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Nonlinear Propagation of Electromagnetic Waves in Antiferromagnet

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1. Introduction

The nonlinearities of common optical materials result from the nonlinear response of their electric polarization to the electric field of electromagnetic waves (EMWs), or $\vec{P}^{NL} = \vec{\chi}^{(1)} \cdot \vec{E} + \vec{\chi}^{(2)} : \vec{E}\vec{E} + \vec{\chi}^{(3)} : \vec{E}\vec{E}\vec{E} + \cdots$. From the Maxwell equations and related electromagnetic boundary conditions including this nonlinear polarization, one can present the origin of most nonlinear optical phenomena.

However, the magnetically optical nonlinearities of magnetic materials come from the nonlinear response of their dynamical magnetization to the magnetic field of EWMs, or the magnetization $\vec{m}^{NL} = \vec{\chi}^{(1)} \cdot \vec{H} + \vec{\chi}^{(2)} : \vec{H}\vec{H} + \vec{\chi}^{(3)} : \vec{H}\vec{H}\vec{H} + \ldots$. From these one can predict or explain various magnetic optical nonlinear features of magnetic materials. The magnetic mediums are optical dispersive, which originates from the magnetic permeability as a function of frequency. Since various nonlinear phenomena from ferromagnets and ferrimagnets almost exist in the microwave region, these phenomena are important for the microwave technology.

In the concept of ferromagnetism(Morrish, 2001), there is such a kind of magnetic ordering media, named antiferromagnets (AFs), such as NiO, MnF_2, FeF_2, and CoF_2 *et. al*. This kind of materials may possess two or more magnetic sublattices and all lattice points on any sublattice have the same magnetic moment, but the moments on adjacent sublattices are opposite in direction and counteract to each other. We here present an example in Fig.1, a bi-sublattice AF structure. In contrast to the ferromagnets or ferrimagnets, it is very difficult to magnetize AFs by a magnetic field of ordinary intensity since very intense AF exchange interaction exists in them, so they are almost not useful in the fields of electronic and electric engineering. But the dynamical properties of AFs should be paid a greater attention to. The resonant frequencies of the AFs usually fall in millimeter or far infrared (IR) frequency regime. Therefore the experimental methods to study AFs optical properties are optical or quasi-optical ones. In addition, these frequency regions also are the working frequency regions of the THz technology, so the AFs may be available to make new elements in the field of THz technology.

The propagation of electromagnetic waves in AFs can be divided into two cases. In the first case, the frequency of an EMW is far to the AF resonant frequency and then the AF can be optically considered as an ordinary dielectric. The second case means that the wave frequency is situated in the vicinity of the AF resonant frequency and the dynamical

magnetization of the AF then couples with the magnetic field of the EMW. Consequently, modes of EMW propagation in this frequency region are some AF polaritons. In the linear case, the AF polaritons in AF films, multilayers and superlattices had been extendedly discussed before the year 2000 (Stamps & Camley, 1996; Camley & Mills, 1982; Zhu & Cao, 1987; Oliveros, et. al., 1992; Camley, 1992; Raj & Tilley, 1987; Wang & Tilley, 1987; Almeida & Tilley, 1990).

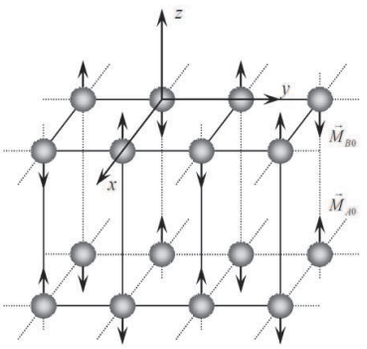


Fig. 1. The sketch of a bi-sublattice AF structure.

