

Two-photon Fourier spectroscopy with femtosecond light pulses

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We demonstrate a Fourier spectrometer that uses intense ultrashort laser pulses. By exciting the $6S_{1/2}-8S_{1/2}$ two-photon transition in atomic cesium vapor, we are able to measure the small hyperfine splitting of the $8S_{1/2}$ excited state. This technique, combining a high spectral resolution with the high peak intensities available to femtosecond laser systems, may offer intriguing opportunities for the study of multiphoton transitions and for spectroscopy in the short-wavelength region. © 1997 Optical Society of America

The current generation of mode-locked laser systems provides ready access to ultrashort light pulses of high intensity, greatly facilitating the study of ultrafast and highly nonlinear phenomena. Unavoidably, these sources have an intrinsic large spectral bandwidth, so their usefulness for high-resolution spectroscopy may appear limited.

In this Letter we demonstrate a Fourier spectrometer that uses femtosecond laser pulses and combines a relatively high resolution with a high peak intensity. By exciting the $6S_{1/2}-8S_{1/2}$ two-photon transition in atomic cesium vapor, we are able to measure the hyperfine splitting of the $8S_{1/2}$ level, which is considerably smaller than the spectral bandwidth of a single laser pulse.

One can realize a simple Fourier spectrometer by spectrally filtering white light with a Michelson interferometer and by recording the absorption or excitation of an atomic sample as a function of the optical delay time T between the two interferometer arms. The interferometer modulates the broadband spectrum of the light source with its comb of sinusoidal transmission maxima. The frequency spacing between adjacent spectral fringes is given by $1/T$. As the optical delay time T is changed, these spectral fringes scan across the spectrum. The spectrum of the atomic sample is obtained by Fourier transformation of the recorded data.

For this type of linear Fourier spectroscopy the white-light source can obviously be replaced with a broadband femtosecond laser with a bandwidth determined by the inverse pulse length $1/\tau$. While the wide spectral bandwidth still makes it possible to record survey-type optical spectra, the peak intensity of this spectrally filtered light is independent of the time T . For a femtosecond laser of high intensity, the light transmitted by a Michelson interferometer is therefore still capable of exciting two-photon and multiphoton transitions in atoms or molecules or of producing high harmonics in a gas jet up into the extreme ultraviolet, regardless of the desired spectral resolution $1/T$.

It is now instructive to look at the Michelson interferometer in the time domain. Each input pulse is split into two mutually phase-coherent output pulses

with a variable delay time T . The response of an atomic resonance to such light can be understood in terms of a Ramsey-type process in which the excitation amplitudes of the two pulses add coherently as long as the pulse delay T remains smaller than the atomic dephasing time.¹ In a simple two-level system the first pulse induces a coherent superposition of the ground and the excited states, so that the atom keeps oscillating at the transition eigenfrequency after the passage of the pulse and before dephasing effects become important. Depending on the phase of the second pulse relative to this induced polarization, the transfer of atomic population from the ground to the excited level can be enhanced or suppressed, and time-domain Ramsey fringes appear in the fluorescence signal from the upper state. If the phases of the pulses are accurately locked, one can obtain the absolute frequency of the transition as the inverse of the fringe period. If two nearby transitions at ω_1 and ω_2 are simultaneously excited by the broadband laser pulses, both transitions generate a fringe pattern with a slightly different period, and one can observe a beat note between the two. In fact, after a time delay equal to $(2n + 1)\pi/|\omega_1 - \omega_2|$ from the first pulse, the two polarizations are exactly out of phase and the second pulse acts on them in opposite ways, with no net transfer of population between the ground and the excited levels. On the other hand, if the second pulse arrives after the full beat period $(2n\pi/|\omega_1 - \omega_2|)$, the two polarizations excited by the first pulse are back in phase and the second pulse can interfere constructively or destructively with both of them (giving maximum or minimum transfer to the excited levels), depending on its phase. As a result, one can observe a sinusoidal modulation of the fringe contrast with a time period equal to the inverse of the frequency separation between the two nearby transitions. A measure of the period of the modulation in the fringe contrast hence yields directly the frequency separation between the investigated lines.

