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High-Resolution Two-Photon Spectroscopy with Picosecond Light Pulses

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We have demonstrated the feasibility of Doppler-free two-photon spectroscopy with a train of picosecond standing-wave light pulses from a synchronously pumped mode-locked cw dye laser. The actively controlled mode spectrum provides a means for accurate measurements of large frequency intervals. From a multipulse spectrum of the sodium $3s$-$4d$ transition we have determined a new value of the $4d$ fine-structure splitting, 1028 ± 0.4 MHz.

Two-photon excitation of gas atoms with a coherent train of standing-wave light pulses can produce fringelike spectra free of first-order Doppler broadening, which resemble the spatial diffraction pattern of a multislit grating or the Ramsey fringes of molecular beam spectroscopy with multiple spatially separated rf field regions.[1-5] Shortly after the possibility of Doppler-free two-photon spectroscopy with coherent pulse trains was first suggested,[1] narrow spectral fringes were observed by exciting the sodium $3s$-$5s$ transition with nanosecond pulses recirculating inside a passive optical resonator.[2]

In this paper we report on experiments which establish the feasibility of high-resolution spectroscopy with a synchronously pumped actively mode-locked cw dye laser. We have recorded Doppler-free spectra of the sodium $3s$-$4d$ transition with pulse ranging in length from 500 psec to less than 1 psec. The actively controlled frequency spacing of the phase-locked laser modes provides a precise frequency calibration scale. This makes it possible to apply accurate electronic frequency counting techniques to the measurement of large line separations, up to high multiples of the pulse repetition rate, where direct modulation or beat-frequency techniques would be difficult or impossible to apply. Using this calibration, we have determined a new value of the sodium $4d$ fine-structure splitting, 1028.5 ± 0.4 MHz.

The origin of the narrow spectral fringes in multipulse excitation is perhaps most easily understood in the frequency domain.[2-4] The pulse train from a mode-locked laser can be described as a superposition of phase-locked oscillating laser modes which are equally spaced in frequency.[6] Similarly, a pulse circulating inside a passive optical resonator transforms into a comb of "shock-excited" cavity modes. Resonant two-photon excitation is possible whenever the sum of two mode frequencies coincides with an atomic transition frequency. Many pairs of modes can satisfy this resonance condition at the same time and contribute to the resonant excitation, and a detailed analysis must consider the spatial field distribution and phases of all these modes.[3,4] If the atoms are in a region where two counterpropagating pulses form a standing-wave field, this excitation can be nearly free of first-order Doppler broadening. The signal magnitude is of the same order as under cw excitation with a single-frequency laser of the same average power.

The multipulse excitation can also be described in the time domain.[1,2,5] Atoms which have been excited to a coherent superposition of ground

state and upper state continue to oscillate at their resonance frequency in the dark period between pulses. The interaction with subsequent pulses will either further excite the atoms or de-excite them by stimulated two-photon emission depending on the relative phase between light field and atomic oscillations. A fringe maximum occurs when all pulses add coherently to the excitation amplitude.

A diagram of our appartus is shown in Fig. 1. A cw argon-ion laser (Spectra-Physics Model No. 171) of cavity length 182 cm was actively mode locked, using an acousto-optic prism modulator (Harris Corporation Part No. 536-940) driven by the amplified output of a stable frequency synthesizer (General Radio Model No. 1164-A). The driving frequency was monitored by a digital frequency counter (Hewlett-Packard Model No. 5381A). The mode-locked argon laser produced pulses of approximately 200 psec duration and 82.32 MHz repetition rate.

This argon laser was used to pump synchronously a modified commerical jet-stream rhodamine-6G dye laser (Spectra-Physics Model No. 375). The dye-laser cavity length was extended to equal half the length of the argon laser (91 cm). The dye laser was therefore only pumped on every second pulse round trip and its repetition rate was twice that of the pump laser. The plane output coupler was replaced by a mirror of 85 cm radius, mounted on a piezotransducer. The cavity length was rigidly maintained by a fused-silica tube and the whole laser was kept in a sealed enclosure and mounted on a vibrationally isolated optical table.

