

Optically pumped Faraday rotation

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Let us assume a material with non-dispersive isotropic third-order nonlinearity. In time domain, the nonlinear polarization density reads

$$\vec{P}_{NL} = \varepsilon_0 \chi^{(3)} \vec{E}^2 \vec{E} \quad (1)$$

We assume that the field is the superposition of a strong pump \vec{E}_p and a weak signal \vec{E}_s :

$$\vec{E} = \vec{E}_p + \vec{E}_s \quad (2)$$

The pump consists of two circularly polarized waves with opposite handedness and slightly different frequencies:

$$\vec{E}_p = E_1 \hat{\mathbf{e}}_+ e^{i\omega_1 t} + E_2 \hat{\mathbf{e}}_- e^{i\omega_2 t} + c.c. \quad (3)$$

where

$$\hat{\mathbf{e}}_{\pm} = \frac{\hat{\mathbf{x}} \pm i\hat{\mathbf{y}}}{\sqrt{2}} \quad (4)$$

It is easy to show that

$$\hat{\mathbf{e}}_+^* = \hat{\mathbf{e}}_- \quad (5)$$

$$\hat{\mathbf{e}}_+^2 = \hat{\mathbf{e}}_-^2 = 0 \quad (6)$$

$$\hat{\mathbf{e}}_+ \cdot \hat{\mathbf{e}}_- = 1 \quad (7)$$

The signal is right-handed circularly polarized and is given by the expression

$$\vec{E}_s = E_s \hat{\mathbf{e}}_+ e^{i\omega_s t} + c.c. \quad (8)$$

We assume that $\omega_1 < \omega_s < \omega_2$ and $\omega_1 \approx \omega_2 \approx \omega_s$.

Inserting Eq. 2 into Eq. 1 yields

$$\begin{aligned} \vec{P}_{NL} = \varepsilon_0 \chi^{(3)} (\vec{E}_p^2 \vec{E}_p + \vec{E}_p^2 \vec{E}_s + 2\vec{E}_p \vec{E}_p \cdot \vec{E}_s \\ + 2\vec{E}_p \cdot \vec{E}_s \vec{E}_s + \vec{E}_s^2 \vec{E}_p + \vec{E}_s^2 \vec{E}_s) \end{aligned} \quad (9)$$

In the following we will keep only the first order terms with respect to \vec{E}_s :

$$\vec{P}_{NL} \approx \varepsilon_0 \chi^{(3)} (\vec{E}_p^2 \vec{E}_s + 2\vec{E}_p \vec{E}_p \cdot \vec{E}_s) \quad (10)$$

We have also neglected the term $\vec{E}_p^2 \vec{E}_p$ since this term describes the self-action for the pump field and it doesn't directly affect the signal's (\vec{E}_s) properties.

Inserting Eqs. 3 and 8 into the first term of Eq. 10 and considering the identities in Eqs. 5-7 yields

$$\begin{aligned}
\vec{P}_{NL,1} &= \varepsilon_0 \chi^{(3)} (E_1 \hat{e}_+ e^{i\omega_1 t} + E_2 \hat{e}_- e^{i\omega_2 t} + c.c.)^2 (E_s \hat{e}_+ e^{i\omega_s t} + c.c.) \\
&= 2\varepsilon_0 \chi^{(3)} [|E_1|^2 + |E_2|^2 + E_1 E_2 e^{i(\omega_1 + \omega_2)t} + c.c.] (E_s \hat{e}_+ e^{i\omega_s t} + c.c.) \\
&= 2\varepsilon_0 \chi^{(3)} [|E_1|^2 + |E_2|^2] (E_s \hat{e}_+ e^{i\omega_s t} + c.c.) \\
&\quad + (E_1 E_2 E_s \hat{e}_+ e^{i(\omega_1 + \omega_2 + \omega_s)t} + c.c.) \\
&\quad + (E_1 E_2 E_s^* \hat{e}_- e^{i(\omega_1 + \omega_2 - \omega_s)t} + c.c.)] \tag{11}
\end{aligned}$$

If we keep only the terms with frequencies close to $\pm\omega_s$, we get

$$\begin{aligned}
\vec{P}_{NL,1} &\approx 2\varepsilon_0 \chi^{(3)} [|E_1|^2 + |E_2|^2] (E_s \hat{e}_+ e^{i\omega_s t} + c.c.) \\
&\quad + (E_1 E_2 E_s^* \hat{e}_- e^{i(\omega_1 + \omega_2 - \omega_s)t} + c.c.)] \tag{12}
\end{aligned}$$

We do the same procedure for the second term in Eq. 10.

$$\begin{aligned}
\vec{P}_{NL,2} &= \varepsilon_0 \chi^{(3)} (E_1 \hat{e}_+ e^{i\omega_1 t} + E_2 \hat{e}_- e^{i\omega_2 t} + c.c.) [E_1 E_s^* e^{i(\omega_1 - \omega_s)t} + E_2 E_s e^{i(\omega_2 + \omega_s)t} + c.c.] \\
&= \varepsilon_0 \chi^{(3)} [E_1^2 E_s^* \hat{e}_+ e^{i(2\omega_1 - \omega_s)t} + E_1 E_2 E_s \hat{e}_+ e^{i(\omega_1 + \omega_2 + \omega_s)t} + |E_1|^2 E_s \hat{e}_+ e^{i\omega_s t} \\
&\quad + E_1^* E_2 E_s \hat{e}_- e^{i(\omega_2 - \omega_1 + \omega_s)t} + E_1 E_2 E_s^* \hat{e}_- e^{i(\omega_1 + \omega_2 - \omega_s)t} + E_2^2 E_s \hat{e}_- e^{i(2\omega_2 + \omega_s)t} \\
&\quad + |E_2|^2 E_s \hat{e}_+ e^{i\omega_s t} + E_1^* E_2 E_s \hat{e}_- e^{i(\omega_2 - \omega_1 + \omega_s)t} + c.c.] \tag{13}
\end{aligned}$$

