Atomic transitions in ultrastrong laser fields*

Joel Gersten and Marvin H. Mittleman
Department of Physics, The City College of the City University of New York, New York, New York, 10031
(Received 16 November 1973)

We have calculated the ionization rate of hydrogen by an intense electromagnetic field in the dipole approximation and in an approximation in which this field dominates the electron-proton interaction in intermediate states. We find the transition rate behaves as E^{-1} for linearly polarized fields and $E^{-2}\ln E$ for circularly polarized fields where E is the amplitude of the electric field. We have also calculated the transition rate for excitation of the $2s$ state and a Raman photon. The result behaves as $(E^{-2}W)$ for small W where W is the energy of the Raman photon. We estimate that the method is a good one for electric field strengths of the order of 2×10^{11} V/cm or higher. This is larger than available currently.

I. INTRODUCTION

Modern laser technology[1] makes the observation of multiphoton-induced atomic transitions possible. The attempts to describe these transitions theoretically may be divided into two classes. The first, higher-order perturbation theory,[2] which expands in the atom-laser coupling Hamiltonian is tedious and not applicable to very-high-intensity lasers. The second, epitomized by the work of Reiss,[3] is an attempt to include the laser to all orders in some way, and then expand in the remaining atom-laser interaction.

The convergence of this procedure is not fully understood, and there have been doubts cast upon its accuracy and basic validity. In this work, we investigate another regime in which the interaction of an electron with the laser field dominates the interaction of the electron with other matter. We shall illustrate with the simplest example of a target hydrogen atom, and make the dipole approximation for the laser field. This is an excellent approximation with any currently available laser.

In Sec. II we briefly describe the method and discuss the range of parameters needed for the applicability of the technique. We find that it is not much beyond present day lasers.

In Sec. III we discuss the ionization rate for both linearly and circularly polarized lasers, and find that they behave as E^{-1}, where E is the electric field intensity. This indicates that there is a value of E at which the rate maximizes, since the rate is known to rise for small E. A brief description of this work has been published earlier.[4]

We also discuss the distribution of angular momentum of the final electron for linearly polarized light and show that, for strong fields and many photons, the excitation process acts as a superposition of angular momentum raising and lowering processes. This gives a random distribution in angular momentum (L) proportional to $\{(2L+1)^{1/2} \times (L)!/2^L[(\frac{1}{2}L)!]^2$.

In Sec. IV we obtain the excitation rate to a final bound state with emission of a single Raman photon to satisfy the energy conservation requirement. The $2s$ state of hydrogen is used as an example.[3]

II. METHOD

Our starting point is the Hamiltonian describing a hydrogen atom in its center-of-mass frame interacting with an intense electromagnetic field treated semiclassically in the dipole approximation.[5]

$$H=P^2/2m+V(r)-(e/m)\vec{A}(t)\cdot\vec{P}, \tag{2.1}$$

where the term $\frac{1}{2}[e^2A^2(t)/2m]$ has been discarded by a contact transformation. The total wave function satisfies

$$\left(i\frac{\partial}{\partial t}-H\right)\psi_i^{(+)}(r,t)=0 \tag{2.2}$$

where the subscript index specifies the initial condition. We may also define a Green's function G, with the operator of Eq. (2.2) in terms of which the solution to (2.2) is

$$\psi_i^{(+)}=\phi_i+GH'\phi_i, \tag{2.3}$$

where

$$H'=(-e/m)\vec{A}(t)\cdot\vec{P}. \tag{2.4}$$

Here ϕ_i is the initial condition for ψ_i, and G is defined to vanish at this initial time. For the linearly polarized case, we take $A(t)$ to be

$$\vec{A}(t)=(\vec{E}/\omega)\sin\omega t, \tag{2.5}$$

where $\vec{E}$ is the electric field strength; note that for convenience the initial time may be made to recede to $-\infty$ by inserting a switch-on–switch-off convergence factor into Eq. (2.5).

The transition amplitude into a final state $\phi_q^{(-)}$ representing an unbound electron in the field of the proton can then be exactly written as

$$\tau_{q0} = \langle \phi_q^{(-)}, H'\phi_0 \rangle + \langle \phi_q^{(-)}, H'G^{(+)}H\phi_0 \rangle , \qquad (2.6)$$

where we have specialized to an initial condition of a ground-state hydrogen atom, ϕ_0. The brackets in Eq. (2.6) are meant to describe a time integration in addition to the usual spatial inner product. The first term of Eq. (2.6) is easily shown to describe single photon absorption, which is impossible for sufficiently low-energy photons. We therefore discard it and subsequent single photon terms. In the remaining term we make our basic assumption, which is to neglect the Coulomb potential $V(r) = -e^2/r$ in the Green's function. The remaining Green's function is then analytically obtainable, and the calculation can proceed. We shall continue this in Sec. III and turn to the validity of the approximation here.

