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Delayed quantum beats as a method of subnatural-linewidth spectroscopy

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The resolution of a quantum-beat experiment, which is basically limited by the atomic natural linewidth, has been improved by observing the fluorescence only from those atoms which have survived in excited states after some time delay. The resulting narrowing enables one to resolve previously overlapping lines in beam-foil quantum-beats experiments using Hα and ^{4}He II (4→3) deexcitation.

Quantum-beat experiments in fast ion beams excited by a thin solid target provide information on excited atomic states and have proved to be a powerful time-resolved fluorescence spectroscopic technique. From quantum-beat modulations, fine and hyperfine structures, g factors, population and coherence cross sections, and Stark splittings have been deduced.[1]

The resolution of a quantum-beat experiment is essentially limited by the atomic linewidth Γ. The frequency spectrum obtained by the Fourier transform of the time modulation of intensity $I(t) \propto \sum_n A_n e^{-\Gamma t} \cos\omega_n t$ exhibits Lorentzian-shaped peaks centered on the frequencies ω_n and resolved only if their separation $\Delta\omega_n$ is more than 2Γ.

In a previous zero-field level-crossing experiment,[2] in which photons were observed after emission from fast particles traversing a gaseous target, we have found a Hanle signal which differs from the classical Lorentzian curve as a result of the limited time integration (0 to $t \sim 1.5\tau$). This time-limited Hanle resonance shows a broadening and oscillations in the wings. Such modifications of the signal shape, including narrowing, have also been discussed and observed in double-resonance[3] and level-crossing experiments.[4] Since

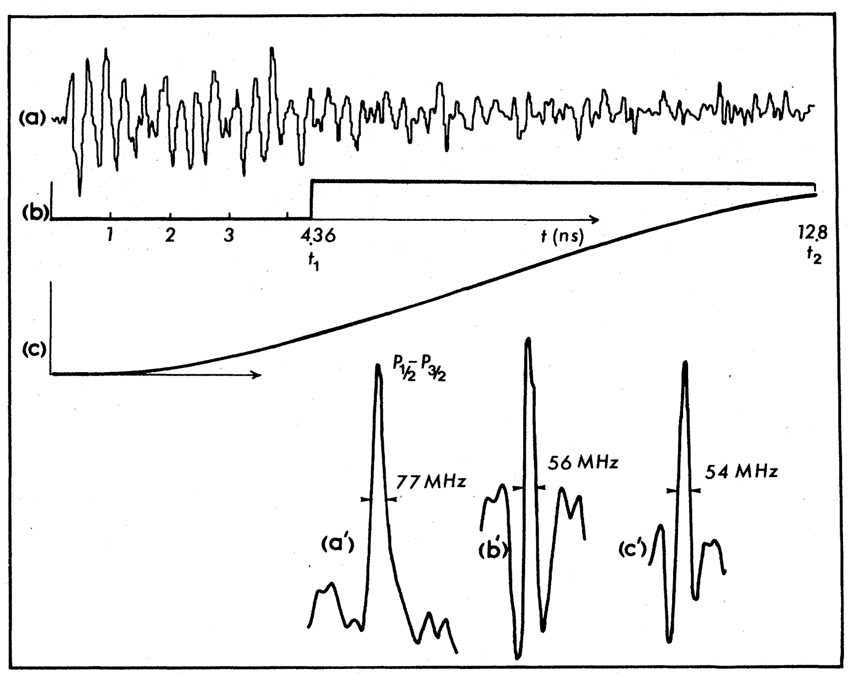

FIG. 1. Experimentally observed zero-field quantum beats in Hα (a). The real part of the Fourier transform of the beat pattern shows a peak at a frequency $\nu = 3.25$ GHz ($P_{1/2} - P_{3/2}$) (a′). Delayed quantum beats are obtained by multiplying the signal (a) by the step function shown in (b). The resulting Fourier-transform peak (b′) is narrowed. Apodisation obtained by multiplying the signal (a) by a half period of a cosine function. As shown in (c′) the Fourier peak remains narrowed but the oscillation amplitudes are reduced.

there exists a close relationship between these radio-frequency methods[5] and the quantum-beat technique (the quantum beats being considered as the inverse Fourier transform of a double-resonance experiment and level crossing as a quantum-beats experiment at zero frequency), the question arises whether one might change and eventually narrow the quantum-beat Fourier lines by observing the fluorescence-decay light not from all excited atoms but from only definite samples of them.