The magnetically nonlinear investigation of AF systems was not given great attention until the 1990s. In the recent years, many progresses have been made in understanding the magnetic dynamics of AF systems (Costa, et. al. ,1993; Balakrishnan, et. al.,1990, 1992; Daniel & Bishop,1992; Daniel & Amuda,1994; Balakrishnan & Blumenfeld,1997). Many investigations have been carried out on nonlinear guided and surface waves (Wang & Awai,1998; Almeida & Mills, 1987; Kahn, et. al., 1988; Wright & Stegeman, 1992; Boardman & Egan,1986), second-harmonic generation (Lim, 2002, 2006; Fiebig et. al, 1994, 2001, 2005), bistability (Vukovic, 1992) and dispersion properties (Wang,Q, 2000). Almeida and Mills first discussed the nonlinear infrared responses of the AFs and explore the field-dependent of transmission through thin AF films and superlattices, where the third-order approximation of dynamical magnetization was used, but no analytical expressions of nonlinear magnetic susceptibilities in the AF films or layers were obtained (Almeida & Mills, 1987; Kahn, et. al., 1988). Lim first obtained the expressions of the susceptibilities in the third-order approximation, in a special situation where a circularly polarized magnetic field and the cylindrical coordinate system were applied in the derivation process (Lim, et. al., 2000). It is obvious that those expressions cannot be conveniently used in various geometries and boundaries of different shape. In analogue to what done in the ordinary nonlinear optics, the nonlinear magnetic susceptibilities were presented in the Cartesian coordinate system by Wang et. al. (Wang & Fu, 2004; Zhou, et. al., 2009), and were used to discuss the nonlinear polaritons of AF superlattices and the second-harmonic generation (SHG) of AF films (Wang & Li, 2005; Zhou & Wang, 2008), as well as transmission and reflection bi-stability (Bai, et. al., 2007; Zhou, 2010).

2. Nonlinear susceptibilities of antiferromagnets

AF susceptibility is considered as one important physical quantity to describe the response of magnetization in AFs to the driving magnetic filed. It is also a basis of investigating dynamic properties and magneto-optical properties. In this section, the main steps and

results of deriving nonlinear magnetic susceptibilities of AFs will be presented in the right-angled coordinate system, or the Cartesian system. The detail mathmetical procedure can be found from our previous works (Wang & Fu, 2004; Zhou, et. al., 2009). The used bi-sublattice AF structure and coordinate system are shown in Fig.1, where we take the AF anisotropy axis and the external magnetic field H_0 along the z axis. The sublattic magnetzation M_0 is the absolute projection value of each total sublattice magnatization to the anisotropy axis. The driving magnetic field $\vec{H}$ changes with time, according to $\exp(-i\omega t)$.

2.1 Basical assumptions, definitions and the first-order susceptibilities

We begin with the assumption that this AF crystal is at a low temperature, or the temperature is much lower than its Neel temperature and the magnetic ordering is properly preserved. Then the magnetization on each sublattice is regarded as saturated without a driving field. In the alternating driving field $\vec{H}$, each sublattice magnetization deviates the AF anisotropy axis and makes a precession with respect to the effective field acting on it. This precession is described by the Bloch's equation with damping,

$$\frac{\partial}{\partial t}\vec{M}_{A(B)} = \gamma \vec{M}_{A(B)} \times \vec{H}^{eff}_{A(B)} - \frac{\tau \vec{M}_{A(B)}}{M_0} \times \frac{\partial}{\partial t}\vec{M}_{A(B)} \tag{2-1a}$$

where γ is the gyromagnetic ratio and τ the damping coefficient, $\vec{M}_A$ and $\vec{M}_B$ are the total sublattice magnetizations and contain two parts, the static part $\pm M_0$ and changing part with time $\vec{m}_{A(B)}$, produced by the driving field,

$$\vec{M}_A = M_0\vec{e}_z + \vec{m}_A,\ \vec{M}_B = -M_0\vec{e}_z + \vec{m}_B \tag{2-1b}$$

$\vec{H}^{eff}_A$ and $\vec{H}^{eff}_B$ are the effective fields acting on sublattices A and B, respectively, and are given by

$$\vec{H}^{eff}_A = (H_0 + \frac{H_a M^z_A}{M_0})\vec{e}_z - \frac{H_e \vec{M}_B}{M_0} + \vec{H},\ \vec{H}^{eff}_B = (H_0 + \frac{H_a M^z_B}{M_0})\vec{e}_z - \frac{H_e \vec{M}_A}{M_0} + \vec{H} \tag{2-1c}$$

where H_a is the AF anisotropy field, H_e is the AF exchange field and $\vec{H}$ indicates the driving field. Substituting (2-1b) and (2-1c) into (2-1a), we have

