Heterodyne-assisted pulsed spectroscopy with a nearly Fourier-transform limited, injection-seeded optical parametric oscillator

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Narrowband pulsed 822 nm signal radiation from an injection-seeded optical parametric oscillator (OPO) system is used to record fluorescence-detected sub-Doppler two-photon excitation (TPE) spectra of atomic cesium. An optical-heterodyne technique is used to monitor the frequency chirp as well as the fluctuating central frequency of successive OPO output pulses, thereby providing a novel way to record sub-Doppler TPE spectra. The measured TPE linewidth approaches the ultimate limit imposed by the Fourier transform of the pulse's temporal profile, demonstrating the utility of this system for pulsed laser spectroscopy applications that require the highest possible resolution.

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Narrowband pulsed coherent light sources are essential for high-precision laser spectroscopy, particularly when it is necessary to perform time-resolved measurements (e.g., lidar and pump-and-probe sequences), to enhance the efficiency of nonlinear-optical spectra (e.g., multiphoton absorption and coherent Raman), or to gain access to wavelength regions where continuous-wave (cw) light sources do not exist (e.g., vacuum ultraviolet and mid infrared), by use of nonlinear-optical frequency conversion. The FWHM optical bandwidth $\Delta\nu$ of radiation from such pulsed coherent light sources can be no better than the Fourier-transform (FT) limit associated with the FWHM of pulse duration Δt. For example, the FT limit for pure Gaussian temporal and power-spectrum profiles is defined by the time–bandwidth product $\Delta t \Delta \nu = 0.441$. Frequency chirp (i.e., variation of the instantaneous frequency during each light pulse) imposes additional spectroscopic limitations, as do fluctuations in the temporal profiles and shifts in the central frequency for successive optical pulses.

In previous papers[1–4] we reported an optical-heterodyne detection (OHD) technique that is able to monitor such optical-bandwidth characteristics of signal output from an injection-seeded pulsed optical parametric oscillator (OPO) system that is continuously tunable in a single longitudinal mode (SLM). We now report a new spectroscopic method that uses OHD and demonstrates the high-resolution capability of such a nanosecond-pulsed OPO system close to the FT limit.

Our OHD technique[2] enables the frequency chirp of the OPO output to be maintained below 10 MHz by matching the wavelength of the cw seed radiation to the peak output wavelength of the free-running phase-matched OPO. We also register the central frequency of each seeded OPO output pulse with megahertz precision and show that these central frequencies conform to a quasi-Lorentzian distribution centered near the seed frequency. Such central-frequency fluctuations are presumed to be due to tiny mechanical instabilities and (or) atmospheric turbulence in the OPO cavity itself, giving rise to inhomogeneous spectroscopic broadening. We turn these fluctuations to advantage by resolving and logging each pulse's central frequency, thereby obtaining finely resolved spectra without needing to scan the seed laser's frequency, OPO cavity length, and (or) OPO phase-matching conditions.

The apparatus used in these experiments is as previously reported[3,4]: a ring-cavity OPO based on periodically poled $KTiOPO_4$ pumped at 532 nm by a long-pulse (27 ns FWHM) SLM Nd:YAG laser, injection seeded by a SLM tunable cw laser, and locked to the cw laser frequency. We use an acousto-optic modulator to separate a fraction of the cw seed beam that is frequency shifted by ~720 MHz and beat this against each OPO signal pulse to record its OHD waveform; a Fourier-transform and sideband-filtering procedure then yields the frequency chirp, central frequency, and reconstructed temporal profile of each pulse.

Modifications of our previous apparatus are described below. The 842-nm SLM cw tunable diode laser previously used as an injection seeder is replaced by a more powerful cw Ti:sapphire (Ti:S) laser (Coherent 899) whose SLM output delivers continuously tunable radiation with submegahertz FWHM optical bandwidth. To demonstrate the OPO's spectroscopic capability, we seed it at 822.47 nm to permit $6S$–$8S$ ($F=4$) two-photon excitation (TPE) of atomic cesium (Cs), with sub-Doppler spectra recorded by detecting $7P$–$6S$ fluorescence at ~460 nm.[5,6] Two Cs vapor cells heated to ~90 °C are incorporated into our apparatus, each with unfocused, retroreflected, counterpropagating light beams: One cell is excited by the cw Ti:S laser and uses a photon-counting photomulti-