For most measurements, the laser bandwidth was restricted by a tuning wedge filter with a free spectral range (FSR) of 100 THz, two uncoated solid quartz intracavity etalons with FSR of 1 THz and 200 GHz, and a temperature-controlled air-spaced piezotuned etalon of 75-GHz FSR and 80% coating reflectivity. The dye-laser

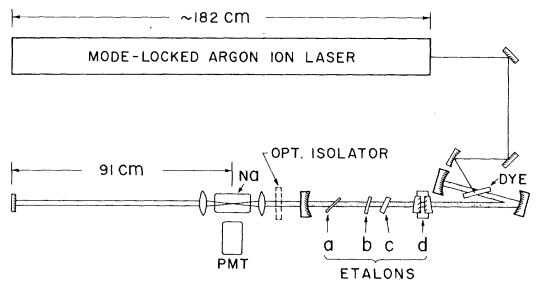

FIG. 1. Scheme of apparatus.

spectrum was monitored with a scanning optical spectrum analyzer. Stable mode locking was obtained when the cavity length was matched to within 2.5 mm of half the argon-laser length. In this case, about 6 to 8 oscillating modes were observed with a stable near-Gaussian intensity distribution of approximately 1 GHz width, corresponding to a transform-limited pulse duration near 500 psec. The pulse length decreased to about 100 psec, if the tunable 75-GHz etalon was replaced by an uncoated fused-silica etalon of 50-GHz FSR. The average dye-laser output power was typically 50 mW. Pulses down to less than 1 psec could be obtained by removing all intracavity etalons except for the tuning wedge, and by carefully adjusting the cavity length. This short pulse length was monitored by measuring the autocorrelation function of the pulses, using crossed-beam second-harmonic generation in a potassium dihydrogen phosphate crystal.[7]

The output of the dye laser was passed through an optical isolator, consisting of a linear polarizer and a quarter-wave plate, to prevent retroreflection. The beam was focused to a 100-μm spot size, and then collimated by a lens and reflected back by a plane mirror. The distance between the focal spot and mirror was adjusted to equal the dye-laser cavity length. This arrangement allowed each pulse to meet its counterpropagating predecessor, thus forming a pulsed standing-wave field at the focus.

An evacuated Pyrex cell containing sodium metal was placed at the focus of the dye-laser beam. The cell was heated to a temperature of 175°C, corresponding to a sodium vapor density near 10^{+12} cm^{-3}. Two-photon excitation of the $3s$-$4d$ transition was monitored by observing the uv fluorescence from the cascade decay of the $4p$ state to the $3s$ state. The output of the photomultiplier (RCA 8850) was amplified and directly fed to a strip chart recorder.

Figure 2 shows a typical multipulse Doppler-free two-photon spectrum of the $3s$-$4d$ transition in sodium, recorded at 500 psec pulse duration. The dye laser was tuned to half the $3s$-$4d$ transition frequency ($\lambda = 578.732$ nm), and the mode spectrum, as observed in the spectrum analyzer, was translated continuously in frequency over several gigahertz by simultaneously scanning the cavity length and the air-spaced intracavity etalon. Imperfect synchronization caused some "slippage" of the modes relative to the envelope which was unimportant for the measurements reported here. (Only the wavelength and the rela-

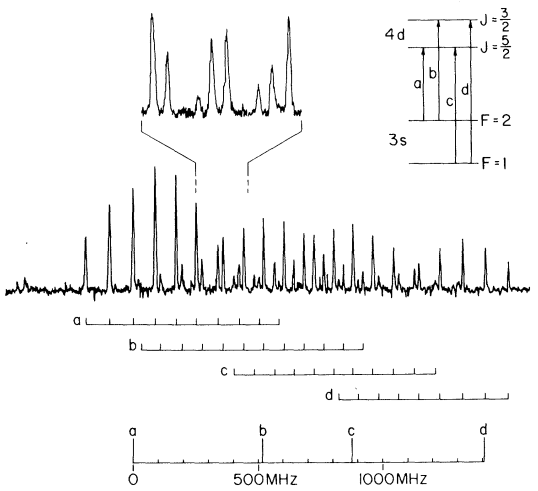

FIG. 2. Multipulse two-photon spectrum of the sodium $3s$-$4d$ transition (center) with expanded portion shown above, recorded with pulses of 500 psec duration. The entire comb of laser modes has been scanned in frequency with the help of piezotranslators. Each of the four line components a to d appears as a comb of narrow fringes, identified by the markers below. The fringe spacing equals half the pulse repetition rate.

tive phase of the light pulses were varied during the scan. The modulation frequency remained fixed, and the delay time between successive pulses did not change, unlike in the earlier experiment employing a passive resonator.[2])

Each of the four line components a to d appear as a comb of resonances, separated by half the laser intermode frequency spacing. The markers below help identify the four sets of fringes. An expanded portion of the spectrum, recorded at a slow scan speed of 5 MHz/sec, is displayed at the top of Fig. 2. The observed linewidth was typically about 4 MHz (full width at the half-maximum in the visible), both at 500-psec and at 100-psec pulse length. The natural linewidth due to the 50-nsec lifetime of the $4d$ state is about 1.6 MHz, for comparison. The larger observed width is ascribed to transit-time broadening and laser frequency jitter. Pressure broadening should be entirely negligible. ac Stark shifts should be the same as under cw excitation with the same average power level, i.e., less than 30 kHz.

