PHYSICAL REVIEW A VOLUME 40, NUMBER 1 JULY 1, 1989

Adiabatic theory of nonresonant multiphoton ionization

Marvin H. Mittleman
Department of Physics, The City College of New York, New York, New York 10031

(Received 12 November 1987; revised manuscript received 15 August 1988)

The Keldysh-Faisal-Reiss theory of nonresonant multiphoton ionization seems to give reasonably good agreement with experiments for production rates in the above-threshold-ionization peaks. The theory has some conceptual difficulties, since physically equivalent Hamiltonians give radically different results when treated by this method. An alternate form of the S matrix is used here in which approximation must be made upon the exact wave function evolving from the initial state rather than the time-reversed form used in the Keldysh-Reiss treatment. The exact initial state is approximated by an adiabatic form which results in an S matrix that eliminates the conceptual difficulties described above.

I. INTRODUCTION

Recent experimental[1,2] and theoretical[3-5] work on multiphoton ionization have brought the field to a very interesting state. For the intensities of interest, $10^{13}-10^{14}$ W/cm^2, and the large number of photons transferred, perturbation theory is both practically impossible and inappropriate. Then other than numerical simulations, an S-matrix approach is the only available theory. The Keldysh[3]-Faisal[4]-Reiss[5] (KFR) version of S-matrix theory gives results for the above-threshold-ionization (ATI) peaks which are almost an order of magnitude smaller than experimental results,[6] and this fact has frequently been quoted as criticism. More recent[7] experiments in which particular ATI peaks are examined for electron energy with high precision show numerous resonance subpeaks within a particular ATI peak. These are interpreted[7] as multiphoton resonance with successive Rydberg states as these states are shifted relative to the ground state by the time (and space) variation of the laser intensity. Typically these subpeaks are a factor of 5 or so above the nonresonant background within the ATI peak under study. In other words, the KFR theory which neglects resonances is not in bad agreement with the nonresonant part of the experimental results. The theory is disturbingly simple and, I claim, has some internal inconsistencies, namely, that equivalent theories give very different results. These are discussed in Sec. II.

The theory starts from the Keldysh approximation which is to replace the exact wave function by the Volkov state[8] in the S matrix, i.e., to neglect the electron-ion interactions. This rough agreement between theory and experiment has led some to try to improve upon the theory by getting a better approximation for the final state of the electron. This has, so far, not been very successful. An equivalent form for the S matrix is used here as a starting point but the approximation procedure is one for the initial rather than the final state. This is done by a low-frequency approximation scheme for the initial state of the atom. The method starts from the fact that the ground state of the atom is isolated from the others by an excitation energy which is large compared to the photon energy. If this were not so, then ATI would not be observable since the atom would be readily ionizable and so would ionize in the leading edge of the laser pulse. The electron would then escape the region of the ion long before the high-intensity part of the pulse arrives and so would absorb no "additional photons." This method is used to write the new approximate S matrix. It is shown in Sec. III that this new form does not give different results for equivalent theories. However, under selected conditions of laser polarization and/or electron production angle this new theory gives very different results from the previous one.

II. DIFFICULTIES WITH PREVIOUS THEORIES

An exact form of the S matrix for multiphoton ionization is (for a single-particle model with $\hbar=1$)

$$S=-i\int dt\, d^3r[\psi_q^{(-)}(\mathbf{r},t)]^* V_i(\mathbf{r},t)\phi_i(\mathbf{r},t)\ , \tag{2.1}$$

where $\psi_q^{(-)}$ is the exact time-reversed wave function for the final state satisfying

$$\left[i\frac{\partial}{\partial t}-H\right]\psi_q^{(-)}=0\ , \tag{2.2}$$

with the boundary condition at $t\rightarrow+\infty$, that $\psi_q^{(-)}$ approaches the final state of the electron with momentum $\mathbf{q}$. Here H is the total Hamiltonian. The state ϕ_i is defined by

