

Observation of Oscillations in Resonance Absorption from a Coherent Superposition of Atomic States*

Theodore W. Ducas, Michael G. Littman, and Myron L. Zimmerman

Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 10 November 1975)

A technique is described for high-resolution spectroscopy using resonance absorption from a laser to monitor the time evolution of a coherently excited superposition of atomic states. As a demonstration a measurement was made of the $3^2P_{1/2}$ hyperfine splitting in sodium.

We have used resonant absorption to monitor the time evolution of a coherent superposition of atomic states, providing a measure of closely spaced structure. A pulsed tunable dye laser prepared a superposition of the two hyperfine components of the sodium $3^2P_{1/2}$ level. Modulations at the hyperfine frequency in the transition rate to a high $n^2S_{1/2}$ level were observed as a function of the delay of a second pulsed laser.

Both this method and quantum beat spectroscopy (QBS),¹⁻⁴ where the modulations of the coherent superposition state are observed in fluorescent decay, eliminate the Doppler width as a spectroscopic limit. In contrast to QBS, probing the modulations by resonant absorption allows structure to be measured in levels whose radiative decay is either difficult to observe or improbable (as in metastable levels). Furthermore with short laser pulses, better resolution can be achieved than that obtained in those QBS

fluorescence techniques which depend upon the timing characteristics of photon detectors. This method (using picosecond lasers) can have resolution comparable to beam-foil spectroscopy,⁴ while providing the additional feature of state-selective laser excitation.

Figure 1 shows the relevant energy levels in Na^{23} , where $\Delta\omega$ is the separation between the $F=2$ and $F=1$ hyperfine components of the $3^2P_{1/2}$ level. Here \vec{F} ($\vec{F}=\vec{I}+\vec{J}$) is the total angular momentum, \vec{I} is the nuclear spin angular momentum ($I=\frac{3}{2}$), and \vec{J} is the electronic angular momentum ($J=\frac{1}{2}$). A laser pulse, tuned to the $3^2S_{1/2}-3^2P_{1/2}$ transition with a duration $\Delta t \ll 2\pi/\Delta\omega$, will excite a coherent superposition of the two hyperfine components of the $3^2P_{1/2}$ level. The bold sublevels in Fig. 1(a) illustrate the creation of a coherent superposition state of the $F=2, m_F=0$ and $F=1, m_F=0$ sublevels by a right-hand-circularly-polarized (rhcp) laser pulse. This state can be written as

$$\psi(t) = [a|1, 0\rangle \exp(-i\omega_1 t) + b|2, 0\rangle \exp(-i\omega_2 t)] \exp(-t/2\tau), \quad (1)$$

where $\hbar\omega_1$ and $\hbar\omega_2$ are the energies of the $F=1$ and $F=2$ levels, respectively, a and b are time-independent constants, and τ is the radiative lifetime of the $3^2P_{1/2}$ level ($\tau \approx 16 \text{ nsec}^5$).

It is convenient to transform to a product representation, $|I, m_I\rangle |J, m_J\rangle$ [see Fig. 1(b)].⁶ In this representation the state in Eq. (1) can be rewritten as

$$\psi(t) = [c \sin(\frac{1}{2}\Delta\omega t) | \frac{3}{2}, +\frac{1}{2} \rangle | \frac{1}{2}, -\frac{1}{2} \rangle + d \cos(\frac{1}{2}\Delta\omega t) | \frac{3}{2}, -\frac{1}{2} \rangle | \frac{1}{2}, +\frac{1}{2} \rangle] \exp[-\frac{1}{2}i(\omega_1 + \omega_2)t] \exp(-t/2\tau), \quad (2)$$

where c and d are constants. The hyperfine coupling is manifest in the coefficients of the product wave function which oscillate with a characteristic frequency of the hyperfine splitting. At $t=0$, $\psi(t)$ is entirely $m_J = +\frac{1}{2}$ in character; at $t=\pi/\Delta\omega$ it has evolved to be completely $m_J = -\frac{1}{2}$ in nature. These oscillations can be monitored by resonant absorption of a short pulse of light ($\Delta t \ll 2\pi/\Delta\omega$) to a still higher $n^2S_{1/2}$ level. This is illustrated in Fig. 1(b) where the probe laser is rhcp. The population of the $n^2S_{1/2}$ level is a measure of the $m_J = -\frac{1}{2}$ character of this inter-

mediate level. We have considered only a single set of sublevels; for a complete analysis we must consider all sublevels connected by the successive excitations. For the case where both the preparation and probe lasers have the same sense of circular polarization the $n^2S_{1/2}$ population is

$$N_{\text{same}} = A \sin^2(\frac{1}{2}\Delta\omega t) e^{-t/\tau}, \quad (3)$$

where t is the delay time between the two laser pulses. If the lasers have the opposite sense of

