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Observation of nonlinear optical phenomenon in vacuum by four waves mixing

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Abstract

In the present paper, we studied possibility of observation and detection of nonlinear optical effects in free space. We studied four wave mixing process in which signal is generated opposite to the direction of probe beam. We observed that large number of photons are generated in signal beam, which should be detectable. Key Words: Nonlinear optics, Maxwell's equations, Four wave mixing, Nonlinear Lagrangian for electromagnetic field.

1. Introduction

Nonlinear interaction between electromagnetic fields in vacuum are well known in Quantum Electrodynamics [1-9]. In quantum electrodynamics the interaction between two photons in vacuum and production of virtual electron positron has been already described [1-6] and the possibility of measuring various signals due to elastic scattering of photons of one another has been investigated theoretically [10-15]. The self-interaction of photon can also be described in terms of Feynman diagram and for two photon interaction the Nonlinear Lagrangian for electromagnetic field [5], which is correct to terms of order e^4 .

$$L = \frac{1}{8\pi} [E^2 - B^2 + K\{(E^2 - B^2)^2 + 7(E \cdot B)^2\}] , \quad K = \frac{e^4}{45\pi^4 m^4} \quad (1)$$

in the natural system of units, $c = \hbar = 1$.

This Lagrangian is identical with the results of Euler and Schwinger [2-3]. Mc. Kenna and Platzmann [6] are first who studied this Lagrangian to study the nonlinear interaction of light in vacuum and they obtained the modified Maxwell's equations. Earlier some authors have been used these modified Maxwell's equation to study Birefringence of vacuum and other effects [7-9, 16-20].

Earlier we used this Lagrangian [4] to solve the problem of birefringence of vacuum in presence of a counterpropagating electromagnetic wave [19]. In earlier work we studied the birefringence of vacuum in presence of a counter propagating electromagnetic wave and found that quite large number of photons are generated in the orthogonal mode. The detection of these photons is however difficult because of non-availability of pure polarized light. The newly generated photons are far too less in number than those present initially in this mode. We note that, this problem arises due to the identity of the generated photons being the same as that of some photons present initially. We remove this in the present paper by giving a new identity to the newly generate photons. In this paper we tried to give an independent identity to the newly generated photons via momentum conservation process such as four wave mixing.

Degenerate four wave mixing was proposed by Hellworth in 1977 [21]. All the four optical fields involved in this mixing process are of the same frequency ω hence the name degenerate. In this process two pump waves injected light of the same frequency ω and wave vectors $\mathbf{k}_1(\omega)$ and $\mathbf{k}_2(\omega)$, from opposite

direction through a nonlinear medium. Then when a third wave of frequency ω (probe wave) and wave vector $\mathbf{k}_3(\omega)$ is injected, phase conjugate light of the same frequency emerged in opposite direction. This emergent light has the wave vector as given below-

$$\mathbf{k}_4(\omega) = \mathbf{k}_1(\omega) + \mathbf{k}_2(\omega) - \mathbf{k}_3(\omega) = -\mathbf{k}_3(\omega)$$

This is the phase matching condition. When the probe frequency is different from that of the pump wave, we have nondegenerate four wave mixing. This is a standard method for generating phase conjugate light by using gases, liquids, dielectrics, semiconductors, organic materials, and non-crystalloids etc. Degenerate four wave mixing was proposed almost the same time by Yariv, et al, [22]. The experiment that first suggested the practical value of degenerate four wave mixing was performed in 1977 [23]. Earlier Four Wave mixing have been used to study some important effects in vacuum [24-31]. Four wave mixing was first used in quantum optics to produce quadrature squeezed light [32]. Later, several authors studied squeezing in Four wave mixing [33-40]. Four wave mixing has some more applications such as, detection of violation of Cauchy-Schwartz and Bell's inequality [41], Quantum effects of the atom cavity interaction [42]. In this paper we study four waves mixing to observe nonlinear optics in vacuum.

Lagrangian (1) leads to the usual Maxwell equations,

$$\nabla \cdot \mathbf{D} = 0, \quad \nabla \cdot \mathbf{B} = 0, \quad \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}. \quad (2a-d)$$

but with the constitutive relations,

$$\mathbf{D} = \mathbf{E} + K [2 (\mathbf{E}^2 - \mathbf{B}^2) \mathbf{E} + 7 (\mathbf{E} \cdot \mathbf{B}) \mathbf{B}], \quad (3)$$

$$\mathbf{H} = \mathbf{B} + K [2 (\mathbf{E}^2 - \mathbf{B}^2) \mathbf{B} - 7 (\mathbf{E} \cdot \mathbf{B}) \mathbf{E}]. \quad (4)$$

for free space.

