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Possibility of High-Resolution Double-Quantum Laser Spectroscopy in Hydrogen*

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We propose experiments for observing double-quantum transitions between the *s* levels in atomic hydrogen. The inherent resolution is considerably higher than in the single-quantum transitions normally used for optical measurements in hydrogen. We present simplified calculations of relevant double-quantum processes, and show how they might be observed in a double-resonance extension of present techniques of precision laser spectroscopy.

The extremely small natural widths[1] of the *s* levels in atomic hydrogen have yet to be utilized in precision optical measurements of the energy levels. On the other hand, the prospects for improved precision in hydrogen and in other atoms have increased considerably in recent years with the successful use of saturated absorption spectroscopy[2,3] to eliminate the Doppler broadening, especially[4] in transitions such as the $2s \rightarrow 3p$ Balmer line in hydrogen. However, such allowed single-photon transitions are limited in resolution by the width of the relatively broad *p* levels. By contrast, a double-quantum transition between two *s* levels would be limited in resolution ultimately only by the *s*-level widths themselves. Thus, for example, the hydrogen $2s \rightarrow 3s$ double-quantum transition is in principle a factor of 30 narrower than the $2s \rightarrow 3p$ Balmer line. As we will see below, it appears to be within present experimental capabilities to use a double-resonance extension of the methods of saturated absorption spectroscopy to observe free of Doppler broadening such transitions as the $2s \rightarrow 3s$ and $2s \rightarrow 4s$ double-quantum transitions in hydrogen. Measurements based on such a method might well yield a significantly improved value of the Rydberg constant,[5] as well as a more accurate determination of small radiative level shifts[6] limited in resolution only by *s*-state lifetimes.

Narrow double-quantum transitions involving a broad intermediate state were observed several years ago,[7] and they have been clarified and analyzed extensively by a number of authors.[8-12] We very briefly review some relevant characteristics of double-quantum transitions and sketch a simple calculation of a coupled three-level system which exhibits the features we wish to emphasize. We then present resonance curves which display the transition rates and linewidths for the $2s \rightarrow 3s$ double-quantum transition in hydrogen, and we discuss experimental ways of making use of this transition and others for precision measurements.

As illustrated in Fig. 1, we consider two narrow levels *A* and *B* that are connected by external oscillating fields to an arbitrarily broad intermediate level *I*. For example, the states *A*, *B*, and *I* could be the hydrogen states 2*s*, 3*s*, and 3*p*, respectively. In a transition between *A* and *B* two photons are emitted or absorbed, one from each field, with the important feature that the energies of the photons are so correlated that the uncertainty in their sum is determined by the widths of states *A* and *B* only. The width of the state *I* determines the widths of the photon energies separately, but not their correlated sum. These conclusions follow directly from basic principles of quantum mechanics.[13]

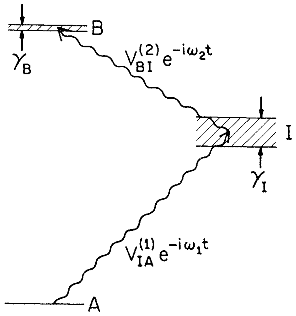

FIG. 1. Representation of the situation analyzed in the text in which a transition between two narrow levels A and B is caused by a pair of external oscillating fields nearly resonant with transitions to a broader intermediate level I.

We take an atom initially in state A where it is assumed for simplicity not to decay at all in the absence of external fields. States B and I are assumed to decay radiatively to states not shown in Fig. 1, although other decays ($I \to A$, for example) can readily be included. The two oscillatory applied fields, which are near resonance with the transitions $A \leftrightarrow I$ and $B \leftrightarrow I$, have transition matrix elements $\hbar V_{IA}\exp(-\omega_1 t) + \text{c.c.}$ and $\hbar V_{BI}\exp(-i\omega_2 t) + \text{c.c.}$ We introduce the notation

$$\hbar\,\Delta\omega_1 \equiv E_I - E_A - \hbar\omega_1, \quad \hbar\,\Delta\omega_2 \equiv E_B - E_I - \hbar\omega_2,$$

where both $\Delta\omega_1$ and $\Delta\omega_2$ are considered small (i.e., much less than either of the transition frequencies). Ignoring all terms far from resonance, we obtain for the amplitudes C_i, for each of the three levels, the coupled equations

$$\begin{aligned}\dot{C}_A &= -iV_{AI}C_I\exp(-i\Delta\omega_1 t),\\ \mathring{C}_I &= -\tfrac{1}{2}\gamma_I C_I - iV_{IA}C_A\exp i(\Delta\omega_1 t) - iV_{IB}C_B\exp(-i\Delta\omega_2 t), \qquad (1)\\ \dot{C}_B &= -\tfrac{1}{2}\gamma_B C_B - iV_{BI}C_I\exp(i\Delta\omega_2 t),\end{aligned}$$

where the effect of the damping by spontaneous radiation is described by effective Hamiltonians $-i\hbar\gamma_B/2$ and $-i\hbar\gamma_I/2$ acting on the two states B and I, respectively.[13]

