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Chiral 매질에서, 전자기파의 비선형성과 여러겹 구조에서의 Coupled-mode theory에 관한 연구

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Theoretical study of Electromagnetic Waves in Chiral media: about Nonlinearity & Multilayers

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Abstract.
We analyze the nonlinearity of chiral media and coupled-mode theory of chiral multilayers. In first topic, second order nonlinear coulped equations are contsructed and a phase matching method is suggested. This approach can be developed to higher order nonlinearity and electric-field-induced second harmonic generation. In second topic, coupled mode equation in chiral multilayers is constructed, and solved for both codirectional coupling and contradirectional coupling. There is a previous formulation about chiral mutilayers[1] with 4×4 matrix but it did not give detail results, so this approach will be compared with that.

1. INTRODUCTION.

Since chiral or optically active media were first recognized and elucidated by Biot, Arago, Pasteur, and Fresnel in the early 19th centry, it has been interest of scientists and detail properties have been elucidated. Recently chiral reflection, refration,[2]-[5] propagation through dielectric-chiral slab,[6],[7] mutilayers,[1] waveguides[8] and etc.,[9],[10],[11] are being investigated theoretically or experimentally. And it is expected that chiral media will received increased attention because of their unique charactistics in cross coupling of elctric and magnetic fields. [12],[13]

Chiral objects[1] are those that cannot be superposed upon their mirror images. Such object appears in either left- or right-handed form. Thus the geometrical notion of chirality is linked to handedness. Chiral materials are composed of handed constituents at the microscopic and/or macroscopic levels, which are embedded in a host medium. In addition to the permittivity and the permeability, a quantity denoted the chiral admittance[9],[14] has been introduced to couple the electric field field to the magnetic field and vice versa. This leads to generalized set of constitutive relations among the electromagnetic-field vectors. The magnitude of the chirality admittance determines the strength of chirality, which controls the cross coupling of electric and magnetic fields in the medium, while its sign indicates the handedness of the medium.[15]

Because of the cross coupling effect, the eigenmodes of chiral media are left- and right-handed circular poarization which have different phase velocities. So there are different aspects although chiral media are isotropic.

There are a lot of material which exhibit chiral effect, for example, biological materials like sugars, aminoacids, DNA, crystals like Quartz, $AgGaS_2$, TeO_2 and recent theories predicted chiral effect in high-temperature supercondoctor.[16] Thus, the analyses of chiral media have importance and are applicable to many regions.

2. CHIRAL MEDIA CHARACTERISTICS.

For a medium which is homogeneous, lossless, isotropic and chiral with permitivity ε, permeability μ and chiral admittance ξ_c, the electromagnetic-field vectors are related by the chiral constitutive relations by Post relations[14] for time harmonic electromagnetic fields with $\exp[i\omega t]$ excitation, such that

$$\mathbf{D} = \varepsilon\mathbf{E} - i\xi_c\mathbf{B}, \quad \mathbf{H} = \mathbf{B}/\mu - i\xi_c\mathbf{E}. \qquad [2.1]$$

This provides direct coupling between electric and magnetic fields owing to the chirality of the medium in addition to that given by Maxwell's equations.

Using the constitutive relations in Eqs.[2.1] with Maxwell's equations, then the source-free chiral Helmholtz equation is

$$\nabla\times\nabla\times\mathbf{E} + \mu\varepsilon\frac{\partial^2\mathbf{E}}{\partial t^2} + 2i\mu\xi_c\frac{\partial}{\partial t}(\nabla\times\mathbf{E}) = 0 \quad . \qquad [2.2]$$

The eigenmode solutions of Eq.[2.2] are circularly polarized waves of either left- or right-handedness such that

$$\mathbf{E}_{F+} = \mathbf{u}_{F+}E_{F+}\mathrm{Exp}[-ik_{F+}z], \quad \mathbf{E}_{F-} = \mathbf{u}_{F-}E_{F-}\mathrm{Exp}[-ik_{F-}z],$$
$$\mathbf{E}_{B+} = \mathbf{u}_{B+}E_{B+}\mathrm{Exp}[ik_{B+}z], \quad \mathbf{E}_{B-} = \mathbf{u}_{B-}E_{B-}\mathrm{Exp}[ik_{B-}z]. \qquad [2.3]$$

where

$$k_{F+} = \omega\mu\{(\varepsilon/\mu + \xi_c^2)^{1/2} - \xi_c\}, \quad k_{F-} = \omega\mu\{(\varepsilon/\mu + \xi_c^2)^{1/2} + \xi_c\},$$
$$k_{B+} = -\omega\mu\{(\varepsilon/\mu + \xi_c^2)^{1/2} - \xi_c\}, \quad k_{B-} = -\omega\mu\{(\varepsilon/\mu + \xi_c^2)^{1/2} + \xi_c\}$$
$$\mathbf{u}_{F+} = \frac{1}{\sqrt{2}}\begin{bmatrix}1\\ i\end{bmatrix}, \quad \mathbf{u}_{F-} = \frac{1}{\sqrt{2}}\begin{bmatrix}1\\ -i\end{bmatrix}, \quad \mathbf{u}_{B+} = \frac{1}{\sqrt{2}}\begin{bmatrix}-1\\ i\end{bmatrix}, \quad \mathbf{u}_{B-} = \frac{1}{\sqrt{2}}\begin{bmatrix}-1\\ -i\end{bmatrix}$$

where [+],[-] denotes left-handedness and right-handedness respectively and F,B denotes forwad-going wave and backward-going wave respectively.

Corresponding to above results, let us define that the medium which exhibits that left circularly polarized wave propagates faster than right circularly polarized wave is left-handed medium, i.e., $\xi_c>0$ and vice versa.

3. NONLINEARITY OF CHIRAL MEDIA.

On concerning of second harmonic generation, the main thing is phase matching[17]. There are some phase mathing methods using anisotropy of materials; Type I, Type II and using waveguid. Thus isotropic materials which lack of centrosymetry cannot be candidates of SHG although they have considerable amounts of second order nonlinearity. But point-group-23 has both second order nonlinearity and chirality[17],[18], which exhibits a possibility of phase mathing by circular dichroism and some assistance.

