High-resolution coherence spectroscopy using pulse trains

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An infinite train of short light pulses is used for periodic creation of a coherent superposition of atomic substates. The coherence is sensitively detected by an optical probe beam using a polarization-selective technique. Narrow resonances are observed whenever the pulse repetition rate or its multiples coincide with an atomic Bohr frequency. Half-widths (full width at half maximum) of 20 kHz have been obtained for the Zeeman splitting of the sodium ground state.

We report a new approach to determine narrow splittings of atomic or molecular levels in Doppler-free laser spectroscopy. It is based on the unperturbed time evolution of a coherent superposition of nearly degenerate states or substates, i.e., on the "free precession of Hertzian coherence.[1]" The coherence is induced in the sample by resonant interaction with pulsed light. In contrast to quantum-beat spectroscopy,[2] however, not a single pulse is used for excitation, but rather an infinite periodic pulse train. Constructive interference of the various contributions to the coherence induced by different pulses gives rise to sharp resonances in the resulting amplitude of coherence whenever the pulse repetition rate or its multiples coincide with the precession frequency, i.e., with the splitting frequency between the nearly degenerate states involved. No temporal coherence between different pulses is needed in this process and the spectrum of the pulses is allowed to be very broad.

The method described here is closely related to experiments on optically driven spin precession using modulated light for excitation.[3] In this context, the possibility of optical pumping by a periodic pulse train has often been mentioned in literature.[4] More recently, optically-induced precessing magnetization in ruby produced by the short pulse train of a pulsed mode-locked ruby laser was observed experimentally.[5] Although the phenomenon of creation of coherence by a pulse train is obviously well known, its potential as a powerful tool in high-resolution spectroscopy seems to be unrecognized so far. In this paper we demonstrate that extremely narrow resonances with half-widths in the kHz range can be obtained without difficulty by application of periodic pulsed excitation in combination with polarization-selective optical detection of coherence.

For a more quantitative description of the method, let us assume an atomic three-level system with a lower state split into substates a and a' and an upper state b connected to a and a' by electric dipole transitions [Λ-type three-level system, see Fig. 1(b)]. Pump pulses short compared to the inverse of the atomic splitting frequency $\omega_{a'a}$ may induce Hertzian coherence in the sample between substates a and a'; this can conveniently be described by an off-diagonal element $\rho_{a'a}$ of the density matrix ρ. Let us assume that in the interaction of the pulse train with the atomic sample steady-state populations $\rho_{aa}, \rho_{a'a'}$, and ρ_{bb} are obtained and that the Hertzian coherence is small ($|\rho_{a'a}| \ll \rho_{aa}, \rho_{a'a'}$). Moreover, we assume that the coherences $\rho_{ab}, \rho_{a'b}$ between the ground state and the excited state can be neglected. In general, this is a good approximation in the case of "broad-line excitation.[2]" Then, with the pulse amplitudes being constant, each pump pulse arriving at time $t_n = -nT$ ($n = 1, 2, \ldots$) will induce a Hertzian coherence $\delta^{(n)}_{a'a} = \delta_{a'a} \cdot \delta_{a'a}$ can be calculated exactly as in quantum-beat spectroscopy.[2] After the pump pulse this coherence oscillates damped at a rate

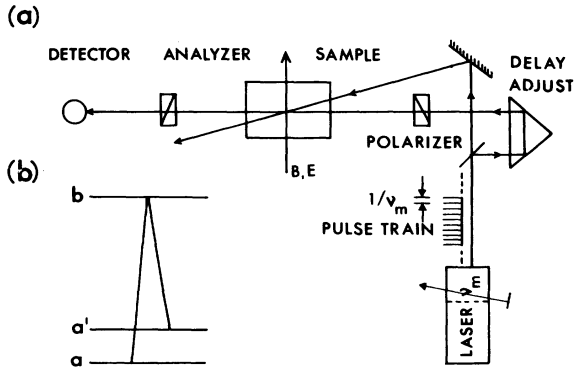

FIG. 1. (a) Experimental scheme. (b) Λ-type three-level system.

$\gamma_{a'a}$ according to

$$\delta^{(n)}_{a'a}(t \geq t_n) = \delta_{a'a} \exp[-i(\omega_{a'a} - i\gamma_{a'a})(t+nT)] \quad (1)$$

and interferes with the contributions $\delta^{(n')}_{a'a}$ $(n' > n)$ already created by preceding pulses. Equation (1) includes the assumption that the time evolution of $\delta^{(n)}_{a'a}$ is not significantly disturbed by succeeding pump pulses; this is expected to be valid for weak pulses with the pulse length being short compared to the pulse interval T. In this case, the total coherence at time t $(0 \leq t < T)$ resulting from a large number n of excitation processes, is obtained by summation:

$$\rho_{a'a}(t) = \sum_{n=0}^{\infty} \delta^{(n)}_{a'a}(t) = \delta_{a'a} Q_{a'a} \exp[-i(\omega_{a'a} - i\gamma_{a'a})t], \quad (2a)$$

$$Q_{a'a} = \sum_{n=0}^{\infty} \exp[-i(\omega_{a'a} - i\gamma_{a'a})Tn]. \quad (2b)$$

For arbitrary times $t' = t + pT$ $(0 \leq t < T$, $p = 0, \pm 1, \pm 2, \ldots)$, the coherence is given by the periodicity condition $\rho_{a'a}(t+pT) = \rho_{a'a}(t)$.

