Two symmetric four-wave mixing signals generated in a medium with anomalous refractive index

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Received 12 November 2020, revised 23 December 2020
Accepted for publication 29 January 2021
Published 26 February 2021

Abstract
We report experimental and theoretical results of two symmetrical signals of degenerate four-wave mixing generated in rubidium vapor. Both nonlinear signals are induced by two almost copropagating laser beams, with $\vec{k}_a$ and $\vec{k}_b$ wave-vectors, and detected simultaneously in the $2\vec{k}_a - \vec{k}_b$ and $2\vec{k}_b - \vec{k}_a$ directions. In each direction, we observe a single peak when the two beams are tuned on the closed transition $^{85}$Rb $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$. The excitation spectra reveal a small frequency separation between the two peaks, which is explained when propagation effects are taken into account. Furthermore, our theoretical analysis shows that a correct description of the frequency position of each peak is achieved with a variable refractive index for both lasers, in which the scanning laser experiences an anomalous window in the refractive index near resonance.

Keywords: four-wave mixing, phase-matching, nonlinear optics, rubidium

1. Introduction
In a four-wave mixing (FWM) process a fourth field is generated as the result of the coherent combination of three electromagnetic fields interacting with a nonlinear sample. This process has been used extensively to investigate a variety of optical phenomena in atomic systems. Since very early studies, different atomic level configurations as, for instance, two-level [1, 2], three-level $\Lambda$ [3, 4], and four-level double-$\Lambda$ schemes [5] have been explored to enhance the efficiency of this nonlinear process. One common characteristic in many of these FWM processes is the possibility to control the refractive index [6] of the medium, and in some conditions cancel the resonant absorption due to the phenomenon of electromagnetically induced transparency (EIT) [7, 8] or enhance the absorption via electromagnetically induced absorption (EIA) [9, 10]. Another interesting feature is the generation of narrow-band photon pairs in atomic ensembles via spontaneous FWM [11] as well as the efficient generation of pairs of intense light beams showing a high degree of intensity squeezing [12, 13].

The present work is concerned with two symmetric FWM signals that are generated together in a sample of thermal rubidium atoms. The nonlinear signals are induced by two independent laser beams, with $\vec{k}_a$ and $\vec{k}_b$ wave-vectors, when both beams are tuned on the same $^{85}$Rb Doppler line $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$. In such a case, where all fields are almost resonant with the same optical transition, the degenerate FWM signals have been analyzed considering a pure [14, 15] or degenerate two-level system [16], with one strong field, and arbitrary polarization of the drive fields [17, 18].

Most of these experiments are performed with a counterpropagating beam configuration, exploring the phase-matching obtained when the generated beam is phase-conjugated with the probe beam. In particular, we employ a copropagating beam configuration and detect simultaneously the transmission of the incident beams and the generated FWM signals at directions $\vec{k}_s = 2\vec{k}_a - \vec{k}_b$ and $\vec{k}_s = 2\vec{k}_b - \vec{k}_a$. In each direction, the excitation spectra show a single FWM peak when the two beams are tuned on the closed
transition $^{85}\text{Rb} \, 5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$. A similar scheme, where two FWM fields are also detected simultaneously, has explored a non-degenerate system and showed that the generated fields with different frequencies, like Stokes and anti-Stokes, can be correlated or anticorrelated depending on the incident beams [19]. In the experiment described here, the degeneracy of the nonlinear process in combination with incident beams of the same intensities leads to two symmetric signals, either in space as in frequency, independent of which beam is scanning.

It is interesting to note that although the two signals are generated by two independent FWM processes, they give information about the dynamic of an ensemble of atoms that interacted simultaneously with the same drive fields. Actually, for a spatially uniform atomic medium, the coherent superposition of the generated fields at different positions along the nonlinear medium leads to the well-known phase-matching condition. This condition determines not only the propagation direction of the outgoing FWM field in terms of the wave-vectors of the incident waves but also the frequencies at which the signals will be maximal. Recent studies [20] in a non-degenerate three-level system, show that the phase-matching condition is responsible for the high efficient FWM signal when the excitation fields are turned off-resonance from the atomic transition. In this case, with counterpropagating beams, the predominant contributions are attributed to the EIA grating effects [21].