2. General consideration for mixing of four waves moving along or against any direction

If we consider that four participating beams travel in the same direction given by, say, the unit vector $\hat{\mathbf{e}}_3$, the electric field can be written as $\mathbf{E} = \hat{\mathbf{e}}_3 E(t - \hat{\mathbf{e}}_3 \cdot \mathbf{x})$ and the magnetic field is $\mathbf{B} = [\hat{\mathbf{e}}_3 \times \mathbf{E}]$. For such a case, $\mathbf{E}^2 - \mathbf{B}^2 = \mathbf{E} \cdot \mathbf{B} = 0$ and no nonlinear effects can be seen.

In view of energy and momentum consideration, the other possible situation is where a pair of beams move forward and the other backward. If we consider a probe beam of frequency ω_p moving along $\hat{\mathbf{e}}_3$ and the two pump beams of frequency ω_0 , one moving along $\hat{\mathbf{e}}_3$ and the other along $-\hat{\mathbf{e}}_3$, the signal beam of frequency ω_s moving along $-\hat{\mathbf{e}}_3$ will be generated. We may now consider cases where either (i) one photon from each of forward and backward moving pump modes is absorbed or (ii) one photon of probe beam and one of backward moving pump beam are absorbed. In the former case generated signal frequency is $\omega_s = 2\omega_0 - \omega_p$ but with a small momentum mismatch given by $k_s - k_p = 2(\omega_p \sim \omega_0)$. In the latter case, again two situations may arise, viz, (A) where emitted pump photons move forward and backward moving signal of frequency $\omega_s = \omega_p$ is generated with a small momentum mismatch equal to $2(\omega_p \sim \omega_0)$, or (B) where the emitted pump photon moves backward and a forward moving signal photon of frequency $\omega_s = \omega_p$ and there is no momentum mismatch. The last case describes a phenomenon in which polarization of the probe beam changes due to presence of counter-propagating pump beam and has been studied earlier

[6]. Because of the momentum mismatch, we have to consider a small interaction volume, which we obtain by considering pulsed pump beams.

For consideration of generation of a signal with a new identity (frequency and direction of propagation), therefore we have to consider following two cases.

Case I: Absorption from pump beams $(\omega_0, \mathbf{e}_3)$ and $(\omega_0, -\mathbf{e}_3)$ and emission into probe beam $(\omega_p, \mathbf{e}_3)$ and a signal beam $(\omega_s = 2\omega_0 - \omega_p, -\mathbf{e}_3)$,

Case II: Absorption from probe beam $(\omega_p, \mathbf{e}_3)$ and pump beam $(\omega_0, -\mathbf{e}_3)$ and emission into pump beam $(\omega_0, \mathbf{e}_3)$ and a signal beam $(\omega_s = \omega_p, -\mathbf{e}_3)$.

3. Semi Classical Theory

Generation of signal by nonlinear interaction of the two pump and a probe beam can be considered through the nonlinear terms in Equations (3) and (4). For case I, therefore, let us consider the electric field

$$\mathbf{E} = \mathbf{E}_+ e^{-i\psi_+} + \mathbf{E}_- e^{-i\psi_-} + \mathbf{E}_p^* e^{i\psi_p} + \text{c.c.}, \quad (5)$$

where c.c. stands for complex conjugate and

$$\psi_{\pm} = \omega_0(t \mp x_3), \quad \psi_p = \omega_p(t - x_3). \quad (6)$$

Here subscripts $\pm$ refer to the directions $\pm \mathbf{e}_3$ of the two pump beams and p to the probe beam.

Obviously, we have to look for the terms involving a product of $\mathbf{E}_+, \mathbf{E}_-, \mathbf{E}_p^*$ in the nonlinear terms of Equations (3) and (4) leading to generation of frequency $\omega_s = 2\omega_0 - \omega_p$, in this case.

Let the generated signal beam be denoted by

$$\mathbf{E}_s = \mathbf{E}_s e^{-i\psi_s} + \text{c.c.}, \quad \mathbf{E}_s = \mathbf{E}_s(x_3), \quad \psi_s = \omega_s(t - x_3), \quad \omega_s = 2\omega_0 - \omega_p \quad (7)$$

We consider x_3 -dependence of $\mathbf{E}_s$ but not of $\mathbf{E}_{\pm}$ and $\mathbf{E}_p$ because the latter three are supposed to be intense and there is no appreciable depletion of these. This leads to

$$\mathbf{B}_s = -[\mathbf{e}_3 \times \mathbf{E}_s] e^{-i\psi_s} - \frac{i}{\omega} [\mathbf{e}_3 \times d\mathbf{E}_s/dx_3] e^{-i\psi_s} \quad (8)$$