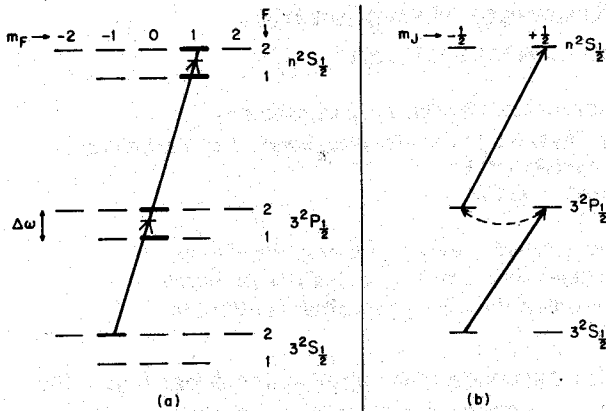


FIG. 1. Energy level diagram for relevant levels in sodium (a) as viewed in the $|F, m_F\rangle$ representation and (b) bold levels as viewed in the $|I, m_I\rangle |J, m_J\rangle$ product representation.

circular polarization, the population is

$$N_{\text{opp.}} = A \left[\cos^2\left(\frac{1}{2}\Delta\omega t\right) + \frac{5}{8} \right] e^{-t/\tau}. \quad (4)$$

A schematic of the experimental apparatus is shown in Fig. 2. The primary features of the arrangement include two pulsed lasers with variable delay time with respect to each other, an atomic beam source, and a detector of the highly excited Na atoms which uses electric field ionization.⁷

The two pulsed tunable dye lasers were pumped by a common N_2 pulsed uv laser. Laser 1 was tuned to the $3^2S_{1/2} \rightarrow 3^2P_{1/2}$ transition in sodium at 5896 Å. Laser 2 was tuned to $3^2P_{1/2} \rightarrow 2^2S_{1/2}$ transition at 4135 Å. Both lasers exhibited peak powers of approximately 100 W, pulse widths of 2 nsec, and spectral widths of about 0.5 cm^{-1} . The low peak power of the lasers was the direct result of making the laser cavities as long as possible in an attempt to reduce the pulse width of the lasers. The pulse width of the lasers was the primary limitation on resolution.

The oscillation period was in the nanosecond regime and a variable optical path length provided a convenient way of accurately delaying the second laser pulse with respect to the first. The delay line consisted of a laser collimator, a long (7 ft) optical bench, and a "roof" prism. Delays as long as 14 nsec were readily obtained, allowing us to observe two cycles in the resonance oscillations of the $3^2P_{1/2}$ level. The collimator served to maintain the image size as the delay was changed.

The light from laser 2 was always rhcp, and the light from laser 1 could be made either rhcp

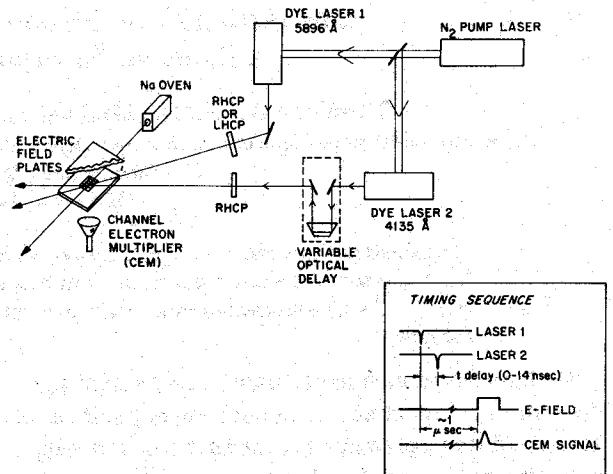


FIG. 2. Schematic diagram of experimental arrangement. Also shown is the timing sequence of laser excitation and electric field ionization.

or lhcp. The two nearly collinear laser beams intersected the atomic beam between electric field plates.

The Na atomic beam provided a density of 10^8 – 10^{10} atoms/cm³ in the interaction region. The background pressure in the apparatus was 10^{-6} Torr. The number of atoms in the $2^2S_{1/2}$ level was measured by an ionization detector. Approximately 1 μsec after the lasers excited the $2^2S_{1/2}$ level in a field-free region, a 4 kV/cm electric field pulse was applied which ionized all atoms in the $2^2S_{1/2}$ state, but none in the $3^2P_{1/2}$ state. The average number of ions for about twenty laser pulses was monitored by a channel electron multiplier followed by a gated integrator.