As is apparent from Fig. 2, the combs of fringes and closely spaced transitions appear interleaved, and the line separations can be determined modulo the fringe period, by measuring the small frequency difference between adjacent fringes. If one has prior knowledge of the line separation to an accuracy better than the fringe spacing, one can determine the actual value of the atomic line splitting. A single scan permits many individual readings, and any systematic nonlinearities of the laser scan are readily revealed by the fringe spacings within a single comb. In this respect, multipulse spectroscopy resembles conventional Fabry-Perot interferometry. However, an actively mode-locked laser has an important advantage over a passive interferometer. If all modes are properly phase locked, their spacing is rigidly determined by the modulation frequency and is not affected by the exact resonator length, by the dispersion of intracavity optical elements, or by phase shift in the mirror coatings. The resulting fringe pattern thus lays down an exact frequency calibration over the bandwidth of a single pulse. Of course this technique will be useful only for sufficiently simple spectra.

To demonstrate the power of this calibration method, we have used it to determine the fine-structure splitting of the Na $4d$ level. The spacing between adjacent fringes in each of the combs is 82.31958 MHz in the visible, as determined from the pulse repetition rate or from the modulation frequency of the acousto-optic prism mode-locking device. The upper-state fine-structure splitting is equal to twice the separation between the a and b fringes as well as between the c and d fringes. This separation is known to be on the order of 500 MHz or $6+\delta n$ fringe intervals. The fractional separation was determined to be $\delta n = 0.247 \pm 0.002$ with a statistical confidence level of 90%, by averaging over 50 readings. This gives a $4d$ fine-structure splitting of 1028.4 ± 0.4 MHz. Our measurement is consistent with, and more accurate than the previous measurement of 1028.3 ± 0.6 MHz, obtained by the level-crossing method.[8] It is almost an order of magnitude more accurate than previous optical measurements by cw two-photon spectroscopy.[9] We have also determined the ground-state hyperfine splitting in the same way from the separation between the a and c fringes and the b and d fringes. The obtained value, 1771.6 ± 0.4 MHz, is in excellent agreement with the well-known result from radio-frequency measurements.[10]

Exciting the sodium $3s$-$4d$ transition with ultrashort pulses, with durations near 1 psec, we have recorded clean multipulse fringes of about 15-MHz bandwidth. The larger width was at least partly caused by increased laser frequency jitter, because the cavity-stabilizing quartz tube was

removed for these preliminary measurements. Slow phase jitter due to external perturbations produces a finite bandwidth for all oscillating modes, which is expected to be the same as the laser bandwidth under single-frequency operation. Electronic feedback stabilization of the laser resonator, for instance by locking one of the modes to a passive reference cavity, should certainly be feasible, and should make it possible to achieve mode linewidths down to the kilohertz region. Such ultrashort pulses should then permit one to make very accurate measurements of line splitting as large as 1000 GHz.

Pulses of 1 psec duration produce a standing-wave region which is only 0.3 mm long. The Doppler-free signal in our experiment appeared reduced because of the resultant small effective volume. Moreover, atoms traversing this field in the longitudinal direction can now contribute significantly to the transit-time line broadening. In the frequency domain, such additional broadening is expected, because the Doppler effect is only partially canceled in two-photon excitation with two modes of widely different frequencies.

The new technique holds particular promise for Doppler-free excitation of the hydrogen 1S-2S transition,[11] using a frequency-doubled synchronously pumped blue dye laser. The signal will be dramatically enhanced over a similar experiment with a cw dye laser, because the efficiency of second-harmonic generation is increased by a factor equal to the inverse duty cycle, and the two-photon excitation rate depends on the square of the average ultraviolet power. At a pulse length of 5 psec and a round-trip time of 10 nsec, this signal enhancement should exeed one million. The broad bandwidth of the ultrashort pulses should allow the hydrogen-to-deuterium isotope shift (168 GHz in the visible) to be measured with high precision, and thereby establish a more accurate value of the ratio of electron mass to proton mass.

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Note added.—By improving the mechanical stability of the mode-locked dye laser we have observed linewidths as narrow as 4 MHz using 1-psec pulses to excite the Na $3s \rightarrow 5s$ two-photon transition.

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