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TWO QUANTUM-BEAT PHENOMENA OBSERVED FOR MAGNETICALLY TUNED ATOMIC SUBLEVELS

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Quantum-beat signals can also be observed when the length of the exciting pulse is not short compared to the Bohr precession period if spontaneous decays during the pulse are important. Experimental demonstration for Zeeman quantum beats in the $6s6p \ ^3P_1 - 6s^2 \ ^1S_0$ ytterbium transition, as well as a simple description of the phenomenon is given. In the same way as quantum beats can be obtained for coherently excited Zeeman sublevels close to the zero-field level crossing, beats can be induced close to a high-field level crossing. We have observed such high-field quantum beats for the $7 \ ^2P_{3/2}$ state of $^{133}$Cs.

The quantum-beat method provides an efficient means of studying the energy splittings between coherently excited atomic substates. The method requires an abrupt excitation and the creation of an atomic orientation or alignment. Superimposed on the exponential decay of the excited state there will be oscillations, beats, at frequencies corresponding to energy sublevel splittings, according to certain selection rules.

The technique was first used by Alexandrov and by Dodd et al. [1], employing shuttered spectral lamps for the excitation. Subsequently it was shown, that quantum beats could also be observed following electron-impact excitation [2] and beam-foil excitation [3], and especially the latter technique has been used extensively. With the availability of tunable lasers, important further developments could be made. Zeeman quantum beats, corresponding to the $\Delta m_f = 2$ energy-level separation in the excited $6s6p \ ^3P_1$ state of Yb, excited by a pulsed dye laser, were first reported by Gornik et al. [4]. Hyperfine-structure quantum beats following pulsed laser excitation were first observed for the $7 \ ^2P_{3/2}$ state of $^{133}$Cs by Haroche et al. [5]. Since then the technique has been used in several investigations. A review is given in [6]. Recently, Silverman et al. have extended the theory of laser-induced quantum-beats to include step-wise excitations and saturation effects [7]. If spontaneous decays during the exciting pulse are neglected it can be shown, that quantum beats will be washed out if the pulse duration is long compared to the excited state Bohr precession period [6]. On the other hand, if such decays are important this is no longer true. In this paper we show, that quantum beats can actually be observed if a low-power, long excitation pulse is used. We have chosen the previously mentioned case of Zeeman quantum beats in the $6s6p \ ^3P_1 - 6s^2 \ ^1S_0$ ytterbium transition. By using the PUMOLS (Pulse-MODulated Laser Spectroscopy) technique, recently used in this laboratory [8,9], the length of the exciting laser pulse could be conveniently varied for the demonstration of the effect.

Using the same experimental techniques we also studied another quantum-beat effect for magnetically tuned atomic sublevels, namely quantum beats close to a high-field level-crossing. From the close theoretical connection between level-crossing- and quantum-beat spectroscopy it is clear, that this phenomenon has the same relation to high-field level-crossing signals as the Zeeman quantum beats have to the Hanle effect. We observed beat signals in the magnetic-field region close to the first level-crossing in the $7 \ ^2P_{3/2}$ state of $^{133}$Cs. The high-field quantum-beat signals can be useful for spectroscopy under specific circumstances to be discussed later.
The long-pulse measurements for Yb were performed with the experimental set-up described in [8,9]. An atomic beam of ytterbium was formed in a vacuum system. The atomic beam was crossed by a multi-mode laser beam from a Rhodamine 110 dye laser, pumped by a CW Ar+ laser. The laser beam was directed perpendicularly to an applied magnetic field and had a linear polarization for \( \sigma \)-excitation \((\Delta m = \pm 1)\). The CW dye-laser beam was pulse-modulated by means of an acousto-optic modulator, which had a rise-time of 10 ns. With a normal pulse generator, controlling the modulator driver unit, suitable pulse lengths and repetition rates could be chosen. Fluorescence light was detected in the direction of the magnetic field, through a linear polarizer, set parallel to the exciting-light polarization plane. Delayed-coincidence electronics were used to record exponential decay curves with superimposed Zeeman quantum beats. The magnetic field was set to yield \( \Delta m = 2 \) quantum beats with a period \( T = 500 \) ns. As the \( g_J \) factor of the \( 6s6p \) \( ^3P_1 \) state is \( 1.49280(4) \) \[10\], the corresponding field was close to 0.48 Gauss. The state has a life-time \( \tau = 880(15) \) ns \[9\], which is conveniently long to enable a recording of several beat periods for the set field. Using a repetition rate of 40 kHz, recordings were now taken for pulse-lengths \( t_p \) starting at 100 ns and increasing each step by 100 ns, up to 1200 ns. Some of these recordings are shown in fig. 1. As expected, the modulation depth first decreases when the pulse-length is increased. For \( t_p = 500 \) ns, equal to the beat period \( T \), the modulation depth is minimum, but when \( t_p \) is further increased, the modulation depth again increases. A second minimum is reached at \( t_p = 1000 \) ns = \( 2T \). The last curve in fig. 1 shows, that for \( t_p = 1200 \) ns a substantial modulation is again present. The observed phenomenon can be easily understood theoretically as an inter-atomic interference effect.

