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Simplification of quantum-beat spectroscopy through optical pumping and multiphoton excitation

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Abstract. Spectra in quantum-beat experiments are shown to be greatly simplified when multiphoton optical excitation occurs from the ground states of atoms which have been spin polarised by optical pumping. The method offers enhanced sensitivity in the determination of hyperfine interactions, and possible extensions of quantum-beat experiments to systems of high angular momentum and nuclear spin which otherwise would provide data intractable to analysis.

1. Introduction

Quantum-beat spectroscopy has been usefully employed to measure small separations in the energies of a variety of kinds of atomic states. In a typical experiment an ensemble of atoms is briefly irradiated by a short pulse of tunable laser light, a subset of atoms is placed into a coherent superposition of sublevels of an atomic excited state, and the fluorescence from the excited state is monitored as a function of time. The intensity of the fluorescence is modulated at frequencies characteristic of the splittings between the sublevels which have been excited; appropriate Fourier transformations yield the spectrum of the component frequencies. The technique has been particularly valuable in the determinations of splittings between close lying hyperfine energy states (Deech *et al* 1975, Dodd and Series 1978, Haroche 1976, Arimondo *et al* 1977).

Analysis of data from an actual experiment is not a simple procedure, however. In an excited atomic state with angular momentum $J = \frac{3}{2}$ and nuclear spin $I = \frac{3}{2}$, for example, there are four hyperfine states, $F = 3, 2, 1, 0$, and five characteristic modulation frequencies corresponding to $\Delta F = \pm 1, \pm 2$ separations between hyperfine states. The experimental data are typically analysed first by dividing by a fitted exponential corresponding to the natural decay rate. A Fourier transform is then made of the residual 'undamped' modulation, with an apodisation applied. Further analysis may involve subtraction of the strongest peak or peaks in the Fourier transform, followed by repeated Fourier analysis. All of these manipulations influence the precision of the numerical results ultimately obtained, but nevertheless are necessary for the deconvolution of the data (see, for example, Zhan-Kui *et al* 1982, Grundevik *et al* 1979).

Recent experiments have demonstrated that optical pumping can produce virtually complete electronic and nuclear spin polarisations in an alkali-metal-atom ground state (Sieradzan *et al* 1983, Cusma and Anderson 1983). It is possible to transfer this polarisation to an excited state via single photon excitation or multiple photon excitation

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(Sieradzan *et al* 1983). In this paper we show that if quantum-beat spectroscopy is combined with optical pumping, a sharp reduction in the number of frequencies modulating the intensity of fluorescence can be achieved, with a concomitant deeper modulation of that fluorescence. The simplifications afforded by the scheme should allow significantly greater sensitivity and accuracy in the determination of hyperfine separations and extensions of quantum-beat spectroscopy to systems which are presently inaccessible for study.

2. Reduction of the number of modulation frequencies in quantum beat spectroscopy

In a typical quantum-beat experiment, a number of alkali-metal atoms might be raised to a $^2P_{3/2}$ state from the $^2S_{1/2}$ ground state by the absorption of a pulse of resonance radiation. If the duration of the pulse is short compared with the hyperfine period, but long compared with the fine-structure period, then particular groups of $|F, m_F\rangle$ sublevels of the excited state are populated coherently at time $t=0$, with subsequent evolution of the state vector being induced by the hyperfine interaction. The total population of the excited state decays at the natural spontaneous emission rate; the evolution of the state vector induces modulations of the intensities of particular polarisations of fluorescence. For $J = \frac{3}{2}$ and $I = \frac{3}{2}$, the intensities of π and σ polarised fluorescence are governed by the following general equation:

$$I(\pi, \sigma) = (A_1 \cos \omega_{32}t + A_2 \cos \omega_{21}t + A_3 \cos \omega_{10}t + a_4 \cos \omega_{31}t + A_5 \cos \omega_{20}t) \exp(-\Gamma t) \quad (1)$$

where ω_{ij} stands for the angular frequency corresponding to the energy separations between hyperfine states F and F' in the $^2P_{3/2}$ state, with $|F - F'| = 1$ or 2 , and Γ is the natural decay rate. The values of the coefficients A_i depend upon the particular modes of excitation and detection. General equations which are helpful in the calculation of these parameters have been given by several authors (Haroche 1975, Luybaert and Van Craen 1977, Giacobino and Cagnac 1980, Dodd and Series 1978).

