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Quantum Beats in Transmission by Time-Resolved Polarization Spectroscopy

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A novel technique is described for high-resolution spectroscopy. The time evolution of a coherent superposition of atomic substates created by a short resonant laser pulse is monitored via changes in polarization of a delayed probe pulse due to stimulated emission. As a demonstration the method was applied to the Zeeman splitting of the 6^3P_1 state of ytterbium.

We report on a new technique of high-resolution laser spectroscopy, which makes use of a slightly modified polarization spectrometer[1] with added time resolution in order to monitor the time evolution of a coherent superposition of atomic states created by a short laser pulse (pump pulse). Just like the well known technique of observing quantum beats in the spontaneously emitted fluorescence[2] it is Doppler free and yields a frequency resolution which is orders of magnitude better than the spectral width of the laser in use. In contrast to conventional quantum beat spectroscopy it does not depend upon the timing characteristics of photon detectors, but it can have resolution comparable to the beam-laser technique.[3] Physically the method relies on time-dependent changes of the complex susceptibility χ of the sample induced by the pump pulse. χ is found to contain oscillating anisotropic contributions, which directly display the time evolution of the superposition of states and can easily be detected by placing the sample between crossed polarizers and transmitting a delayed probe pulse. The frequencies of pump and probe pulse are equal in the simplest case. It should be emphasized that the detector signal is determined by the free propagation of the coherence *between* the pump and probe pulse.

It is well known from optical pumping experiments in the ground state using incoherent light sources[4] and from the experiment of Ducas, Littman, and Zimmermann[5] that the light absorption of a sample is modified by a coherence between sublevels of the absorbing state. The dependence of stimulated emission upon coherence between atomic states has been used extensively in level-crossing experiments.[6] The resulting advantages of and possibilities for detection of *transient* coherence in excited states by a transmission method seem to have gone unexplored, however.

Consider free atoms with a lower state $a = |\alpha jm\rangle$ and an upper state connected by electric dipole transitions to a. The upper state is split into sublevels $b = |\beta jm\rangle$, $b' = |\beta j'm'\rangle, \ldots$. The energies are $\omega_a, \omega_b, \omega_{b'}, \ldots$ ($\hbar = 1$). The pump pulse, whatever its characteristics, will generally leave the atoms in a coherent state at the time t_1. The coherence is conveniently described by the density matrix ρ.[2] A nonvanishing matrix element $\rho_{bb'}$ indicates coherence between the sublevels b and b'. The coherence between the lower and the upper state can be neglected, however, under the

assumption of "broad-line excitation."[2] Therefore we assume $\rho_{ab}(t_1)=0$.

The time development of ρ under the action of the Hamiltonian operator H is given by

$$\dot{\rho}=-i[H,\rho], \tag{1}$$

where $[\cdots]$ indicates the commutator. Here relaxation phenomena are neglected and we set $\hbar=1$. In the time between the pulses, Eq. (1) can be integrated to yield

$$\rho_{bb'}(t)=\rho_{bb'}(t_1)\exp[i(\omega_{b'}-\omega_b)(t-t_1)]. \tag{2}$$

In order to find the complex (linear) susceptibility χ of the coherently excited sample, we have to determine the expectation value $\langle P\rangle$ of the dipole moment under the action of a fictitious weak light field described semiclassically by the perturbation operator V. $\langle P\rangle$ can be calculated by means of the relation

$$\langle P\rangle=\mathrm{Tr}(\rho P). \tag{3}$$

In evaluating Eq. (3) only $\rho_{ab}, \rho_{ab'}, \ldots$ are needed, since the matrix elements of P vanish between b and b'. The time variation of ρ_{ab} is given by Eq. (1), which explicitly written has the form

$$\dot{\rho}_{ab}=-i(\omega_a-\omega_b)\rho_{ab}+iV_{ab}\rho_{aa} - iV_{ab}\rho_{bb}-i\sum_{b'\neq b}V_{ab'}\rho_{bb'}. \tag{4}$$

Since the diagonal elements of ρ are proportional to the population numbers of the levels and the off-diagonal elements are a measure of the coherence, the terms in Eq. (4) can be easily interpreted: The second and the third terms describe the influence of the population of a on the absorption and of b on the stimulated emission, respectively; the last term, however, describes the modifications of stimulated emission originating from the coherence in the upper state. This term is usually neglected without further discussion. For a very weak interaction V, which is the assumption of linear optics, $\rho_{aa}, \rho_{bb}, \rho_{b'b'}, \ldots$, and $\rho_{bb'}$ are not influenced by the perturbation. Equation (2) can then be inserted into Eq. (4) and the integration performed. Because of the exponential time behavior of $\rho_{bb'}$, we finally find that there are contributions of $\langle P\rangle$ oscillating with sidebands of the frequency of the driving field. This phenomenon might be interpreted as "quantum beats in stimulated emission."

Moreover the coherence generally prevents $\langle P\rangle$ from being parallel to the driving field. This may be seen in a special case. In a driving field polarized in the z direction the selection rule $\langle \alpha jm|V|\beta j'm'\rangle=0$ for $m\neq m'$ holds. Without the coherence terms in Eq. (4) we could conclude that ρ is diagonal with respect to the quantum number m. As a consequence Eq. (4) would yield $\langle P_x\rangle=\langle P_y\rangle=0$. With coherence this is no longer true. The coherence $\langle \beta jm|\rho|\beta j'm\pm1\rangle\neq0$, for example, can yield nonvanishing matrix elements $\langle \alpha jm|\rho|\beta j'm\pm1\rangle\neq0$ via Eq. (4). Since usually the matrix elements $\langle \beta jm\pm1|P_x|\alpha jm\rangle$ are nonvanishing, the expectation value of P_x will be nonvanishing by Eq. (3). Because of the oscillation of $\rho_{bb'}$, however, $\langle P_x\rangle$ will be oscillating These phenomena give rise to the anisotropic oscillating contributions of χ mentioned above.

