III-4. Multiple Quantum Frequency Conversion in Extended Interaction Structures

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In 1959, E. T. Jaynes[1] suggested that resonant, multi-quantum processes could provide an efficient method of frequency conversion at high frequency and high powers. This conversion is mediated by a gas or solid composed of molecules having a strong electric-dipole absorption at a frequency Ω. Such a system can resonantly absorb energy from an electromagnetic field at all odd subharmonics of Ω. The $\omega \approx \Omega/(2n+1)$ subharmonic absorption is said to correspond to a $2n+1$ quantum process. The excited (driven) molecular system has electric dipole moments at all odd harmonics of the drive ω. The resonant dipole moment at a frequency $(2n+1)\omega \approx \Omega$ generates an electromagnetic field resulting in the frequency conversion process proposed by Jaynes. Here we report theoretical calculations and experimental observations of this multi-quantum frequency conversion in a traveling-wave structure.

The dynamics of the frequency conversion process are determined by the coupled system of Maxwell's equations describing the electromagnetic field propagating in the molecular medium and the Schrodinger equation describing the molecular system in the presence of the electromagnetic field. The solution of the Schrodinger equation for the dipole response of the molecules provides a constitutive relation relating the polarizability of the molecular medium to the electromagnetic field E.

Analysis shows that only odd harmonics of the drive frequency ω appear. For a three-quantum conversion process in which $\omega \approx \Omega/3$, only the first and third harmonics are important, and we find that[2]

$$P_1 = N\mu \frac{9}{8}\left(1 + \left(\frac{\delta}{\beta}\right)^2\right) x_1, \tag{1}$$

$$P_3 = N\mu \frac{1}{2\beta}\left(\frac{\delta}{\beta} - i\right)\left(x_3 - \frac{9}{16}x_1^3\right). \tag{2}$$

Here P_1 and P_3 are the first and third harmonic amplitudes of the polarizability, and x_1 and x_3 are the first and third harmonic amplitudes of the electric field in units of $(\hbar\Omega/2\mu)$. The effective molecular dipole moment is μ and the differential population of the molecular energy levels associated with the absorption at Ω is denoted by N. The tuning factor

$$\delta = \left[\frac{3\omega}{\Omega}\left(1 - \frac{9}{8}|x_1|^2 - \frac{1}{4}|x_3|^2\right) - 1\right]$$

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is zero "on resonance," and $\beta^{-1} = \tau\Omega$ where τ is the relaxation time associated with the spectral line at Ω.

Using the constitutive equations (1) and (2), we have solved Maxwell's equations for a straight TEM section. We find that the normalized third harmonic field amplitude x_3 builds up with distance, z, down the guide as

$$\frac{x_3(z)}{\frac{9}{16}x_1^3} = 1 - e^{-\frac{\gamma z}{2}}. \tag{3}$$

Here

$$\gamma = \frac{4\pi}{3}\frac{N\mu^2\Omega}{\beta}\ (\mathrm{cm}^{-1})$$

is the absorption coefficient of the molecular system at frequency Ω; and Eq. (3) is valid when $\gamma \gg \alpha_1, \alpha_3$, where α_1 and α_3 are the waveguide attenuation factors measured at ω and 3ω in cm^{-1}. The Poynting vector associated with the energy transport at a frequency 3ω varies as $x_3^2(z)$, and the power converted per unit volume as $dx_3^2(z)/dz$. From Eq. (3), the power converted per unit volume, normalized to the maximum power converted per unit volume, is

$$W(z) = 4(1 - e^{-\gamma z/2})\, e^{-\gamma z/2}. \tag{4}$$

The local "molecular efficiency," defined as the ratio of power absorbed at ω to power emitted at 3ω by the molecular system at z, is

$$\mathcal{E}(z) = 1 - e^{-\gamma z/2}. \tag{5}$$

A fraction $(1 - \mathcal{E})$ of the power absorbed is dissipated in the kinetic (lattice) degrees of freedom of the molecular medium. The average efficiency from $z = 0$ to z is

$$\bar{\mathcal{E}}(z) = \frac{\int_0^z \mathcal{E}(z)\,dz}{z}. \tag{6}$$

The relations (3), (4), (5) and (6) are plotted in Figures 1, 2, 3 and 4. Note that when $x_3/(9/16)x_1^3 = 1/2$ (with $\gamma z \approx 1.4$), the power converted per unit volume is a maximum, and the local molecular efficiency is 1/2. As $x_3/(9/16)x_1^3$ approaches one, $(\gamma z \longrightarrow \infty)$, the power conversion per unit volume approaches zero, while the local molecular efficiency approaches one.

