Coherent Excitation of S and P States of the $n = 2$ Term of Atomic Hydrogen*

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An expression is given for the Stark-induced beat signal in the Lyman-α radiation from hydrogen atoms in a beam-foil experiment. This expression includes possible contributions to the beat signal from coherent excitation of S and P states in the beam-foil process. For the case where the electric field is applied parallel to the beam, the coherent-excitation contributions can be separated experimentally from those resulting purely from coherence produced by the electric field, by simply reversing the direction of the electric field.

The recently developed beam-foil spectroscopic technique[1] has been used to investigate excited states of many atoms and ions. Among the most interesting results of these experiments have been the observations of periodic spatial fluctuations, or "beats," in the intensity of the radiation emitted by light atoms and ions (H, He, Li^+) subsequent to their exit from the exciting foil. Since all the particles in the beam have nearly the same velocity, these spatial beats are equivalent to beats in time measured from the instant of excitation at the foil.

Macek[2,3] has explained these temporal beats as the result of coherent excitation of neighboring fine-structure levels. With the beam direction as the axis of quantization, the symmetry of the experiment about this direction restricts the possibility of coherence to states of the same m_J. Further, the validity of Russell-Saunders coupling for these light atoms requires that only states of the same m_L and m_S can be coherently excited. Thus, beats between L_{J,m_J} states with the same L and m_J but different J (such as $P_{3/2,1/2}$ and $P_{1/2,1/2}$) may more properly be thought of as the result of coherence induced by the fine-structure interaction (proportional to $\vec{L}\cdot\vec{S}$) rather than the excitation process itself.[4] This is implicit in Macek's analysis[3] and has been explicitly stressed by Sellin.[5,6]

Macek[3] has pointed out the possibility of beats between states with the same m_J but different L (such as $S_{1/2,1/2}$ beating with $D_{3/2,1/2}$ or $D_{5/2,1/2}$). Such beats, which cannot be ruled out by any symmetry argument, require true coherence in the excitation process in the sense that the initial relative phases of these states averaged over successive atoms in the beam must not be totally random. There has been only one published report of the observation of such "true" coherence beats, that by Burns and Hancock.[7] In their beam-foil investigation of the Balmer-β line of hydrogen, S-D beat signals appeared to be present along with fine-structure–induced beats between the $P_{3/2}$ and $P_{1/2}$ states and between the $D_{5/2}$ and $D_{3/2}$ states. Extraction of the amplitudes of the component signals required computer fitting of a rather complicated theoretical expression to the observed beat pattern. The purpose of the present Letter is to discuss coherent excitation contributions to the Stark-induced beat signal in the Lyman-α line of hydrogen. We shall show how these contributions can be separated experimentally from those resulting purely from coherence produced by the applied electric field.

To say that the beam-foil process excites a coherent mixture of S and P states of the same m_L $(=0)$ and m_S implies that the initial electronic charge distribution averaged over all atoms lacks reflection symmetry in a plane perpendicular to the axis of quantization and passing thru the atomic nucleus. With the beam direction as the axis of quantization, such an asymmetry of the average initial electronic charge distribution would not be at all surprising in a beam-foil experiment or in the case where the atoms are excited by a unidirectional beam of electrons or ions. One plausible model[8] for the formation of neutral atoms in a beam-foil experiment involves extraction of the neutralizing electron from the foil just after the ion exits, producing an initial electronic charge distribution which protrudes back toward the foil. Thus, it is not really a question of whether there is coherent excitation in such experiments, but rather what is the degree of coherence.

S and P states cannot decay to the same terminal state. Therefore, even if these states are coherently excited, S-P beats cannot be observed unless these states are coupled by an external electric field. The problem is to distinguish be-

tween coherence which is the result of the excitation process itself (nonrandom initial relative phases) and that induced purely by the electric field.