Equations (5-6) lead to

$$\mathbf{B} = [\mathbf{e}_3 \times \mathbf{E}_+] e^{-i\psi_+} - [\mathbf{e}_3 \times \mathbf{E}_-] e^{-i\psi_-} + [\mathbf{e}_3 \times \mathbf{E}_p^*] e^{i\psi_p} + \text{c.c.} \quad (9)$$

and therefore to

$$(\mathbf{E}^2 - \mathbf{B}^2)\mathbf{E} = 4 \left[(\mathbf{E}_+ \cdot \mathbf{E}_-) + (\mathbf{E}_p^* \cdot \mathbf{E}_-) \mathbf{E}_+ \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \quad (10)$$

$$(\mathbf{E}^2 - \mathbf{B}^2)\mathbf{B} = 4 \left[(\mathbf{E}_+ \cdot \mathbf{E}_-) (\mathbf{e}_3 \times \mathbf{E}_p^*) + (\mathbf{E}_p^* \cdot \mathbf{E}_-) (\mathbf{e}_3 \times \mathbf{E}_+) \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \quad (11)$$

$$(\mathbf{E} \cdot \mathbf{B})\mathbf{B} = 2 \left[(\mathbf{e}_3 \cdot [\mathbf{E}_+ \times \mathbf{E}_-]) (\mathbf{e}_3 \times \mathbf{E}_p^*) + (\mathbf{e}_3 \cdot [\mathbf{E}_p^* \times \mathbf{E}_-]) (\mathbf{e}_3 \times \mathbf{E}_+) \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \quad (12)$$

$$(\mathbf{E} \cdot \mathbf{B})\mathbf{E} = 2 \left[(\mathbf{e}_3 \cdot [\mathbf{E}_+ \times \mathbf{E}_-]) \mathbf{E}_p^* + (\mathbf{e}_3 \cdot [\mathbf{E}_p^* \times \mathbf{E}_-]) \mathbf{E}_+ \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \quad (13)$$

where o.t. stands for other terms.

If we substitute the sums of electric fields given by Equations (5) and (7) for $\mathbf{E}$ and the sum of magnetic field given by (8) and (9) for $\mathbf{B}$ in the nonlinear constitutive relations (3) and (4), expressions (10-11) lead to,

$$\mathcal{D} = \mathcal{D}_+ e^{-i\psi_+} + \mathcal{D}_- e^{-i\psi_-} + \mathcal{D}_p e^{-i\psi_p} + \mathcal{D}_s e^{-i\psi_s} + \text{c.c.} \quad (14)$$

$$\mathcal{H} = \mathcal{H}_+ e^{-i\psi_+} + \mathcal{H}_- e^{-i\psi_-} + \mathcal{H}_p e^{-i\psi_p} + \mathcal{H}_s e^{-i\psi_s} + \text{c.c.} \quad (15)$$

with

$$\begin{aligned} \mathcal{D}_s = \mathcal{E}_s + 2K \left[4(\mathcal{E}_+ \cdot \mathcal{E}_-) \mathcal{E}_p^* + 4(\mathcal{E}_p^* \cdot \mathcal{E}_-) \mathcal{E}_+ + 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \right) (e_3 \times \mathcal{E}_+) \right. \\ \left. + 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \right) (e_3 \times \mathcal{E}_+) \right] e^{i\Delta k x_3} \end{aligned} \quad (16)$$

$$\begin{aligned} \mathcal{H}_s [e_3 \times \mathcal{E}_s] - \frac{i}{\omega} [e_3 \times d\mathcal{E}_s/dx_3] + 2K \left[4(\mathcal{E}_+ \cdot \mathcal{E}_-) (e_3 \times \mathcal{E}_p^*) + 4(\mathcal{E}_p^* \cdot \mathcal{E}_-) (e_3 \times \mathcal{E}_+) \right. \\ \left. - 7 \left(e_3 \cdot [\mathcal{E}_+ \times \mathcal{E}_-] \mathcal{E}_p^* \right) - 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \mathcal{E}_+ \right) \right] \end{aligned} \quad (17)$$

where $\Delta k = 2(\omega_0 - \omega_p)$ give the momentum mismatch.