In order to obtain conversion over the entire length of the interaction structure, it is necessary that the phase velocity at ω be the same as that at 3ω. The phase shifts ϕ_1 and ϕ_3 at the first and third harmonics due to the molecular medium are

$$\phi_1 = \frac{3}{8}\beta\gamma z \tag{7}$$

$$\phi_3 = \frac{1}{2}\frac{\delta}{\beta}\gamma z. \tag{8}$$

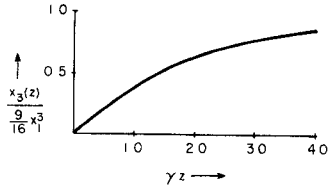

Fig. 1. The field amplitude ratio $x_3(z)/(9/16)\, x_1^3$ vs γz.

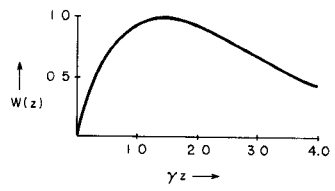

Fig. 2. Normalized power conversion $W(z)$ vs γz.

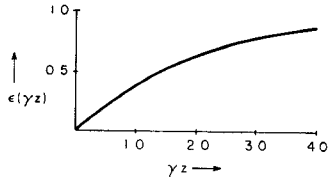

Fig. 3. The local molecular efficiency $\mathcal{E}$ vs γz.

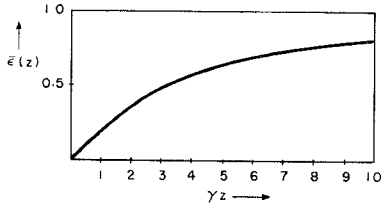

Fig. 4. The average efficiency $\bar{\mathcal{E}}(z)$ for a section of length z vs γz.

From Figs. 1 through 4, it follows that γz values of order 1 to 10 are of interest, so that if β and $\delta/\beta \ll 1$, the correct phase matching is maintained by a simple TEM section.

Experimental confirmation of these theoretical predictions has been obtained. The frequency converter consisted of a coaxial line 2.5 meters in length, inner diameter .555 cm, and impedance 11.3 ohms. This low impedance structure was chosen to minimize the radial variation of the E-field, since the resonance condition $\delta = 0$ depends upon $|x_1|^2$. This line was filled with NH_3 at pressures between 50 and 300 mm Hg. At 300 mm Hg, the structure would support fields of 16 kv/cm corresponding to $x_1 = 1/2$. The strong NH_3 inversion lines at approximately 25 kmc (K-band) were subharmonically driven at 8.3 kmc (X-band), and 3-quantum conversion to 24.9 kmc was observed. The 8.3 kmc drive was obtained from a magnetron source passed through a harmonic filter with a relative attenuation factor of order 20 db at 24.9 kmc. Even without the filter, the third harmonic power level coming from the magnetron is several orders of magnitude below the observed signal level produced in the coaxial region. The field strength at 24.9 kmc builds up as shown in Fig. 1. By taps on the coaxial line at various distances from the X-band input, the conversion at different values of z could be determined. These measurements of K-band power indicate that an estimated K-band power of 100 watts peak exists in the line. Local molecular efficiencies at the end of the structure of the order of 60 per cent were obtained.

Using a C-band source at 5 kmc, 5-quantum conversion to 25 kmc was observed using this same NH_3 filled coaxial structure. These results are also in essential agreement with our theoretical predictions.

REFERENCES

1. E. T. Jaynes, "Theoretical and Experimental Maser Research," Microwave Laboratory Report No. 671, Stanford University, 1959.
2. These constitutive relations are valid when $\beta >> (9/16)\, x_1^3$. For the case in which $\beta < (9/16)\, x_1^3$ we have obtained the constitutive relations by a truncation approximation procedure. (To be published.)