The most extensive investigation of Stark-induced Lyman-α beats in hydrogen was that of Andrä.[9] Hydrogen atoms in the $n=2$ term were produced by protons incident on a thin carbon foil or H_3^+ ions incident on a double foil. These excited atoms were subjected to external electric and magnetic fields. The following are the results relevant to our present discussion.

(1) With an electric field $\vec{E}$ parallel to the beam, a pronounced beat signal was observed for E greater than about 100 V/cm. The frequency of the beat corresponded to the separation between the $S_{1/2}$ and $P_{1/2}$ levels.

(2) With $\vec{E}$ perpendicular to the beam no beats could be detected until E reached 240 V/cm. Below 300 V/cm the beat amplitude was less than 10% of the observed with $\vec{E}$ parallel to the beam.

(3) With magnetic field of 41 G applied perpendicular to the beam the velocity of the beam particles was varied to yield *motional* electric fields (at right angles to the magnetic field and beam directions) in the range from 200 to 400 V/cm. Beat signals were seen with amplitudes comparable to those observed for a field $\vec{E}$ parallel to the beam.

Andrä correctly points out that in the absence of coherent excitation of S and P states the $S_{1/2}$-$P_{1/2}$ beat signal observed for a given electric field strength should be the same for $\vec{E}$ applied either parallel or perpendicular to the beam. In an attempt to explain his data he considers initial states for the particles of the beam which are coherent superpositions of $S_{1/2,\,-1/2}$ and $P_{1/2,\,1/2}$ states or of $S_{1/2,1/2}$ and $P_{1/2,\,-1/2}$ states, the axis of quantization being taken along the beam direction. However, these are precisely the S-P combinations which *cannot* be coherently excited since they involve states with different values of m_J.

In a paper to be published Alguard and Drake[10] present data and calculations which indicate that Andrä's failure to see strong beat signals with $\vec{E}$ perpendicular to the beam may be due to the effects of the fringing field from his Stark plates. Also, Andrä himself[11] seems now to be less than satisfied with the data of Ref. 9.

We have calculated the Lyman-α $S_{1/2}$-$P_{1/2}$ beat signal for the experimental situations investigated by Andrä. Other than neglecting hyperfine structure and possible perturbations of the $S_{1/2}$-$P_{1/2}$ beat signal due to the presence of the $P_{3/2}$ state, the only approximation made was that $(\Gamma/2\omega_0)^2$ is negligible compared to unity. Γ is the radiative width of the P state and ω_0 the zero-field splitting (in angular frequency units) between the $S_{1/2}$ and $P_{1/2}$ states, i.e., the Lamb shift. For the $n=2$ term of hydrogen $(\Gamma/2\omega_0)^2 = 2.2\times10^{-3}$. This calculation, the details of which will be presented elsewhere,[12] follows closely the first part of Wieder and Eck's[13] calculation of anticrossing signals in resonance fluorescence (see Appendix A of Ref. 13).

For $\vec{E}$ parallel to the beam the electric field couples $S_{1/2,1/2}$ to $P_{1/2,1/2}$ and $S_{1/2,\,-1/2}$ to $P_{1/2,\,-1/2}$. These are just the combinations of states which can be coherently excited. The $m_J=+\frac{1}{2}$ and $-\frac{1}{2}$ beat signals are identical in magnitude and sign. The total beat signal is exponentially damped with a damping constant of $\Gamma/2$ and is proportional to

$$(V/\omega)^2[\tfrac{1}{3}(\sigma_{p0}+2\sigma_{p1})-\sigma_s]\cos\omega t + (V\omega_0/\omega^2)\tfrac{1}{3}^{1/2}\langle|f_s\|f_{p0}|\cos\alpha\rangle\cos\omega t + (V/\omega)(\tfrac{1}{3})^{1/2}\langle|f_s\|f_{p0}|\sin\alpha\rangle\sin\omega t$$