The appearance of five modulation frequencies in equation (1) is a consequence of the fact that under customary circumstances numerous sublevels of the excited state are populated, with the fluorescence being a superposition of spontaneous emission from all of these sublevels. For an actual example of a measurement under these normal conditions, see figure 3 in Grundevik *et al* (1979). The number of modulation frequencies can be reduced, however, if the number of excited-state sublevels initially populated can be reduced. This can be achieved if excitation takes place from a single $|F, m_F\rangle$ sublevel of the ground state rather than from the collection of all $|F, m_F\rangle$ sublevels. Such a situation can be brought about by laser optical pumping of the alkali-metal-atom ground state. Electronic and nuclear spin polarisations approaching 100% have been achieved both in cells (Sieradzan *et al* 1983), and in beams (Cusma and Anderson 1983), corresponding to preparation of the system with almost all atoms residing in a particular sublevel of the ground state.

In the following subsection we illustrate the combination of quantum-beat spectroscopy and optical pumping with calculations of the modulation of resonance fluorescence in three sample cases, all based on excitation from a polarised ground state; a $^2S_{1/2}$ ground state with nuclear spin $\frac{3}{2}$ has been assumed.

2.1. Single-photon excitation from a polarised $^2S_{1/2}$ ground state to a $^2P_{3/2}$ excited state

Three modes of excitation from the $|2, 2\rangle$ sublevel of the ground state are possible; σ^+ , σ^- or π . We calculate how the intensities of π and $\sigma^2P_{3/2} \rightarrow ^2S_{1/2}$ fluorescence depend upon time.

Excitation to the $|3, 3\rangle$ sublevel via absorption of σ^+ radiation yields unmodulated fluorescence damped by the natural decay rate of the $^2P_{3/2}$ state. Measurement of the fluorescence can provide a direct determination of the excited-state lifetime, and can serve as a check of the degree of ground-state polarisation.

Excitation to a coherent superposition of the $|3, 2\rangle$ and $|2, 2\rangle$ sublevels via absorption of π radiation yields modulation of fluorescence at just one frequency:

$$I(\pi) \propto \frac{1}{3}(1 + \cos \omega_{32}t) \exp(-\Gamma t) \quad (2a)$$

$$I(\sigma) \propto \frac{1}{3}(2 - \cos \omega_{32}t) \exp(-\Gamma t). \quad (2b)$$

Excitation to a coherent superposition of the $|3, 1\rangle$, $|2, 1\rangle$ and $|1, 1\rangle$ sublevels via absorption of σ^- radiation yields the following modulation of fluorescence:

$$I(\pi) \propto \frac{1}{100}(31 + 10 \cos \omega_{32}t - 6 \cos \omega_{31}t + 15 \cos \omega_{21}t) \exp(-\Gamma t) \quad (3a)$$

$$I(\sigma) \propto \frac{1}{100}(69 - 10 \cos \omega_{32}t + 6 \cos \omega_{31}t - 15 \cos \omega_{21}t) \exp(-\Gamma t). \quad (3b)$$

2.2. Two-photon excitation from a polarised $^2S_{1/2}$ ground state to a $^2D_{3/2}$ excited state

Two-photon π excitation to a superposition of $|3, 2\rangle$ and $|2, 2\rangle$ in the $^2D_{3/2}$ state yields modulation of $^2D_{3/2} \rightarrow ^2P_{1/2}$ fluorescence at one frequency identical to that given by equations (2a) and (2b).

