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

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 \( |1⟩ \) of the ground state to...
the excited level $|3\rangle$ with one-photon detuning $\Delta$ of typically several hundred MHz. The other ‘leg’ of the first $\Lambda$ 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 $\delta$. The pump is sufficiently strong to drive the off-resonant transition starting from $|2\rangle$. The newly created conjugate closes the second $\Lambda$ 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 $\approx 20$ [16, 24] or even $32$ [25] in rubidium, $\approx 32$ in sodium [26] and recently $\approx 2$ 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-$\Lambda$ 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 $^1$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 $E_p$ and the conjugate $E_c$ electric field along the $z$ axis (the propagation direction of the pump beam):

$$\frac{\partial}{\partial z} E_p = \frac{i k_p}{2} \chi_{pp}(\omega_p) E_p + \frac{i k_p}{2} \chi_{pc}(\omega_p) e^{i(\Delta k_z z)} E_c^*$$

$$\frac{\partial}{\partial z} E_c = \frac{i k_c}{2} \chi_{cc}(\omega_c) E_c + \frac{i k_c}{2} \chi_{cp}(\omega_c) e^{i(\Delta k_z z)} E_p^*$$

where, $k_p$ and $k_c$ are the magnitudes of the probe and the conjugate wave vectors, $\Delta k_z$ is the projection of the phase mismatch $\Delta k$ on the $z$ axis, $\chi_{pp}$ and $\chi_{cc}$ are the effective linear susceptibilities for the probe and the conjugate and $\chi_{pc}$ and $\chi_{cp}$ are cross-susceptibilities that give rise to FWM process. The phase mismatch is defined as $\Delta k = 2k_0 - k_p - k_c$ where $k_0$ is the pump wave vector.

Atomic susceptibilities govern the FWM process and affect the gains. The dependence of $|\chi_{pc}|$ 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 $|\chi_{pc}|$ 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 keep all other quantities constant, except the two-photon detuning. The results show that the maximum of $|\chi_{pc}|$ increases as HFS decreases. The model also predicts that the two-photon detuning $\delta$, corresponding to the maximum $|\chi_{pc}|$, also decreases, thus both $\Lambda$ 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-$\Lambda$ scheme on the D1 line of $^{39}$K. Level $|3\rangle$ from figure 1 is $4P_{1/2}$ while two lower levels
The probe gain for the same parameters was $G_p = 58$. The reason for the maximum gains occurring at a particular $\Delta$ is the competition of two effects: amplification and absorption [16, 24]. When $\Delta$ 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 $\Delta$ (figures 4(a) and (b)). At larger $\Delta$, 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-$\Lambda$ 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 $\Delta$ is shown in figure 5. For each $\Delta$ on the graph, we set $\delta$ to maximize the gains of the probe and the conjugate beams. As the concentration of potassium atoms increases, the cross-susceptibilities ($\chi_{cp}$ and $\chi_{pc}$) 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 $\Delta$, 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 break-up. 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 $\Delta$ 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 $\delta$ in the vicinity of Raman resonance ($\delta = 0$) at $\Delta$ 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 $P_\text{in} = 200 \mu$W. 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 $\Delta$ (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 $3.7 \times 10^{11}$ atoms cm$^{-3}$ (at 90 °C) and $1.7 \times 10^{13}$ 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, $P_\text{in} = 200 \mu$W, $\varphi = 2$ mrad.

Figure 6. Dependence of the probe (black squares) and the conjugate (red circles) gain on (a) the angle $\varphi$ 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 $P_\text{in} = 200 \mu$W, $T = 120$ °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-A 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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