The population of the $2^2S_{1/2}$ level was monitored versus the delay of laser 2 with respect to laser 1. Data for N_{same} and $N_{\text{opp.}}$ were recorded for delays in 0.5-nsec steps. By taking the ratio $N_{\text{same}}/N_{\text{opp.}}$, errors due to variations in photon flux, and atomic-beam flux were reduced. From Eqs. (3) and (4), for short laser pulses ($\Delta t \ll 2\pi/\Delta\omega$) the ratio is given by

$$\frac{N_{\text{same}}}{N_{\text{opp.}}} = \frac{\sin^2 \frac{1}{2} \Delta\omega t}{\frac{5}{8} + \cos^2 \frac{1}{2} \Delta\omega t}. \quad (5)$$

Figure 3 shows data from a typical run. The error bar shown represents the limit of error due primarily to fluctuations in the laser intensity. The data were least-squares fitted with the above expression modified for the finite length of the laser pulse. The total system response

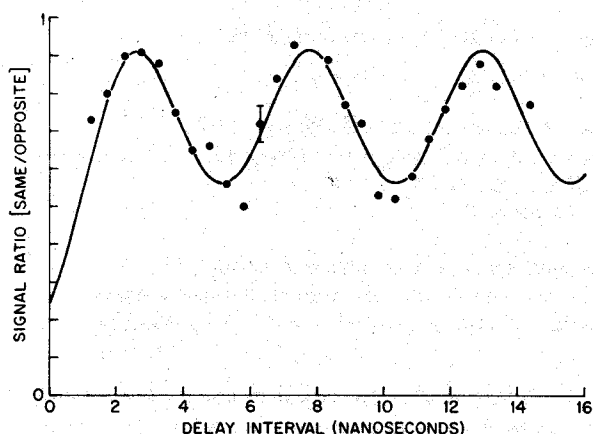


FIG. 3. Experimental results. The points plotted are the ratios of signals for same sense to opposite sense excitation versus delay between preparing and probing laser. The smooth curve is the least squares fit to the data (see text).

function in this case is the convolution of the time profile of the preparing laser with the profile of the probe laser. Thus the population of the $20^2S_{1/2}$ level in time is the convolution of this system response function with Eqs. (3) and (4) for the cases of same and opposite polarization, respectively. We modeled each of the laser pulses with a square-shaped function. The value for the hyperfine frequency for the $3^2P_{1/2}$ state in sodium was found to be 190(5) MHz. This measurement is in excellent agreement with previous experimental determinations of this quantity.⁸⁻¹⁰

We have shown the feasibility of measuring closely spaced structure using resonant absorption from a coherent superposition state. Sodium was studied for experimental convenience, and the $3^2P_{1/2}$ hfs was chosen because it was the simplest structure to analyze. Although we used an atomic beam, this experiment could easily have been performed in a cell with comparable precision since the technique eliminates Doppler broadening and leaves only the natural linewidth

as a spectroscopic limit.

In addition to its applications in atomic spectroscopy, this technique can be used to great advantage in studying molecules because it does not depend upon the observation of fluorescence. This reduces problems encountered in molecular levels with many possible channels of radiative decay, and may allow investigation of systems which decay nonradiatively. These are just some examples of how the use of pulsed radiation to probe the resonance oscillations of coherently excited states can provide a means of measuring structure in systems not amenable to other spectroscopic methods.

We would like to thank Dr. Richard R. Freeman for his many helpful suggestions concerning this work.

*Work supported by the U. S. Air Force Office of Scientific Research (Contract No. F44620-72-C-0057).

¹S. Haroche, J. A. Paisner, and A. L. Schawlow, *Phys. Rev. Lett.* **30**, 948 (1973).

²W. Gornik, D. Kaiser, W. Lange, J. Luther, and H. H. Schulz, *Opt. Commun.* **6**, 948 (1972).

³A. Corney and G. W. Series, *Proc. Phys. Soc., London* **83**, 207 (1964).

⁴H. J. Andr , *Phys. Rev. Lett.* **25**, 325 (1970).

⁵W. L. Wiese, *Atomic Transition Probabilities*, U. S. National Bureau of Standards, National Standards Reference Data Series—4 (U.S. GPO, Washington, D.C. 1966), Vol. 1.

⁶For excitation rapid compared to the hyperfine period, and slow compared to the fine-structure period, the electric dipole selection rule for rhcp excitation is $\Delta m_j = +1$.

⁷T. W. Ducas, M. G. Littman, R. R. Freeman, and D. Kleppner, *Phys. Rev. Lett.* **35**, 366 (1975).

⁸M. L. Perl, I. I. Rabi, and B. Sentzky, *Phys. Rev.* **98**, 611 (1955).

⁹T. W. H nsch, I. S. Shahin, and A. L. Schawlow, *Phys. Rev. Lett.* **27**, 707 (1971).

¹⁰H. T. Duong, P. Jacquinet, S. Liberman, J. L. Picque, J. Pinard, and J. L. Vialle, *Opt. Commun.* **7**, 371 (1973).