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Efficient parametric non-degenerate four-wave mixing in hot potassium vapor

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Abstract

We have observed high gains of the probe and the conjugate beams in non-degenerate four-wave mixing in hot potassium vapor, using a double-Λ configuration at the D1 line of the ^{39}K isotope. Gains of up to 82 for the conjugate beam and 63 for the probe beam have been achieved. Higher gains were obtained than with other alkali atoms under comparable experimental conditions due to lower ground state hyperfine splitting in the potassium atom. Experimental parameters for maximal gain have been determined. Notable gains are achieved at low pump intensities (~10 W cm^{-2}) that are attainable even by conventional laser diodes. Due to their high gains, the probe and the conjugate beams may be suitable for utilization in quantum correlation and relative intensity squeezing experiments.

Keywords: four-wave mixing, potassium, squeezed states, nonlinear optics

(Some figures may appear in colour only in the online journal)

Four-wave mixing (FWM) is a nonlinear interaction of light and a medium accompanied by a characteristic transfer of energy between four modes of the electric field while these modes interact with the medium [1]. FWM in atomic vapors is a valuable tool for the generation of non-classical states of light. Signal and idler beams (here referred to as the probe and the conjugate beams, respectively) generated by this process display intensity correlations and entanglement [2]. These features make them applicable in high-precision spectroscopy [3], sub-shot-noise measurements [4, 5], quantum imaging [6–8], quantum communications and quantum information processing [9, 10].

The first experimental demonstration of squeezed light was made using FWM in an atomic beam of Na [11]. Since such FWM processes generate squeezed light near atomic resonance, the amount of squeezing is limited by other resonant processes such as one-photon absorption and spontaneous emission. Renewed interest in FWM came after predictions [12, 13] that non-degenerate FWM in atomic systems with a double-Λ scheme could overcome these limitations. Experiments that followed confirmed that it was indeed possible to obtain squeezing near atomic resonance [14–16].

Higher gains of the probe and the conjugate beams (also called 'twin' beams) in a non-degenerate FWM process leads to higher relative intensity squeezing and deeper noise reduction [17]. The gains of the probe and the conjugate are defined as $G_p = P_p/P_{in}$ and $G_c = P_c/P_{in}$, respectively, where P_p and P_c are the measured powers of the probe and the conjugate beams, respectively, and P_{in} is initial power of the probe seed inside the amplifying medium. The ability of different mediums to yield large gains of the twin beams was tested with different interaction schemes. So far, all alkali atoms except Fr and Li have been used as the gain medium for FWM [11, 14–26]. In the majority of studies the counter-propagating geometry of two pump beams and one probe beam was used and the degenerate case of FWM process was observed.

However, new beams generated in the aforementioned arrangements are not suitable for applications that require spatially separated beams. The most suitable interaction scheme and experimental arrangement for employing twin beams in relative intensity squeezing experiments was realized by McCormick *et al* [15]. The coupling of hyperfine levels of an alkali atom by a double-Λ scheme is depicted in figure 1. The first Λ scheme consists of a strong pump that couples the lower hyperfine sublevel $|1\rangle$ of the ground state to

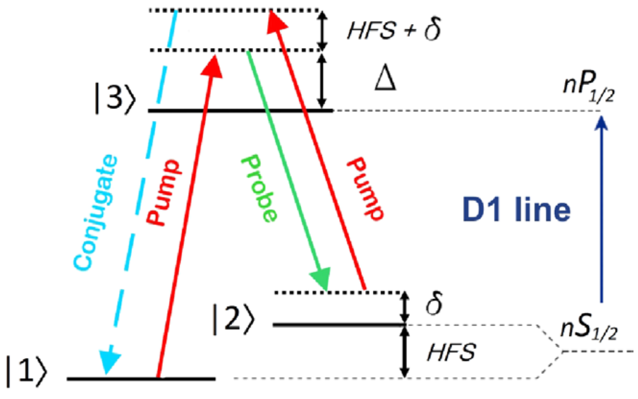

Figure 1. Double-Λ scheme at the D1 line of an alkali atom. HFS—hyperfine splitting, Δ—one photon detuning, δ—two photon detuning. HFS of the $nP_{1/2}$ (i.e. $|3\rangle$) level is negligible in comparison with the ground state HFS.

the excited level $|3\rangle$ with one-photon detuning Δ of typically several hundred MHz. The other 'leg' of the first Λ scheme is the weak probe that stimulates the Stokes scattering from $|3\rangle$ to the higher hyperfine sublevel $|2\rangle$ of the ground state, having two-photon detuning δ. The pump is sufficiently strong to drive the off-resonant transition starting from $|2\rangle$. The newly created conjugate closes the second Λ scheme by stimulating anti-Stokes scattering to the lower hyperfine sublevel.