The essential approximation is that the electron is dominated by the laser field in the intermediate state, rather than by its interaction with the proton. This requires that

$$H' \gg V \sim R_\infty . \qquad (2.7)$$

We may conveniently estimate the magnitude of H' by treating it classically:

$$H' = (e/m)(\vec{E}\cdot\vec{P}/\omega)\sin\omega t = -[(e\vec{E}\cdot\vec{v}/\omega]\sin\omega t . \qquad (2.8)$$

The velocity of a driven oscillator is

$$v \simeq (e/m)[E\omega/(\omega_r^2 - \omega^2)]\sin\omega t + v_{H(t)} , \qquad (2.9)$$

where ω_r is the frequency of the oscillator and v_H is a homogeneous vibration. Note that the forced part of v is in phase with the $A(t)$ occurring in H'. In order for the inequality (2.7) to be satisfied, we take the forced part of v to dominate the homogeneous part, which we estimate as a typical atomic velocity $\sim e^2/\hbar$. This inequality is

$$(e/m)[E\omega/(\omega_r^2 - \omega^2)] \gg e^2/\hbar . \qquad (2.10)$$

The laser frequency is taken to be small compared to the oscillator frequency, which we take as the atomic frequency

$$\omega_r \sim R_\infty/\hbar . \qquad (2.11)$$

Then Eq. (2.10) can be rewritten

$$\bar{E}\bar{\omega} \gg 1 , \qquad (2.12)$$

where $\bar{\omega}$ and $\bar{E}$ are measured in natural units

$$\bar{\omega} = \omega\hbar/R_\infty , \quad \bar{E} = E/(e/2a_0^2) . \qquad (2.13)$$

The inequality (2.7) then may be written

$$\bar{E} \gg 1, \qquad (2.14)$$

which is less stringent than Eq. (2.12), since $\bar{\omega} \ll 1$. The essential restriction required for the method to work is then[6]

$$\bar{E}\bar{\omega} \gg 1 , \qquad (2.12)$$

which for a CO_2 laser ($\bar{\omega} \propto 10^{-2}$) requires field strengths of

$$E \gg 23\times10^{10} \text{ V/cm}$$

while Eq. (2.14) requires only

$$E \gg 27\times10^{8} \text{ V/cm} .$$

Objections can be raised to this harmonic oscillator model on the grounds that it is unrealistic, in that the force increases (in magnitude) indefinitely with distance from the center. For the large amplitude oscillations contemplated here, this may be particularly important. If we remedy this by cutting off the restoring force of the harmonic oscillator at some distance (say, a_0), then we get the inequality $\bar{E}/\bar{\omega} \gg 1$, which is a much less stringent requirement than either Eq. (2.12) or (2.14), and which leads to $E \gg 3\times10^7$ V/cm. Evidently, all these estimates are subject to question, and the only conclusive way of answering the questions posed here is to continue the perturbation series of which we have retained only the first term. However, we emphasize here that the first term of the method outlined below Eq. (2.6) is *exact* in the limit $E \to \infty$.

III. IONIZATION

We now continue from Eq. (2.6). The Green's function, with the approximation discussed in Sec. II, may be written

$$iG^{(+)}(rt, \vec{r}'t') = \Theta(t-t')\int \frac{d^3k}{(2\pi)^3} \times \exp\Big(-i\frac{k^2}{2m}(t-t') + i\vec{k}\cdot(\vec{r}-\vec{r}') + i\vec{k}\cdot[\vec{a}(t)-\vec{a}(t')] - \eta(t-t')\Big), \qquad (3.1)$$

where

$$\vec{a}(t) = (e/m)\int_{-\infty}^{t} dt'\,\vec{A}(t')$$

and η is the usual prescription for encircling a pole. Insertion of Eq. (3.1) into the remaining term of Eq. (2.6) yields, after the spatial integrals are performed,

$$\tau_{q_1 0} = \int \frac{d^3k}{(2\pi)^3}\int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt'\, e^{i(W_q t - W_0 t')} \times e^{-i(k^2/2m)(t-t')}\vec{k}\cdot\dot{\vec{a}}(t)\vec{k}\cdot\dot{\vec{a}}(t') \times e^{i\vec{k}\cdot[\vec{a}(t)-\vec{a}(t')]}\, u_q^{(-)*}(k)u_0(k)e^{-\eta(t-t')} , \qquad (3.2)$$

where the W's are the indicated eigenvalues and the u's are the Fourier transform defined by

$$u_j(k)=\int d^3r\, e^{-i\vec{k}\cdot\vec{r}}\phi_j(r)\ , \tag{3.3}$$

where the ϕ_j are the hydrogenic states in question. Further calculation is facilitated by the identity[7]

$$e^{i\lambda\cos\phi}=\sum_{n=-\infty}^{\infty} i^n J_n(\lambda)e^{in\phi} \tag{3.4a}$$