The experiment described here, with copropagating beams and involving a degenerate process, also reveals a frequency shift from resonance in the FWM signals. Although it is well known that phase-matching conditions play an essential role in determining the characteristics of these generated fields, here we observe a frequency shift that goes in the opposite direction in comparison to the usual cases, where the phase-matching is basically controlled by the geometrical arrangement of the fields. Our results depend heavily on the refractive index that both incident fields experience. In particular, as we detect the two signals simultaneously, we can distinguish a frequency shift toward red or blue from each peak associated with the FWM signals, depending on how the beam whose frequency is sweeping contributes to the observed signal.

Certainly, understanding how to effectively control the properties of the atomic media where the nonlinear process is taking place is of widespread interest not only for fundamental aspects atom-light interaction but also due to the potential for application in quantum communication and quantum information processing. In this sense, this work aims to investigate the main physical mechanisms responsible for these frequency shifts and how they are related to the coherence induced in the atomic system. In section 2, the experimental setup and the principal results are presented. Section 3 is devoted to describe the theoretical model and discuss the different contributions to the FWM spectra calculated for a Doppler-broadened sample. We conclude by summarizing the most important results in section 4.

2. Experimental setup and results

A simplified scheme of the experimental setup is presented in figure 1(a) together with the hyperfine structure of the $D_2$ line of $^{85}\text{Rb}$. Two independent cw diode lasers generate the two beams $E_a$ and $E_b$ that are responsible for driving the FWM process. The beams $E_a$ and $E_b$, with wave-vectors $\vec{k}_a$ and $\vec{k}_b$, respectively, and orthogonal and linear polarization, converge inside a 5 cm long cell containing a natural concentration of rubidium atoms at an angle $2\theta \approx 40$ mrad (see figure 1(b)). To increase the atomic density, the rubidium cell was heated to $\sim 55$ °C. Both beams are tuned on the same Doppler line of $^{85}\text{Rb}$ starting in the hyperfine ground state $F = 3$, as shown in the inset of figure 1(a). To control and monitor the frequency of each laser we use saturated absorption spectroscopy (not shown).

We simultaneously detect four signals: the transmission of both incident beams $T_a$ and $T_b$ and the two FWM signals in the $\vec{k}_s = 2\vec{k}_a - \vec{k}_b$ and $\vec{k}_s = 2\vec{k}_b - \vec{k}_a$ directions, as shown in figure 1(b). In this type of forward geometry, the clean detection of the signals can be a challenge since scattered light from one beam might interfere with another’s detection. To deal with this we take advantage of the linear and cross-polarization of the signals and use polarizing beamsplitters before each detector. The incident beams are typically strong allowing the use of regular photo-diode detectors to acquire $T_a$ and $T_b$. On
the other hand, the FWM signals are very weak so we use avalanche photo-diodes (Thorlabs APD120A) to detect them.

The measurements are performed scanning the frequency of one laser throughout the three allowed hyperfine transitions while the other has a fixed frequency. The intensity of the incident lasers at the cell entrance is 10 to 100 times the saturation intensity of the cyclic transition $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$. The absorption in the medium is increased due to the temperature of the vapor but not enough to completely absorb the incident fields $E_a$ and $E_b$, so we consider it to be operating in a high-intensity regime.

A typical experimental result is shown in figure 2(a) with the transmission of the two beams $T_a$ and $T_b$ and the two generated FWM signals, $2k_a - k_b$ and $2k_b - k_a$, as a function of the detuning ($\delta/2\pi$) of the scanning field $E_u$. The intensities of $E_a$ and $E_b$ at the cell entrance were selected to be approximately the same ($I \sim 50 \text{ mW cm}^{-2}$) and the frequency of $E_b$ beam was fixed near the center of the Doppler broadened spectrum. All curves in figure 2(a) are independently normalized and we chose to measure the frequency detuning with respect to the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$ closed transition.