Substitution in $\dot{\mathbf{D}}$ and $\nabla \times \mathbf{H}$ leads to terms in signal frequency,

$$\begin{aligned} -i\omega_s \mathcal{E}_s - 2i\omega_s K \left[4(\mathcal{E}_+ \cdot \mathcal{E}_-) \mathcal{E}_p^* + 4(\mathcal{E}_p^* \cdot \mathcal{E}_-) \mathcal{E}_+ + 7 \left(e_3 \cdot [\mathcal{E}_+ \times \mathcal{E}_-] \right) (e_3 \times \mathcal{E}_p^*) \right. \\ \left. + 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \right) \mathcal{E}_+ \right] (e_3 \times \mathcal{E}_+) \end{aligned}$$

and

$$\begin{aligned} -i\omega_s \mathcal{E}_s + 2d\mathcal{E}_s/dx_3 + 2i\omega_p K \left[4(\mathcal{E}_+ \cdot \mathcal{E}_-) \mathcal{E}_p^* + 4(\mathcal{E}_p^* \cdot \mathcal{E}_-) \mathcal{E}_+ + 7 \left(e_3 \cdot [\mathcal{E}_+ \times \mathcal{E}_-] \right) (e_3 \times \mathcal{E}_p^*) \right. \\ \left. + 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \right) \mathcal{E}_+ \right] (e_3 \times \mathcal{E}_+) \end{aligned}$$

respectively.

Maxwell's Equations (2 d) then leads to

$$\begin{aligned} \frac{d\mathcal{E}_s}{dx_3} = -i(\omega_s + \omega_p) K \left[4(\mathcal{E}_+ \cdot \mathcal{E}_-) \mathcal{E}_p^* + 4(\mathcal{E}_p^* \cdot \mathcal{E}_-) \mathcal{E}_+ + 7 \left(e_3 \cdot [\mathcal{E}_+ \times \mathcal{E}_-] \right) (e_3 \times \mathcal{E}_p^*) \right. \\ \left. + 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \right) \mathcal{E}_+ \right] (e_3 \times \mathcal{E}_+) e^{i\Delta k x_3} \end{aligned} \quad (18)$$

If the overlap region is of length L and initially $\mathcal{E}_s = 0$, at the exit,

$$\mathcal{E}_s(L) = -i\omega_0 K L F_I \quad \text{f}_{\text{mismatch}} \quad (19)$$

where

$$\begin{aligned} F_I = 2 \left[4(\mathcal{E}_+ \cdot \mathcal{E}_-) \mathcal{E}_p^* + 4(\mathcal{E}_p^* \cdot \mathcal{E}_-) \mathcal{E}_+ + 7 \left(e_3 \cdot [\mathcal{E}_+ \times \mathcal{E}_-] \right) (e_3 \times \mathcal{E}_p^*) \right. \\ \left. + 7 \left(e_3 \cdot [\mathcal{E}_p^* \times \mathcal{E}_-] \right) \mathcal{E}_+ \right] (e_3 \times \mathcal{E}_+) \end{aligned} \quad (20)$$

$$\text{and } f_{\text{mismatch}} = \frac{e^{i\Delta k L} - 1}{ikL} \quad (21)$$

For case II of generation of frequency $\omega_s = \omega_p$, we look for the terms involving product of $\mathcal{E}_-$, $\mathcal{E}_p, \mathcal{E}_+^*$. We start with writing,

$$E = \mathcal{E}_+^* e^{i\psi_+} + \mathcal{E}_- e^{-i\psi_-} + \mathcal{E}_p e^{-i\psi_p} + \text{c.c} \quad (22)$$

$$B = [e_3 \times \mathcal{E}_+^*] e^{i\psi_+} - [e_3 \times \mathcal{E}_-] e^{-i\psi_-} + [e_3 \times \mathcal{E}_p] e^{-i\psi_p} + \text{c.c.} \quad (23)$$

and proceeding exactly in the same way. We are then lead to

$$\begin{aligned} \frac{d\mathcal{E}_s}{dx_3} = & -2i\omega_0 K L \left[4(\mathcal{E}_+^* \mathcal{E}_-) \mathcal{E}_p + 4(\mathcal{E}_p \mathcal{E}_-) \mathcal{E}_+^* + \left[7(e_3 \cdot [\mathcal{E}_+^* \times \mathcal{E}_-]) (e_3 \times \mathcal{E}_p) \right] \right. \\ & \left. + 7(e_3 \cdot [\mathcal{E}_p \times \mathcal{E}_-]) (e_3 \times \mathcal{E}_+^*) \right] e^{i\Delta k x_3} \end{aligned} \quad (24)$$

and therefore to

$$\mathcal{E}_s(L) = -i\omega_0 K L F_I$$

where

$$\begin{aligned} F_{II} = & -2i\omega_0 K L \left[4(\mathcal{E}_+^* \mathcal{E}_-) \mathcal{E}_p + 4(\mathcal{E}_p \mathcal{E}_-) \mathcal{E}_+^* + \left[7(e_3 \cdot [\mathcal{E}_+^* \times \mathcal{E}_-]) (e_3 \times \mathcal{E}_p) \right] \right. \\ & \left. + 7(e_3 \cdot [\mathcal{E}_p \times \mathcal{E}_-]) (e_3 \times \mathcal{E}_+^*) \right] e^{i\Delta k x_3} \end{aligned} \quad (25)$$