3.1. SECOND HARMONIC GENERATION.

Only considering electric second order nonlinear polarization, the constitutive relation Eqs.[2.1] and the chiral wave equation Eq.[2.2] are changed such that

$$\mathbf{D} = \varepsilon\mathbf{E} - i\xi_c\mathbf{B} + \mathbf{P}_{NL} \qquad [3.1.1]$$

$$\nabla\times\nabla\times\mathbf{E} + \mu\varepsilon\frac{\partial^2\mathbf{E}}{\partial t^2} + 2i\mu\xi_c\frac{\partial}{\partial t}(\nabla\times\mathbf{E}) + \mu\frac{\partial^2}{\partial t^2}\mathbf{P}_{NL} = 0 \qquad [3.1.2]$$

The second order nonlinear tensor of point-group-23 has nonvanishing componets $d_{123}=d_{132}=d_{231}=d_{213}=d_{312}=d_{321}$: [17] The tensor d_{ijk} is that of principal coordinate system of crystal, thus it is necessary to transform it to the propagation coordinate system. In the system of propagation axes, z' coincides with the direction of propagation **k**, and x' is chosen so that x'z' plane contains the c axis of the crystal (z axis) and the y' axis is perpendicular to z'and x'. Then transformed second order nonlinear tensor is given by

$$d'_{\alpha\beta\gamma}=R_{\alpha i}R_{\beta j}R_{\gamma k}d_{ijk} \quad [3.1.3]$$

where $R=\begin{bmatrix}\cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta\\ -\sin\phi & \cos\phi & 0\\ \sin\theta\cos\phi & \sin\theta\sin\phi & \cos\theta\end{bmatrix}$

Yet, this is that of linear polarization system, thus one more transformation to circular polarization system is needed. Since the propapation mode is transverse (isotropic), the longitudinal component can be elliminated. Hence, the final results for second order nonlinear tensor is given by

$$C_{\alpha\beta\gamma}=T_{\alpha i}T^{*}_{\beta j}T^{*}_{\gamma k}d'_{ijk},\ \text{where}\ T=\frac{1}{\sqrt{2}}\begin{bmatrix}1 & -i\\ 1 & i\end{bmatrix} \quad [3.1.4]$$

and α,β,γ denotes [+],[-].

Notion ; the second or third term must be complex conjugated when j th or k th electric field is complex conjugated respectively.

Previously assumed that the direction of propagation is along z axis, let us specialize the problem to one dimensional by taking $\partial/\partial x=\partial/\partial y=0$, and also limit the consideration to two frequencies $\omega,2\omega$ and take the corresponding fields to be in the form of traveling plane waves as

$$E^{2\omega}_{\alpha}(z,t)=u_{\alpha}E_{2\alpha}(z)\mathrm{Exp}[i(2\omega t-k_{2\alpha}z)],$$
$$E^{\omega}_{\beta}(z,t)=u_{\beta}E_{1\beta}(z)\mathrm{Exp}[i(\omega t-k_{1\beta}z)] \quad [3.1.5]$$

From the Eq.[3.1.2] assuming that the variation of complex fields amplitudes with z is small enough so that $|kdE/dz|>>|d^2E/dz^2|$ we get following equations,

for second harmonic, 2ω

$$\frac{dE_{2\alpha}}{dz}=-i\frac{4\mu\omega^2E_{1\beta}E_{1\gamma}C_{\alpha\beta\gamma}}{4\mu\xi_{2c}(\alpha)\omega+2k_{2\alpha}}\mathrm{Exp}[i(k_{2\alpha}-k_{1\beta}-k_{1\gamma})z],$$

for fundamental harmonic, ω

$$\frac{dE_{1\beta}}{dz}=-i\frac{2\mu\omega^2E_{2\alpha}E^{*}_{1\gamma}C_{\alpha\beta\gamma^{*}}}{2\mu\xi_{1c}(\beta)\omega+2k_{1\beta}}\mathrm{Exp}[i(k_{1\beta}+k_{1\gamma}-k_{2\beta})z] \quad [3.1.6]$$

Generally in optical region, $\varepsilon(2\omega)>\varepsilon(\omega)$ by dispersion and $\xi_{2c}>\xi_{1c}$; ξ_c is almost propotional to frequency,[12],[13] i.e., $\xi_c\propto\varsigma\partial/\partial t$, to satisfy the phase matching condition, the most reasonable candidates are the pair of $E_-(\omega),E_+(2\omega)$ and $E_+(\omega),E_-(2\omega)$ in left-handed medium and in right-handed medium repectively from refering to Eqs.[2.3].

Assuming the medium is left-handed, i.e., ξ_c>0 with Eqs.[3.1.6], [2.3] we get

$$\frac{dE_{2+}}{dz}=-i\frac{\mu\omega E^2_{1-}C_{+--}}{(\mu\varepsilon_2+(\mu\xi_{2c})^2)^{1/2}}\mathrm{Exp}[i\Delta kz],$$
$$\frac{dE_{1-}}{dz}=-i\frac{\mu\omega E_{2+}E^{*}_{1-}C_{-+-^{*}}}{(\mu\varepsilon_1+(\mu\xi_{1c})^2)^{1/2}}\mathrm{Exp}[-i\Delta kz] \quad [3.1.7]$$

where $\Delta k=k_{2+}-2k_{1-}$

$$=2\omega\{(\mu\varepsilon_2+(\mu\xi_{2c})^2)^{1/2}-(\mu\varepsilon_1+(\mu\xi_{1c})^2)^{1/2}-\mu\xi_{2c}-\mu\xi_{1c}\}$$

Still Δk may be positive, so we must assist Δk to be zero with something, loading static magnetic field along z direction i.e., using Faraday effect[1]. Then the constitutive relation Eq.[2.1] is changed to be

$$\mathbf{D}=\varepsilon\mathbf{E}-i\xi_c\mathbf{B}+i\varepsilon_0\gamma\mathbf{B}_{\bullet}\times\mathbf{E},\ \text{where}\ \mathbf{B}_{\bullet}=B_0z \quad [3.1.8]$$

Together Eq.[3.1.8]and wave equation Eq.[2.2]gives that the eigenmodes are still left- and right-handed circularly polarization but the wave numbers are changed such that

$$k_+=\mu\omega\{((\varepsilon+g)/\mu+\xi_c^2)^{1/2}-\xi_c\},$$
$$k_-=\mu\omega\{((\varepsilon-g)/\mu+\xi_c^2)^{1/2}+\xi_c\} \quad [3.1.9]$$

Thus the external static magnetic field can make k value bigger or smaller to left- and right-handed polarized wave repectively and vice versa. In our case, the purpose is to make k_{2+} smaller and k_{1-} bigger, so the direction of the static magnetic field must be opposite to z axis.

