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Generation of frequency-tunable twin beams and its application in sub–shot-noise FM spectroscopy

Haibo Wang, Zehui Zhai, Shaokai Wang and Jiangrui Gao(*)

State Key Laboratory of Quantum Optics and Quantum Optics Devices
Institute of Opto-electronics, Shanxi University - Taiyuan, Shanxi, 030006, PRC

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Abstract. – Widely frequency-tunable quantum-correlated twin beams have been experimentally demonstrated. The noise of the intensity difference between the signal and idler beams of the twin beams is reduced to more than 3 dB below the shot noise limit (SNL) throughout a wavelength-tuning range of 5.6 nm. A continuously frequency-tuning range of 2.8 GHz appears around the center wavelength of 1041.506 nm, where the maximum noise reduction of about 5.3 dB is observed. The tunable twin beams are used in frequency modulation (FM) spectroscopy of Cs_2 molecules and a spectrum with a sensitivity of about 4 dB beyond that of the SNL is obtained.

In the past twenty years, a variety of nonclassical states of light has been successfully generated and applied to highly sensitive measurements with sensitivities beyond the shot noise limit (SNL). Optical parametric oscillators (OPOs) have turned out to be promising sources for the generation of nonclassical lights and spectroscopy. Particularly, it has been experimentally proven that the quantum noise of the intensity difference between the twin beams produced by a nondegenerate OPO operating above the oscillation threshold can be significantly reduced to far below the SNL [1,2]. In ultra-sensitive spectroscopy measurements, the frequency modulation [3] (FM) spectroscopy technique is used for improving the signal-to-noise ratio (SNR). The measurement sensitivity of FM spectroscopy can reach the limit of the technical noise of the light source in principle [3]. In order to suppress the technical noise, the method of the differential measurement is chosen in which the classical technical noise can be eliminated, so the SNR is limited only by the shot noise of the light source. It is worthwhile exploring a new FM spectroscopy scheme with sensitivity beyond the SNL.

The possibility of improving the sensitivity of weak-absorption spectroscopy by means of the nonclassical lights has been proposed and experimentally demonstrated [4–7]. In order to achieve the sub-SNL absorption spectroscopy, the frequency-tunable sources of squeezed light have to be prepared at first. Polzik *et al.* [5] realized the sub-SNL FM spectroscopy with the

(*) E-mail: jrgao@sxu.edu.cn

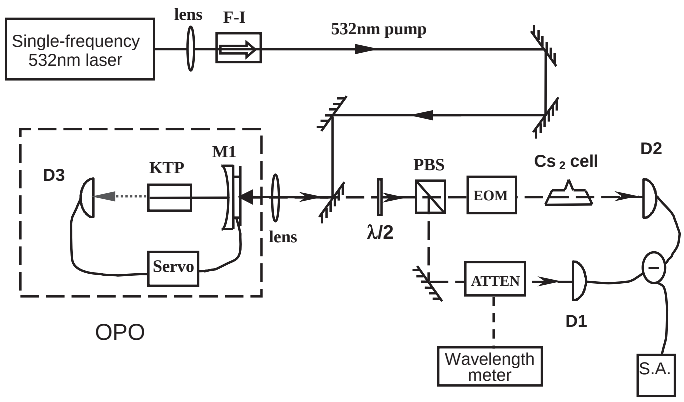

Fig. 1 – Schematic of the experimental setup for the frequency-tunable twin-beam generation and FM spectroscopy measurements.

quadrature vacuum-squeezed-state light generated by an OPO operating below the threshold, in which the SNR was improved by 3.1 dB beyond the SNL. Kasapi *et al.* [7] and Marin *et al.* [8] obtained a frequency-tunable amplitude-squeezed light around 852 nm wavelength with semiconductor lasers which was used to perform FM spectroscopy on the atoms of rubidium and cesium, respectively. Due to the application of amplitude-squeezed light the SNR of the FM spectroscopy was improved by ~ 0.8 dB beyond the SNL. The quantum-correlated twin beams with ~ 9 dB intensity difference squeezing have been experimentally obtained and used for high-sensitivity optical measurements [1, 2]. Frequency-tunable twin beams have also been realized over a 100 nm wavelength-tuning range by a type-I phase-matching LNB OPO [9]. However, the continuously frequency-tunable twin beams have not been applied in the absorption spectroscopic measurements so far to the best of our knowledge.