This result contains a series of interesting features. To begin the analysis with the transmission signals, notice that there are three peaks in each curve. The frequency difference between them reveals that they are related to the hyperfine transitions from the ground state $F = 3$ of $^{85}$Rb. Furthermore, these peaks only appear in the spectra due to the simultaneous interaction of the two incident fields with the atomic medium. These sub-Doppler peaks have been studied when the transmission of a weak beam is measured in the presence of a strong fixed frequency field, and the effect is known as velocity selective optical pumping [22]. Here, we measure the transmission of the two beams, when both are strong.

Notice that the peaks at the transmission curve of the laser with a fixed frequency, $T_b$, appears in the appropriate order of energy growth. The same is not true for the transmission of the scanning laser. To explain this we must look at the different atomic velocity groups that interact with the lasers. The field $E_b$ induces the closed transition $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$ for a group of atoms with velocity $v_1$, as the dashed lines indicate in the box I of figure 2(b). As we scan the frequency of $E_a$, it executes each one of the allowed transitions, resulting in a smaller absorption of the field $E_b$ at these specific frequencies, creating the peaks on the $T_b$ curve.

As for the transmission $T_a$, the field $E_b$ now selects three groups of atoms with velocities $v_1$, $v_2$ and $v_3$, promoting the transition to the excited states $F = 4, F = 3$ and $F = 2$, respectively. Once again, as we scan $E_a$, it will have the resonance frequency of the closed transition, as the box II indicates in figure 2(b). For each velocity group, the optical pumping due to $E_b$ lowers the absorption of $E_a$ and generates the three peaks in the opposite order of energy growth [22–24]. We also note that the transmission of the scanning laser $T_a$ is embedded with the Doppler profile of absorption, while the transmission from the fixed frequency laser $T_b$ is over a flat background. This explains the difference observed in the wings of the two transmissions.

The FWM signal we obtain is due to a degenerate process when both fields interact with the same velocity group $v_1$. In this case, as figure 2(b) shows the two incident fields induce the closed transition to the excited state $F = 4$. The FWM signal can be observed for any frequency position inside the Doppler profile; however, since the intensity is proportional to the population distribution, the maximum signal is observed at the peak of the Doppler profile. Notice that the two FWM signals in figure 2(a) present a small frequency separation. One would expect that since both processes are nearly identical, the output signals should not have different positions in the spectrum. As we discuss in the following section, such features are a result of a steep variation of the refractive index due to the interaction of the medium with both incident beams. This kind of behavior of the refractive index also appears for some coherent effects such as EIA [25].

To highlight the presence of the EIA phenomenon, in some of our experimental configurations, we present one of the transmission curves alongside the two FWM signals in figure 3. In this case, the ratio between the intensities of the incident beams is about three, with $E_b$ being more powerful ($I_b/I_a \sim 3$). In figure 3(a), we scan the frequency of the field $E_a$ while $E_b$ has a fixed frequency and vice versa for figure 3(b).

Since the FWM signals are related to the same incident fields, involving only an interchange of the role of each beam, they present the same intensity relation given by the incident beams. Therefore, the FWM signals are normalized using the highest value between them. As in figure 2(a), these generated signals appear again with a small frequency separation. However, we notice an interchange in the frequency positions depending on which beam is scanning. The relative frequency
Figure 3. Measurements of $T_a$ and FWM signals as a function of the frequency detuning of the scanning beam, for a ratio intensity of incident beams of three. In this case the signals were normalized in respect to the larger one of them. (a) Curves for $E_a$ scanning and $E_b$ fixed in frequency. (b) Curves for $E_b$ scanning and $E_a$ fixed in frequency. Insets, zoom of the peak corresponding to the cyclic transition on the $T_a$ curve in each measurement.