Then, Eqs.[3.1.7] are changed to be

$$\frac{dE_{2+}}{dz}=-i\frac{\mu\omega E^2_{1-}C_{+--}}{((\mu\varepsilon_2-g_2)+(\mu\xi_{2c})^2)^{1/2}}\mathrm{Exp}[i\Delta kz],$$
$$\frac{dE_{1-}}{dz}=-i\frac{\mu\omega E_{2+}E^{*}_{1-}C_{-+-^{*}}}{((\mu\varepsilon_1+g_1)+(\mu\xi_{1c})^2)^{1/2}}\mathrm{Exp}[-i\Delta kz] \quad [3.1.10]$$

where $\Delta k=k_{2+}-2k_{1-}=2\omega\{(\mu(\varepsilon_2-g_2)+(\mu\xi_{2c})^2)^{1/2}$
$-(\mu(\varepsilon_1+g_1)+(\mu\xi_{1c})^2)^{1/2}-\mu\xi_{2c}-\mu\xi_{1c}\}$

The parameter g does not affect the nonlinear equation, and only ε is exchanged with $\varepsilon+g$ or $\varepsilon-g$.

3.1.1 SHG WITH UNDEPLETED INPUT.

Assuming that the amount of power lost from the input (ω) beam (by conversion to (2ω)) is negligible; i.e., $dE_1/dz\approx 0$, so we consider only the first equation of Eqs.[3.1.10]. The solution for $E_{2+}(0)=0$ and for a medium of length L is

$$E_{2+}=-i\frac{\mu\omega E^2_{1-}C_{+--}}{((\mu\varepsilon_2-g_2)+(\mu\xi_{2c})^2)^{1/2}}\frac{\mathrm{Exp}[i\Delta kL]-1}{i\Delta k} \quad [3.1.1.1]$$

3.1.2 SHG WITH DEPLETED INPUT.

If Δk is zero and the length L is long enough, then the input beam is depleted. In this case, we must take some steps to solve the problem as

$$\frac{dA_1}{dz}=-i\kappa^{*}A_2A_1^{*},\ \frac{dA_2}{dz}=-\frac{i}{2}\kappa A_1^2 \quad [3.1.2.1]$$

where

$$A_1=\sqrt{\frac{\alpha_1}{\omega}}E_{1-},\ A_2=\sqrt{\frac{\alpha_2}{2\omega}}E_{2+},\ \kappa=\frac{\omega}{\alpha_1}\sqrt{\frac{2\omega}{\alpha_2}}C_{+--};\ C_{+--}=C^{*}_{-+-^{*}},$$

$\alpha_1^2=(\varepsilon_1+g)/\mu+\xi_{1c}^2,\ \alpha_2^2=(\varepsilon_1-g)/\mu+\xi_{1c}^2$

Assuming there is no SH input, the Eqs.[3.1.2.1] give

$$\frac{d}{dz}(|A_1|^2+2|A_2|^2)=0,\ |A_1|^2+2|A_2|^2=|A_1(0)|^2 \quad [3.1.2.2]$$

We can take A_1 to be real and A_2 to be complex with constant phase. Unless A_2 has constant phase, the SH wave has different phase velocity from the eigenmode,so it is not the propagation mode, thus we take

$$A_2=|A_2|\mathrm{Exp}[i\theta],\ \kappa=|\kappa|\mathrm{Exp}[i\delta] \quad [3.1.2.3]$$

Together Eqs.[3.1.2.1],[3.1.2.2],[3.1.2.3] we get

$$\frac{d|A_2|}{dz}=\frac{1}{2}|\kappa|(\mathrm{Exp}[i(\delta-\theta-\frac{\pi}{2})])(|A_1(0)|^2-2|A_2|^2) \quad [3.1.2.4]$$

since left side is positive real; generation occurs, $\delta-\theta-\pi/2$ must be the integers' multiple of 2π.

We obtain the solution to be

$$|A_2|=\frac{1}{\sqrt{2}}A_1(0)\tanh[\frac{A_1(0)}{\sqrt{2}}|\kappa|] \quad [3.1.2.5]$$

4. COUPLED MODE THEORY OF CHIRAL MULTILAYERS.

The propagation of electromagnetic radiation in periodic media[17] exhibits many interesting and potentially useful phenomena. These phenomena are employed in many optical devices such as diffraction gratings, holograms, free-electron lasers, distributed-feedback lasers, distributed-Bragg-reflected lasers, high-reflectance Bragg mirrors, acousto-optic filters, Solc fillters, and etc.. So chiral mutilayers has a possibility to new applications.

4.1. CHIRAL REFLECTION AND REFRACTION.

For a chiral-chiral interface[1], each of the two circularly polarized

incident waves with opposite handedness is coupled to reflected and transmitted eigenmodes of both handedness. This is because the interface is matched to linear eigenmodes rather than to the circular eigen modes characteristic of chiral media. Thus planar boundaries provide coupling between the chiral eigenmodes. From a mathematical point of view, the boundary condition cannot be matched by waves of a single handedness, so waves of both handedness are needed.
From above results in section 2. we can let the incident, reflected, and refracted waves as