Excitation from $|2, 2\rangle$ to a coherent superposition of $|3, 0\rangle$, $|2, 0\rangle$, $|1, 0\rangle$ and $|0, 0\rangle$ states can occur through two-photon absorption of σ^- polarised light. $D_{3/2} \rightarrow P_{1/2}$ fluorescence takes the following form;

$$I(\pi) \propto \frac{1}{150}(34 - 9 \cos \omega_{31}t - 25 \cos \omega_{20}t) \exp(-\Gamma t) \quad (4a)$$

$$I(\sigma) \propto \frac{1}{150}(116 + 9 \cos \omega_{31}t + 25 \cos \omega_{20}t) \exp(-\Gamma t). \quad (4b)$$

It is noteworthy that in equations (4a) and (4b) only two frequencies, ω_{31} , and ω_{20} , modulate the fluorescence; ω_{32} , ω_{21} and ω_{10} , which might also be expected to be present, in fact are suppressed. The reason for the suppression is clear in explicit calculations. For even F the expansion of $|F, m_F\rangle$ in the $|m_J m_I\rangle$ basis yields coefficients of $|m_J m_I\rangle$ which are equal both in magnitude and sign for the pairs $|\pm \frac{1}{2}, \mp \frac{1}{2}\rangle$, etc, while for odd F the corresponding coefficients have opposite signs. This asymmetry ultimately leads to the cancellation of terms corresponding to $\Delta F = 1$ modulations (ω_{32} , ω_{21} and ω_{10}), and enhancement of modulation at $\Delta F = 2$ (ω_{31} and ω_{20}). The effect thus depends on symmetries inherent in $|F, m_F\rangle$ wavefunctions and is restricted to coherent excitation of $|F, m_F = 0\rangle$ states. Excitation of states of $m_F \neq 0$ generally yields a full complement of allowed modulations.

2.3. Two-photon excitation from a polarised $^2S_{1/2}$ ground state to a $^2D_{5/2}$ excited state

In this case, two-photon σ^- absorption from the $|2, 2\rangle$ sublevel causes coherent excitation of the $|4, 0\rangle$, $|3, 0\rangle$, $|2, 0\rangle$ and $|1, 0\rangle$ sublevels of the $^2D_{5/2}$ excited state.

$D_{5/2} \rightarrow P_{3/2}$ fluorescence takes the following form:

$$I(\pi) \propto \frac{1}{6125} (2894 - 150 \cos \omega_{42} t - 294 \cos \omega_{31} t) \exp(-\Gamma t) \quad (5a)$$

$$I(\sigma) \propto \frac{1}{6125} (3231 + 150 \cos \omega_{42} t + 294 \cos \omega_{31} t) \exp(-\Gamma t). \quad (5b)$$

As in § 2.2 coherent excitation of $|F, m_F = 0\rangle$ sublevels from the polarised ground state yields modulation only at frequencies corresponding to $\Delta F = 2$, those corresponding to $\Delta F = 1$ are suppressed.

Fluorescence under two-photon π excitation to a superposition of $|4, 2\rangle$, $|3, 2\rangle$ and $|2, 2\rangle$ takes the following form:

$$I(\pi) \propto \frac{1}{588} (111 + 105 \cos \omega_{43} t - 15 \cos \omega_{42} t + 91 \cos \omega_{32} t) \exp(-\Gamma t) \quad (6a)$$

$$I(\sigma) \propto \frac{1}{588} (477 - 105 \cos \omega_{43} t + 15 \cos \omega_{42} t - 91 \cos \omega_{32} t) \exp(-\Gamma t). \quad (6b)$$

Excitation to the $|4, 4\rangle$ sublevel via two-photon σ^+ absorption yields unmodulated fluorescence damped by the natural decay rate.

In each of the examples presented above, excitation from an optically pumped ground state results in a sharp reduction of frequencies at which the fluorescence is modulated, yield a substantially simplified spectrum. Measurement of only two modulation frequencies is sufficient to determine the magnetic dipole and electric quadrupole interaction constants, a and b , the ultimate goal of most experiments of this sort. The fluorescence represented by equations (3a) and (3b), (4a) and (4b), (5a) and (5b) and (6a) and (6b) thus contains full information for most purposes. The higher accuracy which this method should allow would also permit a combination of measurements to check for deviations from the interval rule.

