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Atomic Transitions in Intense Fields and the Breakdown of Perturbation Calculations

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Results valid for arbitrarily high intensity of electromagnetic field are presented for induced emission from an excited state of an atom. Comparison with perturbation theory shows that perturbative calculations fail quantitatively and qualitatively at lower intensity than had been expected. Distinctive intense-field features are discussed such as vanishing of the transition probability for certain intensities and photon multiplicities, and appearance of two extrema of the transition amplitude for multiphoton processes at very high intensities.

The purposes of this paper are to report the principal novel features of electromagnetic transitions at very high field intensity, to illustrate the nature of the failure of perturbation calculations in semiclassical electrodynamics, and to establish the intensity of the electromagnetic field at which this failure occurs. This investigation is done within the framework of a simple, specific example—emission induced from the $2s$ state of the hydrogen atom by an intense plane-wave electromagnetic field, such that the energy of a single photon from this field is much less than the $2s$-$1s$ energy difference in hydrogen. The reason for this choice of example is that an intense-field method presented earlier[1] is then unrestrictedly applicable, and a simple comparison with perturbation theory is possible.

Equation (56) of Ref. 1 gives the T matrix for emission from the $2s$ level of hydrogen of $2n+1$ photons of energy ω (we set $\hbar=1$) and one photon of energy ω' as

$$T_{2s,1s}^{(2n+1,1)}=\frac{\sqrt{2}}{m a_0^2}\left(\tfrac{2}{3}\right)^3 e^{i\alpha}\vec{\epsilon}\cdot\vec{\epsilon}'\left(\tfrac{1}{3}ea'a_0\right)(-)^n\left(\tfrac{1}{3}eaa_0\right)^{2n+1}(n+1)^2(n+2)$$
$$\times\sum_{k=0}^{\infty}\binom{n+1+k}{n+1}^2\binom{n+2+k}{n+2}\binom{2n+2k+1}{k}(-)^k\left(\tfrac{1}{3}eaa_0\right)^{2k}, \qquad (1)$$

where a is the amplitude of the vector potential of the inducing field (intense field) of frequency ω and polarization vector $\vec{\epsilon}$; a' is the corresponding amplitude of the field of frequency ω' and polarization $\vec{\epsilon}'$ whose presence is necessary to satisfy energy conservation, i.e., $(2n+1)\omega+\omega'=E_{2s}-E_{1s}$; a_0 is the Bohr radius; and α is the relative phase between the fields of amplitudes a and a'. Equation (1) is not adequate for our purposes because of the presence of the sum, which has a radius of convergence $y\equiv\frac{2}{3}eaa_0<1$ (note that y is the same as the parameter $2b$ employed in Ref. 1). It is possible to perform the sum in closed form and thus eliminate this restriction.[2]

We define

$$\mathcal{T}=\left(\tfrac{1}{2}y\right)^{2n+1}\sum_{k=0}^{\infty}(n+k+1)^2(n+k+2)\binom{2n+2k+1}{k}\left(-\tfrac{1}{4}y^2\right)^k, \qquad (2)$$

which contains all the amplitude dependence of the intense field in Eq. (1). Carrying out the summation in Eq. (2) yields the result

$$\mathcal{T}=\frac{1}{8B^{7/2}}\left(\frac{y}{B^{1/2}+1}\right)^N [-NB^{5/2}-N^2B^2+N(N^2-7)B^{3/2}+6(N^2-2)B+15NB^{1/2}+15], \tag{3}$$

where $B=1+y^2$ is a new intensity parameter, and $N=2n+1$ is the number of intense-field photons emitted in the induced-emission process. It is clear from Eq. (3) that an essential singularity exists at $B=0$, or $y^2=-1$, which causes the radius of convergence of Eq. (2) or (1) to be $y^2<1$. The singularity at $y^2=-1$ is in an unphysical region, and thus Eq. (3) is an analytical continuation of Eq. (2) to arbitrarily large physical values of y, as long as the conditions $\omega \ll E_{2s}-E_{1s}$ and $\omega y \ll E_{2s}-E_{1s}$ are met, as discussed in Ref. 1.