The intriguing opportunities for this type of high-resolution laser spectroscopy with intense femtosecond laser pulses have so far gone largely unexplored, even though the feasibility of Ramsey-type high-resolution

spectroscopy with multiple phase-coherent light pulses was discussed and demonstrated long ago.²⁻⁵ In most previous studies the time delay between the pulses was kept fixed while the central wavelength of the laser was scanned over the investigated transitions. In the present experiment, on the other hand, the emission wavelength of the laser does not need to be controlled or stabilized, and it may drift and fluctuate as long as the investigated transitions still fall within the laser bandwidth. In this case the spectral resolution is merely given by the time delay between the pulses and does not depend on the bandwidth of the source. As long as one is not interested in absolute frequencies but only in frequency splittings or coherence decay times, the relative phase between the two pulses need not be controlled either. It has been shown⁶ that one can introduce a random relative phase jitter without affecting the possibility of measuring the fringe contrast as a function of the delay.

This technique also presents a definite advantage with respect to conventional cw laser spectroscopy in high-resolution studies of multiphoton resonances. In fact, when one is trying to investigate the same transitions with high-power cw laser sources, various intensity-related effects (such as ac Stark shifts) have to be taken into account to extract the spectroscopic data properly. On the other hand, with the technique described above, the essential information comes from the dark period between the two pulses and, during that time, the atoms experience no field and their energy levels are not perturbed by the radiation.

The experimental setup is shown in Fig. 1. We used a commercial Kerr-lens mode-locked Ti:sapphire laser (Spectra-Physics, Tsunami) at an 82-MHz repetition rate to produce 12-nJ pulses of ~ 100 -fs duration. Each pulse is directed to a Michelson interferometer to obtain two time-delayed pulses of the same intensity. We can vary the relative delay between the two pulses by moving one of the arms of the interferometer with a computer-controlled calibrated stepping motor. The two pulses are made to propagate collinearly and are focused by a 100-mm focal-length lens into a 5-cm-long gas cell kept at room temperature. The fluorescence emitted from the excited atoms of the gas is spectrally filtered and collected in a right-angle geometry by a photomultiplier (EMI 9635). The two-pulse fluorescence signal is recorded for each position of the delay line and stored in a computer.

We investigated the two-photon transition between the $6S_{1/2}$ and the $8S_{1/2}$ levels of atomic cesium. Both levels are split into two hyperfine states with quantum numbers $F = 3$ and $F = 4$. While the ground state is split by 9192.631770 MHz,⁷ the hyperfine splitting of the $8S_{1/2}$ excited state is only ~ 877 MHz.^{8,9} The selection rule for the $6S_{1/2}-8S_{1/2}$ two-photon transition is $\Delta F = 0$, and there are thus only two allowed transitions, one from each F level in the ground state to the corresponding F level in the excited state, as shown in Fig. 2.

We tuned the central wavelength of the laser to ~ 822 nm to excite both hyperfine components of the two-photon transition, and we measured the fluorescence radiation emitted around 460 nm as the atoms

spontaneously decayed from the excited levels to the ground state via the $7P$ levels. In Fig. 3 we show the time-domain Ramsey fringes that we measured by performing a fine scan of the delay between the two pulses. With a coarser scan of the delay from 0 to approximately 300 ps we observe the modulation of the fringe contrast as a result of the hyperfine splitting between the $F = 3$ and $F = 4$ levels of the two states under investigation, as shown in Fig. 4.

To measure the period of the fringe contrast modulation, we extracted smoothed rms curves from the recorded data and fitted them to damped sinusoids. The value of 8.37(6) GHz that we obtained as the inverse of such a period corresponds to the difference between the two hyperfine splittings in the ground and the excited states. If we use the known value for the $F = 3-4$ separation in the $6S_{1/2}$ level, we obtain a value for the hyperfine splitting of the $8S_{1/2}$ level of 0.82(6) GHz. This result is in agreement with measurements performed with conventional cw laser techniques^{8,9} and shows that high-resolution two-photon spectroscopy can indeed be performed with ultrashort pulsed sources.

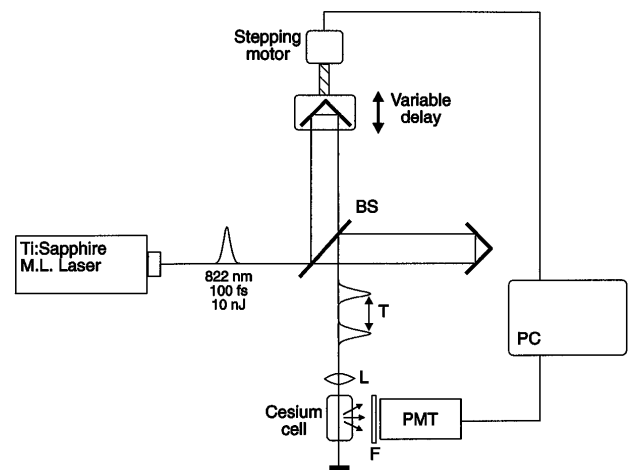


Fig. 1. Experimental setup: BS, 800-nm 50% beam splitter; L, 100-mm focal-length lens; F, filter for 460 nm; PMT, photomultiplier tube; M.L., mode-locked.