$$\left[i\frac{\partial}{\partial t}-H_i\right]\phi_i=0\ , \tag{2.3}$$

with the boundary condition at $t\rightarrow-\infty$ that $\psi_i^{(+)}$ approaches the initial atomic state. The final state $\chi_q^{(-)}$ satisfies (2.7) with the same initial condition and V_f is defined by

$$H=H_i+V_i\ . \tag{2.4}$$

If we define

$$H=\frac{1}{2m}[\mathbf{P}+e\,\mathbf{A}(\omega t)]^2+V(r) \tag{2.5}$$

then the "Keldysh[3] approximation" is to neglect V relative to the electron-field interaction thereby replacing $\psi_q^{(-)}$ by $\chi_q^{(-)}$, where

$$\left[i\frac{\partial}{\partial t}-H_f\right]\chi_q^{(-)}=0\ , \tag{2.6}$$

with

$$H_f=\frac{1}{2m}[\mathbf{P}+e\,\mathbf{A}(\omega t)]^2\ . \tag{2.7}$$

This is the Volkov state.[8] Reiss has applied this for a linear polarization with the choice $H_i=T+V(r)$ with the result

$$S=-2\pi i\sum_N \delta(\varepsilon_q-(N\omega-U_p+W_0))T_N^R\ , \tag{2.8}$$

$$T_N^R=\sum_{a=-\infty}^{\infty} J_a(U_p/2\omega)J_{N+2a}(\boldsymbol{\alpha}\cdot\mathbf{q}) \times\int d^3r\, e^{-i\mathbf{q}\cdot\mathbf{r}}V(r)u_0(r)\ , \tag{2.9}$$

where the parameters are defined by (U_p is the ponderomotive potential)

$$U_p=e^2E^2/4m\omega^2,\quad \boldsymbol{\alpha}=e\mathbf{E}/m\omega^2\ , \tag{2.10}$$

and the bare ground state is given by

$$(W_0-T-V)u_0(r)=0\ . \tag{2.11}$$

This is the form that gives rough agreement with experiment. There is, however, a profound formal difficulty with (2.8). Any meaningful physical result must depend only on the *difference* of energy levels and that remark extends to the "dressed" levels too. The argument of the δ function in (2.8) can be written as

$$(\varepsilon_q+U_p)-W_0-NW\ .$$

The term ε_q+U_p is the dressed or shifted value of the continuum energy,[1] while W_0 is the bare "bare" energy of the initial state. For consistency W_0 should be replaced by $W_0+\Delta W_0$ where ΔW_0 is the shift of the ground-state energy due to the field, the dynamic stark effect, and this should emerge from a consistent calculation. This effect is not numerically significant[1] at the intensities of current experiments.

Suppose that instead of (2.5) we start with the Hamiltonian

$$\overline{H}=\frac{p^2}{2m}+\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\omega t)+V(r)\ , \tag{2.12}$$

which differs from (2.5) by the $A^2(t)$ term, which can be transformed away by a spatially independent contact transformation and so has *no observable effects*. That is, H and $\overline{H}$ differ by a term which is a total time derivative. Then either classically or quantum mechanically they lead to identical observable effects. The preceding equations are then modified:

$$\overline{S}=-i\int dt\, d^3r(\overline{\psi}_q^{(-)})^*\overline{V}_i\phi_i\ , \tag{2.1$'$}$$

where

$$\left[i\frac{\partial}{\partial t}-\overline{H}\right]\overline{\psi}_q^{(-)}=0 \tag{2.2$'$}$$

and

$$\overline{H}=H_i+\overline{V}_i\ , \tag{2.4$'$}$$

with (2.3) unchanged. The Keldysh[3] approximation then yields $\overline{\psi}_q^{(-)}\rightarrow\overline{\chi}_q^{(-)}$ where

$$\left[i\frac{\partial}{\partial t}-\overline{H}_f\right]\overline{\chi}_q^{(-)}=0 \tag{2.6$'$}$$

and

$$\overline{H}_f=\frac{p^2}{2m}+\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\omega t)\ , \tag{2.7$'$}$$

again differing from (2.7) by the A^2 term. The same choice for the initial state yields

$$\overline{S}=-2\pi i\sum_N\delta(\varepsilon_q-(N\omega+W_0))\overline{T}_N\ , \tag{2.8$'$}$$

where

$$\overline{T}_N=J_N(\boldsymbol{\alpha}\cdot\mathbf{q})\int d^3r\, e^{-i\mathbf{q}\cdot\mathbf{r}}V(r)u_0(r)\ . \tag{2.9$'$}$$

