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Spectroscopic study of nonresonant photon absorption by highly excited hydrogen atoms in a strong microwave field

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Hydrogen atoms with principal quantum number $n = 10$ have been laser excited to states between $n = 44$ and 67 in the presence of a strong microwave field whose frequency ω was adjustable between 5.9 and 8.0 GHz. The excitation probability has been studied as a function of n, ω, microwave field strength and the number of microwave photons absorbed.

I. INTRODUCTION

The O(4) symmetry of the hydrogen atom has led to the application of Lie-algebraic methods to determine the spectrum of the atom while exposed to a time-periodic electromagnetic field $\vec{\mathbf{E}}(t+\tau) = \vec{\mathbf{E}}(t)$, $\vec{\mathbf{B}}(t+\tau) = B(t)$.[1] The theory assumes that the field interaction is strong enough to dominate over the fine-structure interaction while being weak enough for the coupling between states of different principal quantum number n to be ignored. If one has only an electric field $\vec{\mathbf{E}}(t)$ that always lies in one plane (designated the XY plane), then on the n^2 multiplet of given n the interaction Hamiltonian H_1 can be written in terms of components of the renormalized Runge-Lenz vector $\vec{\mathbf{K}}$ and electronic angular momentum $\vec{\mathbf{L}}$ as

$$H_1 = -\vec{\mathbf{r}}\cdot e\vec{\mathbf{E}}(t) = \hbar\vec{\mathbf{J}}\cdot\vec{\omega}_s(t), \quad \vec{\omega}_s(t) \equiv \tfrac{3}{2} n a_0 e\vec{\mathbf{E}}(t), \tag{1}$$

$$\vec{\mathbf{J}} \equiv (K_x, L_y, K_z).$$

The quantity $\vec{\omega}_s(t)$ is the instantaneous Stark angular frequency vector. For a static electric field, the plane of the classical orbit of the atomic electron revolves about $\vec{\mathbf{E}}$ with angular frequency ω_s.[2] For the more general time-dependent case, the time evolution operator $U_n(t)$ is a rotation of $\vec{\mathbf{J}}$ about a time-dependent direction[1]

$$U_n(t) = \exp[-i\vec{\mathbf{J}}\cdot\vec{\beta}_n(t)], \tag{2}$$

where $\vec{\beta}_n(t)$ is a unique functional of $\vec{\omega}_s$,

$$\vec{\beta}_n(t) = \vec{\beta}(\vec{\omega}_s),$$

once $\vec{\mathbf{E}}(t)$ is given. Although the functional $\vec{\beta}$ has not yet been published for any specific time dependent $\vec{\mathbf{E}}(t)$, yet the form of Eq. (2) has led to a qualitative description of the spectrum of the atom in a monochromatic field of frequency ω. There are $2n-1$ different frequencies ϵ_{nm_j} in the eigenspectrum of $U_n(t)$, each accompanied by harmonic satellite structure at $\epsilon_{nm_j} \pm k\hbar\omega$, where k is any integer.[1] The frequencies ϵ_{nm_j} are equally spaced by an amount κ_n that depends on $\vec{\beta}$. In the static-field limit the $\hbar\epsilon_{nm_j}$ become the first-order Stark energies and the harmonic satellite structure disappears.

The present paper describes some early experiments designed to observe the spectrum of the excited hydrogen atom in a linearly polarized time-oscillating field

$$\vec{\mathbf{E}}(t) = \vec{\mathbf{E}}_0\cos\omega t.$$

The atoms had a selected high principal quantum number n that was varied between 44 and 67. The frequency ω was in the microwave region, 5.9 to 8.0 GHz. The ratio k_n of free-atom adjacent energy level separation $\hbar\omega_{nn}$ to photon energy $\hbar\omega$ attained values between 5 and 10. The Stark adiabaticity parameter $\alpha \equiv \omega_s/\omega_{nn}$ was between 10^{-3} and 10^{-1}. The experimental field strengths were less than those required for multiphoton transition of order k_n to levels $n' = n \pm 1$.[3] However, the field strength parameter x defined as the ratio of peak Stark interaction energy to photon energy