For the special cases of plane polarized light, both cases I & II lead to

$$\mathcal{E}_s(L) = -i P \omega_s K L \mathcal{E}_+ \mathcal{E}_- \mathcal{E}_p^* e^{f_{\text{mismatch}}} \quad (26)$$

On putting $\mathcal{E}_\pm = \mathcal{E}_\pm \in_\pm$, $\mathcal{E}_p = \mathcal{E}_p \in_p$, we get $F_I = F_{II} = P e$, where $(P, e) = (16, e_x)$, $(-6, e_y)$, $(28, e_y)$ and $(-6, e_x)$ for $(\in_\pm = \in_p = e_x)$, $(\in_\pm = e_x, \in_p = e_y)$, $(\in_+ = e_x, \in_- = e_y, \in_p = e_x)$ and $(\in_+ = e_x, \in_- = e_y, \in_p = e_y)$ respectively. If we consider pulses of radiation, for any pulse of energy $\mathcal{E}$, intensity I area A , pulse length L and duration $\tau = L$, we have

$$I = \frac{1}{2\pi} |\mathcal{E}|^2, |\mathcal{E}| = I \tau A = I L A, \quad (27)$$

which leads to

$$\frac{I_s}{I_p} = \frac{16 \pi^4 P^2 K^2 \epsilon_+ \epsilon_-}{A^2 \lambda_0^2} |f_{\text{mismatch}}|^2 \quad (28)$$

4. Quantum Theory

The Lagrangian density given in equation (1) leads to the Hamiltonian density in radiation gauge,

$$\begin{aligned}\hat{H} &= \hat{H}_0 + \hat{H}_{NL}, \quad \hat{H}_0 = \frac{1}{8\pi} (\hat{E}^2 + \hat{B}^2), \\ \hat{H} &= \frac{1}{8\pi} \left[(\hat{E}^2 + \hat{B}^2) + K \left\{ (\hat{E}^2 - \hat{B}^2)^2 + 2 (\hat{E}^2 + \hat{B}^2)(\hat{E}^2 - \hat{B}^2) + 7 (\hat{E} \cdot \hat{B})^2 \right\} \right]\end{aligned}\quad (29)$$

Where caps on symbols are used to emphasize that they are operators not c-numbers.

For case I we can write in the radiation gauge

$$\hat{A} = \sqrt{\frac{2\pi}{\omega_0 V}} [\hat{\epsilon}_+ \hat{a}_+ e^{-i\psi_+} + \hat{\epsilon}_- \hat{a}_- e^{-i\psi_-}] + \sqrt{\frac{2\pi}{\omega_p V}} \hat{\epsilon}_p^* \hat{a}_p^\dagger e^{i\psi_p} + \sqrt{\frac{2\pi}{\omega_s V}} \hat{\epsilon}_s^* \hat{a}_s^\dagger e^{i\psi_s} + \text{h.c.}$$

where V is the interaction volume and h.c. stands for hermitian conjugate and $\hat{a}_\pm$ and $\hat{a}_{p,s}$ are annihilation operators for the forward and backward moving pump beam, probe beam and signal beam respectively, having phases ψ_+ , ψ_- , ψ_p and ψ_s defined in Equations (6) and (7).

We can calculate $\hat{E} = -\dot{\hat{A}}$, $\hat{B} = \nabla \times \hat{A}$ and then get

$$\begin{aligned}\hat{E}^2 - \hat{B}^2 &= \frac{8\pi\omega_0}{V} (\hat{\epsilon}_+ \cdot \hat{\epsilon}_-) \hat{a}_+ \hat{a}_- e^{-i(\psi_+ + \psi_-)} - \frac{8\pi\sqrt{\omega_p \omega_s}}{V} (\hat{\epsilon}_p^* \cdot \hat{\epsilon}_s^*) \hat{a}_p^\dagger \hat{a}_s^\dagger e^{i(\psi_s + \psi_p)} \\ &\quad + \frac{8\pi\sqrt{\omega_- \omega_p}}{V} (\hat{\epsilon}_- \cdot \hat{\epsilon}_p^*) \hat{a}_- \hat{a}_p^\dagger e^{-i(\psi_- - \psi_p)} + \frac{8\pi\sqrt{\omega_+ \omega_s}}{V} (\hat{\epsilon}_+ \cdot \hat{\epsilon}_s^*) \hat{a}_+ \hat{a}_s^\dagger e^{-i(\psi_+ - \psi_s)} + \text{o.t.}\end{aligned}\quad (30)$$