This result has serious difficulties. The energy dependence in the δ function in (2.8′) is clearly wrong. Both theory[9,10] and experiment[1] require the presence of the ponderomotive potential U_p as it appears in (2.8). Furthermore, the numerical results obtained from (2.9′) give angular distributions which show no agreement at all with experiment.[11]

The question arises as to why two such similar conceptual treatments give such wildly divergent results and how can one make an *a priori* choice between the two. This is discussed below.

III. LOW-FREQUENCY TREATMENT OF THE INITIAL STATE

Another form[12] of the S matrix which is exact is the time-reversed form of (2.1)

$$S=-i\int dt\, d^3r(\chi_q^{(-)})^*V_f\psi_i^{(+)}\ , \tag{3.1}$$

where $\psi_i^{(+)}$ is the exact wave function satisfying

$$\left[i\frac{\partial}{\partial t}-H\right]\psi_i^{(+)}=0\ , \tag{3.2}$$

with the boundary condition at $t\rightarrow-\infty$ that $\psi_i^{(+)}$ approaches the initial atomic state. The final state $\chi_q^{(-)}$ satisfies (2.7) with the same initial condition and V_f is defined by

$$H=H_f+V_f\ . \tag{3.3}$$

In this form the approximation procedure must be applied to the initial form of the exact wave function, $\psi_i^{(+)}$. For the reasons discussed in the Introduction we proceed with a bound-state-low-frequency approximation.[13] To that end let

$$\psi_i^{(+)}=e^{-ie\mathbf{r}\cdot\mathbf{A}(\omega t)}\overline{\Phi}_i^{(+)} \tag{3.4}$$

to obtain

$$\left[i\frac{\partial}{\partial t}-\left[\frac{p^2}{2m}+V(r)+e\mathbf{r}\cdot\mathbf{E}(\omega t)\right]\right]\Phi_i^{(+)}=0\ , \tag{3.5}$$

where the electric field is defined by

$$\mathbf{E}=-\dot{\mathbf{A}}\ . \tag{3.6}$$

We now define an adiabatic set of states

$$\left[W_n(\omega t)-\left[\frac{p^2}{2m}+V(r)+e\mathbf{r}\cdot\mathbf{E}(\omega t)\right]\right]\phi_n(\omega t)=0 \tag{3.7}$$

and note that the ground state, which is well separated from the others, is well described by this method. Strictly speaking, the electric field in (3.7) must be cut off at some large distance since the spatial boundary conditions are unphysical for (3.7). It will, however, only be treated in perturbation theory so no difficulty will arise. Then Φ_i in (3.5) can be well approximated by

$$\Phi_i^{(+)} \simeq \phi_i(\omega t)\exp\left[-i\int^t dt' W_i(\omega t')\right] . \tag{3.8}$$

The lowest-order approximation is sufficient for ϕ_i,

$$\phi_i(\omega t) \simeq u_0(r) \tag{3.9}$$

[see (2.11)] and the second-order approximation for $W_i(\omega t)$ is

$$W_i(\omega t) \simeq W_0 - \tfrac{1}{2}\alpha_s E^2(\omega t) , \tag{3.10}$$

where α_s is the static dipole polarizability of the ground state. The electric field is described with arbitrary polarization

$$\mathbf{E}(\omega t) = E_0[\hat{\mathbf{x}}\cos(\tfrac{1}{2}\xi)\sin(\omega t) - \hat{\mathbf{y}}\sin(\tfrac{1}{2}\xi)\cos(\omega t)] , \tag{3.11}$$

where ξ is the polarization parameter and $\hat{\mathbf{z}}$ is the direction of propagation. This yields