$$x \equiv \frac{\frac{3}{2} n n_e a_0 e E_0}{\hbar\omega} \equiv \frac{\mu E_0}{\hbar\omega} \tag{3}$$

reached values between 0.1 and 10. In Eq. (3) the electric quantum number $n_e \equiv n_1 - n_2$ is the difference in parabolic quantum numbers. For static fields, $\hbar n_e$ is the component of the (renormalized) Runge-Lenz vector along the field direction.[2] As will be discussed below, the harmonic satellite structure can be important when $x \gtrsim 1$. The experiments to be discussed are for selected n, but not selected n_e. Thus k_n and α are precisely defined while x is not. However, the atoms in our atomic beam that contributed most to our experimental signals possessed a fractionally small spread in values of n_e and therefore x also.

We first describe the experiments and their results. Then these will be compared as much

as possible with the prediction of time-dependent perturbation theory.

II. THE EXPERIMENTS

The present experiments were originally proposed in 1975[4] and established as feasible in 1977.[5] They utilized a collinear infrared laser beam and fast atomic beam technique.[6] A mixed-state hydrogen atom beam was produced by $H^+ + Ar$ charge exchange collisions. The atom energy was variable from 4 to 19 keV with a directly measured bound on the energy FWHM of 30 eV. By a proper selection of atom energy and CO_2 laser line, a Doppler-tuned one photon excitation beginning with the $n = 10$ component in the beam could produce atoms in the beam with a selected much higher value of n. Field ionization of the laser-excited atoms then produced a fast proton signal proportional to the laser excitation probability.

What distinguishes the present experimental apparatus from others of this general type is a 1-m long J band microwave TE_{10} waveguide section in the beam line. The superimposed laser and atom beams passed down the center of the waveguide in the direction of microwave propagation. While in the waveguide (and only then), the $n = 10$ atoms in the beam could at some point be excited to a state of some high n, the transition involving the absorption of one IR laser photon *and* k microwave photons.

Figure 1 shows laser excitation spectra produced at two microwave power levels employing the P-22 laser line at 1045.022 cm^{-1} and a fixed microwave frequency ω of 7.829 GHz. The laser frequency in the atom's rest frame was swept by varying the atom velocity v. A large number of microwave-dependent excitation resonances (sidebands) are observed, corresponding to various values of k and n. The 0.5-W spectrum we call an intermediate power spectrum; the 6-W high power spectrum exhibits additional resonances due to higher-order microwave photon absorption, that is, larger observable values of k.

The components of the earth's magnetic field transverse to the atom beam were canceled to within 60 mG when our data was taken. Thus the atoms are exposed to a residual motional electric field $\vec{F}_m = \vec{v} \times \vec{B}$ of at most 0.1 V/cm, to be compared with a peak microwave field strength F