In this letter, we present experimental investigations on the generation of frequency-tunable quantum-correlated twin beams with type-II phase-matched OPO and its application in the FM spectroscopy of the Cs_2 molecules. Exploiting the quantum-correlated twin beams, the SNR of the spectroscopy measurement is improved by ~ 4 dB relative to the SNL of the total twin beams and by ~ 1 dB relative to that of the signal light.

The twin beams with highly correlated intensity fluctuations between the signal and idler modes can be produced from a nondegenerate OPO operating above its oscillation threshold. The intensity-difference noise spectrum between the signal and idler modes is expressed by [10]

$$S(\Omega) = S_{\mathrm{SNL}}\bigl(1 - \eta\xi/\bigl(1 + \Omega^2\tau_{\mathrm{c}}^2\bigr)\bigr), \tag{1}$$

where S_{SNL} is the shot noise limit, τ_{c} is the lifetime of photon in the optical cavity, Ω is the noise frequency, $\Omega\tau_{\mathrm{c}}$ is the frequency normalized to the cavity bandwidth, η is the quantum efficiency of the detector, and $\xi = T/(T+\mu)$ is the output coupling efficiency of OPO (T is the transmission coefficient of the output coupler and μ is the loss coefficient inside the cavity). From eq. (1), we see that the quantum correlation between the signal and idler modes of twin beams is independent of the frequency of the subharmonic waves. If suitable coated-cavity mirrors are used, the OPO operating above the threshold can be developed to be a useful nonclassical light source with the broadband frequency-tunable feature.

The experimental setup for frequency-tunable twin-beam generation and FM spectroscopy measurements is shown schematically in fig. 1. The pump source is a laser diode(LD)-pumped intracavity frequency-doubled ring Nd:YVO_4/KTP laser. The continuous frequency tuning of 5.6 GHz on the output laser (SHG) is achieved by means of scanning the cavity length of the laser with a piezoelectric transducer (PZT) attached to a cavity mirror (M1). By changing the temperature of the laser crystal (Nd:YVO_4), a wider tuning range of 20 GHz is obtained. The doubly resonant OPO consists of a concave mirror M1 with 20 mm radius of curvature and a $2 \times 5 \times 10$ mm flat-flat KTP crystal. A facet of the crystal (inner facet) was coated with an antireflecting film at both wavelengths of the pump wave (532 nm) and of the subharmonic waves (around 1064 nm) to minimize intracavity losses, and the other facet, which served as a cavity mirror, was coated to reach high reflection for both wavelengths. The KTP crystal was cut at the angles $\theta = 90°$ and $\phi = 0°$ (α-cut) that can perform 90° noncritical phase matching for the pump wavelength of 532 nm. With the α-cut KTP crystal, the pump, signal, and idler beams transmit collinearly in OPO and the walk-off effect of the beams is significantly weakened. The M1 serves as the input and output coupler for both pump and subharmonic light fields, the transmissions of which are 95% at 532 nm and 2.3% at the infrared wavelengths of the signal and the idler modes. A PZT was attached to mirror M1 for scanning and locking the cavity length. The crystal temperature can be adjusted with a Peltier thermoelectric (TE) cooler that is driven by a home-made temperature controller with a stability of 0.1%. The stable single-frequency operation of the device was achieved by means of the Pound-Drever-Hall technique [11]. Two electrodes are attached to the KTP crystal, in this case the nonlinear crystal also plays the role of an electro-optical phase modulator (PM) needed in the frequency-locking system. The leakage infrared field from the HR facet of the KTP was used for providing the necessary error signal for the servoloop for locking in the OPO at the double resonance. The signal and idler beams in the output of OPO are separated by a polarizing beam splitter (PBS). The idler beam is directly detected by a photodetector D1, whereas the signal beam is detected by D2 after passing through the electro-optical modulator (EOM) and the sample cell containing the Cs_2 molecule. Both detectors are ETX500. The modulation frequency used in the measurements is 4 MHz and the modulation index $M = 0.01$. The transmission loss of the cell is 13%, which is the main loss mechanism in this system. An intensity attenuator (ATTEN) is inserted before D1 to balance the dc photocurrents in the signal and idler channels. The spectra of noise powers are recorded by a spectrum analyzer (S.A.).