plus three similar terms that are smaller by a factor of $\Gamma/2\omega$. These latter terms give contributions to the beat signal which are less than 5% of those from the terms given above. V is the Stark matrix element (in angular frequency units) coupling the $S_{1/2,1/2}$ and $P_{1/2,1/2}$ states, and $\omega = (\omega_0^2+4V^2)^{1/2}$. The quantities σ_s, σ_{p0}, and σ_{p1} are the beam-averaged excitation cross sections for the S state and the P states with $m_L=0$ and 1. The symbols $|f_s|$ and $|f_{p0}|$ denote the absolute magnitudes of the *amplitudes* for exciting the S state and the P state with $m_L=0$, and α is the relative phase of these amplitudes. Angular brackets around a quantity indicate that it is an average over all the atoms in the beam. A given cross section is related to its corresponding excitation amplitude by $\sigma=\langle|f|^2\rangle$.

The second and third terms in the above expression for the beat signal are those resulting from coherence in the beam-foil excitation process. Previously, for simplicity, we have referred to such coherence as the result of a nonrandom initial relative phase ($\langle\cos\alpha\rangle\neq0$, or $\langle\sin\alpha\rangle\neq0$, or both). However, since it is quite possible that the magnitudes and relative phase of f_s and f_{p0} are correlated, the correct averages are those indicated in terms two and three. The first term in the expression gives the incoherent-excitation contribution to the beat signal. It is proportional, as expected, to the difference be-

tween the total excitation cross sections for the $P_{1/2}$ and $S_{1/2}$ states.

The Stark matrix element V is proportional to E_z, where the subscript z is used to emphasize that while $\vec{E}$ is aligned along the beam direction it may point either parallel or antiparallel to the particle velocity. The incoherent part of the beat signal is an even function of E_z and the coherent part an odd function of E_z. To separate the incoherent and coherent parts one simply takes data with $\vec{E}$ first parallel to the beam and then antiparallel. The sum of these two beat signals gives twice the incoherent part and the difference twice the coherent part. Thus, it should not be too difficult to separate out the coherent-excitation contribution to the beat signal, even if it is a relatively small fraction of the incoherent contribution.

This reversal of sign of the coherent-excitation contribution to the beat signal upon reversal of $\vec{E}$ is not at all surprising if one reflects upon our previous discussion of the fact that coherent excitation of S and P states implies an initial polarization along the beam direction of the average electronic charge distribution. For one direction of $\vec{E}$, the initial effect of the field is to increase this polarization, while for the opposite direction of $\vec{E}$ the initial effect is to decrease it.

For $\vec{E}$ perpendicular to the beam the electric field couples $S_{1/2,1/2}$ to $P_{1/2,-1/2}$ and $S_{1/2,-1/2}$ to $P_{1/2,1/2}$. These combinations cannot be coherently excited and one is left with only the first term in the above expression for the beat signal. Therefore, it is tempting to conclude from the relative weakness of Andrä's $\vec{E}$-perpendicular beat signal that the principal part of the $\vec{E}$-parallel signal comes from coherent excitation. However, the relatively strong beat signal observed by Andrä and others[10,14] with a magnetic field $\vec{B}$ applied perpendicular to the beam contradicts this conclusion and supports Alguard and Drake's assertion that the weakness of Andrä's $\vec{E}$-perpendicular signal was an instrumental effect.

A field $\vec{B}$ perpendicular to the beam couples $S_{1/2,1/2}$ to $S_{1/2,-1/2}$ and $P_{1/2,1/2}$ to $P_{1/2,-1/2}$, where, again, the axis of quantization is taken along the beam. This coupling removes the degeneracy of the S states and of the P states and produces states which are equal-mixture linear superpositions of the base states. These superposition states, which have well-defined values of m_J *with respect to an axis of quantization parallel to* $\vec{B}$, will be designated by $S_{1/2,1/2}'$, etc. The motional electric field (perpendicular to the beam and to $\vec{B}$) couples $S_{1/2,1/2}'$ to $P_{1/2,-1/2}'$ and $S_{1/2,-1/2}'$ to $P_{1/2,1/2}'$. These combinations *can* be coherently excited, and, in fact, give coherent-excitation contributions to the beat signal which are nearly identical in magnitude to those for an equal $\vec{E}$ field parallel to the beam. However, the $S_{1/2,1/2}'$-$P_{1/2,-1/2}'$ and $S_{1/2,-1/2}'$-$P_{1/2,1/2}'$ coherent-excitation contributions are *opposite in sign* and of nearly the same magnitude. To the extent that the Zeeman splitting of the S' states and of the P' states in negligible compared to the Lamb shift ω_0, there is complete cancelation and no net coherent-excitation contribution to the beat signal.

