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Microwave absorption by hydrogen atoms in high Rydberg states

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The behavior of Rydberg atoms in a recent experiment involving selective excitation in combined ac and dc electric fields is shown to be consistent with the predictions of a one-dimensional model incorporating only the extreme level of each Stark manifold. For low microwave powers, the excitation is resonant and is described qualitatively by multiphoton perturbation theory. For higher powers, an alternative stochastic theory predicts rapid ionization at all frequencies, as observed experimentally.

Interest in the dynamics of atoms in strong electromagnetic fields has been enhanced by recent experiments, involving vacuum ultraviolet,[1] infrared,[2,3] and microwave[4] radiation, in which the fields are sufficiently strong that the intervals between the emission and absorption of photons are of the same order as the periods of the electronic orbits. In such situations the intermediate states do not survive long enough for the quantum structure to be fully developed, and quantum theories involve summations over many of these states. For most atoms it would be extremely difficult to compute accurate wave functions and matrix elements for all of the intermediate states. Thus it would be very helpful if theoretical techniques could be developed that do not depend on eigenstate expansions. One possible approach is through the use of classical or semiclassical methods. This is particularly attractive for the study of microwave absorption by atoms in high Rydberg states.

Even for systems with only one active electron, the dynamics is complicated in three dimensions and one-dimensional models are more tractable. This led Jensen[5] and others to study the surface-state-electron (SSE) model in which an electron is bound to a perfectly smooth surface. Jensen has studied the classical mechanics of such a system and Casati, Chirikov, and Shepelyansky[6] have used an eigenvalue-expansion method to study quantum limitations on chaotic excitations with the same Hamiltonian. They have confirmed that quantum diffusion through energy space is slower than in classical mechanics and that there is a threshold field which must be exceeded if significant diffusion is to occur.

The microwave experiments by Bayfield and Pinnaduwage[4] are providing a rich collection of data on which more complete theories can be tested. In an attempt to produce a one-dimensional system, hydrogen atoms are excited in a strong dc electric field by laser light that is tuned to one of the extreme Stark states with parabolic quantum numbers $n_1=0$, $n_2=n-1$, and $m=0$ and n between 60 and 66. The Rydberg atoms pass through a waveguide oriented so that the microwave, laser, and dc electric fields are all perpendicular to the beam direction. For microwave powers below 0.5 W, strong transitions with small changes (Δn) in n are observed at certain resonant frquencies. As the power is increased, transitions with larger values of Δn are seen and the probability of ionization increases, dominating at most frequencies for powers of around 2 W.

The purpose of this Rapid Communication is to discuss the application of standard high-order perturbation theory to this experiment. We will study the applicability of a one-dimensional model and identify quantum barriers to rapid diffusion in energy space. The similarities and differences between these experiments and the computations of Casati *et al.*[6] will be analyzed.

The matrix elements governing dipole transitions between specific Stark states were obtained by Gordon.[7] However, the general expressions in terms of hypergeometric functions mask the simple pattern of the rates of transitions from extreme Stark states. For transitions from the initial state $(n,0,n-1,0)$ to $(n',n_1'=k,\ n'-k-1,0)$, the dipole matrix elements are well approximated by $c(\Delta n,k)n^{2-k}$. Thus for highly excited states there is a strong constraint to remain on the ladder of extreme Stark states. The most important coefficients $c(\pm1,0)$, $c(\pm2,0)$, and $c(+1,1)$ are of magnitude 0.32, 0.11, and 0.6, respectively.

One of the most prominent peaks observed by Bayfield and Pinnaduwage corresponds to a four-photon transition between states with $n=60$ and $n=59$.[4] This leads to a peak at a frequency of 7.5 GHz, with an apparent width of 120 MHz at a power of 0.5 W.