Figure 4. Behavior of the two FWM signals at different intensities of the beams $E_a$ and $E_b$ at the entrance of the Rb cell. All curves are independently normalized.

Figure 5. Three-level theoretical model with (a) and (b) being the processes that generate the two circular components of the signal, with frequency $\omega_s$, in the $2\vec{k}_a - \vec{k}_b$ direction; (c) and (d) are analogous to the $2\vec{k}_b - \vec{k}_a$ direction.
Our treatment of the problem begins by considering an electric dipole coupling as the interaction Hamiltonian

$$\hat{H}_{\text{int}} = -\hbar \sum_{j \neq k} \left( \hat{\Omega}_j e^{i \omega_j t - \delta_j} + \text{c.c.} \right) |j\rangle \langle k|,$$

where $\hat{\Omega}_j = \frac{\mu_j E_j}{2\hbar}$ ($j = a$ or $b$) is the Rabi frequency with $\mu_j$ being the transition dipole moment and $E_j$ the amplitude of the electric field; $\omega_j$ is the optical frequency and $k_j$ is the wave-vector associated to the fields indicated in figure 5. Using this Hamiltonian it is possible to write Liouville’s equation

$$\frac{\partial \rho_{jk}}{\partial t} = -i(\omega_{jk} + \gamma_{jk} + \gamma')\rho_{jk} - \frac{1}{\hbar} \langle j | \hat{H}_{\text{int}}, \hat{\rho} | k \rangle,$$

where $\gamma_{jk}$ is the decay rate of the density matrix element $\rho_{jk}$, $\gamma'$ is the time of flight decay rate and $\omega_{jk}$ is the frequency of the $|j\rangle \rightarrow |k\rangle$ transition.

To model the FWM field, we must solve the Bloch equations (obtained from equation (2)) self-consistently with the Maxwell equations. Since we are interested in the FWM signal at the $2\mathbf{k}_a - \mathbf{k}_b$ and $2\mathbf{k}_b - \mathbf{k}_a$ directions, we can calculate the induced coherence $\rho_{13}$ between the two ground states [1] and [3], in all orders of the incident fields; and then, add the interaction with one of the fields, $E_a$ or $E_b$, to generate the $\rho_{21}'$ or $\rho_{23}'$ coherences at the frequencies $\omega_a = 2\omega_b - \omega_a$ and $\omega_a = 2\omega_b - \omega_a$. In fact, for each direction, both coherences $\rho_{21}'$ and $\rho_{23}'$ contribute to the generated signal, each one leading to a certain circular component of the signal, as figures 5(a) and (b) show.

The joint contribution of the two coherences for the FWM signal in each direction can be equivalently regarded as the scattering of the two circular components of each field $E_a$ or $E_b$ by the coherence $\rho_{13}$. As the fields co-propagate with a small angle, the scattered fields will travel in different directions.

Both signals generated in the experiment might be modeled with the same set of equations. Therefore, we choose to obtain the equations that describe the $2\mathbf{k}_a - \mathbf{k}_b$ process. We also write the equation for only the circular component of figure 5(a), adding both of them at the end of the calculation. Furthermore, we are interested initially in the coherence $\rho_{13}$, so we use a three-level system with a single field in each transition, allowing the use of the rotating wave approximation and a steady-state solution of the equations. In this case, the Bloch equations can be written as:

$$\rho_{11}' = -i\sigma_{12}\rho_{21} + i\sigma_{21}\rho_{12}' + \Gamma_{21}\rho_{22} + \gamma'\rho_{11}',$$

$$\rho_{22}' = i\sigma_{23}\rho_{32} - i\sigma_{32}\rho_{23} + \Gamma_{12} + \gamma'\rho_{22},$$

$$\rho_{33}' = i\sigma_{31}\rho_{13} - i\sigma_{13}\rho_{31} + \Gamma_{23} + \gamma'\rho_{33},$$

$$\sigma_{12} = -i\rho_{12}(\rho_{12} - \rho_{21})\rho_{22}' + i\rho_{13}\Omega_{13}';$$