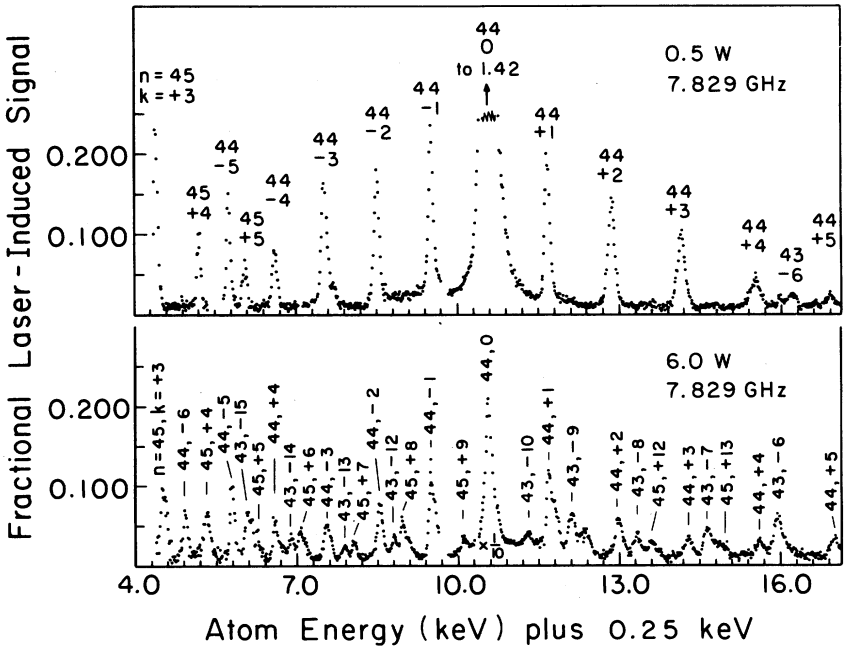

FIG. 1. Excitation spectra for an $n = 10$ hydrogen atom laser-excited to states with $n = 43$, 44, or 45 and with various numbers k of microwave photons absorbed. A fast atom Doppler tuning technique results in laser frequency tuning by varying the atom kinetic energy. The resonances at low atom energies are more intense because the laser-beam atom-beam interaction time varies inversely with atom velocity. The resonances observed at 6 W of microwave power (b) but not at 0.5 W (a) may contain significant contributions from still higher-order photon absorption processes than those indicated. The $k = 0$ resonance contains a large contribution due to laser excitation outside the region of microwave field. When the microwaves are turned off, only $k = 0$ resonances are observed.

of 6.3 V/cm for a microwave power level P of only 0.12 W. When we conducted experiments with $n = 44$ and $k = \pm 1$ with the earth's field only partially canceled ($F_m = 0.2$ V/cm), none of the data was observably different from that with the field canceled, independent of what portion of the motional static-field Stark state distribution was used. Thus we believe that the magnitude of residual motional electric fields played an insignificant role in our experiments, where $F \geq 0.1 F_m$. This cannot be said for the direction of the residual field.

Figure 2 shows the observed variation with microwave power $P \propto E_0^2$ of the intensity of the $k = -1$ sideband of $n = 44$. Complete Doppler scans were taken over the resonance profile at each value of P; what are plotted in Fig. 2 are values always taken at the resonance peak. Let P_1 be the power for the observed maximum. As is discussed in Sec. III of this paper, the observed microwave field dependence is expected to be $J_k(2x)$, where x is the field strength parameter defined in Eq. (3). Looking at the low power $P \ll P_1$ and intermediate power $P \approx P_1$ data, we see that the power dependence does rise, saturate, and then fall. However, the oscillations expected for $P > P_1$ are not present, suggesting that more than one significant value of x is involved there. We find similar microwave field dependences for the other resonances shown in Fig. 1(a). Figure 3(a) shows plots of our

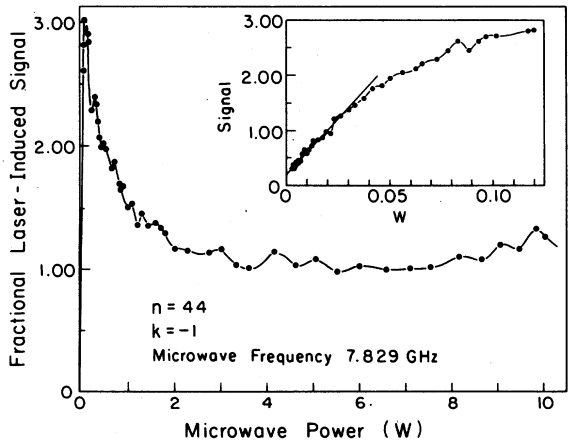

FIG. 2. The relative intensity of the $n = 44$, $k = -1$ resonance, as a function of microwave power. The insert shows the dependence at low powers. The line is hand drawn through the points. Many of the small oscillations in the curve appear to be reproducible, but barely so within the stability of experimental parameters. The rise in the curve above 7.5 W may be due to significant microwave one-photon absorption in the $n = 10$ state. A power level of 0.12 W corresponds to a peak field strength of about 6 V/cm.