The response of $\rho_{a'a}(t)$ due to periodic excitation is described by the quantity $Q_{a'a}$ in Eq. (2). This sum yields

$$Q_{a'a} = \frac{1 - Re^{i\omega_{a'a}T}}{(1-R)^2 + 4R \sin^2(\omega_{a'a}T/2)}. \quad (3)$$

Here R is given by $R = e^{-\gamma_{a'a}T}$. Periodic excitation resonances of $\rho_{a'a}(t)$ occur, whenever $\omega_{a'a}T = 2\pi q$ $(q = 0, 1, 2, \ldots)$. If $1/\gamma_{a'a} \gg T$, the linewidth [full width at half maximum (FWHM)] of these resonances is given by $\delta\nu \simeq \gamma_{a'a}/\pi$. The similarity of Eq. (3) to an Airy function should be noted. Results analogous to those in Eqs. (1)–(3) are obtained for a V-type three-level system.

In principle, any detection technique of the various quantum beat methods[6] can be applied to detect periodic excitation resonances. We make use of the time-dependent optical anisotropy of the sample generally connected with Hertzian coherence. Such effects are well known from magnetic resonance work[7] and have more recently been used to observe quantum beats in transmission.[6,8] The experimental arrangement of the technique is shown in Fig. 1(a). An infinite train of short light pulses is sent into a sample of atoms to optically induce periodic excitation resonances of Hertzian coherence. The resulting sustained oscillation of coherence in the sample gives rise to resonances of anisotropy. They are monitored by an optical probe beam using a polarization selective technique. For convenience a low-intensity fraction of the pump pulse train can be used for this purpose. Periodic excitation resonances can be studied by measuring the intensity of the probe pulse train at the detector as a function of the pulse repetition rate ν_m. For Zeeman or Stark splittings ν_m can be kept fixed while using an external magnetic field B or electric field E to sweep the atomic Bohr frequency through the resonance. Due to the periodicity of the pulse sequence of excitation and detection, it is sufficient to calculate the transmitted probe pulse intensity within the time interval $0 \leq t < T$. This calculation can be performed similarly as in a quantum-beat transmission experiment.[8] For the experimental arrangement as shown in Fig. 1, it is found that the maximum signal occurs for zero delay time between the pump and probe pulses.

As a demonstration we applied this periodic excitation technique to Zeeman coherences of the sodium ground state. A cw pumped cavity-dumped dye laser tuned to the Na D_1 line generates nanosecond pulses (about 5 ns) with a repetition rate of up to 6 MHz and a peak power of 1 W. Its wavelength is only controlled by a birefringent filter. The pulse train is split into a strong circularly polarized pump beam and a weak probe beam with linear polarization. Both beams are adjusted to simultaneously overlap within the sodium vapor which is contained in a heated cell with density numbers of 10^{11} cm^{-3} in an Ar buffer gas pressure of about 10 mbar. The sample is subjected to a magnetic field with the field vector parallel to the probe beam polarization. A photomultiplier monitors the transmitted probe beam intensity behind the crossed polarizer and a multichannel analyzer stores the time-averaged signal.

Two types of experiments were performed: (i) The pulse repetition rate ν_m was kept fixed at 3 MHz while the magnetic field was swept. Ground-state periodic excitation resonances occured whenever the pulse repetition rate ν_m or its multiples $q\nu_m$ equaled an atomic Larmor frequency (see Fig. 2). For $q = 1$, only a single resonance was observed. However, higher-order resonances corresponding to higher magnetic fields showed a substructure due to the nonlinearity of the Zeeman splitting in the sodium ground state. Corresponding to the experimental geometry, the detected resonances could only result from coherences between adjacent Zeeman levels of the hyperfine states with $F = 1$ and $F = 2$, respectively. Six spectral components are expected,[9] all of which could be clearly resolved. Resonances originating from the $3s\ ^2P_{1/2}$ state could not show up since its lifetime ($\tau \simeq 15$ ns) is short compared to $1/\nu_m$. (ii) In a second type of experiment the magnetic field was kept fixed near a value where the 7th order ($q = 7$) Zeeman resonances occured for $\nu_m = 3$ MHz. When the pulse repetition rate was electronically varied around 3 MHz, these high order resonances showed up