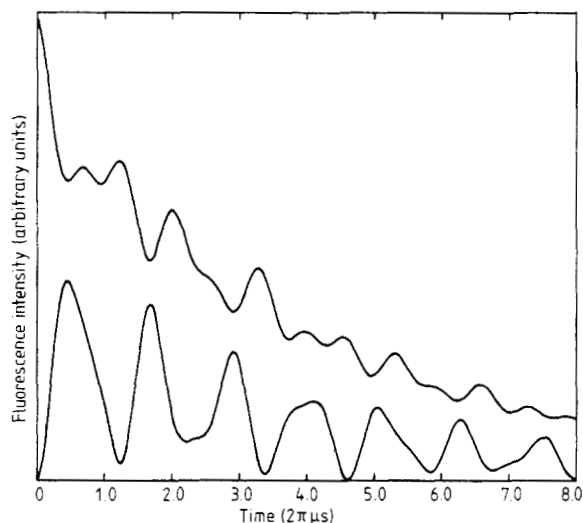


Figure 1. Synthesised quantum-beat spectrum for the $9^2D_{3/2}$ state of ^{87}Rb . The upper curve represents π polarised $2^2D_{3/2} \rightarrow 2^2P_{1/2}$ fluorescence which would be observed in a normal experiment. The lower curve represents similar fluorescence which would be observed if the $2^2D_{3/2}$ state were populated by two-photon σ^- excitation from the spin-polarised $2^2S_{1/2}$ ground state.

A numerical example helps to illustrate the usefulness of the method. The hyperfine constants in the $9^2D_{3/2}$ state of ^{87}Rb are known to be $a = 1.90$ (1) MHz and $b = 0.11$ (3) MHz (Belin *et al* 1975). The lifetime of the state is approximately 5.7×10^{-7} s (Marek and Munster 1980). We have used these values to simulate the results from a normal quantum-beat experiment performed on this state assuming stepwise excitation from a unpolarised $2^2P_{3/2}$ state. We obtain the $2^2D_{3/2} \rightarrow 2^2P_{1/2}$ fluorescent 'signal' shown by the upper curve in figure 1, which involves modulation at the frequencies ω_{32} , ω_{31} , ω_{21} and ω_{20} . Two-photon σ^- excitation of the $9^2D_{3/2}$ state from the polarised ground state, on the other hand, would yield fluorescence described by equation (4a); we illustrate that by the lower curve in figure 1. Two frequencies, ω_{31} and ω_{20} , rather than four are now present, and the spectrum is considerably simplified for analysis.

We have chosen only a few of the simplest examples to illustrate the advantages of combining ground-state optical pumping with multiple photon excitation in quantum-beat spectroscopy. Our results are easily extended to states of higher angular momentum and larger nuclear spin. Indeed, the reduction of modulation frequencies in such cases may well make analyses possible for states which otherwise would provide hopelessly convoluted and intractable data.

References

- Arimondo E, Inguscio M and Violino P 1977 *Rev. Mod. Phys.* **49** 31-75
 Belin G, Holmgren L and Svanberg S 1975 *Phys. Scr.* **13** 351-62
 Cusma J T and Anderson LW 1983 *Phys. Rev. A* **28** 1195-7
 Deech J S, Luypaert R and Series G W 1975 *J. Phys. B: At. Mol. Phys.* **8** 1406-14
 Dodd J V and Series G W 1978 *Progress in Atomic Spectroscopy* Part A ed W Hanle and H Kleinpoppen (Plenum: New York) pp 639-77
 Giacobino E and Cagnac B 1980 *Progress in Optics* vol 17, ed E Wolf (Amsterdam: North Holland) pp 87-161
 Grundevik P, Lundberg H, Martensson A-M, Nystrom K and Svanberg S 1979 *J. Phys. B: At. Mol. Phys.* **12** 2645-54
 Haroche S 1976 *High Resolution Laser Spectroscopy, Topics in Applied Physics* 13 ed K Shimoda (Berlin: Springer) pp 256-313
 Luypaert R and Van Craen J 1977 *J. Phys. B: At. Mol. Phys.* **10** 3627-36
 Marek J and Munster P 1980 *J. Phys. B: At. Mol. Phys.* **13** 1731-41
 Sieradzan A, Krasinski J and Franz F A 1983 *J. Phys. B: At. Mol. Phys.* **16** 43-53
 Zhan-Kui J, Jonsson G and Lundberg H 1982 *Phys. Scr.* **26** 459-61