In this paper, we report both the observation of narrowed Fourier-spectrum lines and the resolution of two previously overlapping lines in time-delayed quantum-beat experiments.

In our experiments, a 0.25-MeV/amu H^+ (or He^+) beam was excited by passage through a thin carbon foil (7 μg/cm^2). The foil was moved in steps of 200 μm (100 μm) along the beam axis. The light emitted perpendicular to the ion beam axis was detected by a spectrometer-photomultiplier combination. A 200-μm (100-μm) spatial resolution was obtained using suitable lenses and slits.[6,7]

For the zero-field quantum-beats study (in Hα), the target chamber was shielded from the earth's magnetic field to 0.01 gauss in the transverse direction (motional electric field 7 V/m). Decay curves [Fig. 1(a)] were recorded over a beam length of 9 cm (~12.8 nsec) using a linear polarizer placed parallel to the ion beam. The signal is of the form

$$I=\sum_n [a_n + b_n \cos(\omega_n t - \varphi_n)]e^{-\Gamma_n t} + C .$$

The cascade term (C) is negligible in the present experiment as we did not succeed in observing any characteristic frequency due to cascading from upper levels. The Fourier spectrum of the intensity I is composed of four peaks resulting from fine structures and coherences $\Delta l = 2$. Of interest in this experiment is the shape of the $P_{1/2} - P_{3/2}$ resonance at $\omega_0/2\pi = 3250$ MHz. The natural width of the $3p$ level in hydrogen is $\Gamma_{3p} = 190$ MHz ($\Gamma/\omega_0 \simeq 1\%$). Thus the Fourier-transform line shape reduces to the Lorentzian curve $F(\omega, 0, \infty) = \Gamma/[\Gamma^2 + (\omega - \omega_0)^2]$ corresponding to the line of nat-

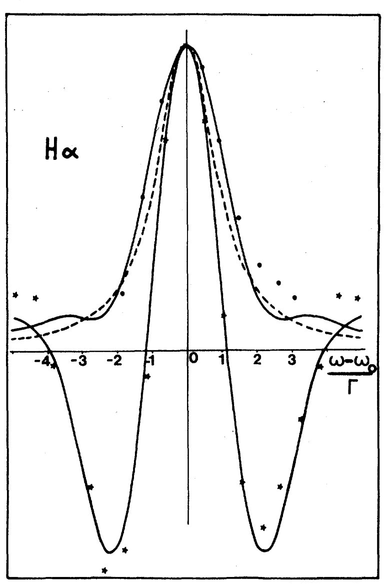

FIG. 2. The real part of the Fourier transform of the experimentally obtained Hα beat signal for the limits from $t_1 = 0$ to $t_2 = 2.46/\Gamma$ and from $t_1 = 0.84/\Gamma$ to $t_2 = 2.46/\Gamma$ are shown by points in this figure. Full curves represent the corresponding theoretical calculations. The dashed curve indicates the theoretical Lorentzian curve for the limits $t_1 = 0$ and $t_2 = \infty$.

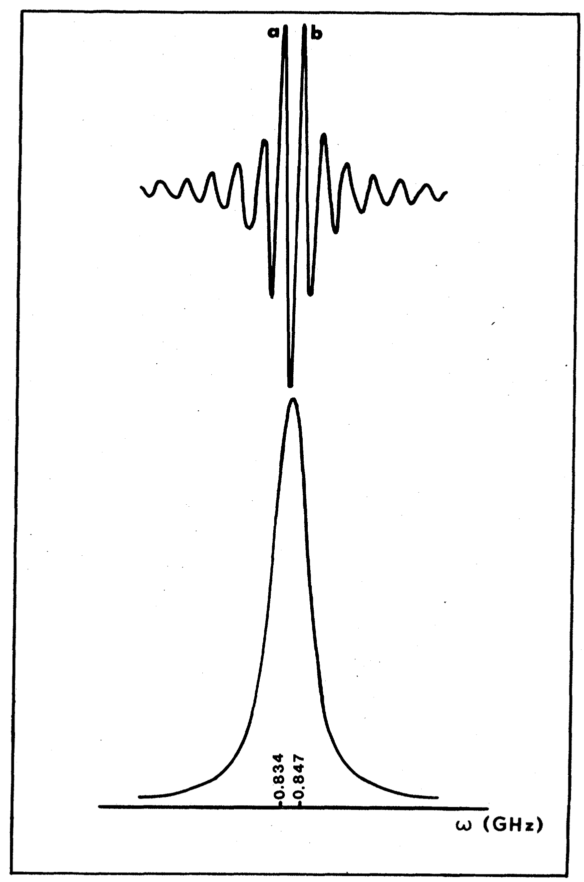

FIG. 3. Real part of the Fourier transform of the function y defined by $y = e^{-0.015x}(\cos 0.834x + \cos 0.847x)$ in the interval $x \in [x_1, 512]$ and by $y = 0$ for x outside of this interval. The single, broadened Fourier peak for the case $x_1 = 0$ (lower curve) is resolved into two components a and b for the case $x_1 = 280$ (upper curve).

ural width 2Γ centered on $\omega_{1/2-3/2}$ [Fig. 1(b)′].