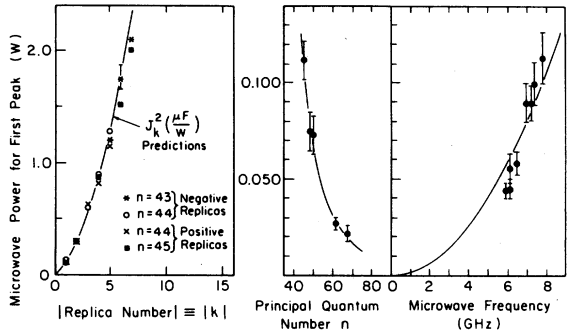

FIG. 3. (a) The dependence of the microwave power P_k required for maximum excitation of the kth order resonance ("sideband"), as a function of k; (b) the variation of P_1 with principal quantum number n; and (c) the variation of P_1 with microwave frequency ω, compared with the ω^2 dependence expected for the induced moment μ independent of ω.

measured values P_k for the power required to maximize the intensity of a kth order sideband. For the values of n and ω fixed and the value of μ assumed constant, reasonable agreement with the predictions of $J_k[2\mu E_0/(\hbar\omega)]$ is found.

For the case of $k = -1$, we have studied the above phenomena by varying n and ω. Figures 3(b) and 3(c) show the results. Within experimental error P_1 varies as $\omega^{(3.0 \pm 0.5)}\, n^{(-4.0 \pm 0.2)}$ for $44 \leq n \leq 67$ and the limited frequency range 5.9–8.0 GHz; the corresponding μ varies as $n^2\omega^{-1/2}$. The n^2 dependence would be expected if the important values of n_e in Eq. (3) scale proportional to n.

The data shown in Fig. 1 are for the "off resonance" case, i.e., where the $k = -5$, $n = 44$ peak is separated from the $k = +5$, $n = 45$ peak. These peaks would coincide for a frequency on resonance for a 10 photon transition from $n = 44$ to $n = 45$. At a microwave power of 0.5 W, we have measured the sum of the areas under the $k = \pm 5$ peaks as a function of resonance detuning and find no resonant enhancement to within ±20%. The intensities of the other peaks with $|k| < 5$ similarly show no strong resonance effects.

III. INTERPRETATION OF THE EXPERIMENTAL RESULTS

The problem of an excited hydrogen atom perturbed by an external linearly polarized oscillating electric field was first considered by D. Blochinsew.[7] Taking the perturbation to be $e\vec{r} \cdot \vec{E}_0 \cos \omega t$, he searched for a perturbed wave function of the form

$$\Psi(r,t)=\left(\sum_{i=1}^{f_n} C_i\Psi_{ni}(\vec{r})+u(\vec{r})e^{i\omega t}+v(\vec{r})e^{-i\omega t}\right)\times\exp\left[i\left(\omega_n t+\Delta\omega_n\int_0^t\cos\omega\tau\,d\tau\right)\right]. \quad (4)$$

Here the unperturbed time-independent Schrödinger equation is solved by $H_0\psi_{ni}(\vec{r})=\hbar\omega_n\psi_{ni}$, and $u(\vec{r})$, $v(\vec{r})$ and $\Delta\omega_n$ are to be found to first order. If the f_n degenerate states Ψ_{ni} are chosen to be the parabolic states with axis of quantization along E_0, then one can write

$$\Psi(\vec{r},t)=\sum_{i=1}^{f_n} C_i\phi_{ni}(\vec{r},t), \quad (5)$$

where the first-order solution for each perturbed parabolic state taken alone is

$$\phi_{ni}(\vec{r},t)=\psi_{ni}(\vec{r})\exp\left[i\left(\omega_n t+\Delta\omega_{ni}\int_0^t\cos\omega\tau\,d\tau\right)\right]+u_{ni}\exp\left[i\left((\omega_n+\omega)t+\Delta\omega_{ni}\int_0^t\cos\omega\tau\,d\tau\right)\right]+v_{ni}\exp\left[i\left((\omega_n-\omega)t+\Delta\omega_{ni}\int_0^t\cos\omega\tau\,d\tau\right)\right], \quad (6)$$

