure the transition probability per unit time of the multiphoton detachment process can be obtained. This is Keldysh's result.

As a next step we show that if one evaluates (1) in the radiation gauge then the result is also (12). Using the rules of the gauge transformation (5a,5b) and formulae (6) and (7) we obtain the initial and final state wave functions and the operator of perturbation in the radiation gauge as

$$\psi^R(\mathbf{r},t) = T_{ER}(\mathbf{r},t)\ \psi^E(\mathbf{r},t) \tag{13a}$$

and

$$P^R(\mathbf{r},t) = T_{ER}(\mathbf{r},t)\ P^E(\mathbf{r},t)\ T^+_{ER}(\mathbf{r},t) \tag{13b}$$

with

$$T_{ER}(\mathbf{r},t) = \exp\ (ie\mathbf{r}.\mathbf{A}(t)/\hbar c), \tag{14}$$

where the index R refers to the radiation gauge. As the operators T_{ER} and P^E are commuting, i.e. $T_{ER}\ P^E = P^E\ T_{ER,}$ and T_{ER} is unitary

$$P^R(\mathbf{r},t) = P^E(\mathbf{r},t). \tag{15}$$

Putting (15) into (1), using(13a) and again the unitarity of T_{ER} the equivalence of the result with (12) becomes obvious.

Finally, in order to point out the inconsistencies in the calculation of Reiss [20] we discuss the problem once more in detail.

We start with the problem of what kind of state can be considered as a bound, initial state of definite energy in the radiation gauge. In view of the considerations given at the beginning of this comment it seems obvious that the eigenfunctions of the instantaneous energy operator (3) are appropriate for this. Their eigenvalues are identical with the eigenvalues of H_0 and the wavefunctions in the radiation gauge can be obtained from (9) applying transformation (13a) with (14)

$$\psi^R_n(\mathbf{r},t) = \exp\ (ie\mathbf{r}.\mathbf{A}(t)/\hbar c)\ \psi^E_n(\mathbf{r},t). \tag{16}$$

These wave functions obey the equation

$$\epsilon^R(t)\ \psi^R_n(\mathbf{r},t) = E_n\ \psi^R_n(\mathbf{r},t). \tag{17}$$

However, in the radiation gauge Reiss [20] uses an incorrect initial state of the form

$$\psi_i(\mathbf{r},t) = \Phi_n(\mathbf{r})\ \exp\ (-iE_nt/\hbar), \tag{18}$$

which is exactly the initial wave function ψ^E_n in the electric-dipole gauge given by (9),

Applying again transformation (13a) with (14) on (11) we obtain the final state in the radiation gauge which is of Volkov type [21]

$$\psi_f^R(\mathbf{r},t) = \exp\frac{i}{\hbar}\left\{\mathbf{p}\cdot\mathbf{r} - \int_0^t \frac{1}{2m}\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\sin(\omega\tau)\right)^2 d\tau\right\}. \tag{19}$$

This is used also by Reiss [20] as final state. Thus the initial and final states of Reiss are inconsistent as they are taken at different gauges.

The incorrect use of the initial state wave function by Reiss results in a perturbation term [20]

$$P^R(\mathbf{r},t) = -\frac{e\mathbf{A}(t).(-i\hbar\nabla)}{m\,c} + \frac{e^2\mathbf{A}(t)^2}{2mc^2}, \tag{20}$$

which is also incorrect. This can be shown in the following way. In the radiation gauge $U^R = 0$ and $\mathbf{A}^R = \mathbf{A}(t)$, thus the Schrödinger equation (2) has the form

$$\left\{\varepsilon^R(t) - i\hbar\,\partial/\partial\,t\right\}\,\psi^R(\mathbf{r},t) = 0, \tag{21}$$

with

$$\varepsilon^R(t) = \frac{1}{2m}\left(\mathbf{p} - \frac{e}{c}\mathbf{A}(t)\right)^2 + V(r). \tag{22}$$

We make a formal modification of this equation adding to and subtracting from it the same quantity $e\mathbf{r}.\mathbf{E}(t)$. Then (21) can be written as

$$\left\{[\varepsilon^R(t) - i\hbar\,\partial/\partial\,t + e\mathbf{r}.\mathbf{E}(t)] - e\mathbf{r}.\mathbf{E}(t)\right\}\,\psi^R(\mathbf{r},t) = 0. \tag{23}$$

Substituting (16) into (23) and using (17) and

$$\mathbf{E}(t) = -\frac{\partial}{c\partial t}A(t), \tag{24}$$

we can recognize that the effect of the square bracket on the wave function (16) gives null. Thus, contrary to Reiss [20], the last term in (23) has to be considered as perturbation in the radiation gauge

$$P^R = -e\mathbf{r}.\mathbf{E}(t), \tag{25}$$

which, in accordance with (15), is the same as in the case of electric-dipole gauge (10). This follows from the correct choice of the initial state (16).

Summarizing, our statements are as follows:

Reiss [20] uses improper initial state wave function and improper perturbation term (Eq. (20) in this note) in the radiation gauge.

As a consequence, formulae (11), (12) and (26), (42) for $(S-1)_{fi}$ and the transition probabilities per unit time (31), (32) and (45) of Reiss [20] are not correct. Formula (45), which describes the case of linear polarization, significantly differs from the correct result of Keldysh [17], [18].

Finally the results of recent calculations [18], [19] are also questionable because these are based on the formulae criticized above.

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