Such an arrangement, yielding non-degenerate FWM and spatially separated twin beams, was employed with achieved gains ≈20 [16, 24] or even 30 [25] in rubidium, ≈32 in sodium [26] and recently ≈ 2 in cesium [27]. The theoretical explanations for this arrangement were also provided [24, 28, 29]. Apart from relative intensity squeezing experiments, this scheme is used in other applications such as slow light [25, 30–32], storage of light [33, 34], and heralded state density matrix reconstruction [35], and is also proposed for all-optical quantum networks [36–39].

In this paper we report FWM in a double-Λ scheme in hot potassium vapor. There are very few works on FWM in potassium vapor [19, 20] and all of them are done with counter-propagating pumps. Ground state hyperfine splitting (HFS) in ^{39}K (461 MHz [40]) is lower than in any other alkali atom, both for lighter atoms such as ^{7}Li (803 MHz [41, 42]) or ^{23}Na (1772 MHz [42, 43]) and heavier atoms, like ^{85}Rb (3036 MHz [42–44]) or ^{133}Cs (9193 MHz [43, 45]). In addition, all the transitions of the D1 line of ^{39}K completely overlap due to Doppler broadening. This affects the dynamics of pumping and repopulating ground state hyperfine sublevels in a different way than in other alkali atoms. All of the aforementioned properties of ^{39}K make it interesting as a medium for FWM and other applications.

The influence of ground state HFS on the efficiency of FWM can be estimated from the theoretical model given by Turnbull *et al* [24]. In the model, the following equations describe the change of the probe $E_{\rm p}$ and the conjugate $E_{\rm c}$ electric field along the z axis (the propagation direction of the pump beam):

$$\frac{\partial}{\partial z}E_{\rm p} = \frac{{\rm i}k_{\rm p}}{2}\chi_{\rm pp}(\omega_{\rm p})E_{\rm p} + \frac{{\rm i}k_{\rm p}}{2}\chi_{\rm pc}(\omega_{\rm p}){\rm e}^{{\rm i}\Delta k_z z}E_{\rm c}^{*} \qquad (1)$$

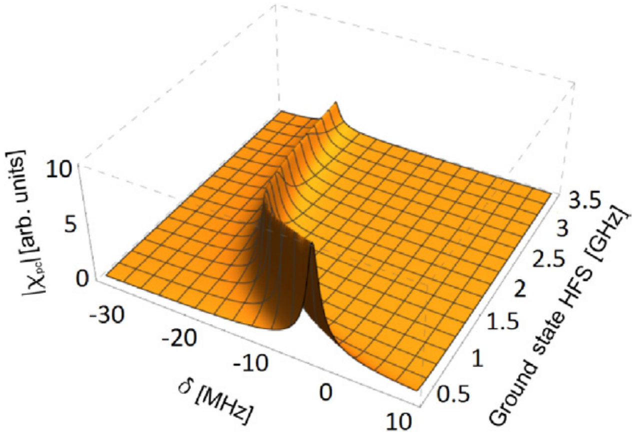

Figure 2. Dependence of $|\chi_{\rm pc}|$ on ground state HFS and two-photon detuning δ. The one-photon detuning ($\Delta = 700$ MHz) and dipole matrix elements of the double-Λ scheme transitions were kept constant.

$$\frac{\partial}{\partial z}E_{\rm c} = \frac{{\rm i}k_{\rm c}}{2}\chi_{\rm cc}(\omega_{\rm c})E_{\rm c} + \frac{{\rm i}k_{\rm c}}{2}\chi_{\rm cp}(\omega_{\rm c}){\rm e}^{{\rm i}\Delta k_z z}E_{\rm p}^{*} \qquad (2)$$

where, $k_{\rm p}$ and $k_{\rm c}$ are the magnitudes of the probe and the conjugate wave vectors, Δk_z is the projection of the phase mismatch $\Delta\boldsymbol{k}$ on the z axis, $\chi_{\rm pp}$ and $\chi_{\rm cc}$ are the effective linear susceptibilities for the probe and the conjugate and $\chi_{\rm pc}$ and $\chi_{\rm cp}$ are cross-susceptibilities that give rise to FWM process. The phase mismatch is defined as $\Delta\boldsymbol{k} = 2\boldsymbol{k}_0 - \boldsymbol{k}_{\rm p} - \boldsymbol{k}_{\rm c}$ where $\boldsymbol{k}_0$ is the pump wave vector.