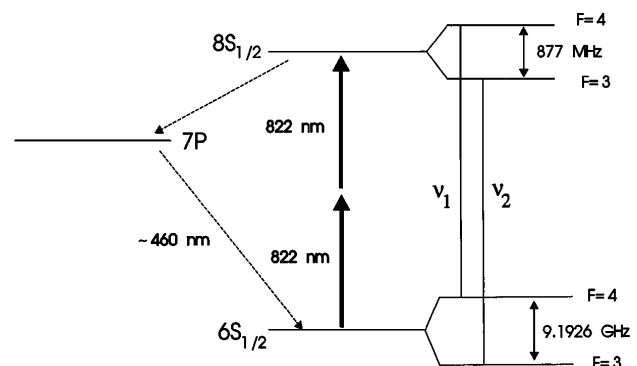


Fig. 2. Schematic drawing of the levels involved in the $6S_{1/2}-8S_{1/2}$ two-photon transition in cesium. The fluorescence signal at 460 nm is emitted as the excited atoms decay to the ground state through the $7P$ levels. The hyperfine structure of the $6S_{1/2}$ and $8S_{1/2}$ levels is also shown.

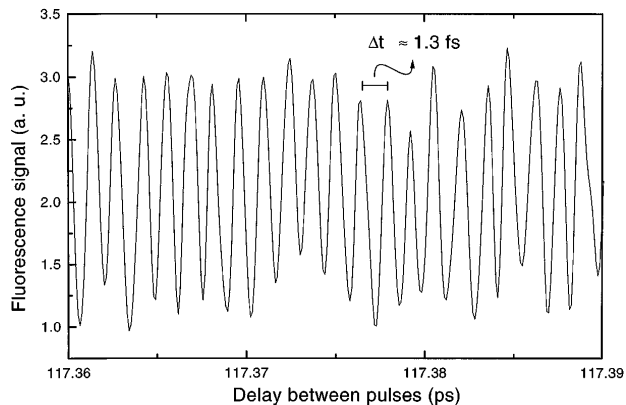


Fig. 3. Time-domain Ramsey fringes in the fluorescence signal emitted by the excited cesium atoms as a function of the time delay T between the pulses. From a measurement of the fringe period one can readily obtain a rough estimate of the atomic frequency.

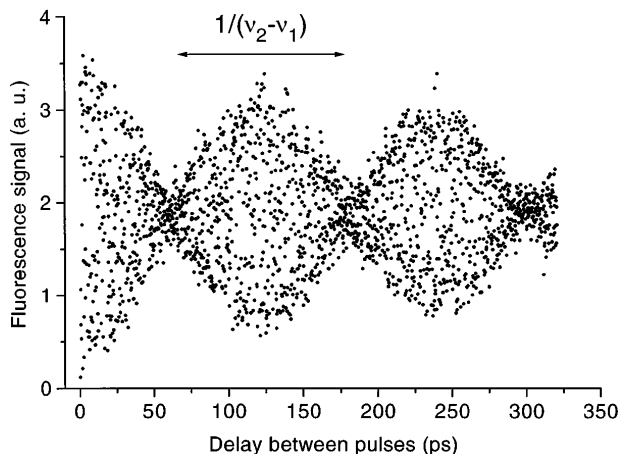


Fig. 4. Fluorescence signal as a function of the time delay T between the pulses. The periodic modulation in the fringe contrast is clearly visible and was used to determine the frequency splitting between the levels.

The main source of uncertainty in our measurement is related to the short distance that can be traveled in our delay line, which allows us to record just a few oscillations of the fringe contrast and thus limits the

precision in the determination of the period. Better accuracies could be obtained with a longer delay line and with improvements in detection efficiency, for example, by heating the cesium cell to increase the atomic density in the vapor.

Our simple experiment illustrates the potential of high-resolution multiphoton spectroscopy with ultra-short light pulses. With the higher intensities available from amplified laser systems one can extend this technique to the study of higher-order multiphoton transitions. By focusing the two time-delayed laser pulses into a gas jet, one can generate high harmonic radiation up into the extreme ultraviolet¹⁰ and then extend high-resolution Ramsey spectroscopy into an interesting spectral region that has not been accessible to laser spectroscopy in the past.

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