Figure 1 shows $\mathcal{T}$ plotted against y for the case $N=1$. The dashed curve shown in the same figure is the perturbation-theory result, which can be found from Eq. (3) by taking the limit $y\to 0$ (or $B\to 1$). The perturbation result is simply $\mathcal{T}_{N=1} \underset{y\to 0}{\longrightarrow} y$. It was stated on general grounds in Ref. 1 that perturbation theory could not be correct when y is of the order unity. Figure 1 shows that the limitations of perturbation theory are even more severe. Major quantitative deviations begin to occur at $y\approx 0.2$ or $y^2\approx 0.04$,[3] rather than $y^2\approx 1$. A value of $y^2=0.04$ corresponds to about 6.5×10^{12} W/cm^2 of 1.06-μm radiation. Such an intensity, while high, is presently achievable with a focused laser beam.

The qualitative deviations from perturbation theory shown in Fig. 1 are striking. A maximum in $\mathcal{T}$ occurs at $y=0.34$, followed by a zero at $y=2[(1.5)^{1/2}-1]^{1/2}=0.948$. Thus, not only does the transition probability (proportional to $\mathcal{T}^2$) fail to rise monotonically as the intensity increases, it actually declines and passes through zero for purely dynamical reasons.[4] The zero occurs at about 1.5×10^{14} W/cm^2 of 1.06-μm radiation.[5] Beyond this zero, another extremum of $\mathcal{T}$ occurs, followed by a slow decline as the intensity parameter gets very large. Thus we have the seemingly paradoxical result that extremely high field intensities lead to smaller transition probabilities than much more modest intensities.

What happens as the intensity gets very high, and the lowest-order process (shown in Fig. 1) gets less probable, is that higher-order processes become increasingly important. This can be seen in Fig. 2, where $\mathcal{T}$ is plotted against N, the number of photons of inducing-field type that the atom emits. For convenience, N is treated as a continuous parameter although, of course, only odd-integer values are physical. For low intensities, one would expect $\mathcal{T}$ to be much greater for a single-photon process than for higher-order processes. In Fig. 2, this is seen to be true for $y=0.1$, which is a rather high intensity experimentally. As intensity increases further, the probability for higher-order processes builds up relative to the lowest order. Thus, at $y=1$, not only is a three-photon process much more probable than a one-photon induced emission, but so also are five- and seven-photon processes. At $y=5$, two extrema in $\mathcal{T}$ occur, for three- and nineteen-photon transitions, with the second extremum the larger one. Note also the flatness of the curve, so that 15-, 17-, and 21-photon processes are almost equally probable.[6] The $y=10$ case is typical of very high-intensity behavior. The extrema for $N=7$ and 37 bracket a near zero in the transition probability at $N=17$.

The curves of Fig. 2 exhibit features which can be shown to exist in general. As a function of N for fixed y, $\mathcal{T}$ has no zeros for $0\leq y<0.948$, and exactly one zero for $y>0.948$. Hence, never more than two extrema can occur. Also, $\mathcal{T}$ as a

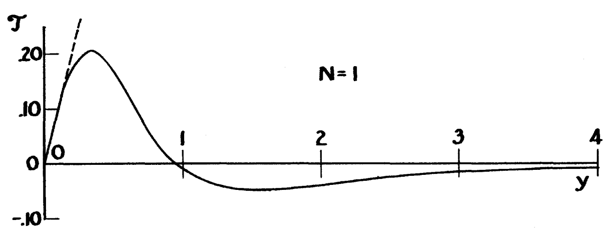

FIG. 1. "Reduced" transition amplitude as a function of the intensity parameter for $N=1$. The dashed line is the perturbation-theory result. The solid curve is the intense-field result.