Atomic susceptibilities govern the FWM process and affect the gains. The dependence of $|\chi_{\rm pc}|$ on HFS and two-photon detuning is shown in figure 2 and is calculated according to equations A12–A20 given in the appendix of [24].The equations enable the calculation of the stationary values of $|\chi_{\rm pc}|$ as a function of the relevant experimental parameters: one-photon detuning, two-photon detuning, ground state HFS, pump laser Rabi frequencies, and the concentration of the atoms, i.e. the temperature. The equations are given under the assumption that Rabi frequencies for both pump transitions in figure 1 are equal. The probe and conjugate fields are assumed to be weak and their contribution is kept only to the first order. Since we want to estimate the influence of ground state HFS of alkali atoms on the efficiency of FWM we kept all other quantities constant, except the two-photon detuning. The results show that the maximum of $|\chi_{\rm pc}|$ increases as HFS decreases. The model also predicts that the two-photon detuning δ, corresponding to the maximum $|\chi_{\rm pc}|$, also decreases, thus both Λ schemes are closer to Raman resonance.

Motivated by the above analysis, the present work investigates the properties of FWM in hot potassium vapor using the non-degenerate scheme of figure 1 and a co-propagating geometry of the pump and probe beams. To the best of our knowledge there are no previous investigations of this kind in potassium. Exceptionally high gains could make potassium vapor the preferred medium for relative intensity squeezing experiments [16] and other applications utilizing highly efficient FWM [3–10]

We have performed the double-Λ scheme on the D1 line of ^{39}K. Level $|3\rangle$ from figure 1 is $4P_{1/2}$ while two lower levels

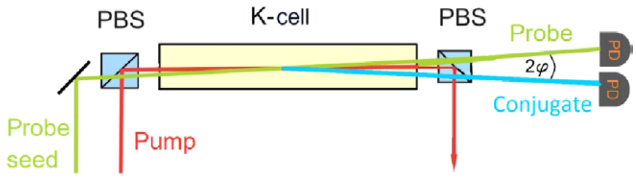

Figure 3. Experimental setup. Pump (red) and probe seed (green) beams are combined at a polarization beam splitter (PBS). They intersect at a small angle φ inside the potassium vapor cell (K-cell) yielding the conjugate beam (blue) and amplified probe beam (green) via the FWM process. The probe and the conjugate beams are detected by two photodiodes (PD). Note that angle between the conjugate and the probe beams is 2φ.

$|1\rangle$ and $|2\rangle$ are $4S_{1/2}$, $F = 1$ and $4S_{1/2}$, $F = 2$ respectively. The simplified scheme of the experimental setup is presented in figure 3. A single-mode frequency stabilized Ti:Saphire laser was used in the experiment. It delivers 600 mW at the 770 nm D1 line of ^{39}K and it is used for both the pump and probe seed beams. The probe seed ($\approx$200 μW) is obtained by picking up a small fraction of the pump at the 90:10 beam splitter and sending it through the two acousto–optic modulators (AOMs). The first AOM produces a tunable frequency shift (170–200 MHz) and it operates in a double-pass configuration. The second AOM has a fixed frequency shift (80 MHz), making the overall frequency offset between the pump and probe seed close to the HFS of the ^{39}K ground state. Two-photon detuning δ is scanned by changing the RF frequency fed to the first AOM.

The pump and the probe seed have mutually orthogonal linear polarizations. The beams are combined at a polarization beam splitter and sent through the heated, 50 mm long, natural-abundance vacuum potassium vapor cell, where they intersect at a small angle that we change in the range of 2–10 mrad. Both beams, the pump and the probe seed, are focused at the intersection and their waists are 1.05 mm and 0.8 mm, respectively. The windows of the cell are Brewster angled and the cell is rotated to provide the maximal pump transmission ($\approx$95% per window). Since the probe seed is polarized perpendicularly to the pump, its transmission is lower ($\approx$70% per window).

After passing through the vapor cell, the pump beam is rejected by the second polarizing beam splitter. The conjugate beam (which has the same polarization as the probe seed) and the amplified probe beam are detected by two photodiodes.

We have investigated the dependence of the probe and the conjugate gains on two-photon detuning δ with one-photon detuning Δ as a parameter. The δ step was 2 MHz. The results for various values of Δ are shown in figure 4.