$$\hat{E}^2 + \hat{B}^2 = \frac{8\pi\sqrt{\omega_+ \omega_p}}{V} (\hat{\epsilon}_+ \cdot \hat{\epsilon}_p^*) \hat{a}_+ \hat{a}_p^\dagger e^{-i(\psi_+ - \psi_p)} + \frac{8\pi\sqrt{\omega_+ \omega_s}}{V} (\hat{\epsilon}_- \cdot \hat{\epsilon}_s^*) \hat{a}_- \hat{a}_s^\dagger e^{-i(\psi_- - \psi_s)} + \text{o.t.}\quad (31)$$

and

$$\begin{aligned}\hat{E} \cdot \hat{B} &= -\frac{4\pi\omega_0}{V} (\mathbf{e}_3, \hat{\epsilon}_+, \hat{\epsilon}_-) \hat{a}_+ \hat{a}_- e^{-i(\psi_+ + \psi_-)} - \frac{4\pi\sqrt{\omega_0 \omega_s}}{V} (\mathbf{e}_3, \hat{\epsilon}_+, \hat{\epsilon}_s^*) \hat{a}_+ \hat{a}_s^\dagger e^{-i(\psi_+ - \psi_s)} \\ &\quad + \frac{4\pi\sqrt{\omega_- \omega_s}}{V} (\mathbf{e}_3, \hat{\epsilon}_-, \hat{\epsilon}_p^*) \hat{a}_- \hat{a}_p^\dagger e^{-i(\psi_- - \psi_p)} + \frac{4\pi\sqrt{\omega_p \omega_s}}{V} (\mathbf{e}_3, \hat{\epsilon}_s, \hat{\epsilon}_p^*) \hat{a}_+ \hat{a}_s^\dagger e^{i(\psi_p + \psi_s)} + \text{o.t.}\end{aligned}\quad (32)$$

In case I and II we look for terms involving $\hat{a}_+ \hat{a}_- \hat{a}_p^\dagger \hat{a}_s^\dagger$ and $\hat{a}_+^\dagger \hat{a}_- \hat{a}_p \hat{a}_s$ respectively.

For interaction volume $V = AL$, we get the Hamiltonian

$$\begin{aligned}\mathcal{H}_{NL} &= \frac{2\pi\omega_0\sqrt{\omega_p \omega_s}}{V} K \left[4(\hat{\epsilon}_+ \cdot \hat{\epsilon}_-) (\hat{\epsilon}_p^* \cdot \hat{\epsilon}_s^*) + 4(\hat{\epsilon}_- \cdot \hat{\epsilon}_p^*) (\hat{\epsilon}_+ \cdot \hat{\epsilon}_s^*) - 7(\mathbf{e}_3 \cdot [\hat{\epsilon}_+ \times \hat{\epsilon}_-]) \right. \\ &\quad \left. (\mathbf{e}_3 \cdot [\hat{\epsilon}_s^* \times \hat{\epsilon}_p^*]) - 7(\mathbf{e}_3 \cdot [\hat{\epsilon}_+ \times \hat{\epsilon}_s^*]) (\mathbf{e}_3 \cdot [\hat{\epsilon}_- \times \hat{\epsilon}_p^*]) \right] \hat{a}_+ \hat{a}_- \hat{a}_p^\dagger \hat{a}_s^\dagger + \text{f}_{\text{mismatch}} + \text{o.t.}\end{aligned}\quad (33)$$

Equation of motion of $\hat{a}_s$ is

$$\begin{aligned}i\dot{\hat{a}}_s &= [\hat{a}_s, \mathcal{H}_{NL}] \\ &= \frac{2\pi\omega_0\sqrt{\omega_p \omega_s}}{V} K \left[4(\hat{\epsilon}_+ \cdot \hat{\epsilon}_-) (\hat{\epsilon}_p^* \cdot \hat{\epsilon}_s^*) + 4(\hat{\epsilon}_- \cdot \hat{\epsilon}_p^*) (\hat{\epsilon}_+ \cdot \hat{\epsilon}_s^*) - 7(\mathbf{e}_3 \cdot [\hat{\epsilon}_+ \times \hat{\epsilon}_-]) \right. \\ &\quad \left. (\mathbf{e}_3 \cdot [\hat{\epsilon}_s^* \times \hat{\epsilon}_p^*]) - 7(\mathbf{e}_3 \cdot [\hat{\epsilon}_+ \times \hat{\epsilon}_s^*]) (\mathbf{e}_3 \cdot [\hat{\epsilon}_- \times \hat{\epsilon}_p^*]) \right] \tau \hat{a}_+ \hat{a}_- \hat{a}_p^\dagger + \text{f}_{\text{mismatch}}\end{aligned}\quad (34)$$

The total number of photons N are given by $N = \frac{\varepsilon}{\omega_0} = \frac{I \tau A}{\omega_0}$. This identification leads to the signal photons

N_s given in terms of photons $N_{\pm}$, N_p (each $\gg 1$) of the pump and probe beams by

$$\frac{N_s}{N_p} = \frac{16 \pi^4}{V^2 \lambda_0^2} P^2 K^2 \tau^2 \omega_s \omega_p N_+ N_- |\text{fmismatch}|^2 \quad (35)$$

which is identical with classical results of $\frac{I_s}{I_p}$.