It is important to realize that coherent excitation of states with different L values can be detected also in time-averaged experiments. If hydrogen atoms are excited at a steady rate from the ground state to the $n=2$ term by bombardment by an electron beam, and if the $S_{1/2,-1/2}$ state is tuned to degeneracy with the $P_{1/2,-1/2}$ state by a magnetic field parallel to the electron beam, the change in the Lyman-α signal produced by an electric field parallel to the electron beam should contain a contribution that is an odd function of E_z from coherent excitation as well as the familiar contribution that is an even function of E_z from the population difference of these two states. Square-wave modulation of the electric field and lock-in detection at the modulation frequency could be used to detect only the coherent-excitation part of the signal. Similarly, in experiments such as those performed by Lamb and his coworkers,[15] where states of opposite parity are coupled by a resonant rf electric field, coherent excitation would manifest itself by a contribution to the rf signal which, for low rf power, would be proportional to the first power of the oscillating electric field strength. This contribution would be present only for those signals induced by an $\vec{E}_{rf}$ *parallel* to the bombarding electron beam.

The exact expression for the time-averaged signal, for the cases of coupling both by a dc electric field and by an rf electric field, can be given in a relatively simple form[12] by slight modification of Wieder and Eck's[13] expression for anticrossing signals. An important result of this analysis is that the coherent-excitation contribution to the time-averaged signal is down from the contribution from the population difference by at least a factor of the order of $(\tau_a/\tau_b)^{1/2}$, where τ_b is the lifetime of the longer lived of the two coupled states. Thus, for the $n=2$ term of hydro-

gen the coherent-excitation contribution to the time-averaged signal would be quite small.

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Spin-Exchange Shift and Narrowing of Magnetic Resonance Lines in Optically Pumped Alkali Vapors*

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We observe very narrow (~200 Hz) magnetic resonance lines in alkali vapors with atomic densities much greater than usual and with cell dimensions much smaller than usual. The Zeeman resonance frequency is shifted downward by a large factor, but the spin-exchange broadening vanishes in the high-density limit. This new regime affords important new possibilities for investigating basic interactions in dense alkali vapors, and it also opens the way to miniaturized optically pumped devices.

Spin-exchange collisions[1] are known to broaden the magnetic resonance lines of an alkali vapor. The line broadening has always been found to be equal to the product of the spin-exchange rate and a numerical constant on the order of unity. Measurements of the broadening as a function of the alkali vapor density have been used to deduce thermally averaged spin-exchange cross sections.[2] Since the broadening has been found to be approximately equal to the spin-exchange rate in all experiments so far, one might conclude that sharp magnetic resonance lines could not be observed when the spin-exchange rate is comparable to or greater than the magnetic resonance frequency. However, in this paper we show that in sufficiently dense alkali vapors, the spin-exchange broadening of the Zeeman resonance line vanishes, and the sole effect of spin-exchange collisions is to diminish the apparent gyromagnetic ratio of the atoms by a large factor. Thus, there is an important new high-density regime for optically pumped alkali vapors. We can now see narrow (~200 Hz) magnetic resonance lines in cells with 1 mm thickness and atomic densities greater than 10^{14} cm^{-3}. The potential for miniaturization of optically pumped devices is obvious. It is also clear that the high-density regime affords completely new possibilities for the investigation of spin-relaxation phenomena in dense alkali vapors. Since the spin-exchange broadening vanishes, any self-broadening of the Zeeman resonance line must be due to other, as