$$\sigma_{13} = -i\rho_{13}\rho_{12} + i\rho_{23}\Omega_{23}';$$

$$\sigma_{23} = -i\rho_{23}(\rho_{23} - \rho_{32})\rho_{33}' + i\rho_{13}\Omega_{13}',$$

where $\gamma_{jk}$ terms are the coherence from equation (2) in the rotating frame, whereas $\Gamma_{jk}$ are the decay rates of the populations; the Rabi frequency has been redefined to include the spatial phase $\Omega_{jk} = \Omega_j e^{-i\delta_j}$; $\delta_j$ is the detuning of each laser with respect to the $|j\rangle \rightarrow |k\rangle$ transition; $\rho_{jk}'$ are the populations in the absence of the fields and represent the terms that compensate the loss of atoms from the interaction region with an arrival of new atoms in the ground state at a rate $\gamma'$. The missing coherence equations are the complex conjugate of the ones presented.

We obtain the coherence $\sigma_{13}$ with the help of a linear algebra suite. The final solution is intricate, but a simpler version with a dependency on the population terms of the matrix density is shown:

$$\sigma_{13} = \frac{-\Omega_{12}^*\rho_{21} + (\rho_{12} - \rho_{21})}{\rho_{12} + \gamma_{12} + \gamma'} e^{-i(\mathbf{k}_a - \mathbf{k}_b)},$$

$$\sigma_{23} = \frac{i\sigma_{12}^*\rho_{23} e^{-i(\mathbf{k}_a - \mathbf{k}_b)}}{-i\delta_a + \gamma_{32} + \gamma'}.$$

With the full solution of equation (4), we look for the coherence $\sigma_{23}'$. This density-matrix element, responsible for the FWM field with frequency $\omega_{23} = 2\omega_b - \omega_a$, wave-vector $\mathbf{k}_a$ in the direction of $2\mathbf{k}_a - \mathbf{k}_b$ and a circular polarization, is described by:

$$\sigma_{23}' = \frac{i\sigma_{12}^*\rho_{23} e^{-i(\mathbf{k}_a - \mathbf{k}_b)}}{-i\delta_a + \gamma_{32} + \gamma'}.$$

The asymmetry and frequency separation observed in the experimental spectra is not yet present in the theoretical results. This response indicates that we must include the propagation of the generated light in the atomic medium, with special attention to the phase-matching conditions of each signal. Therefore, our goal is to obtain the generated electric field $E_g(\mathbf{r}, t) = E_g e^{-i\omega_{g}\mathbf{r} + \mathbf{k}_a}$, given by the wave equation obtained from Maxwell’s equations

$$\frac{\partial^2 E_g(\mathbf{r}, t)}{\partial z^2} - \frac{n^2 c^2}{\epsilon_0 c^2} \frac{\partial^2 E_g(\mathbf{r}, t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 P(\mathbf{r}, t)}{\partial t^2}.$$
where we write the macroscopic polarization as \( P = P_{\text{linear}} + P_{\text{NL}} \). We followed the treatment developed in reference [14], assuming that both input fields \( E_a \) and \( E_b \) are strong enough to allow us to neglect their absorption. As the FWM processes occur near resonance, we include the linear term to describe the absorption of the generated field. This term is modeled by the first-order susceptibility \( \chi^{(1)} \)

\[
P_{\text{linear}} = \frac{\hbar \epsilon_0}{\rho_{12}} \chi^{(1)} \tilde{\Omega}_s e^{-i\omega_r + \tilde{k}_r r} = \frac{iN\mu_{12}\rho_{11}}{\hbar} \frac{\chi^{(1)} \tilde{\Omega}_s e^{-i\omega_r + \tilde{k}_r r}}{-i\delta_s + \gamma_1 + \gamma^*},
\]

(7)

where \( N \) is the atomic density; \( \delta_s \) is the detuning of the generated signal; \( \tilde{\Omega}_s \) is the Rabi frequency of the FWM field.