The rates of multiphoton transitions can be computed by perturbation theory, provided that one allows for the permanent dipole moments of the Stark states.[8] For a transition involving s photons between neighboring states on the extreme Stark ladder $(\Delta n=1)$, the dominant terms are those involving one n-changing matrix element and $(s-1)$ diagonal elements. These can be summed to give the Rabi flopping frequency (in atomic units)

$$M^{(s)}=\frac{0.32n^2}{(s-1)!}F^s\left(\frac{3n}{2\omega}\right)^{s-1}, \tag{1}$$

in which F is the field strength and ω is the photon frequency. At a power of 0.5 W, $F=2.3\times10^{-9}$ a.u., and the resonant frequency given above corresponds to 1.14×10^{-6} a.u. The four-photon process has a rate of 2.8×10^{-9} a.u. or $1.3\times10^{8}\ \mathrm{s}^{-1}$. Since the Rydberg atoms are in the microwave cavity for about 400 ns, this transition is well saturated. The width of this stimulated emission line should be approximately 40 MHz, which is only one-third of the observed width. The experimental data suggest that the width varies linearly with the microwave power, which is also inconsistent with our theory. These discrepancies suggest that the observed peak may contain contributions from several Stark states, slightly removed from the extreme level with $n_1=m=0$. This admixture could arise from the weak transitions that lead to nonzero values of n_1, from m-changing transitions caused by imperfect alignment of the microwave field or from nonadiabatic transitions due to the changing of the combined electric field as its value passes through zero and the Stark states become momentarily degenerate.

The transition from $n=60$ to 59 is also observed as a five-photon process at 6 GHz. The calculated rate for this process is 1.5×10^7 s^{-1}, suggesting that the five-photon transition is also saturated. As the photon frequency is reduced further and the value of s is increased, resonant transitions will be rapid providing that the ratio $(1.5n^5F/s)$ is not very much less than 1.

Let us now consider multiphoton ionization from the extreme Stark state with $n=60$. The experiments show that at many frequencies between 6 and 8 GHz considerable ionization occurs at power levels of 1 W. Taking into account the combined effect of the dc and microwave fields, the maximum electric field is 4.5×10^{-9} a.u. Extreme Stark levels with $n>71$ are then classically unstable and thus are very short lived. Because of the rapid rate of multiphoton transitions, the states with n just less than this critical value will have very large widths and almost all transitions will be on resonance. The rate-limiting steps will then be the initial transitions upwards from $n=60$.

The most rapid onset of ionization occurs near 6.8 GHz. Although this frequency is not close to resonance for transitions with $\Delta n=1$ or $\Delta n=2$, it is well matched for a 12-photon transition from $n=60$ to $n=63$. The computation of this rate can be simplified by assuming that the intermediate states following the fourth and eighth steps are the levels $n=61$ and 62, respectively. The transition rate can then be written as

$$M^{(12)}=M^{(4)}\frac{1}{2D_1}M^{(4)}\frac{1}{2D_2}M^{(4)} \quad , \tag{2}$$

in which $M^{(4)}$ is the rate of the four-photon process with $\Delta n=1$, as computed above. D_1 and D_2 are the energy defects associated with the two intermediate levels, $n=61$ and 62. This rate is proportional to the sixth power of the microwave power and at 1 W is of the order of 10^6 s^{-1}. The next step is a seven-photon transition to $n=65$. Although fewer photons are involved here, the intermediate energy defects are greater and more cancellation occurs in the summation over the various pathways from $n=63$ to $n=65$. The rate is approximately of the same order. From $n=65$, a 12-photon transition to $n=69$ is near resonance and will proceed very rapidly on this microsecond time scale.

Accurate calculations are underway to check these results from perturbation theory. However, these estimates confirm the experimental findings that power levels around 1 W are sufficient to drive processes involving large changes in n.

Each of the calculations reported above involves the summation over a large number of terms and there is a high degree of cancellation. For example, in the calculation of the 12-photon transition with $\Delta n=3$, the largest single term is of the order 10^3 a.u., which is 10^{14} greater than the sum. Such destructive interference seems to be an inherent feature of this perturbation theory, except when the photon frequency is close to an integral multiple of the electron orbital frequency. As the power of the applied field is increased, the effects of the discreteness of the quantum energy spectrum are washed out by power broadening, but traces of this interference may remain.

