Studies of Tunable Four-Photon Parametric Fluorescence of Crystals in the Visible Region

H. ITO, H. INABA
Research Institute of Electrical Communication, Tohoku University, Sendai, Japan

Received 20 May 1970

Tunable four-photon parametric fluorescence, which is valuable for investigating material nonlinearities and parametric devices, was studied theoretically and experimentally. Theoretical analysis leads to the conclusion that the emitted power depends directly on the focusing of the pumping beam, unlike the process of three-photon parametric fluorescence which is independent of the focusing of the pumping beam. The calculation of the phase-matching conditions for nonlinear crystals such as KDP, ADP, TiO$_2$ and CdS shows that collinear phase-matching can be realised over the visible region using a ruby laser for pumping. Experimentally, tunable emission was observed in the visible range from 4200 Å to 5300 Å with KDP and ADP crystals pumped by a Q-switched ruby laser.

1. Introduction
Tunable optical parametric fluorescence (OPF) due to the second-order nonlinearity in crystals has been observed by several authors [1-4]. The emission is produced spontaneously by parametric scattering when a pumping photon splits into signal and idler photons. This phenomenon is inherently different from parametric oscillation [5] since it may still be observable even at quite low levels of pumping far from the threshold for oscillation. Alternatively, the effect can be interpreted as the quantum noise associated with zero-point fluctuations of the signal and idler modes. Kleinman [6] has theoretically analysed this spontaneous parametric process involving three photons, and has concluded that focusing of the pumping beam does not enhance the interaction.

Recently, observations of four-photon OPF in water [7] and calcite [8] have been reported in conjunction with a Q-switched ruby laser. According to our analysis of four-photon OPF based on the third-order nonlinearity of the material, the emitted power depends on the focusing as in the second harmonic generation (SHG) process. The main advantage of this OPF is that using the direct pumping of a ruby or neodymium laser, the frequency region of the signal emission is distributed over the visible range. The four-wave parametric interaction also provides a useful means of measuring the nonlinear susceptibility tensor of the fourth rank (such as that responsible for third harmonic generation (THG)) for various materials over a wide range of wavelengths. This yields the great potentiality of knowing the third order nonlinearity in materials where strong absorption in the uv region prevents the detection of THG, and also avoids the complexity of measuring powers at different wavelengths which in the case of THG are far apart. Furthermore, this technique offers valuable information on the phase-matching condition and the pumping energy to be used in four-photon optical parametric oscillators, which can be operated in the frequency region close to or even on the shorter wavelength side of the pumping field.

From this point of view, we are carrying out the analysis of and experiments on, four-photon OPF using centrosymmetric as well as noncentrosymmetric materials. This paper reports a part of these results on collinearly phase-matchable and tunable interactions in some crystals customarily used in nonlinear optics and optical electronics.
2. Analysis and Calculation of Phase-Matching Condition

The energy and momentum conservations for the four-photon OPF require the following conditions:

\[ 2\omega_3 = \omega_1 + \omega_2 \]  
\[ 2k_3 = k_1 + k_2 \]

where suffixes 1, 2 and 3 correspond to signal, idler, and pumping waves, respectively. Here we are considering the degenerate type of four-photon coupling which gives rise to the absorption of a couple of pumping photons \( \hbar \omega_3 \), accompanied simultaneously by the emission of signal and idler photons, \( \hbar \omega_1 \) and \( \hbar \omega_2 \).

Theoretical analysis [9] performed by extending Kleinman's theory to the four-photon interaction gives the fluorescence intensity of the signal,

\[ \Delta P_1 = \Delta \nu_1 \Delta \Omega_1 \frac{144\pi^2 h n_2 V_2 \omega_1 \omega_2}{n_3 c^6 n_2 V_2z} \frac{P_3^2}{\lambda s^2 L^3} \frac{\sin^2 (4kL/2)}{(4kL/2)^2} \]

where the pumping beam is assumed to be a plane wave, \( \Delta \nu_1 \) and \( \Delta \Omega_1 \) are bandwidth and solid angle of the monochromator, \( n \) the refractive index, \( A \) the beam cross section, \( L \) the length of the nonlinear material, \( P_3 \) the pumping power, \( 4k (= 2k_3 - k_1 - k_2) \) is the wave-vector mismatch taken in the direction of the pumping, \( V_2 \) the group velocity of the idler wave, and \( V_2z \) is component of \( V_2 \) along the pumping beam. It is seen that the spontaneously emitted power is proportional to \( P_3^2/\lambda s \), as is the case for SHG. This indicates that the signal intensity depends on the coherent properties of the pumping, unlike the process of three-photon OPF which is independent of the focusing of the pumping beam.

The momentum conservation or phase-matching condition can be realised either collinearly or noncollinearly by controlling temperature, angle, or electro- or acousto-optic character of the crystal for the tuning. Conceivable collinear phase-matching conditions which are achievable for a ruby laser pumping beam have been numerically analysed for various nonlinear crystals by taking account of possible combinations of ordinary and extraordinary rays. No consideration is given, however, to