which is used as

$$\vec{k}\cdot\dot{\vec{a}}(t)\vec{k}\cdot\dot{\vec{a}}(t')e^{i\vec{k}\cdot[\vec{a}(t)-\vec{a}(t')]}=\sum_{n,n'} nn'\omega^2 J_n(\vec{k}\cdot\vec{\epsilon})J_{n'}(\vec{k}\cdot\vec{\epsilon})i^{-n+n'}e^{i\omega(nt-n't')}\ , \tag{3.4b}$$

where

$$\vec{\epsilon}=(e/m\omega^2)\vec{E}\ . \tag{3.5}$$

The time integrations may now be performed:

$$\tau_{q,0}=2\pi i\sum_{nn'=-\infty}^{\infty}\delta(W_q-W_0+n\omega-n'\omega)i^{n'-n}nn'\omega^2 \times\int\frac{d^3k}{(2\pi)^3}\frac{u_q^{(-)*}(k)u_0(k)J_n(\vec{k}\cdot\vec{\epsilon})J_{n'}(\vec{k}\cdot\vec{\epsilon})}{W_0-k^2/2m+n'\omega+i\eta}\ . \tag{3.6}$$

In order to identify the photon number l we make the transformation $n=n'-l$ and rewrite Eq. (3.6) as

$$\tau_{q,0}=-2\pi i\sum_{l=-\infty}^{\infty}\delta(W_q-W_0-l\omega)T_{q,0}(l) \tag{3.7}$$

so that

$$T_{q,0}(l)=-i^l\omega^2\sum_{n=-\infty}^{\infty}n(n-l)\int\frac{d^3k}{(2\pi)^3} \times\frac{u_q^{(-)*}(k)u_0(k)J_{n-l}(\vec{k}\cdot\vec{\epsilon})J_n(\vec{k}\cdot\vec{\epsilon})}{W_0-k^2/2m+n\omega+i\eta}\ . \tag{3.8}$$

It is now convenient to use the identity

$$\frac{n(n-l)}{\epsilon_0+n\omega}=\frac{1}{\omega}\left(n-l-\frac{\epsilon_0}{\omega}+\frac{\epsilon_0}{\omega}\frac{\epsilon_0+\omega l}{\epsilon_0+\omega n}\right)\ , \tag{3.9}$$

where

$$\epsilon_0=W_0-k^2/2m+i\eta\ .$$

The identities[7]

$$\sum_n J_{n-l}(x)J_n(x)=\delta_{l,0}\ ,\qquad \sum_n nJ_{n-l}(x)J_n(x)=\tfrac{1}{2}x(\delta_{l,1}+\delta_{l,-1}) \tag{3.10}$$

allow us to drop the first three terms in Eq. (3.9) when substituted into Eq. (3.8), since they do not contribute to multiphoton transitions. The remaining expression for $T_{q,0}$ is

$$T_{q,0}(l)=-i^l\sum_{n=-\infty}^{\infty}\int\frac{d^3k}{(2\pi)^3}u_q^{(-)*}(k)u_0(k) \times J_{n-l}(\vec{k}\cdot\vec{\epsilon})J_n(\vec{k}\cdot\vec{\epsilon})\frac{\epsilon_0(\epsilon_0+\omega l)}{\epsilon_0+\omega n}\ . \tag{3.11}$$

Now $u_q^{(-)}$ is the transform of the final state of the electron which to lowest order in the Coulomb interaction is a plane wave. We have been expanding in this interaction, so we retain this leading term only, so that

$$u_q^{(-)}(k)=(2\pi)^3\delta(\vec{k}-\vec{q})\ . \tag{3.12}$$

Then the last factor in Eq. (3.11) becomes

$$\epsilon_0+\omega l=W_0-k^2/2m+\omega l =W_0-q^2/2m+\omega l=W_0-W_q+\omega l\ ,$$

which vanishes because of the energy-conservation δ function in Eq. (3.7). Then the only term of Eq. (3.11) which survives is the $n=l$ term where the denominator cancels this zero. The result is

$$T_{q,0}(l)=(-i)^l u_0(q)J_0(\vec{q}\cdot\vec{\epsilon})J_l(\vec{q}\cdot\vec{\epsilon})l\omega\ . \tag{3.13}$$

It has been shown[3] that transitions with different l's are incoherent, so the transition rate for a given photon multiplicity is given by

$$W_l=2\pi\int\frac{d^3q}{(2\pi)^3}\delta(W_q-W_0-l\omega)\,|T_{q,0}(l)|^2\ . \tag{3.14}$$

Then using the ground state of hydrogen

$$u_0(q)=8\pi^{1/2}a_0^{-5/2}/[q^2+a_0^{-2}]^2\ , \tag{3.15}$$

where a_0 is the Bohr radius, the result may be written

$$\overline{W}_l\equiv\frac{W_l}{R_\infty}=\frac{16}{\overline{\epsilon}l^2}\int_0^{\lambda_l}dx\,J_0^2(x)J_l^2(x), \tag{3.16}$$

where

$$\lambda_l=(2\overline{\epsilon}/\overline{\omega}^2)(l\overline{\omega}-1)^{1/2}\ . \tag{3.17}$$

This remaining integral is essentially the integral over the angle between $\vec{k}$ and $\vec{\epsilon}$.