Although the wavelength-tuning characteristic of singly resonant OPO (SROPO) is much better than that of doubly resonant OPO (DROPO) [12,13], the DROPO has to be used for obtaining quantum-correlated twin beams because the simultaneous resonance of the signal and idler modes is the necessary requirement for keeping the original quantum correlation between them [10]. A widely tuning range of wavelengths from 1091.2 nm to 1088.2 nm for the signal beam and from 1039.8 nm to 1042.6 nm for the idler beam is experimentally accomplished under the doubly resonant operation of OPO by tuning the temperature of the KTP crystal. Figure 2 shows the change of the wavelengths of the output signal and idler lights from the OPO when the temperature of the α-cut KTP is continuously tuned for a given pump wavelength at 532 nm. The solid curves indicate the ideal phase-matching condition calculated from Sellmeier equations. The dots and squares are the experimental points for the idler and signal beams recorded by a wavelength meter during the temperature tuning of the KTP crystal from 10 °C to 50 °C. The perfect agreement between the theoretical calculation and experimental values obviously demonstrates the good temperature tuning property of the output twin beams from the OPO. Continuous frequency tuning in the range of 2.8 GHz without mode hops is achieved by means of tuning the frequency of the pump laser, which is also demonstrated in fig. 3. The tuning range is measured by a Fabry-Perot interferometer, which is also used for monitoring the single-axial-mode operation of the signal and idler modes inside the OPO.

It is clear from eq. (1) that the output coupling efficiency $\xi = T/(T + \mu)$ is crucial for the

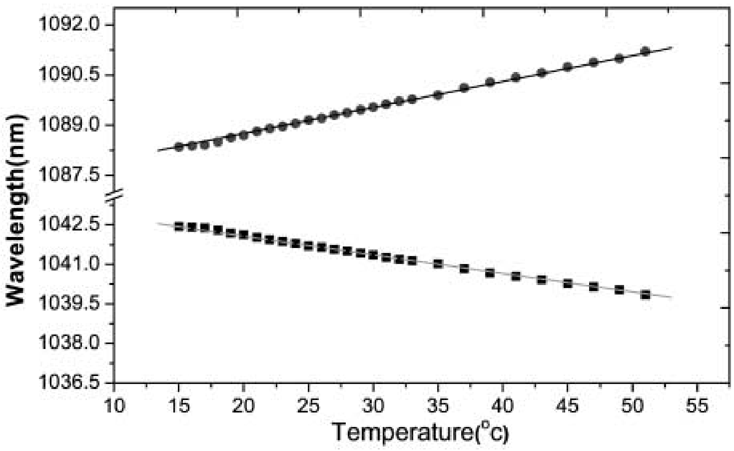

Fig. 2 – The output wavelengths of OPO *vs.* the temperatures of KTP. The solid curves are the theoretical values calculated from Sellmeier equations under the ideal phase-matching conditions. The dots and squares are the experimentally measured points for the idler and signal beams, respectively.

quantum correlation of frequency-tunable twin beams. In our case, the transmission (T) for the output coupler is 2.3% in the wavelength range from 1000 to 1100 nm that sufficiently covers the entire spectrum range of the output light of OPO. The measured oscillation threshold of the pump power was $\sim$ 106 mW without significant change in the wavelength-tuning range of 2.8 nm for the output signal beam. The intracavity loss μ deduced from the measured thresholds is about 0.6%. In our system, T and μ, therefore ξ, are almost constant throughout all the frequency-tuning range, which provides the physical base for obtaining approximately same quantum correlation beams in a broad tuning range.