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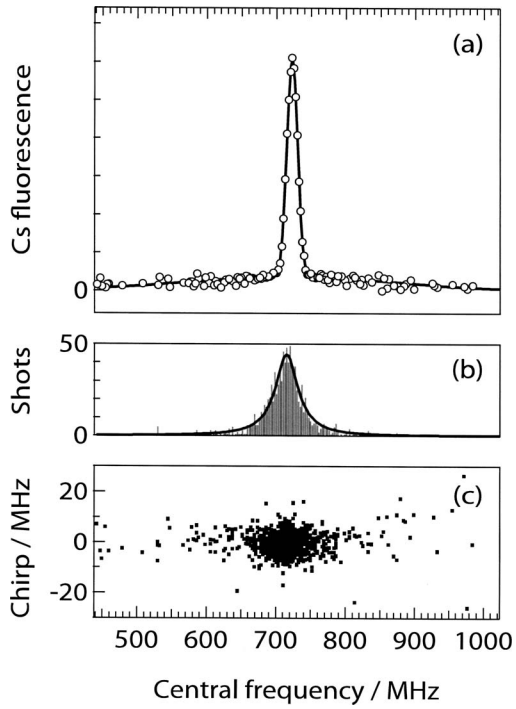

Fig. 1. (a) Cs TPE fluorescence as a function of OHD-determined central frequency with double-Gaussian fit, (b) histogram of the number of OPO shots recorded with a Lorentzian fit, (c) total linear frequency chirp between FWHM temporal power-profile limits.

plier for fluorescence detection, while the other is excited by the pulsed OPO and uses current-mode photomultiplier detection. An optical parametric amplifier (OPA) stage, based on birefringently phase-matched $LiNbO_3$, can be employed for higher-power measurements. The OPA process also happens to reduce the pulse duration.

As the SLM Ti:S laser is tuned, the cw spectrum yields a linewidth on the Ti:S frequency scale of ~2 MHz FWHM, consistent with previous TPE observations[5,6] By holding the cw seed frequency at the peak of the 822.47 nm line, we can adjust the phase-matching conditions of the periodically poled $KTi:OPO_4$ OPO[1–4] to vary the frequency chirp of the OPO signal output. The results presented in Fig. 1 were obtained under zero-chirp conditions (i.e., perfectly phase matched), with the OPO pumped by ~20 μJ of 532 nm energy (2.5 times its free-running threshold) and delivering a total of ~4.5 μJ/pulse of SLM signal output.

Figure 1(a) shows an averaged *pulsed* TPE spectrum that we obtained by binning the central frequency of each OPO pulse as determined by our OHD technique. Two Gaussian functions with a common peak frequency satisfactorily fit the prominent, narrow sub-Doppler peak and its weak underlying Doppler-limited pedestal. On the OHD-beat frequency scale, the TPE peak is centered at 722.7 ± 0.1 MHz, and its sub-Doppler FWHM linewidth $\Delta\nu_b$ is 18.0 ± 0.2 MHz. It is particularly significant that the spectrum in Fig. 1(a) is recorded without active scanning of the seed wavelength or variation of the OPO operating parameters, apart from the naturally occurring minor instabilities that give rise to the central-frequency fluctuations. Consequently, the attainable spectroscopic resolution is not limited by the inhomogeneous broadening that these fluctuations would impose if the OPO frequency had simply been tuned without using our novel OHD-based binning approach. It would be possible to extend the range of such spectra by, for example, using two stable cw SLM lasers: one as a reference locked to the atomic resonance and the other as the OPO seed with tunable frequency.

The histogram in Fig. 1(b) displays the corresponding distribution of central frequencies for successive OPO signal pulses. The average central frequency is 713 MHz (i.e., ~10 MHz below the sideband frequency generated by the acousto-optic modulator) with a 53 MHz 1σ Lorentzian distribution, which is sufficiently wide to span the Cs TPE spectrum, as in Fig. 1(a).

The corresponding total frequency chirp (measured by our linear fit approach[1–4] between the FWHM intensity limits) is plotted in Fig. 1(c) for more than 10^3 successive OPO signal pulses, as a function of each pulse's OHD-determined central frequency. The frequency chirp values in Fig. 1(c) are scattered ($1\sigma = 5.6$ MHz) about a mean of −0.25 MHz (i.e., effectively zero chirp).

We performed a series of similar experiments with the OPO system operated under assorted chirp conditions. We also employed a single $LiNbO_3$ OPA stage, pumped either near half-power (subsequently designated OPA/2) or at full power; the signal pulse energies measured after the OPA stage are ~1.5, ~2.7, and ~6.0 μJ, respectively. Figure 2(a) depicts the averaged temporal power profiles reconstructed from the Fourier transformation, recorded under zero-chirp conditions for these three OPO–OPA configurations. We used these profiles in the time do-