$$\begin{aligned}\psi_i^{(+)} \simeq {} & e^{-ie\mathbf{r}\cdot\mathbf{A}(\omega t)}u_0(r) \\ & \times\exp\left[-i\left[(W_0 - \tfrac{1}{4}\alpha_s E_0^2)t \right.\right. \\ & \left.\left. - \frac{1}{8\omega}\alpha_s E_0^2\cos\xi\sin(2\omega t)\right]\right] ,\end{aligned} \tag{3.12}$$

where $A(\omega t)$ is obtained from (3.6) and (3.11).

The final state in (3.1) can be obtained from (2.6) and (2.7) as

$$\begin{aligned}\chi_q^{(-)} = {} & \exp\{i[\mathbf{q}\cdot\mathbf{r} - (\varepsilon_q + U_p)t + \boldsymbol{\alpha}(\omega t)\cdot\mathbf{q} \\ & + (U_p/2\omega)\cos\xi\sin(2\omega t)]\} ,\end{aligned} \tag{3.13}$$

where $\dot{\boldsymbol{\alpha}} = e\,\mathbf{A}/m$ or

$$\boldsymbol{\alpha}(\omega t) = \frac{e}{m\omega^2}\mathbf{E}(\omega t) . \tag{3.14}$$

Substitution of (3.12) and (3.13) into (3.1) with $V_f = V(r)$ yields, after some manipulation,

$$S = -2\pi i\sum_N \delta(\varepsilon_q - N\omega + U_p - W_0 + \tfrac{1}{4}\alpha_s E_0^2)T_N^A , \tag{3.15}$$

with (up to a phase factor)

$$\begin{aligned}T_N^A = {} & \sum_a J_a((U_p - \tfrac{1}{4}\alpha_s E_0^2)\cos(\xi)/2\omega) \\ & \times\int d^3r\, e^{-i\mathbf{q}\cdot\mathbf{r}}V(r)u_0(r)J_{n+2a}(R)e^{i(N+2a)\Omega} ,\end{aligned} \tag{3.16}$$

where

$$\begin{aligned}R = {} & \alpha_0\{[q_x\cos(\tfrac{1}{2}\xi) - m\omega y\sin(\tfrac{1}{2}\xi)]^2 \\ & + [q_y\sin(\tfrac{1}{2}\xi) + m\omega x\cos(\tfrac{1}{2}\xi)]^2\}^{1/2}\end{aligned} \tag{3.17}$$

and

$$\tan\Omega = \frac{[q_y\sin(\tfrac{1}{2}\xi) + m\omega x\cos(\tfrac{1}{2}\xi)]}{[q_x\cos(\tfrac{1}{2}\xi) - m\omega y\sin(\tfrac{1}{2}\xi)]} , \tag{3.18}$$

where $\alpha_0 = eE_0/m\omega^2$. Notice that both R and Ω are dependent on $\mathbf{r}$, the coordinate of integration, which makes the numerical evaluation (3.16) somewhat tedious in this form.

Equation (3.16) can be simplified by using

$$J_{2a+N}(R) = \int_{-\pi}^{\pi} d\lambda\frac{1}{2\pi}\exp\{i[(N+2a)\lambda - R\sin\lambda]\} . \tag{3.19}$$

When this is substituted into (3.16) the sum can be performed,

$$\sum_a J_a(Y)e^{i2a(\lambda+\Omega)} = e^{iY\sin[2(\lambda+\Omega)]} . \tag{3.20}$$