The maximal conjugate gain ($G_c = 82$; peak value in figure 4(b)) was obtained at $\Delta = 700$ MHz and $\delta = -6$ MHz. The probe gain for the same parameters was $G_p = 58$. The reason for the maximum gains occurring at a particular Δ is the competition of two effects: amplification and absorption [16, 24]. When Δ increases, the amplification of the probe and the conjugate beams decreases, but so does one-photon absorption. The trade-off is in our case for $\Delta = 700$ MHz (figure 4(c)). Since the frequency offset between the probe and the conjugate beams is $\approx$920 MHz (approximately double the HFS) and the probe beam is tuned closer to the resonance, one-photon absorption is stronger for the probe beam. This is the reason why we observe different G_p and G_c for smaller Δ (figures 4(a) and (b)). At larger Δ , one-photon absorption becomes smaller, thus G_p and G_c get closer (figure 4(d)), but are rather small due to detuning far from resonance.

According to our expectations, qualitatively supported by results in figure 2, we have obtained higher gains than in other alkali atoms under comparable experimental conditions. For more detailed theoretical study and quantitative comparison between experimental and theoretical results one might consider adjusting the theoretical model from [24] for particular properties of potassium. Unlike rubidium, all the transitions forming the double-Λ scheme in potassium are overlapped due to large Doppler broadening at specified temperatures. Moreover, one might also consider the geometry and intensity profiles of overlapping laser beams and their spectral properties.

The dependence of G_p and G_c on the temperature for various values of Δ is shown in figure 5. For each Δ on the graph, we set δ to maximize the gains of the probe and the conjugate beams. As the concentration of potassium atoms increases, the cross-susceptibilities (χ_{cp} and χ_{pc}) also increase [24]. On the other hand, large susceptibilities lead to large values of the refractive index and its transverse gradient that cause beam focusing and beam filamentation [1, 24]. Stars in figure 5 indicate the highest temperatures for particular values of Δ, above which these effects prevent the proper measurement of the intensities of the probe and the conjugate beams. At high vapor temperatures and/or pump intensities self-focusing of the probe and conjugate beams appears gradually, ending up with beam breakup. As the pump intensity and/or vapor temperature increases the probe and the conjugate beams become more divergent due to self-focusing. This makes the beams partially overlapped and hinders proper measurement of the powers independently.

Varying the temperature and Δ we have determined that the values of $T = 140$ °C and $\Delta = 1500$ MHz provide the highest probe gain, $G_p = 63$ (we found $G_c = 69$ for the same set of parameters).

The dependence of G_p and G_c on the mutual angle between the pump and the probe beam is presented in figure 6(a). While in rubidium [24] the dependence on this angle has a maximum at 5 mrad, in potassium it monotonically decreases. This is in accordance with Glassner *et al* [19] where, in their configuration of counter-propagating pumps and degenerate FWM, the probe reflectivity can be considered as an analogue to the probe gain, since both are affected by atomic susceptibility.

The dependence of the probe and the conjugate gains on the pump power is shown in figure 6(b). We found that the lowest pump intensity, at which we were able to detect the conjugate beam, is about 10 W cm^{-2} corresponding to laser power of $\approx$100 mW. This, relatively low, laser power can easily be attained with conventional lasers diodes. We were able to measure even higher gains (96 for the conjugate, 73 for the probe) at a pump intensity of 51 W cm^{-2} but the laser becomes unstable at high powers.