As a demonstration of the method a Zeeman quantum beat experiment was performed on the 6^3P_1 state of atomic ytterbium, which has a suitable lifetime of ~1 μsec.[7] The experimental scheme is shown in Fig. 1. A linearly polarized

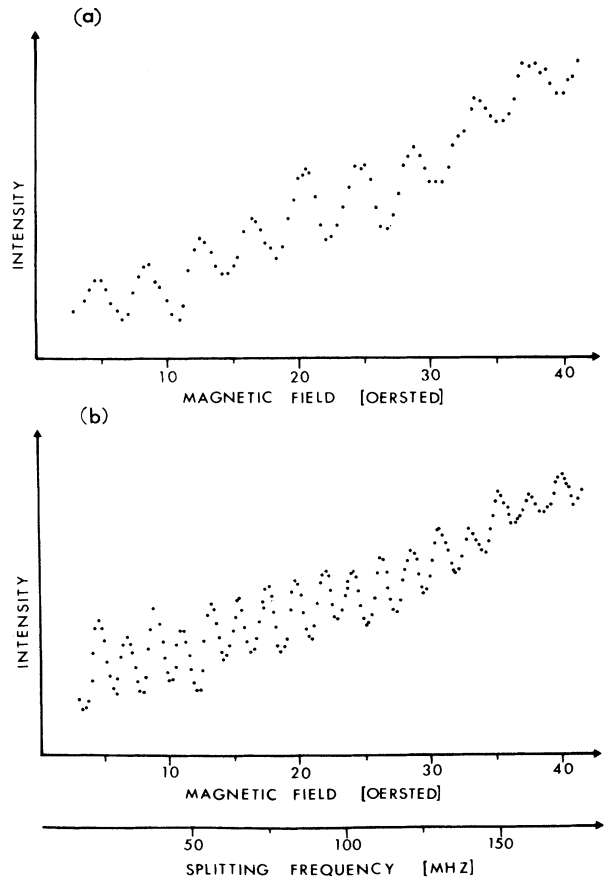

FIG. 2. Zeeman quantum beats in stimulated emission. The magnetic field is scanned for constant time delay τ of the probe pulse. (a) τ = 61 nsec. (b) τ = 113 nsec.

N_2-laser–pumped dye-laser pulse ($\lambda = 557$ nm, peak power 1 kW, pulse length 5 nsec, spectral width 2 Ghz) is sent on a heated stainless steel cell containing metallic Yb. The cell temperature is 830 K, corresponding to a number density of 10^{14} cm^{-3}. The interaction length is approximately 3 cm; contamination of the windows is prevented by a small amount of helium ($p \sim 0.3$ Torr). The direction of the pump pulse and of its polarization are perpendicular to a constant magnetic field H generated by a pair of Helmholtz coils. Thus the pump pulse induces coherence between the Zeeman sublevels $m = +1$ and $m = -1$. The probe pulse is delayed by an amount τ by means of an optical delay line[8] and sent nearly collinearly with the pump pulse through the cell; it is polarized at an angle of 45° with respect to the pump pulse. Without the pump pulse the probe pulse is blocked from the photomultiplier by a crossed polarizer. With the pump pulse present the probe pulse is partially transmitted because of the induced anisotropy. A variation of the detector signal corresponding to $\cos(2\omega_L \tau)$ is expected when the Larmor frequency ω_L is changed by sweeping the magnetic field H. The multiplier pulse is stretched and digitized by standard means.

Experimental results are given in Fig. 2 for two delay times τ.[9] Each point represents the average of 10 and 30 pulses in Figs. 2(a) and 2(b), respectively. Though the experimental setup was fairly curde, the beat structure is clearly resolved even at splittings of 175 MHz, which are not usually accessible to conventional quantum beat spectroscopy.

The method presented here compares favorably with conventional quantum beat spectroscopy and it has all advantages of a transmission method, while avoiding the problem of finding a small signal against a large background. It may be preferable to the method of Ducas, Littman, and Zimmermann, since it requires one laser only. There are, however, further modifications of the quantum beat method recently discussed in literature. As far as they rely on coherence between states of different parity coupled by the light field,[10] they also require some coherence of the exciting laser, which is not easily established by pulsed lasers. Pulsing the laser source may sometimes be avoided by the ingenious techniques of Stark switching[11] or frequency switching.[12] The technique presented in this paper, however, requires a broadband laser source only, and thus seems to be extremely versatile and simple.

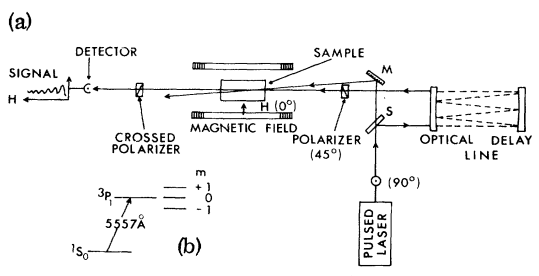

FIG. 1. (a) The experimental setup. (b) Level scheme of Yb.

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