with

$$\Delta\omega_{ni}=\langle ni|e\vec{r}\cdot\vec{E}_0|ni\rangle/\hbar,$$
$$u_{ni}=-\frac{1}{2\hbar}\sum_{\substack{m=1\\ m\neq n}}^{\infty}\sum_{k=1}^{f_n}\frac{\langle ni|e\vec{r}\cdot\vec{E}_0|mk\rangle}{\omega_{nm}+\omega}\psi_{mk}, \quad (7)$$
$$v_{ni}=-\frac{1}{2\hbar}\sum_{\substack{m=1\\ m\neq n}}^{\infty}\sum_{k=1}^{f_n}\frac{\langle ni|e\vec{r}\cdot\vec{E}_0|mk\rangle}{\omega_{nm}-\omega}\psi_{mk},$$

and $\omega_{nm}\equiv\omega_n-\omega_m$. The amplitude for a transition between two such perturbed parabolic states of different principal quantum number n involves the matrix element

$$\langle\phi_{ni}|e\vec{r}|\phi^*_{n'i'}\rangle=\exp[i(\Delta\omega_{ni}-\Delta\omega_{n'i'})\sin\omega t/\omega]\times\{\langle ni|e\vec{r}|n'i'\rangle e^{i\omega_{nm}t}+e^{i(\omega_{nm}+\omega)t}(\langle u_{ni}|e\vec{r}|n'i'\rangle+\langle ni|e\vec{r}|v^*_{n'i'}\rangle)+e^{i(\omega_{nm}-\omega)t}(\langle v_{ni}|e\vec{r}|n'i'\rangle+\langle ni|e\vec{r}|u^*_{n'i'}\rangle)\}. \quad (8)$$

The adiabatically developing phase factor in Eq. (6) and in Eq. (8) must be present for the perturbed wave function to have the correct static-field (zero-frequency) limit. This has been discussed term by term to arbitrary order of perturbation theory by Langhoff *et al.*[8] To second order there must be the phase factor

$$\exp\left[i\Delta\omega_{ni}\frac{\sin\omega t}{\omega}+i\tfrac{1}{2}\alpha_{ni}(\omega)E_0^2\left(t+\frac{\sin2\omega t}{2\omega}\right)\right], \quad (9)$$

where $\alpha_{ni}(\omega)$ is the polarizibility of the state ψ_{ni}. Langhoff *et al.* emphasize that for a single state ψ_{ni} the phase factor is "secular" in that it does not affect observable quantities, for the latter involve the modulus square of operators.

In the present experiments, however, it was unlikely that the $n=10$ atoms entering the microwave field at time $t=0$ were in single parabolic states with quantization axis along the microwave field direction. Residual static electric fields were in a direction different from this, both inside and outside the microwave field region. Thus superpositions of Stark states along the microwave field direction [see Eq. (5)] enter into our experiments.

A solution of the superimposed parabolic-state problem must take degeneracy properly into account through a simultaneous solution of a set of time-dependent secular equations. This has been done for the simple case of $n=2$ by Zon and Sholokhov,[9] where there are only the two spherical (field-free) states $2s$ and $2p_0$ involved. Even then an exact solution must be obtained numerically. However, an approximate solution for the time-dependent amplitudes of the two spherical states is

$$A_{2s,2p}=\exp\left\{i\frac{\bar{\alpha}E_0^2}{4}\left(t+\frac{\sin2\omega t}{2\omega}\right)\right\}\left[C_1\exp\left(-i\Delta\omega_{sp}\frac{\sin\omega t}{\omega}\right)\pm C_2\exp\left(+i\Delta\omega_{sp}\frac{\sin\omega t}{\omega}\right)\right], \quad \bar{\alpha}\equiv\tfrac{1}{2}(\alpha_{2s}+\alpha_{2p}). \quad (10)$$

If the atom were in the $2s$ state before exposure to the oscillating field, then $C_1=C_2=\frac{1}{2}$. Note that each parabolic state has its own different first-order oscillating phase factor. We can now see why for the

superimposed parabolic state case, the presence of these phase factors becomes observable. Let us consider an induced resonant optical transition from the 1s state up to the perturbed $n=2$ states, followed by some detection transition to a final continuum state f. The overall transition amplitude from the 1s state to the final state is second order, and thus involves a sum over a complete set of intermediate states which we choose to be spherical states. Ignoring all states except $1s$, $2s$, $2p_0$, and f, the electric dipole selection rule $\Delta l = \pm 1$ for the optical transition leads to just one nonzero term in the sum, with the overall amplitude then being proportional to

$$\langle 1s \,|ez\,|A_{2p}\psi_{2p}\rangle\langle A_{2p}\psi_{2p}\,|H_{\rm det}\,|f\rangle .$$