$$
\begin{aligned}
{}^{i}\mathbf{E} &= \frac{1}{\sqrt{2}}\left[\cos{}^{i}\theta_{+} \quad i \quad -\sin{}^{i}\theta_{+}\right]^{T}\,{}^{i}E_{+}\,e^{-ik_{+}(x\sin{}^{i}\theta_{+}+z\cos{}^{i}\theta_{+})} \\
&+\frac{1}{\sqrt{2}}\left[\cos{}^{i}\theta_{-} \quad -i \quad -\sin{}^{i}\theta_{-}\right]^{T}\,{}^{i}E_{-}\,e^{-ik_{-}(x\sin{}^{i}\theta_{-}+z\cos{}^{i}\theta_{-})}, \\
{}^{i}\mathbf{H} &= -\frac{i}{\eta}{}^{i}\mathbf{E}_{+} + \frac{i}{\eta}{}^{i}\mathbf{E}_{-}, \\
{}^{r}\mathbf{E} &= \frac{1}{\sqrt{2}}\left[-\cos{}^{r}\theta_{+} \quad i \quad -\sin{}^{r}\theta_{+}\right]^{T}\,{}^{r}E_{+}\,e^{-ik_{+}(x\sin{}^{r}\theta_{+}-z\cos{}^{r}\theta_{+})} \\
&+\frac{1}{\sqrt{2}}\left[-\cos{}^{r}\theta_{-} \quad -i \quad -\sin{}^{r}\theta_{-}\right]^{T}\,{}^{r}E_{-}\,e^{-ik_{-}(x\sin{}^{r}\theta_{-}-z\cos{}^{r}\theta_{-})}, \\
{}^{r}\mathbf{H} &= -\frac{i}{\eta}{}^{r}\mathbf{E}_{+} + \frac{i}{\eta}{}^{r}\mathbf{E}_{-}, \\
{}^{t}\mathbf{E} &= \frac{1}{\sqrt{2}}\left[\cos{}^{t}\theta_{+} \quad i \quad -\sin{}^{t}\theta_{+}\right]^{T}\,{}^{t}E_{+}\,e^{-ik'_{+}(x\sin{}^{t}\theta_{+}+z\cos{}^{t}\theta_{+})} \\
&+\frac{1}{\sqrt{2}}\left[\cos{}^{t}\theta_{-} \quad -i \quad -\sin{}^{t}\theta_{-}\right]^{T}\,{}^{t}E_{-}\,e^{-ik'_{-}(x\sin{}^{t}\theta_{-}+z\cos{}^{t}\theta_{-})}, \\
{}^{t}\mathbf{H} &= -\frac{i}{\eta'}{}^{t}\mathbf{E}_{+} + \frac{i}{\eta'}{}^{t}\mathbf{E}_{-}
\end{aligned}
\qquad [4.1.1]
$$

where η, η' are wave impedences of media as
$1/\eta = (\varepsilon/\mu + \xi_c^{2})^{1/2}$, $1/\eta' = (\varepsilon'/\mu' + \xi_c'^{2})^{1/2}$ and
${}^{i}\theta_{+}, {}^{i}\theta_{-}\ {}^{r}\theta_{+}, {}^{r}\theta_{-}, {}^{t}\theta_{+}, {}^{t}\theta_{-}$ are arbitrary yet.
From the boundary conditions, i.e., continuities of tangential components of $\mathbf{E}$ and $\mathbf{H}$ fields, we get from one of those to satisfy the boundary condition for whole boundary region,
$k_{+}\sin{}^{i}\theta_{+} = k_{-}\sin{}^{i}\theta_{-} = k_{+}\sin{}^{r}\theta_{+} = k_{-}\sin{}^{r}\theta_{-} = k'_{+}\sin{}^{t}\theta_{+} = k'_{-}\sin{}^{t}\theta_{-}$
thus, let $\theta_{+} \equiv {}^{i}\theta_{+} = {}^{r}\theta_{+},\ \theta_{-} \equiv {}^{i}\theta_{-} = {}^{r}\theta_{-},\ \theta'_{+} \equiv {}^{t}\theta_{+},\ \theta'_{-} \equiv {}^{t}\theta_{-}$ [4.1.2]
This is known as chiro-Snell' law. Thus we get those in matrix form as

$$
\begin{bmatrix} -1 & 1 & 1 & -1 \\ \cos\theta_{+} & \cos\theta_{-} & \cos\theta'_{+} & \cos\theta'_{-} \\ -1 & -1 & \eta_r & \eta_r \\ \cos\theta_{+} & -\cos\theta_{-} & \eta_r\cos\theta'_{+} & -\eta_r\cos\theta'_{-} \end{bmatrix}
\begin{bmatrix} {}^{r}\mathbf{E}_{+} \\ {}^{r}\mathbf{E}_{-} \\ {}^{t}\mathbf{E}_{+} \\ {}^{t}\mathbf{E}_{-} \end{bmatrix}
=
\begin{bmatrix} 1 & -1 \\ \cos\theta_{+} & \cos\theta_{-} \\ 1 & 1 \\ \cos\theta_{+} & -\cos\theta_{-} \end{bmatrix}
\begin{bmatrix} {}^{i}\mathbf{E}_{+} \\ {}^{i}\mathbf{E}_{-} \end{bmatrix}
$$

where $\eta_r = \eta/\eta'$ [4.1.3]
This result correponds with the privious result by D.L.Jaggard et al.,[1] so the final result needs not be written. Because the result is so long and complicated. The important thing is the couplings among the modes.

4.2. COUPLED EQUATIONS OF MULTILAYERS.

We represent the problem of chiral multilayers with four modes,i.e.,forward-going waves of both handedness and backward-going waves of both handedness, by well known coupled-mode theory.[8],[17]
We start from the Lorentz reciprocity.

$$\nabla\cdot(\mathbf{E}\times\mathbf{H}^{*} + \mathbf{E}^{*}\times\mathbf{H}) = 0 \qquad [4.2.1]$$

We verified the Lorentz reciprocity, i.e., Eq.[4.2.1] is held for eigenmodes of chiral media. It is trivial because chiral media are reciprocal media[14].
Assuming there is no change in permeability, i.e., the media are non magnetic, and the perturbations are given as $\Delta\varepsilon$, $\Delta\xi_c$, after some mathematical manipulations together the perturbed mode (it is represented with prime superscript.) and unperturbed mode with Eq.[4.2.1]and neglecting only the term $\mu\Delta\xi_c^{2}$;it is much smaller than others since we know the orders as $\varepsilon \approx 10^{-12}, \mu \approx 10^{-7}, \xi_c \approx\leq 10^{-5}$
Then we obtain

$$\nabla\cdot(\mathbf{E}'\times\mathbf{H}_n^{*} + \mathbf{E}_n^{*}\times\mathbf{H}') = -i\omega\,\{i\mu\Delta\xi_c(\mathbf{H}_n^{*}\cdot\mathbf{E}' - \mathbf{E}_n^{*}\cdot\mathbf{H}') + \Delta\varepsilon\mathbf{E}^{*}\cdot\mathbf{E}'\}$$

