

Nonlinear Effects in Single-photon Resonant Excitation of Atomic Hydrogen

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Abstract

The first nonperturbative calculation of the amplitude for the excitation of a *discrete* atomic state by a classical electromagnetic field is described. Specifically, the excitation amplitude for the $H(1s) \rightarrow H(2p)$ transition involving the net absorption of only one photon is shown to exhibit a nonlinear dependence on the field amplitude beyond an intensity $I \approx 10^{13} \text{ W cm}^{-2}$. In this region, the rate of change of the transition amplitude decreases with increasing field strength, finally becoming negative after $I \approx 4 \times 10^{14} \text{ W cm}^{-2}$. Degeneracy of the excited state is shown to be largely responsible for the early onset of such behaviour, thereby picking out atomic hydrogen as the most suitable target for experimental observations.

1. Introduction

Investigations of atomic transitions induced by intense electromagnetic fields, which require a nonperturbative formalism for their theoretical description, have so far been confined to multiphoton ionisation (MPI) (Muller *et al.* 1992). It would be of great interest to see whether transitions between bound states are also affected in a similar fashion; however, theoretical analyses of these processes to date have not gone beyond lowest order perturbation theory (Power 1990). The recently developed Floquet theory of the S matrix (Unnikrishnan 1993) now offers a nonperturbative method which is ideally suited for calculations under conditions where the effect of the atomic continuum on the excitation of the bound state of interest is not of much consequence. Excitation of a discrete level through the net absorption of a *single* photon is representative of just such a situation, since the initial and final states have to be in exact resonance with the applied frequency, and therefore, at moderate intensities and in the absence of any degeneracy, one practically has to deal with only a two-level atom. Of course, if degeneracy exists, all states degenerate with, and coupled by the interaction to, either the initial or the final state must be retained in a realistic calculation. By studying the effect of nearby states on the absorption process, one may then restrict the calculations to intensities where the final results are not significantly affected by couplings with higher levels. This paper describes such a calculation of the transition amplitude for the excitation of $H(2p)$, which highlights the influence of degeneracy on this process.

2. Theory

Since the basic theory and general computational procedures have been presented in Unnikrishnan (1993), only the details relevant to this particular calculation are given here. The amplitude for the transition $H(1s) \rightarrow H(2p)$ by the absorption of a single photon from an electromagnetic field described by $\mathbf{E} = \mathbf{E}_0 \cos \omega t$, $\omega = 0.0375$ (all quantities in atomic units, $\hbar = m_e = |e| = 1$), and which is sufficiently intense for spontaneous decay during the excitation process to be negligible, is given by

$$T_{2p,1s}^{(1)} = (\Delta_{1s} - \Delta_{2p}) \alpha_{0,2p}^{2p*} \alpha_{1,2p}^{1s}, \quad (1)$$

where the level shift Δ_a and the Floquet coefficients $\alpha_{N\mu}^a$ corresponding to a reference (unperturbed) state $|a\rangle$, $a = 1s, 2p$ are determined by numerically solving the set of eigenvalue equations

$$(\epsilon_a + \Delta_a + N\omega - \epsilon_\mu) \alpha_{N,\mu}^a = \sum_{\nu} (\alpha_{N-1,\nu}^a + \alpha_{N+1,\nu}^a) V_{\mu\nu},$$

$$N = -\infty, \dots, -1, 0, 1, \dots, \infty, \quad (2)$$

$$V_{\mu\nu} = \langle \mu | \mathbf{E}_0 \cdot \mathbf{r} | \nu \rangle / 2. \quad (3)$$

In equations (2) and (3), ν and μ denote the atomic states having eigenenergies ϵ_ν and ϵ_μ respectively. The label 'a' on various quantities in equation (2) specifies the initial condition which renders the solution unique, namely

$$\alpha_{N,\mu}^a \xrightarrow[E_0 \rightarrow 0]{} \delta_{N0} \delta_{\mu a}.$$

The interaction matrix elements $V_{\mu\nu}$ are available from Bethe and Salpeter (1977).

As explained in Unnikrishnan (1993), the calculation proceeds by limiting $|N|$ to, say, N_p and similarly the number of atomic states to N_a and checking for the convergence of the results as N_p and N_a are increased. In the present calculation, convergence in N_p was achieved, while the maximum value of N_a used was 6, corresponding to principal quantum numbers 1, 2 and 3, since the object was to ensure that the $n = 3$ manifold only caused a small perturbation. In fact, as would be evident from the results presented below, nonlinear effects are very much dominated by the $2s \leftrightarrow 2p$ coupling.

To determine Δ_{1s} and $\alpha_{1,2p}^{1s}$, one starts by solving (2) with a small value of E_0 and choosing the eigenvalue whose eigenvector is such that the element labelled by $N = 0$ and $\mu = 1s$ is the largest. This eigenvector represents the interacting state which has evolved from $H(1s)$. The eigenvalues and eigenvectors at progressively higher E_0 are then determined by requiring their variation to be smooth, i.e. the system evolves adiabatically. Similarly Δ_{2p} and $\alpha_{0,2p}^{2p*}$ are determined by starting another series of calculations with small E_0 , but this time choosing that eigenvalue which has an eigenvector with the element labelled by $N = 0$ and $\mu = 2p$ as the dominant one. The transition amplitude at any E_0 is then given by (1).