When the inequality (2.12) is satisfied, λ_l is large except for l's very near threshold ($l\sim\overline{\omega}$). For large λ_l

$$\overline{W}_l=(16/\overline{\epsilon})(N_l/l^2)\ , \tag{3.18a}$$

where

$$N_l=\int_0^\infty dx\,J_0^2(x)J_l^2(x)\ . \tag{3.18b}$$

We see from this the $\overline{\epsilon}^{-1}$ behavior for large fields. Since $\overline{W}_l$ is known to be a rising function of

$\bar{\epsilon}$ for small fields ($\sim\bar{\epsilon}^{2l}$), this shows that there is a field for which $\overline{W}_l$ is a maximum.[3] Equation (3.16) also gives an $(\bar{\epsilon})^{2l}$ behavior for $\overline{W}_l$, but the coefficient is probably incorrect because the criterion for the validity of the method may not be satisfied. We may, however, use Eq. (3.16) to estimate the field at which the transition rate is a maximum. It is obtained from

$$\lambda_l J_0^2(\lambda_l)J_l^2(\lambda_l)=\int_0^{\lambda_l} dx\, J_0^2(x)J_l^2(x) \ . \qquad (3.19)$$

This may be solved with sufficient accuracy by approximating the Bessel functions by the first two terms of their power series with the result

$$\lambda_l \simeq \left(\frac{2l}{l+2}\,\frac{2l+3}{2l+1}\right)^{1/2} \ . \qquad (3.20)$$

The use of this theory for values of $\lambda\sim\sqrt{2}$ is no doubt incorrect, but the validity can really only be tested by theories which are known to be more accurate in this range.

For large photon multiplicity, λ_l again becomes large ($\lambda_l\sim\sqrt{l}$), but not as fast as the order of the Bessel function in Eq. (3.16). We must therefore use the asymptotic form[7] for $l>x$,

$$J_l(x)\to\frac{e^l}{(2\pi l)^{1/2}}\left(\frac{x}{2l}\right)^l e^{-x^2/4l}[1+O(l)+O(x^2/l^3)]. \qquad (3.21)$$

Then Eq. (3.17) becomes

$$\overline{W}_l=\frac{16}{\bar{\epsilon}l^2}\int_0^{a\sqrt{l}} dx\, J_0^2(x)\left(\frac{x}{2l}\right)^{2l} e^{-x^2/2l}\,\frac{e^{2l}}{2\pi l}, \qquad (3.22)$$

where

$$a=2\bar{\epsilon}\,\bar{\omega}^{-3/2}.$$

The substitution $x=(2ly)^{1/2}$ changes this to

$$\overline{W}_l=\frac{4\sqrt{2}}{\pi\bar{\epsilon}}\,l^{-5/2}\left(\frac{e^2}{2l}\right)^l\int_0^{a^2/2} dy\, J_0^2((2ly)^{1/2})y^{l-1/2}e^{-y}. \qquad (3.23)$$

Then J_0 may be replaced by its asymptotic form[7]

$$J_0^2((2ly)^{1/2})=(1/\pi)(2ly)^{-1/2}[1+\sin(8ly)^{1/2}]. \qquad (3.24)$$

The sine term oscillates rapidly and may be dropped, so that we get

$$\overline{W}_l=\frac{4}{\pi^2\bar{\epsilon}l^3}\left(\frac{e^2}{2l}\right)^l\int_0^{a^2/2} dy\, y^{l-1}e^{-y} \ . \qquad (3.25)$$

Evaluation for large l yields

$$\overline{W}_l=\frac{4}{\pi^2\bar{\epsilon}l^4}\left(\frac{\bar{\epsilon}^2}{\bar{\omega}^3}\,\frac{e^2}{l}\right)^l e^{-2\bar{\epsilon}^2/\bar{\omega}^3}, \qquad (3.26)$$

which is an extremely rapid falloff with l. The result is valid for $l^{1/2}\gg\bar{\epsilon}/\bar{\omega}^{3/2}$.