5. Result and Discussion

If four beams move on the same line, the principal task is their separation required for incidence and for detection. Two co-propagating beams having orthogonal polarization can be separated by use of a polarizing beam splitter. If the two copropagating beams of different frequency have the same polarization, separation may be affected by a simple prism using dispersion. If the two beams are counterpropagating and have orthogonal polarizations, a polarizing beam splitter can be used for separation. For two counter propagating waves having the same polarization, one can use a Faraday-rotation-cell giving rotation by $\pi/4$ and a polarizing beam splitter for which the two directions of polarization have the direction of polarization of the given two beams as the bisector.

To make $\frac{I_s}{I_p}$ large, one may consider the two pump beams having intense light pulses of ultra-

short duration. For duration $\sim 10\text{fs}$ fmismatch can be ~ 1 . If we divide a 10KJ, 10fs pulse into two and get the counter propagating pump beams focused on area $(10\mu^2)$, we have

$$\frac{I_s}{I_p} \sim 7.74 \times 10^{-16},$$

for the maximum value of P . For this case if the probe beam has energy 1J in the interaction volume, a pulse of signal of energy $0.774 \times 10^{-15}\text{J}$ will be produced which shall have 2×10^3 photons. This should be detectable easily.

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8. References

- 1 O. Halpern, Scattering process produced by electrons in negative energy states, Phys Rev 44 (1933) 855.
- 2 W. Heisenberg and H. Euler, Consequences of the Dirac Theory of Positrons, Z. Phys 98, 714 (1936).
- 3 J. Schwinger, On Gauge invariance and vacuum polarization, Phys Rev 82 (1951) 664.
- 4 R. Karplus, M. Neuman, Non linear interactions between electromagnetic fields, Phys Rev 80 (1950) 380.
- 5 R. Karplus, M. Neuman, The scattering of light by light. Phys Rev 83 (1951) 776.
- 6 J. Mc. Kenna, P. M. Platzman, Nonlinear interaction of light in a vacuum, Phys Rev 129 (1963) 2354.
- 7 T. Heinzl et al., On the observation of vacuum birefringence, Opt. Commun. 267(2006) 31821.
- 8 J. Klein and B.P. Nigam, Birefringence of the vacuum, Phys. Rev. 135 (1964) B1279.