We take V_{IA} as a small perturbation,[14] which amounts to assuming $C_A \simeq 1$ in Eq. (1). We place no restrictions on the size of V_{BI}. After a time $t \gg \gamma_I^{-1}$, we can ignore the transient terms, and we are left with

$$C_I \simeq -V_{IA}K^{-1}[i(\Delta\omega_1 + \Delta\omega_2) + \tfrac{1}{2}\gamma_B]\exp(i\Delta\omega_1 t), \quad C_B \simeq iV_{BI}V_{IA}K^{-1}\exp[i(\Delta\omega_1 + \Delta\omega_2)t], \qquad (2)$$

where

$$K \equiv |V_{IB}|^2 + \tfrac{1}{4}\gamma_I\gamma_B - \Delta\omega_1(\Delta\omega_1 + \Delta\omega_2) + \tfrac{1}{2}i[(\Delta\omega_1 + \Delta\omega_2)\gamma_I + \Delta\omega_1 V_B].$$

The transition rates R_B and R_I "through" states B and I are, respectively,

$$\begin{aligned}R_B &= \gamma_B|C_B|^2 = \gamma_B|V_{BI}|^2|V_{IA}|^2|K|^{-2},\\ R_I &= \gamma_I|C_I|^2\\ &= \gamma_I|V_{IA}|^2[(\Delta\omega_1 + \Delta\omega_2)^2 + (\tfrac{1}{2}\gamma_B)^2]|K|^{-2}, \qquad (3)\end{aligned}$$

and the total transition rate out of state A is $R_A = R_B + R_I$. When $\gamma_I \gg \gamma_B$, the transition rates show sharp resonances near $\Delta\omega_1 + \Delta\omega_2 = 0$ and broader resonances for $\Delta\omega_1$ and $\Delta\omega_2$ individually near zero, as expected. In Figs. 2 and 3 we display some of these features for the case mentioned earlier when the states A, B, and I are the hydrogen states $2s$, $3s$, and $3p$, respectively.

With the states thus identified we now try to get a qualitative idea of specific ways a transition such as $2s \leftrightarrow 3s$ might be observed free of Doppler broadening. Suppose hydrogen atoms in the $2s$ state are acted upon by a radio-frequency field at the $3p \leftrightarrow 3s$ transition frequency[16] ($\Delta\omega_2 = 0$ as in Fig. 2). This field has negligible Doppler broadening. At the same time these atoms are irradiated by two oppositely directed pulsed laser beams tuned to a common frequency near the $2s \leftrightarrow 3p$ Balmer line. By inspection of Fig. 2(a), we see that a laser pulse in one direction will burn a broad single-quantum hole[2] in the velocity distribution of $2s$ atoms except for a sharp peak where the double-quantum effects take place.[17] The laser pulse in the opposite direction would then cause some of the remaining $2s$ atoms to make double-quantum transitions to the $3s$ state, again with a sharp peak in the velocity distribution, as follows from Fig. 2(b). As

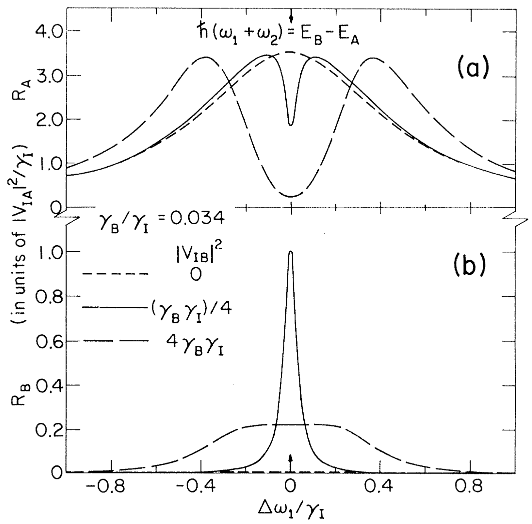

FIG. 2. (a) Transition rate R_A from state A when $\Delta\omega_2=0$ plotted as a function of ω_1 for three different strengths of the transition amplitude V_{BI}. The dip in the single-quantum rate occurs at the double-quantum transition resonance (Ref. 15). (b) Transition rate R_B through state B for the same conditions as in (a).