We may also investigate the angular momentum distribution of the final electron by returning to the expression for the T matrix, Eq. (3.11), and retaining only the $n=l$ term as above. Dropping phases, this is

$$T_{q,0}(l)=\int\frac{d^2k}{(2\pi)^3}\,u_q^{(-)*}(k)u_0(k) \times J_0(\vec{k}\cdot\vec{\epsilon})J_l(\vec{k}\cdot\vec{\epsilon})\left(W_0-\frac{k^2}{2m}\right) \ . \qquad (3.27)$$

The relevant angular momentum part of $u_q^{(-)}(k)$ is

$$[u_q^{(-)}(k)]_L=[\tfrac{1}{2}(2L+1)]^{1/2}P_L(\hat{q}\cdot\hat{\epsilon})P_L(\hat{k}\cdot\hat{\epsilon}) \ . \qquad (3.28)$$

Then the L dependence of the T matrix can be obtained from

$$I_L=[\tfrac{1}{2}(2L+1)]^{1/2}\int_{-1}^{+1} d\mu\, P_L(\mu)J_0(\beta\mu)J_l(\beta\mu), \qquad (3.29)$$

where

$$\mu=\hat{k}\cdot\hat{\epsilon}$$

and

$$\beta=2\bar{\epsilon}q/\bar{\omega}^2\gg1 \ .$$

The first observation one can make from Eq. (3.29) is that $l+L$ must be even. Since β is large, it is useful to write I_l as

$$I_L=[2(2L+1)]^{1/2}\int_0^{\beta} dx\,P_L\left(\frac{x}{\beta}\right)J_0(x)J_l(x) \qquad (3.30)$$

and to extract the leading terms in β

$$I_L=\frac{1}{\beta}[2(2L+1)]^{1/2}\times\left[\int_0^1 dx\,P_L\left(\frac{x}{\beta}\right)J_0(x)J_l(x)+\int_1^{\beta} dx\,P_L\left(\frac{x}{\beta}\right)[J_0(x)J_l(x)-J_0^A(x)J_l^A(x)]+\int_1^{\beta} dx\,P_L\left(\frac{x}{\beta}\right)J_0^A(x)J_l^A(x)\right], \qquad (3.31)$$

where the subscript A on J_0, J_l indicates that the asymptotic form for large x is to be used.

The integrals must be treated separately for the cases L, even or odd. For even L, the first integral is finite for $\beta\to\infty$, and the second vanishes in this limit. For L odd the first integral vanishes as β^{-1}, and the second is finite. The remaining integral is

$$\int_1^{\beta} dx\,P_L\left(\frac{x}{\beta}\right)\frac{2}{\pi x}\cos(x-\tfrac{1}{4}\pi)\cos(x-\tfrac{1}{4}\pi-\tfrac{1}{2}l\pi) =\frac{1}{\pi}\int_{\beta-1}^{1}\frac{d\mu}{\mu}\,P_L(\mu)[\cos\tfrac{1}{2}l\pi+\sin(2k\mu-\tfrac{1}{2}l\pi)] \ .$$

For L (and l) even, the first term in the bracket dominates, and the result for large β is

$$[(-)^{l/2}/\pi]\,P_L(0)\ln\beta\,.$$

For L (and l) odd, the integral vanishes for large β. Therefore, the dominant behavior comes from even values of L where

$$I_L \sim \frac{(-)^{l/2}}{\pi}\,\frac{1}{\beta}\ln\beta[2(2L+1)]^{1/2}P_L(0) = \frac{(-)^{l+L/2}}{\pi}\,\frac{[2(2L+1)]^{1/2}\ln\beta}{\beta}\,\frac{L!}{2^L[(\frac{1}{2}L)!]^2}\,. \tag{3.32}$$

The above calculation can be repeated for circularly polarized radiation, in which case

$$\vec{A}(t) = (\epsilon/\omega)[\hat{a}_x\cos\omega t + \hat{a}_y\sin\omega t] \tag{3.33}$$

and Eq. (3.4b) is changed to

$$\vec{k}\cdot\dot{\vec{a}}(t)\vec{k}\cdot\dot{\vec{a}}(t')\exp\{i\vec{k}\cdot[\vec{a}(t)-\vec{a}(t')]\} = \sum_{nn'} nn'\omega^2 J_n(k\epsilon\sin\theta)J_{n'}(k\epsilon\sin\theta)i^{\,l} \times\exp[-i(n-n')\phi + i(n\omega t - n'\omega t')]\ , \tag{3.34}$$

where θ and ϕ are, respectively, the polar and azimuthal angles of $\vec{k}$ with respect to the axis defined along the direction of propagation of the laser, and to an arbitrary χ axis perpendicular to this direction. The remaining operations are identical with those described for the previous calculation. In particular, replacement of $u_q^{(-)}(k)$ by a δ function [Eq. (3.12)] makes the ϕ dependence of Eq. (3.34) an over-all phase factor in the T matrix. Therefore, the only difference in the two results is contained in the arguments of the Bessel functions. The final result is

$$W_l = \frac{32(l\bar{\omega}-1)^{1/2}}{(l\bar{\omega})^2}\int_0^1 \frac{d\mu\,\mu}{(1-\mu^2)^{1/2}}\,J_0^2(\lambda_l\mu)J_l^2(\lambda_l\mu)\,. \tag{3.35}$$