In fact the decay is studied along a finite path from x_1 to x_2 which corresponds to a time duration t_1 to t_2 (Fig. 1). Writing $\delta = \omega - \omega_0$ and neglecting the antiresonance (less than 1% under the condition $t_2 - t_1 > 1/\Gamma$), the real part of the Fourier transform is developed as

$$\sum_{1,2} (-1)^{n+1} e^{-\Gamma t_n} \frac{\Gamma \cos\delta_n t - \delta \sin\delta t_n}{\Gamma^2 + \delta^2} .$$

The peak remains centered on ω_0, and symmetric oscillations are observed in the wings. These can also be used to determine the centroid of the resonance. The central peak is broadened if one terminates data taking after a finite time t_2, but the peak is again narrowed by a factor of $1 + \Gamma t_1$ if the initial time t_1 is delayed from zero (the time of initial excitation after the foil). In our experiment in which $t_2 = 2.46\Gamma^{-1}$ and $t_1 = 0.84\Gamma^{-1}$, the observed narrowing and wing oscillations agree very well with a truncated Fourier calculation (Fig. 2). Doppler broadening is less than 4 MHz. By an apodisation technique, obtained by multiplying the signal by a cosine function, it is possible to lower the oscillatory wing amplitudes without broadening the central peak [Figs. 1(c), (c′)].

The next step consists of studying the possibility of resolving previously blended lines by this technique. By computer simulation we have studied the Fourier spectrum of the sum of two close-lying cosine curves with frequency difference $\Delta\omega$ slightly smaller than the damping constant Γ. In this case the full-time Fourier spectrum consists of only one broadened peak. By Fourier-transform analysis of the delayed signal, both peaks appear well defined (Fig. 3).

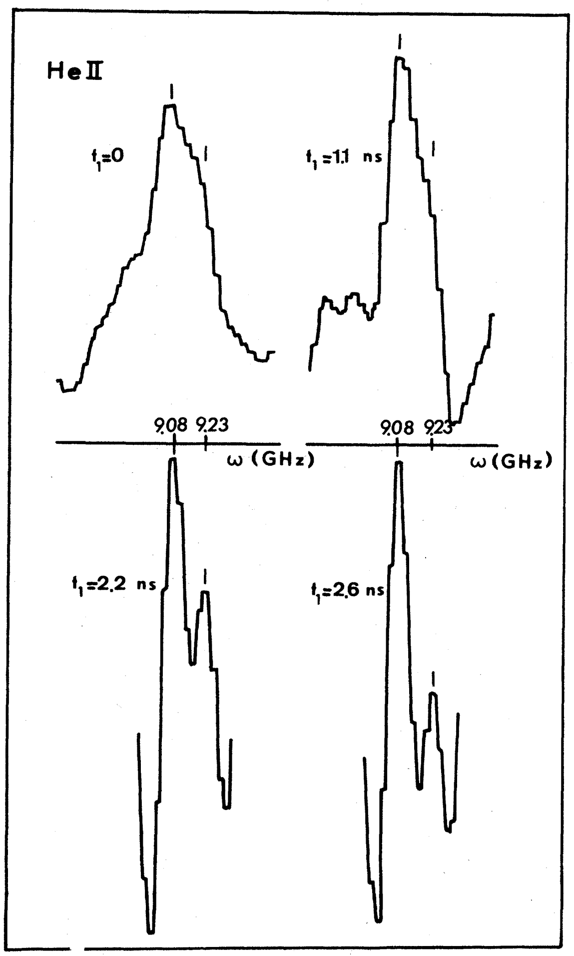

FIG. 4. Resolution of two close-lying lines by the method of delayed Stark quantum beats in He II (n = 4→3) at 4685 Å. The applied field strength is 730 V/cm. The real part of the Fourier transform of the beat pattern corresponds to the limits from t_1 to t_2 = 7.2 nsec. The lines which are unresolved for both delays t_1 = 0 and 1.1 nsec begin to become resolved for the delay t_1 = 2.2 nsec and are completely resolved for the delay t_1 = 2.6 nsec.