Let the quantum-beat signal for true abrupt excitation be
\[
i(t) = a e^{-t/\tau} (1 + b \cos \omega t).
\]
(1)

with
\[
\omega = \frac{2\pi}{T} = \frac{4\pi \mu_B}{h} g_J \mathcal{B}.
\]
(2)

Here \( \mu_B \), \( h \), and \( \mathcal{B} \) are the Bohr magneton, the Planck constant and the magnetic field strength, respectively. For the chosen geometry, \( b \) should be equal to 1 for a sample of only spin-zero ytterbium isotopes \[6\]. However, the experimental value for \( b \) is less than 1 as shown by the first curve in fig. 1, due to the presence of 30 percent isotopes with a nuclear spin, non-perfect polarizers etc.

For excitation with a square pulse of length \( t_p \) we obtain
\[
I(t) \propto \int_0^{t_p} e^{-t/u} (1 + b \cos \omega (t-u)) du,
\]
(3)

(236)
for the time region after the light has been switched off. Integrating by parts and using trigonometric formulae one obtains

\[ I(t') = A e^{-t'/\tau} (1 + B \cos (\omega t' + \varphi)), \quad (4) \]

with

\[ t' = t - t_p \geq 0, \]

and

\[ B = b \frac{(e^{2t_p/\tau} - 2e^{t_p/\tau} \cos \omega t_p + 1)^{1/2}}{(1 + \tau^2 \omega^2)^{1/2} (e^{t_p/\tau} - 1)}. \quad (5) \]

Eq. (4) is of the same form as eq. (1) apart from the introduced phase shift \( \varphi \).

In fig. 2 the value of \( B/b \) has been plotted as a function of \( t_p/T = \omega t_p/2\pi \), for \( \omega T = 11.06 \), corresponding to the experimental situation. The function has minima with the same value, \( (1 + \tau^2 \omega^2)^{-1/2} \), for \n \[ t_p = nT, \quad n = \ldots, 2, 1 \]

Experimental results with estimated error bars are also included in the figure. Clearly, the simple theory well describes the occurrence of quantum beats in connection with long-pulse excitation.

Our observation of high-field quantum-beat signals close to a level crossing were performed for the \( 7^2 \)\( \pm \)\( 2 \) state of \(^{133}\)Cs. The energy-level substructure of such a state is illustrated, e.g. in fig. 1 of ref. [11]. An atomic-beam apparatus, similar to the one just described, was used. A coumarine 47 dye laser, tuned to 4555 Å, was used to induce the \( 6^2 S_{1/2} \rightarrow 7^2 P_{3/2} \) transition. The laser beam was pulse-modulated to yield a train of pulses of the shortest length (30 ns) attainable with the used modulator. A homogeneous magnetic field was produced by a Helmholtz coil system, calibrated by observing optical-pumping signals in the ground state of cesium. Delayed-coincidence techniques were used for the detection. The same geometrical arrangement as described in the previous experiment was used. It is the arrangement normally used for the observation of \( \Delta m = 2 \) level-crossing signals of Lorentzian line-shape. When the magnetic field was set close to the field position for the first \( \Delta m = 2 \) level crossing, occurring at 28.782(7) Gauss [11], weak quantum-beat signals could be observed superimposed on the exponential decay curve. \( (b \text{ eq. (1)} \text{ was about 0.03}) \text{. The beat structure could be enhanced by turning the detection polarizer 90 degrees after the first half of the planned measuring time to induce a 180 degrees phase-shift for the beats, and then subtracting out the exponential decay (the incoherent background) while attaining the double beat amplitude after the second half of the measuring time. In fig. 3 such curves are shown for three magnetic-field settings close to the crossing.}

\[ \text{Field [Gauss]} \]

\[ 26.34 \]

\[ 26.83 \]

\[ 30.23 \]

Fig. 3. Quantum-beat signals recorded for the \( 7^2 P_{3/2} \) state of \(^{133}\)Cs close to the first \( \Delta m = 2 \) level crossing, occurring at 28.78 gauss. The exponential decay has been suppressed by subtracting curves displaying beats which are 180 degrees out of phase with respect to each other. The dashed lines are drawn for the guidance of the eye. The total measuring time for each curve was about 30 minutes.
From the beat frequencies obtained for known magnetic fields, the field for a zero beat frequency (the crossing position) could be extrapolated. It was found to occur at 28.8(1) gauss, yielding a magnetic-dipole interaction constant for the $7^2P_{3/2}$ state of 16.62(6) MHz. The value, though less accurate, is in agreement with the previous results [11]. Clearly, the same precision as in level-crossing measurements can in principle be attained by quantum-beat observations.

A few comments on the observed quantum-beat phenomena should be made. The possibility of obtaining a substantial Zeeman-beat modulation also when long-pulse excitation is used should be noted when making precautions for unwanted deviations from an exponential decay in lifetime measurements [9]. The spectroscopic method, where quantum-beat signals are observed close to the position of high-field level-crossings can be advantageous in certain types of experiments. E.g., if two CW multi-mode lasers are used in a step-wise excitation scheme, the intensity fluctuations in the fluorescence light may be substantial due to mode drifts, and a DC detection of normal level-crossing signals may be difficult. On the other hand, if a time-resolved technique is used, such intensity fluctuations do not seriously affect the signal-to-noise ratio in a curve displaying quantum-beat signals. The detection of low-frequency beats close to a crossing point puts much lower demands on the electronics than the observation of high-frequency beats at zero magnetic field or at fields far from a crossing point. The technique will be tried in connection with fine-structure determinations for highly excited F-states in Cs.

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