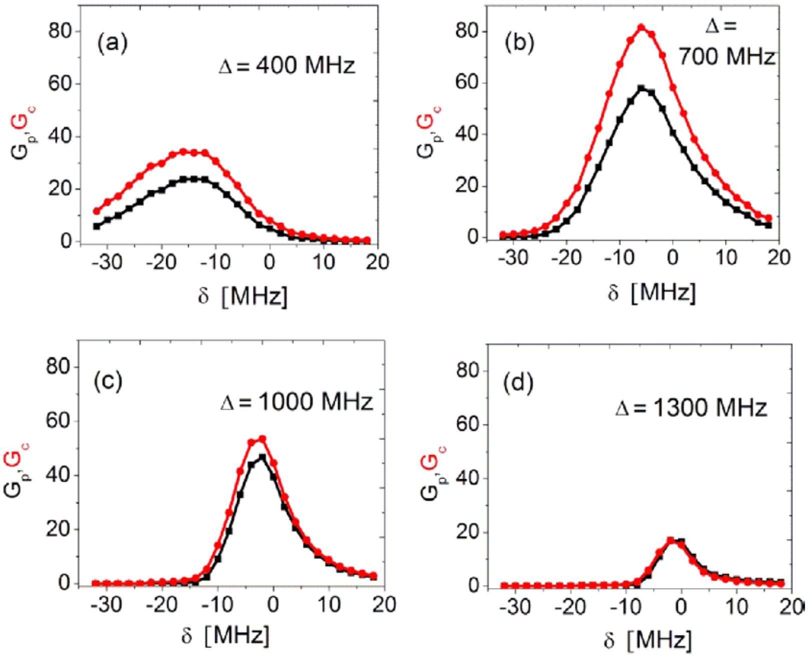

Figure 4. The probe (black squares) and the conjugate (red circles) gain curves versus two-photon detuning δ in the vicinity of Raman resonance ($\delta = 0$) at Δ equal to (a) 400 MHz, (b) 700 MHz, (c) 1000 MHz and (d) 1300 MHz. The pump power was $P_0 = 400$ mW and the probe seed power was $P_{\text{in}} = 200$ μW. Vapor temperature was kept constant at 120 °C ($\approx 3 \times 10^{12}$ atoms cm^3), and angle between the pump and the probe was $\varphi = 3$ mrad. The lines are to guide the eye.

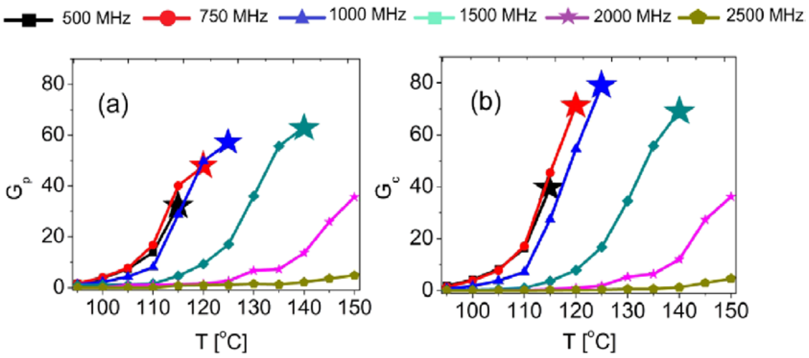

Figure 5. Temperature dependence of (a) the probe gain G_{p} and (b) the conjugate gain G_{c}. Different colors correspond to the different values of Δ (given in the legend). For the given range of the temperature of potassium vapor the number density of the atoms, calculated according to Ticke [40], is between 3.7×10^{11} atoms cm^{-3} (at 90 °C) and 1.7×10^{13} atoms cm^{-3} (at 150 °C). Stars denote the temperatures at which filamentation and self-focusing of the probe and the conjugate beams occur. Parameters are $P_0 = 400$ mW, $P_{\text{in}} = 200$ μW, $\varphi = 2$ mrad.

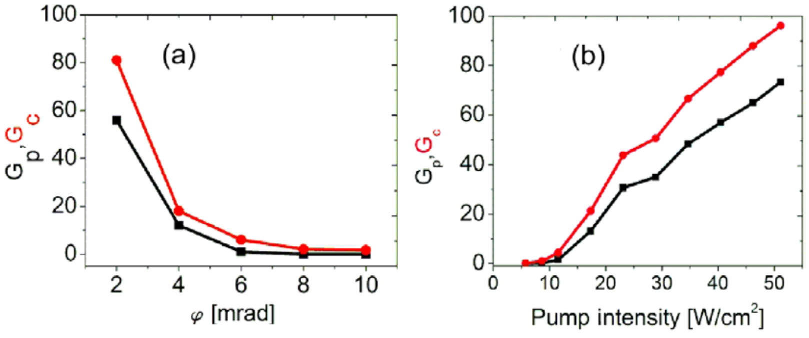

Figure 6. Dependence of the probe (black squares) and the conjugate (red circles) gain on (a) the angle φ between the pump and the probe for $P_0 = 400$ mW and (b) the pump intensity for $\varphi = 2$ mrad. Parameters for both cases are $P_{\text{in}} = 200$ μW, $T = 120$ °C, $\Delta = 700$ MHz.

In conclusion, we have observed non-degenerate FWM in hot potassium vapor at the D1 line using co-propagating pump and probe beams and a double-Λ coupling scheme. In accordance with simple qualitative theoretical considerations, the obtained gains are among the highest in alkali atoms. This is due to the high atomic susceptibilities caused by the lowest ground state HFS in potassium. We expect that the obtained high gains might find useful application in experiments for relative intensity squeezing, sub-shot-noise measurements and other applications requiring efficient FWM.

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