The square modulus of this has its time dependence contained in the factor $|A_{2p}|^4$:

$$\left\{|C_1|^2+|C_2|^2+2\,\mathrm{Re}\left[C_1^* C_2 \exp\left(2i\Delta\omega_{2p}\frac{\sin\omega t}{\omega}\right)\right]\right\}^2 . \tag{11}$$

With $C_1 = C_2 = \frac{1}{2}$ as in the example mentioned above, this would become $\cos^2(\Delta\omega_{sp}\sin\omega t/\omega)$. Since $\exp\times(\pm i2\Delta\omega_{sp}\sin\omega t/\omega) = \sum_{k=-\infty}^{+\infty}(-1)^{\pm k}J_{\pm k}(2\Delta\omega_{sp}/\omega)e^{-ik\omega t}$,

$$\begin{aligned}|A_{2p}|^2 &= C_1^2+C_2^2+C_1C_2\sum_{k=-\infty}^{\infty}\left[(-1)^k J_k\left(\frac{2\Delta\omega_{sp}}{\omega}\right)+(-1)^{-k}J_{-k}\left(\frac{2\Delta\omega_{sp}}{\omega}\right)\right]e^{-ik\omega t}\\ &= C_1^2+C_2^2+2C_1C_2\sum_{k=-\infty}^{+\infty}(-1)^k J_k\left(\frac{2\Delta\omega_{sp}}{\omega}\right)e^{-ik\omega t}\end{aligned} \tag{12}$$

and so the transition amplitude for the kth Fourier component or sideband is proportional to

$$2C_1C_2J_k\left(\frac{2\Delta\omega_{sp}}{\omega}\right)|\langle 1s\,|ez\,|2p\rangle|^2 . \tag{13}$$

Returning to the present experiments, a similar but more complicated situation qualitatively explains our results. Again the electric dipole selection rule favors certain final $n=44$ spherical states over others, for a given initial $n=10$ state. One thus expects for each initial state that the sum over final states will yield a few important interference terms that lead to Bessel functions $J_k(2\Delta\omega_{ni}/\omega)$ appearing in the final overall transition probability for a given sideband. The data show that even although many initial and final states may generally be involved, the signals at lower microwave field strengths are dominated by a relatively few final states having about the same values of $\Delta\omega_{ni}$. The detailed reasons for this remain to be investigated.

It is evident that better experiments might use atoms initially prepared in a single $n=10$ Stark state. Such experiments are being attempted in our laboratory. One interesting question concerns the alternation of the sidebands when large static fields are added to the microwave field in the interaction region. If degeneracy were to be completely lifted, the sidebands should greatly weaken in intensity.

It is interesting that Eq. (8) brings out the existence of two types of microwave photon absorption. The first type is the familiar one of nondegenerate state perturbation theory, with one-photon microwave absorption represented by the second and third terms within curly brackets in Eq. (8). The second type arises from the Fourier decomposition Eq. (12) of the oscillating phase factor in Eq. (8). Since $\omega \ll \omega_{nm}$ for our experiments, there is very little frequency dependence in the amplitudes for type-one photon absorption *alone*; this is evident from Eqs. (7) and (8). Thus the strong frequency dependence experimentally observed and shown in Fig. 3(c) clearly establishes the presence of strong photon absorption of type two for the present system of degenerate high Rydberg states in a microwave field.

Returning briefly to Eq. (8), one can see from Eq. (7) that the second and third terms in square brackets are smaller than the first term by a factor of order $n\omega_s/\omega_{nm}$. For the 0.5 W $n=44$ data of Fig. 1 this quantity is of order 10^{-1}, while for 6 W, $n=67$ data it is much closer to unity. Thus further experiments can be carried out at microwave field strengths above the perturbative region.

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