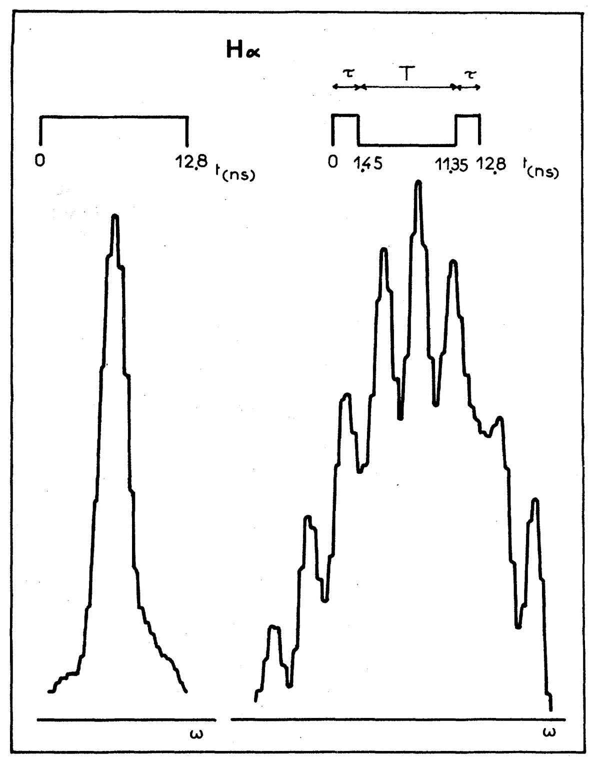

FIG. 5. Fourier transform of Hα experimental modulations, for the transition $P_{1/2} - P_{3/2}$ at 3250 MHz. On the left: square modulus of the Fourier transform of the quantum beats from t = 0 to 12.8 nsec. On the right: pseudo-Ramsey fringes; modulus of the Fourier transform of the quantum beats from t = 0 to 1.45 nsec and from t = 11.35 to 12.8 nsec.

To investigate experimentally this result, we have chosen, as a convenient case, the Stark quantum-beat signal in ^{4}He II (4–3) since, by changing the applied electric-field strength, it is a simple matter to bring resonance peaks closer together until they become unresolvable by the usual process of observation of photons emitted by the total ensemble of atoms. This situation arises, for example, when the electric field is fixed at 730 V/cm for the transitions $\frac{1}{2}$, $1 \to \frac{1}{2}$, -1 and $\frac{1}{2}$, $2 \to \frac{1}{2}$, 0 at $\nu_1 = 9.08$ GHz and $\nu_2 = 9.23$ GHz ($\nu_1 - \nu_2 = 150$ MHz). Natural linewidths are ~225 and ~130 MHz, respectively, and these lines are not resolved. When one uses a delay of 1.1 nsec, the narrowing $(t_1 + 1/\Gamma)^{-1}$ ~180 and 120 MHz is not sufficient to observe the line separation effect, but for a delay of 2.2 nsec the widths are narrowed to 150 and 110 MHz, and the lines are well resolved (Fig. 4).

Another well-known method for narrowing spectroscopic signals is that of the Ramsey fringes which are utilized in rf spectroscopy of atomic and molecular beams.[8,9] We modified this technique for application to modulated decays, in order to compare with our delayed method. The quantum beats are analyzed during two equal-time intervals τ separated by an interval T for which the signal is not recorded. Owing to the finite lifetime of upper levels, the fringe contrast considerably decreases as the interval T is increased. Figure 5 gives an example of modified Ramsey fringes for the transition $P_{1/2} - P_{3/2}$ of Hα. The narrowing is less important than in the delayed quantum-beat method. Furthermore, we do not succeeed in resolving the two previously studied overlapping lines.

In spite of the loss of intensity inherent in the delayed quantum-beat method, a narrowing factor of 1.5 has been obtained which is in agreement with the theoretical treatment for a delay of the order of one lifetime. As an intuitive explanation of the method, we can observe that if the spontaneous decay of the whole ensemble of excited atoms has the natural width, narrowed width curves are obtained from the signals of a selected subensemble of atoms whose mean lifetime is longer than that of the whole ensemble. Such narrowing seems to be attainable in many cases and can be especially helpful in experiments where, under normal conditions, the lines strongly overlap but have a sufficient signal strength for this method to be used.

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