where [4.2.2]
the subscript n denotes one of the unperturbed eigenmodes, i.e., in our case, 1 indicates forward-going [+] handed wave, 2 indicates forward-going [-] handed wave, 3 indicates backward-going [+] handed wave, 4 indicates backward-going [-] handed wave.
Using the method of variation constant with eigenmodes, we take the perturbed mode as

$$
\begin{aligned}
\mathbf{E}' &= \sum_m a_m(z)\mathbf{E}_m, \qquad [4.2.3] \\
\mathbf{H}' &= \mathbf{B}'/\mu - i(\xi_c + \Delta\xi_c)\mathbf{E}' \\
&= \sum_m a_m(z)\mathbf{H}_m + \frac{i}{\omega\mu}\frac{da_m(z)}{dz}(\mathbf{z}\times\mathbf{E}_m) - i\Delta\xi_c a_m(z)\mathbf{E}_m
\end{aligned}
$$

At this point we define the eigenmodes refering to the above result in section 4.1. such that

$$
\begin{aligned}
&\mathbf{E}_1 = \mathbf{e}_1\,\mathrm{Exp}[-i\beta_1 z],\quad \mathbf{H}_1 = \mathbf{h}_1\,\mathrm{Exp}[-i\beta_1 z], \\
&\mathbf{E}_2 = \mathbf{e}_2\,\mathrm{Exp}[-i\beta_2 z],\quad \mathbf{H}_2 = \mathbf{h}_2\,\mathrm{Exp}[-i\beta_2 z], \\
&\mathbf{E}_3 = \mathbf{e}_3\,\mathrm{Exp}[-i\beta_3 z],\quad \mathbf{H}_3 = \mathbf{h}_3\,\mathrm{Exp}[-i\beta_3 z], \\
&\mathbf{E}_4 = \mathbf{e}_4\,\mathrm{Exp}[-i\beta_4 z],\quad \mathbf{H}_4 = \mathbf{h}_4\,\mathrm{Exp}[-i\beta_4 z] \qquad [4.2.4]
\end{aligned}
$$

where $\beta_1 = -\beta_3 = k_{+}\cos\theta_{+},\ \beta_2 = -\beta_4 = k_{-}\cos\theta_{-}$

$$
\begin{aligned}
\mathbf{e}_1 &= \frac{1}{\sqrt{2}}[\cos\theta_{+} \quad i \quad -\sin\theta_{+}]^{T}, & \mathbf{h}_1 &= -\frac{i}{\eta}\mathbf{e}_1 \\
\mathbf{e}_2 &= \frac{1}{\sqrt{2}}[\cos\theta_{-} \quad -i \quad -\sin\theta_{-}]^{T}, & \mathbf{h}_2 &= \frac{i}{\eta}\mathbf{e}_2 \\
\mathbf{e}_3 &= \frac{1}{\sqrt{2}}[-\cos\theta_{-} \quad i \quad -\sin\theta_{-}]^{T}, & \mathbf{h}_3 &= -\frac{i}{\eta}\mathbf{e}_3 \\
\mathbf{e}_4 &= \frac{1}{\sqrt{2}}[-\cos\theta_{-} \quad -i \quad -\sin\theta_{-}]^{T}, & \mathbf{h}_4 &= \frac{i}{\eta}\mathbf{e}_4
\end{aligned}
$$

Substituting Eqs.[4.2.3] with Eqs.[4.2.4] for perturbed mode in both sides of Eq.[4.2.2]
The result of left side is given to be
Left side = L(1) + L(2) + L(3)

$$
\begin{aligned}
L(1) &= \sum_m \nabla\cdot\{a_m\mathbf{E}_m\times\mathbf{H}_n^{*} + \mathbf{E}_n^{*}\times a_m\mathbf{H}_m\} \qquad [4.2.5] \\
&= \sum_m \frac{da_m}{dz}\mathbf{z}\cdot(\mathbf{e}_m\times\mathbf{h}_n^{*} + \mathbf{e}_n^{*}\times\mathbf{h}_m)\mathrm{Exp}[-i(\beta_m-\beta_n)z]
\end{aligned}
$$

in which the Lorentz reciprocity was applied for eigenmodes.

$$
\begin{aligned}
L(2) &= \sum_m \nabla\cdot\{\mathbf{E}_n^{*}\times[\frac{i}{\omega\mu}\frac{da_m}{dz}(\mathbf{z}\times\mathbf{E}_m)]\} \\
&= \sum_m \frac{1}{\omega\mu}(\beta_m-\beta_n)\frac{da_m}{dz}\mathbf{z}\cdot\{\mathbf{e}_n^{*}\times(\mathbf{z}\times\mathbf{e}_m)\} \\
&\quad \times\mathrm{Exp}[-i(\beta_m-\beta_n)z] \qquad [4.2.6]
\end{aligned}
$$

where we eliminate the second order derivative term assuming $|(\beta_m-\beta_n)da_m/dz| \gg |d^2a_m/dz^2|$.

$$
\begin{aligned}
L(3) &= \sum_m \nabla\cdot\{\mathbf{E}_n^{*}\times(-i\Delta\xi_c a_m\mathbf{E}_m)\} \\
&= \sum_m -i\{\frac{d\Delta\xi_c}{dz}a_m - i\Delta\xi_c(\beta_m-\beta_n)a_m\}\,\mathbf{z}\cdot(\mathbf{e}_n^{*}\times\mathbf{e}_m) \\
&\quad \times\mathrm{Exp}[-i(\beta_m-\beta_n)z] \qquad [4.2.7]
\end{aligned}
$$

where we eliminate the second order term $\Delta\xi_c\,da_m/dz$ assuming $|\Delta\xi_c| \ll |(\beta_m-\beta_n)/\omega\mu|$, it is identical the elimination in Eq.[4.2.2].