For large λ_l, a method similar to that used for Eq. (3.29) yields

$$W_l = \frac{32}{\pi^2}\,\frac{(l\bar{\omega}-1)^{1/2}}{(l\bar{\omega})^2}(\tfrac{1}{2}+\cos^2\tfrac{1}{2}l\pi)\frac{1}{\lambda_l^2}\,\ln\tfrac{1}{2}\lambda_l = \frac{8}{\pi^2}(1+\tfrac{1}{2}\cos l\pi)\,\frac{\bar{\omega}^2}{l^2(l\bar{\omega}-1)^{1/2}}\,\frac{1}{\bar{\epsilon}^2} \times\ln\left(\frac{\bar{\epsilon}}{\bar{\omega}^2}(l\bar{\omega}-1)^{1/2}\right)\,. \tag{3.36}$$

Note that this is smaller than the analogous result (3.17) for linear polarization by a field-dependent factor of $(1/\bar{\epsilon})\ln\bar{\epsilon}$. Presumably, this can be explained from angular momentum considerations. Absorption of each linearly polarized photon can either raise or lower the electron's angular momentum by unity, yielding a statistical distribution of angular momentum for the final electron, (3.32). On the other hand, absorption of a circularly polarized photon must raise the angular momentum by unity each time, so that the final electron will have a very high angular momentum. The high-angular-momentum components of the final wave function have small amplitudes in the vicinity of the atom, which reduces the matrix element and yields a smaller transition rate.

We should also point out the different results for even and odd values of the photon multiplicity. This has been previously noted by Reiss.[8]

IV. BOUND-BOUND TRANSITIONS

We shall use a method similar to that used above to calculate the transition rate to the $2s$ state with emission of a photon W, as required by energy conservation. We treat the radiation operator for this photon, H_r, in lowest-order perturbation theory. The transition amplitude in this approximation is

$$\tau_{fi} = \langle\psi_f^{(-)}, H_r\psi_i^{(+)}\rangle \simeq \langle\phi_f\,,(1+H'G)H_r(1+GH')\phi_i\rangle, \tag{4.1}$$

where G is now taken to be Eq. (3.1). The emission operator for a photon of energy W, and for polarization λ in the dipole approximation is

$$H_r = (e\hbar/m)(2\pi/WV)^{1/2}\,\vec{\epsilon}_\lambda\cdot\vec{p}e^{iWt}, \tag{4.2}$$

where $\vec{\epsilon}_\lambda$ is the polarization vector and V is the quantization volume of this field. The calculation will be done for linear polarizations only, since the angular momentum considerations described above preclude multiphoton transitions of this kind for circularly polarized lasers.

Straightforward substitution of G [Eq. (3.1)] into Eq. (4.1), and performance of the spatial integrals, yields

$$\tau_{fi} = i\gamma\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\int\frac{d^3k}{(2\pi)^3}\,u_f^*(k)\left(W_f-\frac{k^2}{2m}\right) \times\vec{\epsilon}_\lambda\cdot\vec{k}\left(W_i-\frac{k^2}{2m}\right)u_i(k)e^{iWt'}\exp i\left(W_f-\frac{k^2}{2m}\right)t - i\left(W_i-\frac{k^2}{2m}\right)t'' + i\vec{k}\cdot[\vec{a}(t)-\vec{a}(t')] - \eta(t-t''), \tag{4.3}$$

where

$$\gamma = (e\hbar/m)(2\pi/WV)^{1/2}.$$

We again use the Bessel-function expansion theorem (3.4) and perform the time integrations with the result

$$\tau_{fi}=-2\pi i\sum_{n,n'=-\infty}^{\infty}\delta(W_f+W-W_i+(n-n')\omega)i^{n-n'}\int\frac{d^3k}{(2\pi)^3}\frac{u_f^*(k)u_i(k)J_n(\vec{k}\cdot\vec{\epsilon})J_{n'}(\vec{k}\cdot\vec{\epsilon})(W_f-k^2/2m)(W_i-k^2/2m)\gamma\vec{\epsilon}_\lambda\cdot\vec{k}}{(W_i-k^2/2m+n'\omega+i\eta)(W_f-k^2/2m+n'\omega+i\eta)} \tag{4.4}$$

The substitution $n'=n-l$ allows us to write

$$\tau_{fi}=-2\pi i\sum_{l=-\infty}^{\infty}\delta(W_f+W-W_i-l\omega)\,T_{fi}(l) \tag{4.5}$$

and interpret $T_{fi}(l)$ as the l-photon transition matrix. We obtain

$$T_{fi}(l)=-(-i)^l\sum_{n=-\infty}^{\infty}n(n-l)\int\frac{d^3k}{(2\pi)^3}u_f^*(k)u_i(k)J_n(\vec{k}\cdot\vec{\epsilon})J_{n-l}(\vec{k}\cdot\vec{\epsilon})$$
$$\times\frac{(W_f-k^2/2m)(W_i-k^2/2m)\gamma\vec{\epsilon}_\lambda\cdot\vec{k}}{(W_i-k^2/2m+n\omega+i\eta)[W_f-k^2/2m+(n-l)\omega+i\eta]}, \tag{4.6}$$

where the symbols have the same meaning as in Sec. III.