Then the substitution $\lambda = \beta - \Omega$ and the use of the periodicity of the resulting integrand yields

$$T_N^a = \int_{-\pi}^{\pi} d\beta\frac{1}{2\pi}\exp(i\{N\beta + Y\sin(2\beta) - \alpha_0[q_x\cos(\tfrac{1}{2}\xi)\sin\beta - q_y\sin(\tfrac{1}{2}\xi)\cos\beta]\})\int d^3r\, e^{-i\mathbf{Q}\cdot\mathbf{r}}V(r)u_0(r) , \tag{3.21}$$

where

$$\begin{aligned}Y &= \frac{1}{2\omega}(U_p - \tfrac{1}{4}\alpha_s E_0^2)\cos\xi , \\ \mathbf{Q} &= \mathbf{q} - m\omega\alpha_0[\hat{\mathbf{y}}\sin(\tfrac{1}{2}\xi)\sin\beta + \hat{\mathbf{x}}\cos(\tfrac{1}{2}\xi)\cos\beta] .\end{aligned} \tag{3.22}$$

The spatial integral can be done analytically when an analytic form of $V(r)u_0(r)$ is available. As an example, for the ground state of hydrogen we get

$$\begin{aligned}T_N^A = {} & -4\sqrt{\pi}e^2 a_0^{-3/2}\int_{-\pi}^{\pi} d\beta\frac{1}{2\pi}\exp(i\{N\beta + Y\sin(2\beta) - \alpha_0 q(N)\sin\theta[\sin\beta\cos(\tfrac{1}{2}\xi)\cos\phi - \cos\beta\sin(\tfrac{1}{2}\xi)\sin\phi]\}) \\ & \times(Q^2 + a_0^{-2})^{-1} ,\end{aligned} \tag{3.23}$$

where $q(N)$ is obtained from the δ function in (3.15) and (θ,ϕ) are the usual spherical angular coordinates of the $\mathbf{q}$ vector. The laser propagation direction is the $\hat{\mathbf{z}}$ axis and the polarization (3.11) fixes the axes in the perpendicular direction. From (3.22)

$$Q^2 = q^2(N) + \tfrac{1}{2}(m\omega\alpha_0)^2[1 + \cos\xi\cos(2\beta)] - 2m\omega\alpha_0 q(N)\sin\theta[\cos(\tfrac{1}{2}\xi)\cos\phi\cos\beta + \sin(\tfrac{1}{2}\xi)\sin\phi\sin\beta] . \tag{3.24}$$

Note that for arbitrary polarization the transformation $\phi \rightarrow -\phi$ (i.e., $q_y \rightarrow -q_y$) does not leave T_N^A unchanged but that this transformation, accompanied by the reversal of the laser polarization ($\xi \rightarrow -\xi$) does. This is not inconsistent with existing experiment[2] and may be consistent with more recent work.[11] The ϕ dependence is not present in (2.9). For circular polarization, $\xi = \pi/2$, the ϕ dependence of T_N^A disappears.

The δ function in (3.15) yields the dependence of the electron energy upon the dynamic Stark shift of the ground state, a fact predicted by a simple model calculation[11] or by a general analysis applicable to any atom.[12] A return to $\xi = 0$, linear polarization, permits a comparison with the Reiss result.[5] One difference in the results is the absence of the ground-state energy shift in the previous result. Another is the presence of this shift in the first Bessel function in (3.16) and not in (2.9). These are numerically small for a typical experiment: For Xe,[14] $\alpha_s \approx 30 a_0^3$ so that for a yttrium aluminum garnet (YAG) laser

$$\tfrac{1}{4}\alpha_s E^2 / U_p = \tfrac{1}{4}\frac{\alpha_s}{a_0^3}\left[\frac{\hbar\omega}{\mathcal{R}}\right]^2 \simeq \tfrac{1}{10} ,$$

so the numerical difference is small. Finally the $\mathbf{r}$ dependence in R and Ω is absent in (2.9). Its general importance is measured by the ratio

$$m\omega r / q = (\mathcal{R}/\varepsilon_q)^{1/2} (\hbar\omega/\mathcal{R})(r/2a_0) .$$

This can be small for the higher ATI peaks where $\varepsilon_q \sim \mathcal{R}$ but could be significant for the lowest peaks.