As for the nonlinear part, given by \( P_{\text{NL}} = N\text{Tr}(\rho\rho) \) [29], it is connected to the right-hand side of equation (5). Moreover, we neglect the transversal components, projecting the fields in the \( \hat{z} \) direction, taken to be the bisector between the incident fields (see figure 1(b)).

We solve the wave equation for the Rabi frequency, resulting in

\[
\tilde{\Omega}_s = \frac{\kappa^* \gamma_3 \gamma_{31}}{-i\delta_s + \gamma_3 + \gamma^*} \left( -i\delta_s + \gamma_3 + \gamma^* \right) ^{-1},
\]

(8)

where the constant \( \kappa = \frac{\omega_0 \gamma_3^2}{\hbar \epsilon_0} \) and \( \tilde{k}_r \) is the wavenumber.

From figure 1(b) we may write the phase-matching conditions, \( \Delta k = |\tilde{k}_r| \), for both FWM processes, generated in the \( 2\tilde{k}_a - \tilde{k}_b \) and \( 2\tilde{k}_b - \tilde{k}_a \) directions, with the field \( E_b \) with a fixed frequency while we scan the field \( E_a \)

\[
\Delta k_{2\tilde{k}_a - \tilde{k}_b} = \frac{2\omega_a}{c} \left[ n_a \cos(\theta) - n_b \cos(3\theta) \right] - \frac{\omega_b n_b}{c} \cos(\theta) - \cos(3\theta) \right];
\]

(9)

\[
\Delta k_{2\tilde{k}_b - \tilde{k}_a} = \frac{2\omega_b}{c} \left[ \cos(\theta) - \cos(3\theta) \right] - \frac{\omega_a n_a}{c} \cos(\theta) - n_b \cos(3\theta). \]

We write only the phase-matching in the bisector direction between the incident fields, since the perpendicular direction might be neglected as \( \theta \) is small. This leads to a rather symmetrical result. We also consider the index of refraction for the generated signal as the index of the field with a fixed frequency.

To model the refractive indexes \( n_a \) or \( n_b \) we use the real part of the electric susceptibilities of the transition in which laser is. Since these susceptibilities are essentially the coherences that we may extract solving the Bloch equations (equation (3)) to all orders, we write the refractive indexes for both beams in the \( 2\tilde{k}_a - \tilde{k}_b \) process as

\[
n_a = 1 + \frac{N\mu_{23}^2}{2\hbar\epsilon_0} \frac{\text{Re}(\sigma_{21})}{\Omega_a};
\]

\[
n_b = 1 + \frac{N\mu_{23}^2}{2\hbar\epsilon_0} \frac{\text{Re}(\sigma_{21})}{\Omega_b}.
\]

(10)

The curves for the refractive index as a function of the detuning of the \( E_a \) field are presented in figure 6(a) for a stationary atom. It is important to notice that while \( n_b \) behaves as one would expect, i.e. it grows with frequency but has an abnormal dispersion around the resonance, \( n_a \) does not follow the usual behavior. On the other hand, it is interesting that \( n_a \) is, in fact, a function of the detuning of the \( a \) laser, an indicator of the interaction between the fields due to the atomic medium.

It is in the behavior of \( n_a \) that lies the key factor for the spectral position of the FWM signals. In the place of the anomalous dispersion, there is a window with two of such dispersion and an inflection point on the resonance. This is a typical feature of the EIA process [25], observed in the transmission of the input beams. We can only achieve these refractive index curves by solving the Bloch equations, equation (3), in all orders. With
this, we allow for high-intensity coherent effects to play a role in the model.

With the refractive index modeled, we present the phase-matching condition for both FWM signals in figure 6(b), again for an atom with no velocity. Notice that the $\Delta k$ for the $2\vec{k}_a - \vec{k}_b$ signal is much closer to zero below the resonance and therefore the FWM signal itself should appear on the same region of the spectrum, as presented in the solid curves of figure 6(c) (orange/light curve). The same argument applies to the other signal, $2\vec{k}_b - \vec{k}_a$ (wine/black curve). If one considers $\Delta k = 0$, i.e. no phase mismatch, the result is shown in the dashed curve of figure 6(c). In this case, both FWM spectra are overlapping, broad, and identical.