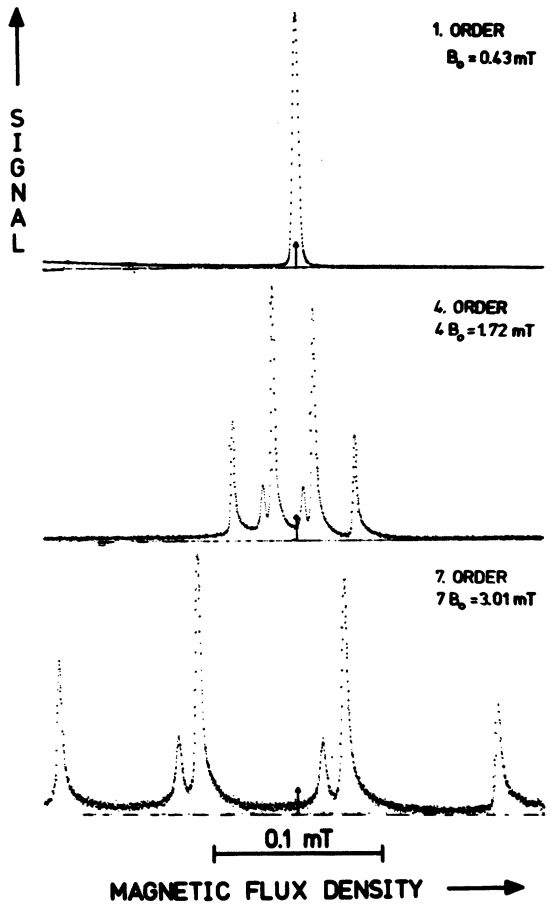

FIG. 2. Normalized intensities of the periodic excitation spectra of the qth-order Zeeman resonances. The arrows indicate the positions of the magnetic flux densities qB_0 for $q=1$, 4, and 7. The pulse repetition rate is 3 MHz.

as a function of ν_m. This is shown in Fig. 3 where the abscissa represents $7\nu_m$. The linewidth of the resonances is smaller than 20 kHz (FWHM), and seems to be mainly determined by transit time broadening and magnetic field inhomogeneties. No influence of the pulse intensity on the linewidth was observed so far. The accuracy in the determination of the position of the resonances suffers from a drift of the magnetic field, which was a few parts per thousand during the measurement.

It should be noted that experiments with pulse trains have been recently performed in ladder-type three-level systems.[10,11] These experiments made use of coherence between states connected by a two-photon transition. The method presented here might be discussed in terms of two-photon coherence in V- and Λ-type three-level systems and therefore is related to the experiments in Refs. 10 and 11. In the ladder-type three-level system, however, the oscillation frequency of

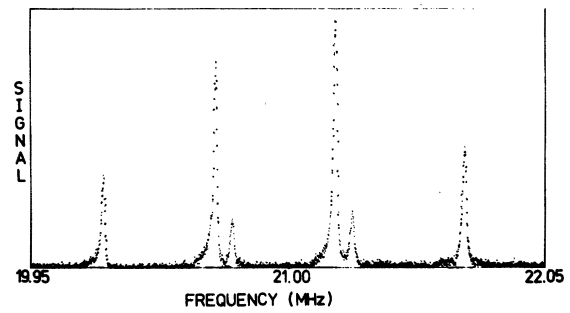

FIG. 3. 7th-order Zeeman resonances of the sodium ground state as a function of pulse repetition rate ν_m for a fixed magnetic flux density of 3.01 mT. The frequency scale represents $7\nu_m$. The following parentheses identify the Zeeman sublevels of the hyperfine levels involved in the resonances from left to right: $(F=2;\ m=2,\ m'=1)$, $(2;1,0)$, $(1;0,1)$, $(2;0,-1)$, $(1;-1,0)$, $(2;-1,-2)$.

the coherence is in the optical range and the interaction of the sample with subsequent pulses depends critically on the relative phase between the light field and the atomic oscillation at the two-photon transition frequency. In contrast, the process of creation of coherence in our experiment can be discussed in terms of resonant Raman-type two-photon processes between nearly degenerate levels; in the limit of short pulses these processes depend on the absolute square of the electric light field and consequently are not sensitive to the optical phase.

The experiments reported here demonstrate the feasibility of periodic excitation spectroscopy using infinite pulse trains. Most remarkably, resonance widths orders of magnitude smaller than the bandwidth of the light source can be obtained. Thus the method should prove especially useful in the determination of very narrow energy splittings; in this application the requirements on the laser light source are very modest. The experimental simplicity of the method is combined with high sensitivity; moreover, it is applicable to fluorescing and nonfluorescing states. We expect that this technique will prove useful in different branches of spectroscopy.

Note added in proof. In the meantime resonance halfwidths (FWHM) below 1 kHz have been obtained. Moreover, very recently, we observed high order ($q>400$) periodic excitation resonances originating from levels separated by 1.8 GHz; in this case a picosecond pulse train of only a few MHz repetition rate was used (H. Harde, H. Burggraf, unpublished).

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[1]Coherence between closely spaced states is referred to as "Hertzian" throughout this report; it includes coherence between Zeeman or hyperfine structure substates.

[2]See, e.g., S. Haroche, in *High Resolution Laser Spectroscopy*, edited by K. Shimoda (Springer, Heidelberg, 1976), p. 253.

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