We return to the problem discussed in Sec. II. How does the method of this section respond to the change of the Hamiltonian of which the deletion of the A^2 term is a prototype? The removal of A^2 from H and H_f simply removes the two terms containing U_p in (3.13). Therefore

$$\bar{\chi} = \chi_f \exp\left[i \int^t dt' \frac{e^2}{2m} A^2(\omega t') \right] . \tag{3.25}$$

The only other change in the S matrix results from the change in (3.12) since $\bar{\psi}_i^{(+)}$ no longer satisfies (3.2) but instead

$$\left[i\frac{\partial}{\partial t} - \bar{H} \right] \psi_i^{(+)} = 0 , \tag{3.2$'$}$$

where $\bar{H}$ is given by (2.12). The effect of the removal of the A^2 term from H shows up in (3.7) by changing it to

$$\left[W_n(\omega t) - \left[\frac{p^2}{2m} + V(r) + e\mathbf{r}\cdot\mathbf{E}(\omega t) - \frac{e^2}{2m} A^2(\omega t) \right] \right] \Phi_n(\omega t) = 0 . \tag{3.7$'$}$$

This results in relation between $\psi_i^{(+)}$ and $\bar{\psi}_i^{(+)}$ which is

$$\bar{\psi}_i^{(+)} = \psi_i^{(+)} \exp\left[i \int^t dt' \frac{e^2}{2m} A^2(\omega t') \right] , \tag{3.26}$$

which when substituted into the S matrix along with (3.25) exactly restores the original form, (3.1), since the phase factors in (3.25) and (3.26) cancel. Evidently this proof would be repeated with a much more general transformation.

The fact that H and $\bar{H}$ yield the same results for the T matrix when the approximation scheme presented in this section is applied and not in the KFR theory stems from the fact that a *dressed* energy of the ground state is used here, whereas the bare energy appears in (2.3) and (2.8). The difference between H and $\bar{H}$ provides a shift in energies but not a shift in the *difference* of dressed energies. It is the fact that (2.8) does not contain a difference of dressed energies that leads to the difficulties ascribed to the KFR theories presented in Sec. II.

The glaring flaw in all these S-matrix theories is the omission of intermediate resonances which for much of the (I,ω) parameter space seems to be[7] very important. It was, however, important to describe the nonresonant result correctly before proceeding to the next problem.

ACKNOWLEDGMENTS

This research was supported by a grant from the National Science Foundation and by City University of New York Grant No. 664268.

[1] P. Kruit, J. Kimman, H. G. Muller, and M. J. Van der Wiel, Phys. Rev. A **28**, 248 (1983).

[2] P. H. Bucksbaum, M. Bashkansky, R. R. Freeman, T. J. McIrath, and L. F. DiMauro, Phys. Rev. Lett. **56**, 2590 (1986).

[3] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys.—JETP **20**, 1307 (1965)].

[4] F. H. M. Faisal, J. Phys. B **6**, L89 (1973).

[5] H. R. Reiss, Phys. Rev. A **22**, 1786 (1980); J. Phys. B **20**, L79 (1987).

[6] M. D. Perry, O. L. Landen, A. Szoke, and E. M. Campbell, Phys. Rev. A **37**, 747 (1988).

[7] R. Freeman and L. Van Woerkom (private communication).

[8] D. M. Volkov, Z Phys. **94**, 250 (1935).

[9] I. J. Berson, J. Phys. B **8**, 3078 (1975).

[10] M. H. Mittleman, Phys. Rev. A **24**, 2245 (1984).

[11] M. Bashkansky, P. Bucksbaum, and D. W. Schumacher, Phys. Rev. Lett. **60**, 2458 (1988).

[12] See, for example, M. L. Goldberger and K. M. Watson, *Collision Theory* (Wiley, New York, 1965).

[13] This low-frequency approximation to the wave function is essentially the form proposed by H. Reiss, Phys. Rev. A **1**, 803 (1970) and Phys. Rev. A **23**, 3019 (1981) for subsequent references to the "momentum translation approximation." The idea has also been used in the scattering context by N. M. Kroll and K. M. Watson, Phys. Rev. A **8**, 804 (1973) as well as by many others.

[14] T. M. Miller and B. Bederson, Adv. At. Mol. Phys. **13**, 1 (1977).