$$
\begin{aligned}
\text{Right side} &= \sum_m -i\omega\{\Delta\varepsilon(\mathbf{e}_n^{*}\cdot\mathbf{e}_m) + i\mu\Delta\xi_c(\mathbf{h}_n^{*}\cdot\mathbf{e}_m - \mathbf{e}_n^{*}\cdot\mathbf{h}_m)a_m\} \\
&\quad \times\mathrm{Exp}[-i(\beta_m-\beta_n)z] \qquad [4.2.8]
\end{aligned}
$$

where we eliminate the terms,$\Delta\xi_c\,da_m/dz$ and $\mu\Delta\xi_c^{2}$ as we did in Eqs.[4.2.2].[4.2.7].
Finally, from the equality of both sides, we obtain the coupled equation as

$$
\begin{aligned}
&\sum_m \frac{da_m}{dz}\mathrm{Exp}[-i\beta_m z]\{\mathbf{z}\cdot(\mathbf{e}_m\times\mathbf{h}_n^{*} + \mathbf{e}_n^{*}\times\mathbf{h}_m) \\
&+\frac{1}{\omega\mu}(\beta_m-\beta_n)\,\mathbf{z}\cdot\{\mathbf{e}_n^{*}\times(\mathbf{z}\times\mathbf{e}_m)\}\} \\
&= \sum_m a_m\mathrm{Exp}[-i\beta_m z]\{-i\omega\Delta\varepsilon(\mathbf{e}_n^{*}\cdot\mathbf{e}_m)
\end{aligned}
$$

to be continued

$$+\Delta\xi_c[(\beta_m-\beta_n)z\cdot(e_n^*\times e_m)+\omega\mu(h_n^*\cdot e_m-e_n^*\cdot h_m)]$$
$$+i\frac{d\Delta\xi_c}{dz}z\cdot(e_n^*\times e_m)\} \quad [4.2.9]$$

This equation is valid for n = 1,2,3,4, so there are four independent equations,thus with n=1,2,3,4, the Eq[4.2.9] can be constructed in matrix form after further mathematical manipulation as

$$\begin{bmatrix} a_1'e^{-i\beta_1 z} \\ a_2'e^{-i\beta_2 z} \\ a_3'e^{i\beta_1 z} \\ a_4'e^{i\beta_2 z}\end{bmatrix} = \{-i\Delta\varepsilon\begin{bmatrix} M & N \\ -N & -M\end{bmatrix}+i\Delta\xi_c\begin{bmatrix} P & Q \\ -Q & -P\end{bmatrix}-\frac{d\Delta\xi_c}{dz}\begin{bmatrix} S & T \\ T & S\end{bmatrix}\} \cdot\begin{bmatrix} a_1e^{-i\beta_1 z} & a_2e^{-i\beta_2 z} & a_3e^{i\beta_1 z} & a_4e^{i\beta_2 z}\end{bmatrix}^T$$

where [4.2.10]

$$\begin{bmatrix} M & N \\ -N & -M\end{bmatrix}=\omega\{[\mathbf{ehv}_{nm}]+\frac{1}{\omega\mu}[(\beta_m-\beta_n)\mathbf{ezev}_{nm}]\}^{-1}\cdot[\mathbf{eed}_{nm}]$$

$$\begin{bmatrix} P & Q \\ -Q & -P\end{bmatrix}=-i\{[\mathbf{ehv}_{nm}]+\frac{1}{\omega\mu}[(\beta_m-\beta_n)\mathbf{ezev}_{nm}]\}^{-1}\cdot\{[(\beta_m-\beta_n)\,\mathbf{eev}_{nm}]+\omega\mu[\mathbf{ehd}_{nm}]\}$$

$$\begin{bmatrix} S & T \\ T & S\end{bmatrix}=-i\{[\mathbf{ehv}_{nm}]+\frac{1}{\omega\mu}[(\beta_m-\beta_n)\mathbf{ezev}_{nm}]\}^{-1}\cdot[\mathbf{eev}_{nm}]$$

where the prime denotes derivative to z and $\ldots_{nm}$]'s denote matrices after the initial of terms in Eq.[4.2.9]and the eliments of matrices are submatrices in 2×2 form and all of the matrices have real values, thus we can manipulate the problem according to the amount of perturbations $\Delta\varepsilon, \Delta\xi_c$. (we can simplify the Eq.[4.2.10] further or not, such that the terms of muliple $(\beta_1-\beta_2)$, $(\beta_3-\beta_4)$ can be eliminated since the terms smaller than others by the order 0^{-3} or the smaller.)

With the coupled equation provided, we can solve the problem of chiral multilayers for arbitrary perturbations numerically or analytically for special cases.

4.3. ANALYTICAL SOUTIONS FOR SPECIAL CASES: PERIODIC LAYERS.

In the periodic layers, the perturbation of permittivity, $\Delta\varepsilon$ and the perturbation of chiral admittance, $\Delta\xi_c$ can be represented with their Fourier series such that

$$\Delta\varepsilon=\sum_{p\neq0}\varepsilon_p \mathrm{Exp}[-ip\frac{2\pi}{\Lambda}],\quad \Delta\xi_c=\sum_{p\neq0}\xi_{cp}\mathrm{Exp}[-ip\frac{2\pi}{\Lambda}] \quad [4.3.1]$$

where Λ is the period of layer and p is any integer number except for zero .

After further mathematical manipulation, together Eqs. [4.2.10], [4.3.1] we get [4.3.2]

$$\begin{bmatrix} a_1'e^{-i\beta_1 z} \\ a_2'e^{-i\beta_2 z} \\ a_3'e^{i\beta_1 z} \\ a_4'e^{i\beta_2 z}\end{bmatrix} = \sum_{p\neq0} e^{-ip\frac{2\pi}{\Lambda}}\{-i\varepsilon_p\begin{bmatrix} M & N \\ -N & -M\end{bmatrix}+i\xi_{cp}(\begin{bmatrix} P & Q \\ -Q & -P\end{bmatrix}-p\frac{2\pi}{\Lambda}\begin{bmatrix} S & T \\ T & S\end{bmatrix})\} \cdot\begin{bmatrix} a_1e^{-i\beta_1 z} & a_2e^{-i\beta_2 z} & a_3e^{i\beta_1 z} & a_4e^{i\beta_2 z}\end{bmatrix}^T$$

By the rule of well-known coupled-mode theory, we can consider only the terms which satisfy the resonant coupling condition, i.e., the phase term is near zero.