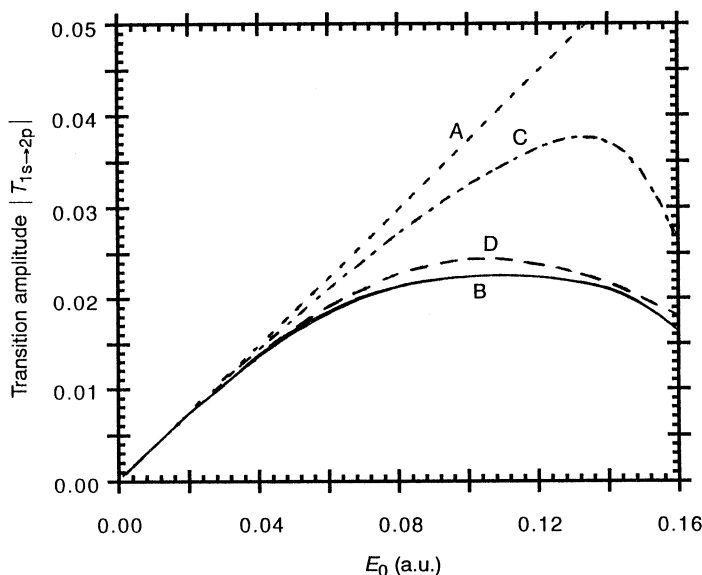


Fig. 1. Amplitude for the transition $H(1s) \rightarrow H(2p)$ as a function of the field amplitude E_0 : A—perturbation theory (golden rule); B—nonperturbative calculation including all couplings among states of principal quantum number $n = 1, 2$ and 3 ; C—same as B, but excluding the $2s \leftrightarrow 2p$ coupling; and D—only $n = 1$ and 2 included.

3. Discussion of Results

As is well known (Heitler 1954), the transition rate between two discrete states depends on whether the incident beam has a sharp or broad frequency spectrum around ω , relative to the natural linewidth of the excited state. In either case, however, the rate is proportional to $|T_{1s,2p}|^2$, which in turn is proportional to E_0^2 , according to the golden rule. Therefore, in order to investigate the possibility of a nonlinear dependence of the transition rate on the intensity, it is enough to examine the variation of $|T_{1s,2p}|$ with E_0 , which is displayed in Fig. 1. The linear variation predicted by the golden rule is represented by the dotted line A, while the solid curve B is the result of taking full account of the virtual photon exchanges among the $n = 1, 2$ and 3 manifolds. Also shown are the results obtained by dropping the $2s \leftrightarrow 2p$ coupling (C), and ignoring the $n = 3$ manifold completely but retaining the $2s \leftrightarrow 2p$ coupling (D). (The result of neglecting all but the two resonant states, i.e. the two-level atom model including counter-rotating terms, is well represented by curve A in the range shown.)

From Fig. 1 we see that the golden rule holds good up to $E_0 \approx 0.02$ ($I = 10^{13} \text{ W cm}^{-2}$), beyond which higher order interactions begin to be increasingly effective in arresting the linear growth of the excitation amplitude, until finally, after $E_0 \approx 0.11$ ($I \approx 4 \times 10^{14} \text{ W cm}^{-2}$), $|T_{1s,2p}|$ becomes a decreasing function of E_0 . A comparison of curves B and D shows that the behaviour of $|T_{1s,2p}|$ in the range of intensity covered in the figure is essentially governed by the $1s, 2s$ and $2p$ states. However, it is evident from the behaviour reflected in curve C that, but for the presence of the degenerate $2s$ level, the golden rule would have practically held sway until $E_0 \approx 0.06$ and the departure from linearity would have been

less dramatic thereafter. Such pronounced nonlinear effects can, therefore, be expected only at somewhat higher intensities, in the excitation of nonhydrogenic atoms. It may be noted here that the existence of analogous effects in MPI, e.g. 'peak suppression', is now well established (Muller *et al.* 1992).

To summarise, the degeneracy of the excited states of hydrogen markedly enhances the occurrence of nonlinear effects in single photon excitation. In particular, departures from the golden rule should be observable at intensities of the order of $10^{13} \text{ W cm}^{-2}$ in the case of the $\text{H}(1s) \rightarrow \text{H}(2p)$ transition. The calculations reported in this paper can be improved upon by including the continuum as described in Unnikrishnan (1995). However, as mentioned in the Introduction, coupling to the energetically far removed states would not be expected to be important for the transition considered here, since the initial and final states are discrete and resonantly coupled. Furthermore, the main conclusion regarding the significant influence of the degeneracy of the final state remains unaltered.

References

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