- 9 B. King, A. Di Piazza and C. H. Keitel, Double-self vacuum polarization effects in ultra-intense laser field, *Phys. Rev. A* 82 (2010) 032114.
- 10 A. Ferrando et al, Nonlinear phase shift from photon photon scattering in vacuum, *Phys. Rev. Lett.* 99 (2007) 150404.
- 11 B. King, A. Di Piazza, c.H. Keitel, A matterless double slit, *Nature Photon*, 4 (2010) 92-94.
- 12 D. Tommasini and H Michinel, Light by light diffraction in vacuum, *Phys. Rev. A* 82 (2010) 011803.
- 13 Y. Monden and R. Kodama, Enhancement of laser interaction with vacuum for a large angular aperture, *Phys. Rev. Lett.* 107 (2011) 073602.
- 14 G.Y. Kryuchkyan and K. XZ. Hatsagortsyan, Bragg scattering of light in vacuum structured by strong periodic fields, *Phys. Rev. Lett.* 107 (2011) 053604.
- 15 B. King, C.H. Keitel, Photon –photon scattering in collisions of intense laser pulses, *New J. Phys.* 14 (2012) 053604.
- 16 R. Baier and P. Breitenlohner, Photon propagation in external fields, *Acta Phys. Austriaca* 25 (1967) 212.
- 17 Z. Bialynicka- Birula and I Bialynicki-Birula, Photon propagation and photon splitting in an external field, *Phys. Rev. D* 2 (1970) 2341.
- 18 E. Brezin and C. Itzykson, Polarization phenomenon in vacuum Nonlinear electrodynamics, *Phys. Rev. D* 3 (1971) 618.
- 19 R. Prakash, P. Shukla, Birefringence of vacuum in the presence of a counterpropagating electromagnetic field, *International Journal of Modern Physics B* 21 (2007) 3045.
- 20 D. Bakalov, et al, Experimental method to detect the magnetic birefringence of vacuum, *Quantum Semiclass. Opt.* 10 (1980) 239-250.
- 21 R. W. Hellwarth, Generation of time reversed wave fronts by nonlinear refraction, *J. Opt. Soc. Am* 67 (1977) 1.
- 22 A. Yariv, D. M. Peeper, Amplified reflection, phase conjugation and oscillation in degenerate four wave mixing, *Opt. Lett.* 1 (1997) 16.
- 23 D. M. Bloom, G. C. Bjorklund, Conjugate wave fronts generation and image reconstruction by four wave mixing, *App. Phys. Lett* 31 (1977) 592.
- 24 Marcus Beutler, Masood Ghotbi, Frank Noack, and Ingolf Volker Hertel, Generation of sub -50—fs vacuum ultraviolet pulses by four wave mixing in argon, *Optics Letters*, 35 (2010) 1491-3.
- 25 João L. Silva, Helder M. Crespo, and Rosa Weigand, Generation of high energy vacuum ultraviolet femtosecond pulses by multiple beam cascaded Four wave mixing in a transparent solid, *Applied optics*, 50 (2011) 1968-73.
- 26 I. Babushkin, F. Noack, J. Herrmann, Sub-10-fs pulse generation in vacuum ultraviolet using Chirped Four wave mixing in hollow fibers, *chemical physics* 92(2009)762-764.
- 27 Jan Renger, Romain Quidant, Niek van Hulst, Stefano Palomba, and Lukas Novotny, Free space excitation of propagating surface plasmon polaritons by nonlinear Four wave mixing, *Phys Rev. Lett.* 103(2009) 266802.
- 28 C. Dorman, J. P. Marangos & J.C. Petch, Investigation of vacuum ultraviolet generation in a resonant four wave mixing scheme in krypton with electromagnetically induced transparency, *Journal of Modern optics* 45(1998) 1123-1135.
- 29 F. Moulin, D. Bernard , Four wave mixing interaction in gas and vacuum. Definition of a third order nonlinear effective susceptibility in vacuum: χ^3_{vacuum} , *Optics Communication* 164(1999) 137-144.
- 30 J. Lundin et al, Detection of elastic photon- photon scattering through four wave mixing using high power lasers, *Phys. Rev. A* 74(2006) 043821.
- 31 D. Bernard, F. Moulin, F. Amiranoff, A. Braun, J.P. Chambaret, G. Darpentigny, G. Grillon S. Ranc, F. Perrone, Search for stimulated photon-photon scattering in vacuum, *Eur. Phys. J. D.* 10 (2000) 141.
- 32 H. P. Yuen, J. H. Shapiro, Generation and detection of two-photon coherent states in degenerate four-wave mixing, *Opt. Lett.* 4 (1979) 334.
- 33 P.Kumar, J. H. Shapiro, Squeezed state generation via four wave mixing, *Phys. Rev. A.* 30 (1984) 1568.
- 34 M. D. Ried, D. F. Walls, Quantum statistics of degenerate four wave mixing, *Opt. Comm.* 50 (1984) 406.

- 35 M. D. Ried, D. F. Walls, Generation of squeezed states via degenerate four wave mixing, Phys. Rev. A. 31 (1985) 1622.
- 36 B. Yurke, Squeezed coherent state generation via four wave mixing in an optical cavity Phys. Rev. A. 32 (1985) 300.
- 37 Pramila Shukla, Ranjana Prakash, Ordinary and Amplitude squared squeezing in four wave mixing process, Modern Physics Letter B 27 (2013)135086.
- 38 Ranjana Prakash, Pramila Shukla, Detection of Sum and Difference Squeezing, IOSR Journal of Applied Physics 1 (2012) 43.
- 39 Ranjana Prakash, Pramila Shukla, Radiation Squeezing for M two-Level Atoms Interacting with a Single Mode Coherent Radiation, Chinese Journal of Physics 53 (2015) 100901-8.
- 40 D. K. Giri, P. S. Gupta, The squeezing of radiation in four wave mixing process J. Opt. B.: Qum. Sem. Opt. 6 (2004) 91.
- 41 A. Nadeem, Ansari, M. S. Zubery, Violation of Cauchy Schwartz inequality and Bell's inequality in four wave mixing Phys. Rev. A. 38 (1988) 2380.
- 42 G. V. Varda, M.S. Kumar, G. S. Agarwal Quantum effects of the atom cavity interaction on four wave mixing Opt. Comm. 62 (1987) 328.

9. Declaration:

- The author have no relevant financial interests to disclose
- The authors have no conflict of interest to declare that are relevant to content of this article.
- All authors certify that they have no affiliations with or involvement in any organization or entity with nay financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.
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