4.3.1. CONTRADIRECTIONAL COUPLING IN PERIODIC LAYERS.

Assuming $2\pi/\Lambda >> |\beta_1-\beta_2|$, so resonant coupling can occur between mode1,2 and mode 3,4 only, we can rewrite Eq.[4.3.2] for resonant couplings assuming that the resonant coupling occurs at p = m, as [4.3.1.1]

$$\begin{bmatrix} a_1'e^{-i\beta_1 z} \\ a_2'e^{-i\beta_2 z} \\ a_3'e^{i\beta_1 z} \\ a_4'e^{i\beta_2 z}\end{bmatrix} = -i\begin{bmatrix} e^{-im\frac{2\pi}{\Lambda}} & 0 \\ 0 & e^{-im\frac{2\pi}{\Lambda}}\end{bmatrix}\cdot\{\begin{bmatrix} \varepsilon_m & 0 \\ 0 & \varepsilon_{-m}\end{bmatrix}\cdot\begin{bmatrix} 0 & N \\ -N & -0\end{bmatrix} -\begin{bmatrix} \xi_{cm} & 0 \\ 0 & \xi_{c-m}\end{bmatrix}\cdot(\begin{bmatrix} 0 & Q \\ -Q & 0\end{bmatrix}-\begin{bmatrix} m\frac{2\pi}{\Lambda} & 0 \\ 0 & -m\frac{2\pi}{\Lambda}\end{bmatrix}\cdot\begin{bmatrix} 0 & T \\ T & 0\end{bmatrix})\} \cdot\begin{bmatrix} a_1e^{-i\beta_1 z} & a_2e^{-i\beta_2 z} & a_3e^{i\beta_1 z} & a_4e^{i\beta_2 z}\end{bmatrix}^T$$

Assuming the lossless medium, i.e., $\varepsilon_m=\varepsilon_{-m}^*$, $\xi_{cm}=\xi_{c-m}^*$ and combining the second line gives

$$\begin{bmatrix} a_1'e^{-i\beta_1 z} \\ a_2'e^{-i\beta_2 z} \\ a_3'e^{i\beta_1 z} \\ a_4'e^{i\beta_2 z}\end{bmatrix} = -i\begin{bmatrix} e^{-im\frac{2\pi}{\Lambda}} & 0 \\ 0 & e^{-im\frac{2\pi}{\Lambda}}\end{bmatrix}\cdot\{\begin{bmatrix} \varepsilon_m & 0 \\ 0 & \varepsilon_m^*\end{bmatrix}\cdot\begin{bmatrix} 0 & N \\ -N & 0\end{bmatrix}-\begin{bmatrix} \xi_{cm} & 0 \\ 0 & \xi_{cm}^*\end{bmatrix}\cdot\begin{bmatrix} 0 & R \\ -R & 0\end{bmatrix} \cdot\begin{bmatrix} a_1e^{-i\beta_1 z} & a_2e^{-i\beta_2 z} & a_3e^{i\beta_1 z} & a_4e^{i\beta_2 z}\end{bmatrix}^T$$

where [4.3.1.2]

$$\begin{bmatrix} 0 & R \\ -R & 0\end{bmatrix}=\begin{bmatrix} 0 & Q \\ -Q & 0\end{bmatrix}-\begin{bmatrix} m\frac{2\pi}{\Lambda} & 0 \\ 0 & -m\frac{2\pi}{\Lambda}\end{bmatrix}\cdot\begin{bmatrix} 0 & T \\ T & 0\end{bmatrix}$$

Let the trial solution as

$$a_1\equiv A_1(z)e^{i\frac{\Delta\beta_1}{2}z}, a_2\equiv A_2(z)e^{i\frac{\Delta\beta_2}{2}z}, a_3\equiv A_3(z)e^{-i\frac{\Delta\beta_1}{2}z}, a_4\equiv A_4(z)e^{-i\frac{\Delta\beta_2}{2}z} \quad [4.3.1.3]$$

where $\Delta\beta_1\equiv2\beta_1-m\frac{2\pi}{\Lambda}$, $\Delta\beta_2\equiv2\beta_2-m\frac{2\pi}{\Lambda}$

Then,

$$\begin{bmatrix} A_1' \\ A_2' \\ A_3' \\ A_4'\end{bmatrix}=\begin{bmatrix} \Delta & W \\ W^* & \Delta^*\end{bmatrix}\cdot\begin{bmatrix} A_1 \\ A_2 \\ A_3 \\ A_4\end{bmatrix}\equiv X\cdot\begin{bmatrix} A_1 \\ A_2 \\ A_3 \\ A_4\end{bmatrix} \quad [4.3.1.4]$$

where

$$W=-i\{\varepsilon_m N-\xi_{cm}R\},\quad \Delta=\begin{bmatrix} -i\frac{\Delta\beta_1}{2} & 0 \\ 0 & -i\frac{\Delta\beta_2}{2}\end{bmatrix}$$

and prime denotes derivative to z.

Hence, the problem is terminated in the eigenvalue problem of linear differential equation system. The characteristic equation, i.e., $|X-\lambda I|=0$ gives eigenvalues, $\lambda_1,\lambda_2,-\lambda_1,-\lambda_2$ and corresponding eigenvectors, V_1,V_2,V_3,V_4

Thus, the solution is given by

$$[a_1\ \ a_2\ \ a_3\ \ a_4]^T=P_{phase}\cdot V_{eigenvec}\cdot S_{sol}\cdot C_{coeffi}\cdot[a_1(0)\ \ a_2(0)\ \ 0\ \ 0]^T$$

where [4.3.1.5]

$$P_{phase}=\begin{bmatrix} e^{\Delta' z} & 0 \\ 0 & e^{\Delta z}\end{bmatrix}, V_{eigenvec}=[V_1\ \ V_2\ \ V_3\ \ V_4],$$

$$S_{sol}=\begin{bmatrix} \Sigma & 0 \\ 0 & \Sigma'\end{bmatrix}, \text{ where } \Sigma=\begin{bmatrix} e^{\lambda_1 z} & 0 \\ 0 & e^{\lambda_2 z}\end{bmatrix}, \Sigma'=\begin{bmatrix} e^{-\lambda_1 z} & 0 \\ 0 & e^{-\lambda_2 z}\end{bmatrix},$$

$$C_{coeffi.}=\{\begin{bmatrix} I & 0 \\ 0 & 0\end{bmatrix}\cdot V_{eigenvec.}+\begin{bmatrix} 0 & 0 \\ 0 & I\end{bmatrix}\cdot V_{eigenvec.}\cdot S_{sol.}|_{z=L}\}^{-1}$$

where I denotes identity matrix and the value of coefficient matrix is found from the boundary condition which is $a_1(0), a_2(0)$ at entrance and $a_3(L)=a_2(L)=0$ at exit.