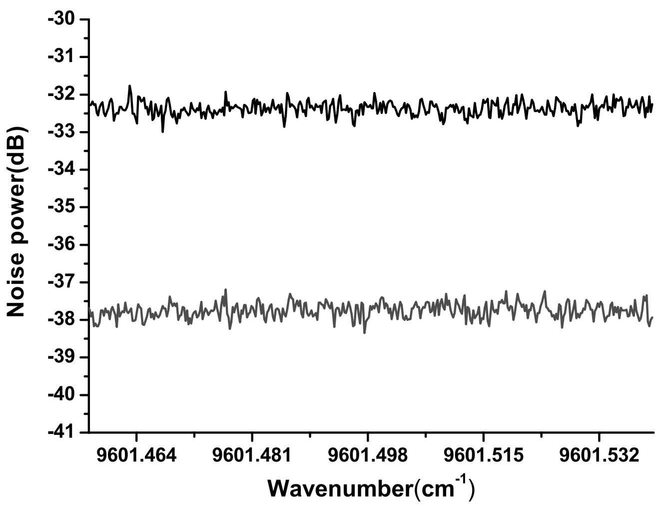

Fig. 3 – The measured quantum noise squeezings of the intensity difference *vs.* the wavelengths of the output signal wave at the analysis frequency of 2 MHz. The upper and lower traces are the SNL and the squeezed quantum fluctuation of the intensity difference between the signal and idler beams.

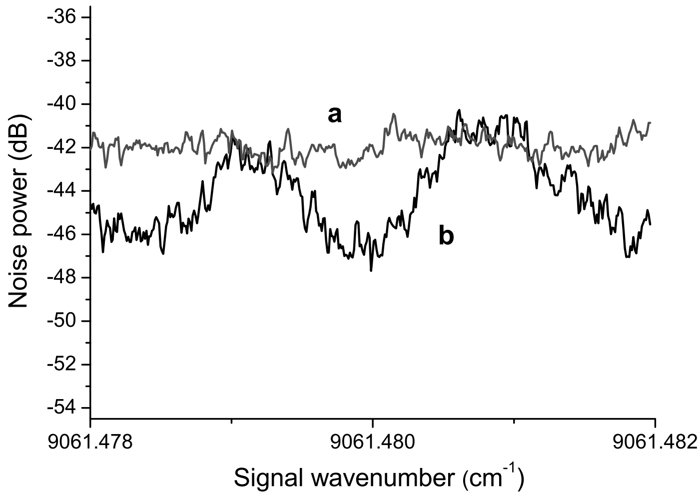

Fig. 4 – The sub-SNL FM spectroscopy of the Cs_2 molecule absorption measured with frequency-tunable twin beams. Curve a is the SNL of the twin beams. Curve b is the noise power of the intensity difference of the twin beams. The analyzer RBW is 10 kHz.

The intensity correlation between the signal and idler beams and the standard quantum limit are measured with a self-homodyne detection system [10]. Figure 3 is the measured quantum noise squeezing of the intensity difference fluctuation *vs.* the wave numbers (cm^{-1}) of the output subharmonic waves at the analysis frequency of 2 MHz. The range of the abscissa in the unit of wave number corresponds to the continuously tunable range of 2.8 GHz of the output field from the OPO. During the measurements of squeezing, the Cs_2 cell, the EOM and the attenuator in fig. 1 were removed. The upper and lower traces are the SNL and the squeezed quantum fluctuation of the intensity difference between the signal and idler beams, respectively. The measurement principle and scheme are same with that described in ref. [10]. A noise reduction of 5.3 ± 0.3 dB below the SNL is accomplished at a tuning range of 2.8 GHz around the center wavelength of 1041.506 nm (KTP crystal at room temperature, 28 °C). The experimental result is in good agreement with the theoretically expected value of 5.7 dB from eq. (1) within the experimental errors, the detector efficiency is 93%.