The modeling of the refractive index that goes in equation (10) is critical to understand the features of the experimental signal. If this refractive index behaves as it usually does in a medium with a resonance, i.e. it grows with the frequency with an anomalous dispersion window around the resonance, then the FWM signal $2\vec{k}_b - \vec{k}_a$ would only exist above resonance, in complete disagreement with the experiment.

If one chooses to neglect any dispersion effects in the phenomenon, the phase mismatch due to the angle could be obtained, however for a large detuning (of hundreds of MHz). Therefore, angle alone does not provide the proper $\Delta k$ to be compensated with only the laser detuning. It could be the case if the ground states were not degenerate and therefore, lasers $E_a$ and $E_b$ had different wavenumbers [20].

One must take into account the Doppler broadening due to the high temperature of the vapor. Consequently, to obtain the FWM spectra in figure 6(c) we integrate equation (8) with the Maxwell–Boltzmann velocity distribution. There is a small difference in amplitude between the two results, but we cannot confirm if this matches the experiment since there are intensity fluctuations that prevent us from clearly seeing the amplitude relation between signals. In these curves, the atomic density is of the order of $10^{12}$ cm$^{-3}$ and the intensity of both input fields is ten times the saturation intensity of the transition.

The chosen intensity and atomic density are in agreement with the experimental range of the parameters. However, it is important to state that while we can change the intensity in the experiment by a factor of ten and still obtain the same FWM process, the theoretical model fails after a change in intensity by a factor of two. So, although our model does not provide a complete description of the process, it points in the direction of an explanation for the main features of our FWM experimental result.

The hypothesis supported by our model is, therefore, that the nonlinear interaction between the fields in the medium leads to two fundamental responses: (i) a window with two dispersive curves in the behavior of the refractive index of the scanning laser, like in the EIA process; (ii) a variable refractive index for the laser with a fixed frequency. These two effects combine to form the appropriate phase-matching condition to generate one signal below resonance and the other above.

4. Conclusions

We have investigated the excitation spectra of two symmetrical FWM signals generated in rubidium vapor, using a copropagating laser beams configuration. The nonlinear signals were induced by two independent lasers when both were tuned on the closed transition $^{85}\text{Rb} \ 5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F = 4)$, resulting in a single peak in each spectrum. Although this degenerate FWM process is well known, we have detected the two signals simultaneously and explored the symmetry between them. Noteworthy, the results have revealed some anomalies in the index of refraction of the atomic medium induced by the interaction with both fields.

An interesting point is that, even though the two signals are generated by two independent FWM processes, they provide information about the dynamic of an ensemble of atoms that interacted simultaneously with the same excitation fields. This feature can be used to investigate the interaction between the atoms within the excitation region or to explore some correlation induced by the atomic system. In particular, the degeneracy of the nonlinear process in combination with the configuration of the fields leads to a symmetry in the signals, both spatial and in frequency, regardless of which beam is used to probe the excitation spectrum.

The anomalies of the refractive index are unveiled by two experimental features: (i) an absorption dip in the transmission of the beam that is scanning in frequency, like in an EIA process, and (ii) a frequency shift of both FWM signals, in opposite directions, determined by the phase-matching condition. Our theoretical analysis, applied to a three-level system, shows how the index of refraction seen by each beam can change during the interaction process. Most importantly, the correct description of the frequency position of each peak is supported by a variable refractive index for both lasers, in which the scanning laser experiences an anomalous window in the refractive index near resonance.

Acknowledgments

This work was supported by CAPES (PROEX 534/2018, No. 23038.003382/2018-39). A C de Almeida acknowledges financial support by CNPq (141103/2019-1).

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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