$$\frac{\partial \vec{M}_A}{\partial t} = \gamma\{m_{Ay}[H_0 + \frac{H_a M_{Az}}{M_0} + H_z - \frac{H_e(m_{Bz} - M_0)}{M_0}] - (M_0 + m_{Az})[H_y - \frac{H_e}{M_0}m_{By}]\}\vec{e}_x$$
$$+\gamma\{(M_0 + m_{Az})[H_x - \frac{H_e}{M_0}m_{Bx}] - m_{Ax}[H_0$$
$$+\frac{H_a M_{Az}}{M_0} + H_z - \frac{H_e(m_{Bz} - M_0)}{M_0}]\}\vec{e}_y + \gamma\{m_{Ax}[H_y(\omega) - \frac{H_e}{M_0}m_{By}]$$
$$-m_{Ay}[H_x(\omega) - \frac{H_e}{M_0}m_{Bx}]\}\vec{e}_z + \frac{\tau}{M_0}\{[m_{Ay}\frac{\partial}{\partial t}(M_0 + m_{Az})$$

$$-(M_0+m_{Az})\frac{\partial}{\partial t}m_{Ay}]\vec{e}_x+[(M_0+m_{Az})\frac{\partial}{\partial t}m_{Ax}-m_{Ax}\frac{\partial}{\partial t}(M_0+m_{Az})]\vec{e}_y$$
$$+[m_{Ax}\frac{\partial}{\partial t}m_{Ay}-m_{Ay}\frac{\partial}{\partial t}m_{Ax}]\vec{e}_z\} \tag{2-2a}$$

$$\frac{\partial}{\partial t}\vec{M}_B=\gamma\{m_{By}[H_0+\frac{H_aM_{Bz}}{M_0}+H_z-\frac{H_e(M_0+m_{Az})}{M_0}]-(m_{Bz}-M_0)[H_y-\frac{H_e}{M_0}m_{Ay}]\}\vec{e}_x$$
$$+\gamma\{(m_{Bz}-M_0)[H_x-\frac{H_e}{M_0}m_{Ax}]$$
$$-m_{Bx}[H_0+\frac{H_aM_{Bz}}{M_0}+H_z-\frac{H_e(M_0+m_{Az})}{M_0}]\}\vec{e}_y+\gamma\{m_{Bx}[H_y-\frac{H_e}{M_0}m_{Ay}]$$
$$-m_{By}[H_x-\frac{H_e}{M_0}m_{Ax}]\}\vec{e}_z+\frac{\tau}{M_0}\{[m_{By}\frac{\partial}{\partial t}(-M_0+m_{Bz})$$
$$-(-M_0+m_{Bz})\frac{\partial}{\partial t}m_{By}]\vec{e}_x+[(-M_0+m_{Bz})\frac{\partial}{\partial t}m_{Bx}-m_{Bx}\frac{\partial}{\partial t}(-M_0+m_{Bz})]\vec{e}_y$$
$$+(m_{Bx}\frac{\partial}{\partial t}m_{By}-m_{By}\frac{\partial}{\partial t}m_{Bx})\vec{e}_z\}. \tag{2-2b}$$

We shall use the perturbation expansion method to derive nonlinear magnetizations and susceptibilies of various orders. We take M_0 and H_0 as the 0-order magnetization and the 0-order field. $\vec{H}$ is considered as the first-order field and we note that the complex conjugation of this field should be included in higher-order mathmetical procesures higher than the first-order one. In the third-order aproximation, the induced magnetizations $\vec{m}_{A(B)}$ are divided into the first-, second- and third-order parts, or

$$\vec{m}_{A(B)}=\vec{m}^{(1)}_{A(B)}+\vec{m}^{(2)}_{A(B)}+\vec{m}^{(3)}_{A(B)}+c.c. \tag{2-3}$$

where *c.c.* indicates the complex conjugation. In practice, one needs the AF magnetization rather than the lattice magnetizations, so we define $\vec{m}=\vec{m}_A+\vec{m}_B$ as the AF magnetization and $\vec{n}=\vec{m}_A-\vec{m}_B$ as its supplemental quantity. In the linear case, $M_{Az}=M_0$, $M_{Bz}=-M_0$ and considering that the linear magnetizations should change with time according to $\exp(-i\omega t)$, Eqs.(2-2) can be simplified as