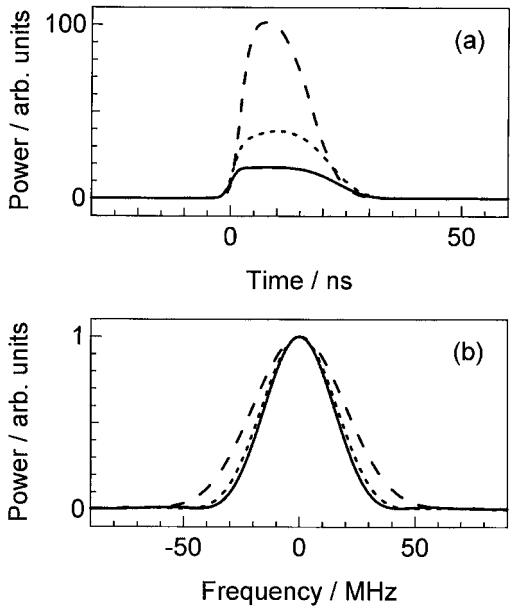

Fig. 2. (a) Reconstructed averaged-power, zero-chirp temporal profiles for an OPO alone (solid curve), for OPA/2 (short-dashed curve), and at full OPA power (long dashes); (b) calculated FT-limited TPE power spectra from (a), with peaks normalized to unity (see text).

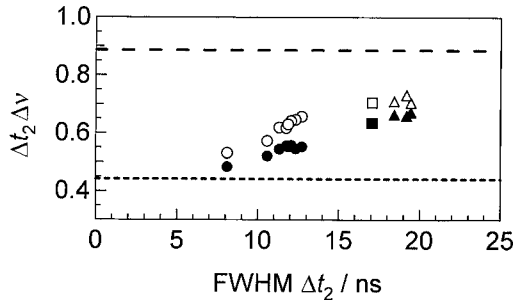

Fig. 3. Comparison of observed and calculated time–bandwidth products ($\Delta t_2 \Delta \nu$) plotted as a function of the FWHM Δt_2 of the square of the temporal power profile (as appropriate for TPE). Open symbols, FWHM $\Delta \nu = 2\Delta \nu_b$ of the Cs TPE linewidths; filled symbols, $\Delta \nu$ = FWHM of the FT-limited TPE power spectra as calculated, e.g., in Fig. 2(b). Triangles, OPO only (varying chirp); squares, OPA/2 (zero chirp); circles, full OPA (varying chirp). Also shown are the idealized FT limits for Gaussian (short dashes) and rectangular (long dashes) temporal power profiles.

main (t) to calculate power spectra in the frequency domain (ν), corresponding to the effective FT limit for these TPE measurements. To represent the nonlinear-optical polarization that gives rise to TPE we derive a signal proportional to $E^2(t)$ (the square of the incident optical field) by directly Fourier transforming the temporal *power* profiles rather than their square root. We thus obtain the power spectra depicted in Fig. 2(b). The corresponding FT-limited TPE optical bandwidths are 34.4, 37.2, and 46.3 MHz FWHM for zero-chirp OPO, OPA/2, and OPA pulses with FWHM durations of 23.0, 20.7, and 15.5 ns, respectively. In this procedure we assume that the reconstructed pulse's temporal profile is the same as the actual pulse profile and that the TPE process is not saturated; detailed examination of the OHD data validates these assumptions.

In Fig. 3 we provide a direct comparison between the time–bandwidth products ($\Delta t_2 \Delta \nu$) expected from this procedure (filled symbols) and those determined from the Cs TPE linewidth [FWHM $\Delta \nu = 2\Delta \nu_b$ (open symbols)]. The relevant time duration in both cases is the FWHM Δt_2 of the *square* of the temporal power profile (corresponding to the nonlinear-optical polarization for TPE); this is also the abscissa used in Fig. 3. Note that amplification causes the output pulse to shorten such that the FT-limited linewidth increases.

Figure 3 helps to resolve long-recognized difficulties[7,8] in relating sub-Doppler TPE line shapes to the optical power spectrum of radiation used to record them. The experimental data compare favorably with the predicted FT-limited linewidth values. Also shown, as upper and lower limits, are time–bandwidth products for Gaussian (0.441) and rectangular (0.886) idealized laser time profiles. Note that, as well as decreasing in width, the pulse shape changes from being more nearly rectangular (OPO) to more nearly Gaussian (OPA), consistent with these limits.

Finally, we note the effect in Fig. 3 of varying the chirp by controlling the periodically poled $KTi:OPO_4$ crystal temperature to tune the free-running (phase-matched) OPO wavelength away from the seed laser wavelength. The narrowest TPE linewidths occur for the zero-chirp measurements—the rightmost points in each OPO or OPA data set in Fig. 3. There is a direct correlation between increasing chirp and decreasing pulse duration, accompanied by increasing linewidth.

However, one can easily control the chirp to be close to zero simply by matching the seed laser's wavelength to the free-running OPO's wavelength.[1–4] Therefore this system has great potential for applications to pulsed laser spectroscopy that require very high resolution, close to the FT limit.

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