As before, we keep only the terms with frequencies close to $\pm\omega$:

$$\begin{aligned}
\vec{P}_{NL,2} &\approx \varepsilon_0 \chi^{(3)} [E_1^2 E_s^* \hat{e}_+ e^{i(2\omega_1 - \omega_s)t} + |E_1|^2 E_s \hat{e}_+ e^{i\omega_s t} \\
&\quad + E_1^* E_2 E_s \hat{e}_- e^{i(\omega_2 - \omega_1 + \omega_s)t} + E_1 E_2 E_s^* \hat{e}_- e^{i(\omega_1 + \omega_2 - \omega_s)t} + \\
&\quad + |E_2|^2 E_s \hat{e}_+ e^{i\omega_s t} + E_1^* E_2 E_s \hat{e}_- e^{i(\omega_2 - \omega_1 + \omega_s)t} + c.c.] \tag{14}
\end{aligned}$$

From Eqs. 12 and 14, the right- and left-handed polarization densities are found as

$$\begin{aligned}
P_{NL,+} &\approx \varepsilon_0 \chi^{(3)} [3(|E_1|^2 + |E_2|^2) E_s e^{i\omega_s t} + 3E_1^* E_2^* E_s e^{-i(\omega_1 + \omega_2 - \omega_s)t} \\
&\quad + 2E_1 E_2^* E_s^* e^{-i(\omega_2 - \omega_1 + \omega_s)t} + E_1^2 E_s^* e^{i(2\omega_1 - \omega_s)t}] \tag{15}
\end{aligned}$$

$$\begin{aligned}
P_{NL,-} &\approx \varepsilon_0 \chi^{(3)} [3(|E_1|^2 + |E_2|^2) E_s^* e^{-i\omega_s t} + 3E_1 E_2 E_s^* e^{i(\omega_1 + \omega_2 - \omega_s)t} \\
&\quad + 2E_1^* E_2 E_s e^{i(\omega_2 - \omega_1 + \omega_s)t} + (E_1^*)^2 E_s e^{-i(2\omega_1 - \omega_s)t}] \tag{16}
\end{aligned}$$

Observe that $P_{NL,-} = P_{NL,+}^*$.

If the polarization of the probe signal was left-handed, the corresponding expressions would be

$$P_{NL,-} \approx \varepsilon_0 \chi^{(3)} [3(|E_1|^2 + |E_2|^2) E_s e^{i\omega_s t} + 3E_1^* E_2^* E_s e^{-i(\omega_1 + \omega_2 - \omega_s)t} + 2E_1^* E_2 E_s^* e^{-i(\omega_1 - \omega_2 + \omega_s)t} + E_2^2 E_s^* e^{i(2\omega_2 - \omega_s)t}] \quad (17)$$

$$P_{NL,+} \approx \varepsilon_0 \chi^{(3)} [3(|E_1|^2 + |E_2|^2) E_s^* e^{-i\omega_s t} + 3E_1 E_2 E_s^* e^{i(\omega_1 + \omega_2 - \omega_s)t} + 2E_1 E_2^* E_s e^{i(\omega_1 - \omega_2 + \omega_s)t} + (E_2^*)^2 E_s e^{-i(2\omega_2 - \omega_s)t}] \quad (18)$$

Assume now a cavity, e.g., a Fabry-Perot resonator, with degenerate right and left-handed modes. The coupled-mode equations for these modes are

$$\dot{a}_+ = i\omega_0 a_+ \quad (19)$$

$$\dot{a}_- = i\omega_0 a_- \quad (20)$$

where ω_0 is the resonance frequency. For simplicity, we have neglected the possible cavity loss. If the cavity is filled with a chi-3 nonlinear material and pumped as in the previous analysis, it can be found from Eqs. 15-18 that the coupled-mode equations become:

$$\dot{a}_+ = i\omega_0 a_+ + \kappa_1 e^{i2\omega_1 t} a_+^* + \lambda e^{i(\omega_1 + \omega_2)t} a_-^* + \mu e^{-i(\omega_2 - \omega_1)t} a_- \quad (21)$$

$$\dot{a}_- = i\omega_0 a_- + \kappa_2 e^{i2\omega_2 t} a_-^* + \lambda^* e^{i(\omega_1 + \omega_2)t} a_+^* + \mu^* e^{i(\omega_2 - \omega_1)t} a_+ \quad (22)$$

where $\kappa_1 \propto E_1^2$, $\kappa_2 \propto E_2^2$, $\lambda \propto 3E_1 E_2$, and $\mu \propto 2E_1 E_2^*$. In the rotating frame with frequency ω_0 , $a_{\pm} = A_{\pm} e^{i\omega_0 t}$ and Eqs. 21,22 are transformed as

$$\dot{A}_+ = \kappa_1 e^{-i2\delta t} A_+^* + \lambda A_-^* + \mu e^{-i2\delta t} A_- \quad (23)$$

$$\dot{A}_- = \kappa_2 e^{i2\delta t} A_-^* + \lambda^* A_+^* + \mu^* e^{i2\delta t} A_+ \quad (24)$$

where we have made the assumption $\omega_1 = \omega_0 - \delta$, $\omega_2 = \omega_0 + \delta$. The first two terms in Eqs. 23,24 are typical terms that give parametric amplification. The last term, is of the same form as in a spatiotemporally modulated ring with the angular-momentum approach (Nat. Commun. paper), so it's expected to give nonreciprocity.