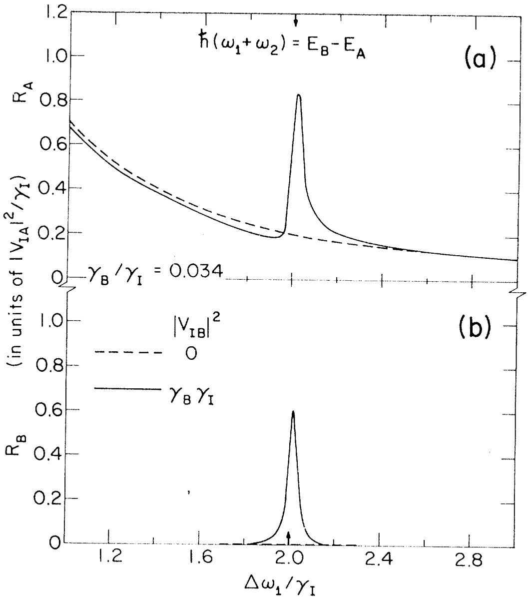

FIG. 3. (a) Transition rate R_A from state A when $\Delta\omega_2 = -2\gamma_I$, plotted as function of ω_1. The double-quantum peak is superposed on single-quantum background rate (Ref. 15). (b) Transition rate R_B through state B for same conditions as in (a).

the laser frequency is varied, these two double-quantum peaks in the velocity distribution will pass through each other, causing a sharp resonance in the appearance of $3s$ atoms coincident with the laser pulse. Thus any indicator of a change in $3s$ population, such as the appearance in cascade of Lyman-α photons, would show a sharp resonance as a function of laser frequency.[18] Another way the effect of the double-quantum transition should show up is in a sharp but small feature in the absorption profile of one of the laser beams when used as a probe. This narrow double-quantum feature could be isolated from the broad single-photon absorption by mistuning the rf field from resonance.[19,20]

The ideas and calculations above can be applied to many transitions in hydrogen and to transitions in other elements as well. It is clear that the transition rates will be largest under the condition of double resonance with an intermediate state. Nevertheless, precision measurements may be possible when double-quantum transitions are generated without such a double-resonance condition. We conclude by noting an important example of such a case, namely that in which there is only a single-frequency field tuned to one half the double-quantum transition frequency. As has been pointed out earlier,[21] when this field is composed of oppositely directed waves, the double-quantum transition occurs free of Doppler broadening. Thus the intriguing possibility exists of observing the ultranarrow $1s \rightarrow 2s$ transition in hydrogen by using oppositely directed laser beams set near one-half the Lyman-α frequency.

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[14] This method of approximation is similar to one used in Ref. 8. A more exact solution can be obtained from the steady-state density-matrix equations which are similar to those set up in Ref. 8.

[15] The curves in Figs. 2 and 3 include the small ($\approx 12\%$) effect of the decay $3p \rightarrow 2s$, which is not contained in Eq. (3).

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[17] We describe this hole-burning method mainly for illustration. A correct analysis of more standard saturated absorption techniques requires a nonperturbative treatment, with two simultaneous laser fields included.

[18] The magnitude of this resonance signal is directly proportional to the double-quantum resonance width, because it depends upon the number of atoms with Doppler shifts lying within this resonance width.

[19] However, $\Delta\omega_2$ must be larger than the Doppler width of the Balmer line in order to eliminate the single-photon absorption.

[20] Our simple calculation will not apply when $\Delta\omega_2 \ll \omega_2$ no longer holds. An interesting such situation is when $\omega_2 = 0$, i.e., a static field for V_{BI}.

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Breakdown of the Statistical-Equilibrium Hypothesis in Channeling*

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New computer-simulation results reveal the source of a previously found difference between analytical and simulation calculations of channeling minimum yield. The difference stems from a focusing effect on ion trajectories in the simulation method which cannot be present if statistical equilibrium exists as is assumed in the analytical method. It is concluded that statistical equilibrium is generally an unsound basis for channeling calculations.

When the direction of a beam of high-energy ions is near a major crystallographic axis in a solid, the ions are steered by the rows of atoms into the regions between the rows and have a reduced probability of making close nuclear encounters. An ion in such a beam interacts with the crystal very much as if the charge on the atoms in each row were distributed continuously along that row. For exact alignment with the axis, the steering or channeling effect is strongest and the yield of close encounters has its lowest value, this value being referred to as minimum yield. The first calculation[1] of the minimum yield was by analytical methods and started by treating the two components of ionic motion transverse to the rows as if the ions move in the continuum potential of the rows. It then assumed that statistical equilibrium or ergodicity prevailed after sufficient depth of penetration into the crystal was reached. This assumption meant that each ion would have equal probability of being anywhere in its constant-energy region of phase space. By virtue of the two-dimensional nature of the motion, there is the further implication that an ion has equal probability of being found at any point within the physical area accessible to it under conservation of energy. This last result produces a great simplification of the calculation.

A second calculation[2] used Monte Carlo techniques in a computer simulation of large numbers of trajectories and gave a value about 3 times as large as the analytical result. It also showed a depth variation of the minimum yield with two very distinct peaks below the surface peak, the deeper of the two being the more prominent. Some limited study[2] of the source of difference between the two methods showed that trajectories whose closest approach to the rows was about 1 or 2 times the thermal-vibration amplitude were making a much larger contribution in the simulation calculations than in the analytical ones. Measurements of minimum yield have