Let us now turn to the computations of Casati *et al.*,[6] in which the time-dependent wave function for a hydrogen atom under the influence of a microwave field is expanded in terms of the unperturbed eigenfunctions. The problem is reduced to one dimension through the use of the SSE Hamiltonian and the coupled differential equations are solved numerically. Because of the absence of a dc electric field, the state expansion must extend beyond $n=200$. Detailed results are reported for an initial state of $n=66$, which is close to that of the experiments analyzed above. However, the photon frequency is considerably higher, being defined as 1.2 times the natural orbital frequency, which gives a value of 27.5 GHz. Two values of the peak field strength are used. For the smaller value, of 1.6×10^{-9} a.u., the distribution of states after 100 field periods (4 ns) is mainly confined to a narrow band between $n=58$ and 70. Significantly more diffusion is observed at the higher field strength of 2.1×10^{-9} a.u.

The frequency used is very close to the natural frequency for atomic states with $n=62$. Hence, transitions downward from the initial state are very rapid. For example at the lower field strength, the eight-photon transition between $n=66$ and $n=58$ proceeds at a rate of 3.5×10^{-6} a.u., or 1.4×10^{11} s^{-1}. The next downward step which is close to resonance is the four-photon transition to $n=55$. Here there is significant cancellation. Perturbation theory predicts that, exactly on resonance, the four-photon process with $\Delta n=3$ has a rate of approximately 5×10^7 s^{-1} which is very slow on the time scale of these computations.

For upward diffusion, the first step appears to be a three-photon transition from $n=66$ to 70, which has a rate of 4×10^9 s^{-1}. Further progress is even slower, the two-photon transition from $n=70$ to 73 having a rate of 2×10^9 s^{-1}. Thus the decline in state populations is less precipitous than on the lower edge of the high-population plateau and part of the state population is able to flow to higher n. As n is increased further, the level spacing decreases and one finds regions where the ratio of photon frequency to orbital frequency is close to an integral value and the transition rate increases once more.

For multiphoton transitions at low powers, large transition rates arise only from the coherent superposition of small transition probabilities over many periods of the orbital motion. At higher fields, with $n^5F>1$, the time interval between n-changing transitions is less than the orbital period and coherence effects are greatly reduced. In this regime a stochastic model of mode coupling might be more appropriate. If the wave function is expanded in terms of bound-state eigenfunctions with energies E_j, the expansion coefficient $a_j(t)$ given by

$$i\dot{a}_j(t)=\sum_k a_k(t)F\cos(\nu t)Z_{jk}\exp[i(E_j-E_k)t] \quad , \tag{3}$$

in which ν is the frequency of the external ac field and Z_{jk} is the dipole matrix element between states with principal quantum numbers j and k. If the difference δ between j and k is large, then the exponent on the right-hand side of Eq. (3) varies rapidly with t. Thus little contribution to $a_j(t)$ would result if $a_k(t)\cos\nu t$ were constant. However, rapid changes in $a_k(t)$ do result in similar changes in $a_j(t)$. The rate of change in $a_j(t)$ due to the coupling with state k, when averaged over the oscillating exponential, is of magnitude

$$\langle|\dot{a}_j(t)|\rangle=\left|\frac{FZ_{jk}}{E_j-E_k}\frac{d}{dt}[a_k(t)\cos\nu t]\right| \quad . \tag{4}$$

Rapid changes in the population of state k thus result in a change of the population of all nearby states. Let us assume that the coherence time associated with this coupling is determined by the Rabi frequency for transitions with neighboring levels, and that beyond such a time interval the coupling leads to changes of the probability amplitude with arbitrary phase that must be combined incoherently. Then the time scale for the transfer of population between two states on the Stark ladder is of the order of 1 μs for coupling with $\delta = 8$ for field strengths of 25 V/cm and $n_0 \approx 60$. Thus, states with $n = 65$ and $n = 73$ should be coupled on this time scale at all frequencies. Since the upper of these two states is prone to rapid autoionization, ionization of the lower state should result.

These analyses have shown that at field strengths F such that $n^5 F \lesssim 1$ the qualitative features of the microwave experiments[4] and the numerical computations[6] can be understood in terms of perturbation theory and that the one-dimensional model is an appropriate starting point for the description of the experiments performed in the presence of a dc field. At such field strengths, the energy exchange rate is sensitive to the frequency of the radiation, as suggested by Shepelyansky,[9] and quantum effects lead to significant destructive interference in multiphoton processes at most frequencies. As the microwave power is increased, a stochastic model is more appropriate. Whether traces of the quantum interference remain will be studied through detailed numerical computation.

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