A vacuum-sealed glass cell including the solid cesium with a purity of 99.9% (vacuum degree 10^{-6} Pa at room temperature) is prepared for the FM spectroscopy measurement. Figure 4 shows a sub-SNL FM spectroscopy of Cs_2 molecules at 40 °C measured with the system of fig. 1. Curve a is the SNL of the twin beams in logarithmic power units set by the difference photoncurrent i_- and it is 3 dB larger than the SNL of the only signal beam (without idler beam) [2]. Curve b is the noise power of the intensity difference of the twin beams which shows the absorption feature of the Cs_2 molecules, it is a typical FM spectrum of "M" shape formed by two peaks [5,14]. The resolution bandwidth (RBW) of the spectrum analyzer used in this measurement is 10 kHz. The measured power of both signal and idler wave is 7 mW. It is shown in fig. 4 that the SNR of FM spectroscopy measurements is improved to about 4 dB beyond the SNL due to using the intensity quantum-correlated twin beams. When the temperature of the Cs cell is increased, the intensity of the absorption increases too. The higher the temperature, the stronger the absorption, that just is the characteristic of the Cs_2 absorption spectrum [15, 16]. When the temperature is over 240 °C, the absorption is strong enough and can be directly detected with a detector. At 40 °C, it only can be recorded with the

FM spectroscopy. When the temperature is decreased to below 30 °C, the FM spectroscopy disappears also in our experiment.

According to the traditional FM spectroscopy theory, the SNR of the absorption signal generated by a photodetector of quantum efficiency η is given by [14, 17]

$$S/N = \overline{i_S^2(\Omega)}/\overline{i_N^2(\Omega)} = \frac{\eta\left(\frac{P_0}{\hbar\omega}\right)\Delta\delta^2 M^2}{8S(\Omega)\Delta f}\,, \tag{2}$$

where M is the modulation index, Ω is the modulation frequency, P_0 is the optical power incident on the photodetector, and $\Delta\delta$ is the absorption coefficient of the sample, Δf is the bandwidth of detection electronics, $S(\Omega)$ is the intensity-difference noise spectrum between the signal and idler modes. The detectable minimum absorption $\Delta\delta_{\min}$ limited by $S/N = 1$ is $\Delta\delta_{\min} = 2[\eta M^2 \frac{P_0}{2\hbar\omega S(\Omega)\Delta f}]^{-\frac{1}{2}}$. In fig. 4, the SNR is improved beyond the SNL about 4 dB which means $S(\Omega) = 0.39$. Taking $\eta = 1$, $P_0 = 7\times10^{-3}$ W, $M = 0.01$, and RBW$\Delta f = 10$ kHz, the minimum detectable absorption using the coherent state light is $\Delta\delta = 1.5\times10^{-4}$. However, the detectable minimum absorption with tunable twin beams obtained in the above experiment reduces to $\Delta\delta_{\min} = 6.5\times10^{-5}$ under the same conditions. If $\Delta f = 1$ Hz, which has been used in the experiment of ref. [6], the sensitivity can be improved to $\Delta\delta_{\min} = 6.5\times10^{-7}$.

In summary, frequency-tunable twin beams with intensity quantum correlation were generated by a nondegenerate DROPO operating above threshold. The intensity difference squeezing more than 3 dB was obtained in a tuning range of 5.6 nm. The maximum noise reduction in the intensity difference was about 5.3 dB below the SNL at the wavelength of 1041.506 nm around which a continuously frequency-tuning range of 2.8 GHz is demonstrated. The SNR of the FM spectroscopy of Cs_2 was improved by about 4 dB beyond the SNL. Since the experimental generation of twin beams with high-intensity correlation is relatively easier than the quadrature vacuum-squeezed-state light, and the self-homodyne detection used in our experiment is also relatively simpler than usual balance-homodyne detection systems due to the fact that the local beam is not needed [5]. On the other hand, the frequency nondegenerate intensity quantum-correlated twin beams provide more selectable wavelengths of light compared with quadrature vacuum-squeezed-state light. The presented spectroscopy using frequency-tunable twin beams would be a valuable scheme for enhancing the sensitivity of absorption spectroscopy to a higher level beyond the SNL.

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