Instead of attempting the formidable task of evaluating Eq. (4.6) in general, we again restrict ourselves to the situation of very large $\vec{\epsilon}$. By symmetry, we replace $\vec{\epsilon}_\lambda\cdot\vec{k}$ by $\vec{\epsilon}_\lambda\cdot\hat{\vec{\epsilon}}\,\vec{k}\cdot\hat{\vec{\epsilon}}$ and perform the angular part of the $\vec{k}$ integral

$$\int d\hat{k}\,\vec{k}\cdot\hat{\vec{\epsilon}}\,J_n(\vec{k}\cdot\vec{\epsilon})J_{n-l}(\vec{k}\cdot\vec{\epsilon})=2\pi k\int_{-1}^{+1}d\mu\,\mu\,J_n(k\epsilon\mu)J_{n-l}(k\epsilon\mu),$$

which vanishes for even values of l. This selection rule is just a consequence of the fact that the initial and final states chosen here have the same parity. We now insert the explicit forms of the u's in Eq. (4.6); u_i is given by Eq. (3.15) and

$$u_f=u_{2s}=(8\pi)^{1/2}a_0^{-5/2}[(k^2-1/4a_0^2)/(k^2+1/4a_0^2)^3]. \tag{4.7}$$

If we now scale k by a_0^{-1} and measure all energies in Rydbergs, the T matrix (omitting an over-all phase) becomes

$$T_{fi}=\frac{\gamma\vec{\epsilon}_\lambda\cdot\hat{\vec{\epsilon}}}{a_0}\frac{4\sqrt{2}}{\pi}\sum_{n=-\infty}^{\infty}n(n-l)\int_0^\infty dk\,k^3$$
$$\times\int_{-1}^{+1}d\mu\,\mu\,J_n(\alpha k\mu)J_{n-l}(\alpha k\mu)\frac{k^2-\frac{1}{4}}{(k^2+\frac{1}{4})^2(k^2+1)(k^2+1-n\overline{\omega}-i\eta)[k^2+\frac{1}{4}-\overline{\omega}(n-l)-i\eta]}, \tag{4.8}$$

where $\overline{W}=W/R_\infty$,

$$\alpha=2\overline{\epsilon}/\overline{\omega}^2, \tag{4.9}$$

and the energy conservation δ function in Eq. (4.5) relates l and $\overline{W}$ by

$$l\overline{\omega}=\tfrac{3}{4}+\overline{W}. \tag{4.10}$$

The integrals and sum in Eq. (4.8) are evaluated in the Appendix for the case $\alpha\gg 1$. For small $\overline{W}$ the result may be written

$$T_{fi}(l)=-i(32\sqrt{2}/9\pi)(\gamma/a_0)(\epsilon_\lambda\cdot\hat{\epsilon}/\alpha\overline{\omega}^2). \tag{4.11}$$

The transition probability per unit time for l-photon absorption can then be obtained from

$$\overline{W}_l=\frac{W_\lambda}{R_\infty}=\frac{2\pi V}{R_\infty}\int\frac{d^3W}{(2\pi\hbar c)^3}$$
$$\times\delta(W_f+W-W_i-l\omega)\sum_\lambda|T_{fi}(l)|^2, \tag{4.12}$$

where the sum is over the two polarization directions of the emitted photon. The result is

$$\overline{W}_l=(2^{11}/3^5\pi^2)(\alpha_F^3/\overline{\epsilon}^2)(l\overline{\omega}-\tfrac{3}{4}), \tag{4.13}$$

where $\alpha_F=\frac{1}{137}$ is the fine-structure constant. Again, the temptation to perform a sum over l must be resisted, since the approximations are not valid for large l.

APPENDIX

We consider the integral and sum in Eq. (4.8):

$$I_l=\sum_{n=-\infty}^{\infty}n(n-l)\int_0^\infty dk\,k^3\int_{-1}^{+1}d\mu\,\mu$$
$$\times J_n(\alpha k\mu)J_{n-l}(\alpha k\mu)\Gamma(k^2), \tag{A1}$$

where

$$\Gamma(k^2)=\frac{k^2-\frac{1}{4}}{(k^2+\frac{1}{4})^2(k^2+1)(k^2+1-n\bar{\omega}-i\eta)(k^2+1-n\bar{\omega}+\overline{W}-i\eta)},\tag{A2}$$

and where Eq. (4.10) has been used to eliminate l in $\Gamma(k^2)$. We first replace the J's by their integral representations

$$J_m(x)=\frac{1}{2\pi}\int_{-\pi}^{\pi}d\theta\, e^{\pm i(m\theta+x\sin\theta)}$$

and then perform the μ integrations. After some rearrangement, I_l can then be written