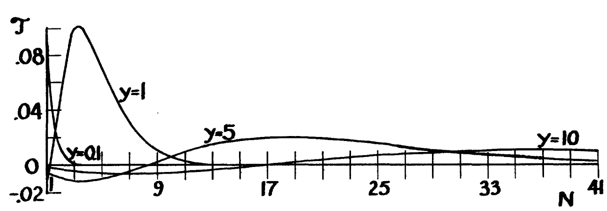

FIG. 2. "Reduced" transition amplitude as a function of photon multiplicity for several fixed values of the intensity parameter. Multiplicity N is regarded as a continuous parameter for convenience. Only odd-integer values of N are physical.

function of N gets more and more flat as y increases, so at very high intensity a large number of photon multiplicities become almost equally probable.

[1]H. R. Reiss, Phys. Rev. A 1, 803 (1970).

[2]It is also possible to perform the derivation in Ref. 1 in such fashion as to achieve a closed-form result directly. This work, and further details of the present paper, will be published elsewhere.

[3]y^2 is more significant physically than y, since field intensity is proportional to y^2.

[4]We wish to emphasize the dynamical origin of this zero transition probability, which distinguishes it from zeros which arise from conservation and symmetry principles, such as angular momentum and parity.

[5]A complete vanishing of the transition probability would not be observed experimentally because a focused laser beam is not truly a plane-wave field as assumed here, and because of approximations inherent in the intense-field method employed here.

[6]When speaking of large values of N, one must keep in mind that the results have meaning only when the constraint $N\omega + \omega' = E_{2s} - E_{1s}$ is satisfied, i.e., $N\omega < E_{2s} - E_{1s}$.

Time-Dependent Photoelectric Counting Statistics for a Q-Switched Laser Near Threshold*

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The time-dependent variation of the photoelectric counting probability has been measured for a Q-switched single-mode laser, whose steady state is in the neighborhood of the threshold of oscillation. The mean and the variance of the instantaneous light intensity, at various times and for various pump parameters, are derived and found to be in very good agreement with the values predicted by Risken and Vollmer.

The problem of the time evolution of the optical field produced when a laser is switched on has been the subject of a number of theoretical[1-5] and experimental[6-11] investigations. Of the experiments, only one was directly concerned with the time dependence of the laser statistics.[9] But while most of the theoretical treatments are applicable to situations where the final steady state is not too far above the threshold of oscillation, very few dynamical measurements appear to have been carried out in this region. Yet it is in the neighborhood of the threshold that the results are likely to be most sensitive to the predictions of the theory. We wish to report the first measurements of the time development of the photon counting statistics, for a laser which is switched from a point far below to a point in the neighborhood of the threshold of oscillation. Within the statistical errors, the results are in very good agreement with the calculations of Risken and Vollmer,[2] which are based on a rotating-wave van der Pol–oscillator model of the laser.

The light source for these experiments was a 10-cm long, single-mode He:Ne laser, whose cavity length could be controlled by a mirror mounted on a piezoelectric ceramic, with the help of a feedback arrangement, as described previously.[12-14] The optical pumping was held constant throughout. By varying the cavity length we were able to vary the atomic amplification and thereby maintain the laser in any steady state from well below to far above the threshold of oscillation. In order to extinguish the laser we introduced a third, external mirror, aligned so as to form a stable Fabry-Perot cavity with the laser mirror nearest to it. A Pockels cell optical shutter was placed between the laser and this third mirror. By controlling the spacing of the external Fabry-Perot cavity, it was possible to extinguish the laser when the shutter was opened and to switch the laser on when the shutter was closed.[15] The effective switching time (about 50 nsec) turned out to be negligible compared with the characteristic rise time of the laser towards its steady state, which was of order 100 μsec. The laser was allowed to dwell in its steady state for about 20 msec following the turnon, when it was again extinguished, and the switching cycle was repeated about 30 times per sec. The feedback loop controlling the laser intensity was operative only during the 20-msec dwell time in the steady state.

The light beam produced by the laser was al-