4.3.2. CODIRECTIONAL COUPLING IN PERIODIC LAYERS.

Assuming $2\pi/\Lambda \approx |\beta_1-\beta_2|$, so resonant coupling can also occur between the mode 1 and the mode 2, and between the mode 3 and the mode 4,in this case, the coupling constants for between the mode 1,2 and the mode 3,4 are too small by the order, $\approx\leq 10^{-3}$, thus we can manipulate the problem with $\mathbf{M,P,S}$ in Eq.[4.3.2]and only with a_1, a_2. The procedures are similar to contradirectional coupling. Then, the coupling equation is given by

$$\begin{bmatrix} a_1' e^{-i\beta_1 z} \\ a_2' e^{-i\beta_2 z} \end{bmatrix} = -i \begin{bmatrix} e^{-im\frac{2\pi}{\Lambda}} & 0 \\ 0 & e^{-im\frac{2\pi}{\Lambda}} \end{bmatrix} \left\{ \begin{bmatrix} \varepsilon_m & 0 \\ 0 & \varepsilon_m^* \end{bmatrix} \cdot \begin{bmatrix} 0 & M_{12} \\ M_{21} & 0 \end{bmatrix} - \begin{bmatrix} \xi_{cm} & 0 \\ 0 & \xi_m^* \end{bmatrix} \cdot \begin{bmatrix} 0 & L_{12} \\ L_{21} & 0 \end{bmatrix} \right. \cdot \begin{bmatrix} a_1 e^{-i\beta_1 z} & a_2 e^{-i\beta_2 z} \end{bmatrix}^T \quad [4.3.2.1]$$

where

$$\begin{bmatrix} 0 & L_{12} \\ L_{21} & 0 \end{bmatrix} = \begin{bmatrix} 0 & P_{12} \\ P_{21} & 0 \end{bmatrix} - \begin{bmatrix} m\frac{2\pi}{\Lambda} & 0 \\ 0 & -m\frac{2\pi}{\Lambda} \end{bmatrix} \cdot \begin{bmatrix} 0 & S_{12} \\ S_{21} & 0 \end{bmatrix}$$

and the subscript of matrices' eliments indicates the component of that matrices.

Let the trial solution as

$$a_1 \equiv A_1(z) e^{i\frac{\Delta\beta}{2}z}, \quad a_2 \equiv A_2(z) e^{-i\frac{\Delta\beta}{2}z} \quad [4.3.2.2]$$

where $\Delta\beta \equiv \beta_1 - \beta_2 - m\frac{2\pi}{\Lambda}$

Then,

$$\begin{bmatrix} A_1' \\ A_2' \end{bmatrix} = \begin{bmatrix} -\frac{i\Delta\beta}{2} & W_{12} \\ W_{21}^* & \frac{i\Delta\beta}{2} \end{bmatrix} \cdot \begin{bmatrix} A_1 \\ A_2 \end{bmatrix} \equiv \mathbf{X} \cdot \begin{bmatrix} A_1 \\ A_2 \end{bmatrix} \quad [4.3.2.3]$$

where

$$\begin{bmatrix} 0 & W_{12} \\ W_{21}^* & 0 \end{bmatrix} = -i \left\{ \begin{bmatrix} 0 & \varepsilon_m M_{12} \\ \varepsilon_m^* M_{21} & 0 \end{bmatrix} - \begin{bmatrix} 0 & \xi_{cm} L_{12} \\ \xi_m^* L_{21} & 0 \end{bmatrix} \right\}$$

and prime denotes derivative to z.

Thus,

$$\begin{bmatrix} a_1 \\ a_2 \end{bmatrix} = \mathbf{P}_{phase} \cdot \mathbf{V}_{eigenvec.} \cdot \mathbf{S}_{sol.} \cdot \mathbf{C}_{coeffi.} \cdot \begin{bmatrix} a_1(0) \\ a_2(0) \end{bmatrix} \quad [4.3.2.4]$$

where

$$\mathbf{P}_{phase} = \begin{bmatrix} e^{i\frac{\Delta\beta}{2}z} & 0 \\ 0 & e^{-i\frac{\Delta\beta}{2}z} \end{bmatrix}, \quad \mathbf{S}_{sol.} = \begin{bmatrix} e^{\lambda_o z} & 0 \\ 0 & e^{-\lambda_o z} \end{bmatrix}$$

$$\mathbf{V}_{eigenvec.} = \begin{bmatrix} V_1 & V_2 \end{bmatrix}, \quad \mathbf{C}_{coeffi.} = \mathbf{V}_{eigenvec.}^{-1},$$

where $\lambda_o, -\lambda_o$ are eigenvalues of characteristic equation $|\mathbf{X} - \lambda \mathbf{I}| = 0$, and V_1, V_2 are corresponding eigenvectors.

In this format, the absolute values of W_{12}, W_{21} are slightly different because of slightly different charateristics of transmission of mode 1and mode 2. If you have a question why the intensity is not conserved properly, the answer is that in chiral media, there must be additional two modes, i.e., mode 3,4 to describe properly, Thus 4×4 analysis is needed.

5.CONCLUSIONS.

In this paper, we investigate the nonlinearity of chiral media and coupled-mode theory of chiral multilayers. We propose a possibility that SHG can occurs with the assistance of external static magnetic field and this approach will be applicable to THG and EFISHG. There is a possibility that the chiral admittance may have nonlinearity, thus it is necessary to elucidate the property theoretically and experimentally. And coupled-mode theory of chiral multilayer provide analytical results of periodic layers and in arbitrary layered structure, the solution will be given by some numerical assistance with the equation provided. The results suggest a possibility to obtain circular polarizer, chiro-Bragg reflector, mode converter, and etc.. And it is also applicable to some perturbation theories.

We expect that further research will bring out a lot of interesting and useful stories into the world from the secret novel of chiral media.

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