$$I_l=\left(\frac{\alpha}{2\pi}\right)^2\sum_n\int\int_{-\pi}^{\pi}d\theta\, d\theta' \times\cos\theta\cos\theta' C_n(\beta)e^{i[n\theta-(n-l)\theta']}\tag{A3}$$

where

$$C_n(\beta)=\frac{\partial^2}{\partial\beta^2}\frac{|\beta|}{\beta}\frac{\partial}{\partial|\beta|}B(|\beta|),\tag{A4}$$

where

$$B(|\beta|)=\int_{-\infty}^{\infty}dk\, k\Gamma(k^2)\frac{e^{ik|\beta|}}{|\beta|}\tag{A5}$$

and

$$\beta=\alpha(\sin\theta-\sin\theta').\tag{A6}$$

The range of integration on θ and θ' can be compressed from $-\pi$ to π to 0 to $\frac{1}{2}\pi$ by judicious substitutions with the result

$$I_l=\frac{\alpha^2 i}{\pi^2}\sum_{n=-\infty}^{\infty}\int_0^{\pi/2}d\theta\int_{-\theta}^{\theta}d\theta'\cos\theta\cos\theta' C_n(\beta) \times\{\sin[n\theta-(n-l)\theta']-\sin[n\theta-(n-l)\theta] +(-)^n\sin[n\theta+(n-l)\theta'] -(-)^n\sin[n\theta'+(n-l)\theta]\}.\tag{A7}$$

We now note that $\beta\geqslant 0$ in the integration range. Let $\beta'=\alpha(\sin\theta+\sin\theta')$ and express the integral as over a domain in the $\beta-\beta'$ plane. Then

$$I_l=\frac{i}{\pi^2}\sum_n\int_0^{2\alpha}d\beta\int_0^{2\alpha-\beta}d\beta' C_n(\beta) \times[\sin(n-\tfrac{1}{2}l)(\theta-\theta')\cos\tfrac{1}{2}l(\theta+\theta') +(-)^n\sin\tfrac{1}{2}l(\theta-\theta')\cos(n-\tfrac{1}{2}l)(\theta+\theta')].\tag{A8}$$

In the limit of large α we have, approximately, $\theta=(\beta+\beta')/2\alpha$ and $\theta'=(\beta'-\beta)/2\alpha$. Then I_l reduces to

$$I_l\simeq\frac{i}{\pi^2}\sum_n\int_0^{2\alpha}d\beta\int_0^{2\alpha-\beta}d\beta' C_n(\beta) \times\left((n-\tfrac{1}{2}l)\frac{\beta}{\alpha}+(-)^n\frac{l\beta}{2\alpha}\right).\tag{A9}$$

Integration with the use of Eqs. (A4) and (A5) leads to the form

$$I_l=\frac{-2}{\pi^2\alpha}\sum_{n=-\infty}^{\infty}\{n-\tfrac{1}{2}l[1-(-)^n]\}\int_{-\infty}^{\infty}dk\, k^2\Gamma(k^2).\tag{A10}$$

Evaluation of the sum is straightforward if we replace the sum by an integral,

$$\sum_{n=-\infty}^{\infty}\{n-\tfrac{1}{2}l[1-(-)^n]\} \{(k^2+1-n\bar{\omega}-i\eta)[k^2+\tfrac{1}{4}-(n-l)\bar{\omega}-i\eta]\}^{-1}=-\frac{i\pi}{\bar{\omega}^2}.\tag{A11}$$

The remaining k integration is then straightforward

$$I_l=\frac{2i}{\pi\alpha\bar{\omega}^2}\int_{-\infty}^{\infty}dk\frac{k^2(k^2-\frac{1}{4})}{(k^2+1)(k^2+\frac{1}{4})^2}=\frac{8i}{9\alpha\bar{\omega}^2}.\tag{A12}$$

Thus we obtain a simple expression for the desired integral.

---

*Research sponsored by CUNY Faculty Research Awards No. 10083 and 10215.

[1]R. Alfano, Sci. Amer. 228, 42 (1973).

[2]See for example, Y. Gontier and M. Trahim, Phys. Rev. 172, 83 (1968).

[3]H. R. Reiss, Phys. Rev. D 4, 3533 (1971).

[4]M. H. Mittleman, Phys. Lett. 47A, 55 (1974).

[5]We use units in which $\hbar=c=1$.

[6]We have been quite conservative here. Equation (2.12) comes from the dominance of the first term of Eq. (2.9) over the second term. The homogeneous oscillation of v will have a frequency ω_r which is very different from ω, so that its contribution to H' [Eq. (2.8)] may well be drastically reduced by a time average. In that case, Eq. (2.12) is unnecessary, and the less stringent Eq. (2.14) is the only restriction.

[7]I. Gradsteyn and I. Ryzhik, *Table of Integrals, Series, and Products* (Academic, New York, 1965).

[8]H. R. Reiss (private communication).