$$-i\omega\vec{m}^{(1)}_A=[(\omega_0+\omega_a+\omega_e+i\tau\omega)m^{(1)}_{Ay}+\omega_em^{(1)}_{By}-\omega_mH_y]\vec{e}_x$$
$$-[(\omega_0+\omega_a+\omega_e+i\tau\omega)m^{(1)}_{Ax}+\omega_em^{(1)}_{Bx}-\omega_mH_x]\vec{e}_y \tag{2-4a}$$

$$-i\omega\vec{m}^{(1)}_B=[(\omega_0-\omega_a-\omega_e-i\tau\omega)m^{(1)}_{By}-\omega_em^{(1)}_{Ay}+\omega_mH_y]\vec{e}_x$$
$$-[(\omega_0-\omega_a-\omega_e-i\tau\omega)m^{(1)}_{Bx}-\omega_em^{(1)}_{Ax}+\omega_mH_x]\vec{e}_y \tag{2-4b}$$

where the special frequencies are defined with $\omega_0=\gamma H_0$, $\omega_a=\gamma H_a$, $\omega_e=\gamma H_e$ and $\omega_m=4\pi\gamma M_0$. The first-order z-components of the sublattice magnetizations are

vanishing. From the definitions $m_i^{(1)} = \sum_j \chi_{ij}^{(1)} H_j$ and $n_i^{(1)} = \sum_j N_{ij}^{(1)} H_j$, we have the nonzero elements of the first-order magnetic susceptibilty and supplementary susceptibility

$$\chi_{xx}^{(1)} = \chi_{yy}^{(1)} = \chi_1 = 2A\omega_m \omega_a' Z_{--}(\omega)\,,\ \chi_{xy}^{(1)} = -\chi_{yx}^{(1)} = i\chi_2 = -4iA\omega_m \omega_a' \omega_0 \omega \tag{2-5a}$$

$$N_{xx}^{(1)} = N_{yy}^{(1)} = -2A\omega_0 \omega_m Z_{-+}(\omega)\,,\ N_{xy}^{(1)} = -N_{yx}^{(1)} = 2iA\omega\omega_m Z_{+-}(\omega) \tag{2-5b}$$

where $\omega_r'^2 = \omega_a'(2\omega_e + \omega_a')$ and $Z_{\pm\pm}(\omega) = \omega_r'^2 \pm \omega_0^2 \pm \omega^2$ with $\omega_a' = \omega_a + i\tau\omega$, and $A = \{[\omega_r'^2 - (\omega - \omega_0)^2][\omega_r'^2 - (\omega + \omega_0)^2]\}^{-1}$. The linear magnetic permeability often used in the past is $\vec{\mu} = \mu_0[1 + \vec{\chi}^{(1)}]$, or $\mu_{xx} = \mu_{yy} = \mu_0(1 + \chi_1) = \mu_0\mu_1$ and $\mu_{xy} = -\mu_{yx} = i\mu_0\chi_2 = i\mu_0\mu_2$.

2.2 The second-order approximation

Similar to the second-order electric polarization in the nonlinear optics, the second-order magneizations also are divided into the dc part unchanging with time and the second-harmonic part varying with time according to $\exp(-2i\omega t)$. Here we first derive the dc susceptibility, which will appear in the third-order ones. Neglecting the linear, third-order terms and the second-harmonic terms in (2-2), reserving only the second-order 0-frequency terms, we obtain the following equations

$$0 = \omega_0 m_y^{(2)}(0) + \omega_a n_y^{(2)}(0) + \gamma H_z m_y^{(1)*} + \gamma m_y^{(1)} H_z^{\ *} \tag{2-6a}$$

$$0 = \omega_0 n_y^{(2)}(0) + (\omega_a + 2\omega_e) m_y^{(2)}(0) + \gamma H_z n_y^{(1)*} + \gamma n_y^{(1)} H_z^* \tag{2-6b}$$

$$0 = -\omega_0 m_x^{(2)}(0) - \omega_a n_x^{(2)}(0) - \gamma H_z^* m_x^{(1)} - \gamma H_z m_x^{(1)*} \tag{2-6c}$$

$$0 = -\omega_0 n_x^{(2)}(0) - (\omega_a + 2\omega_e) m_x^{(2)}(0) - \gamma H_z n_x^{(1)*} - \gamma H_z^* n_x^{(1)} \tag{2-6d}$$

In addition, the z component of the dc magnetization can be obtained from the conservation of each sulattice magnetic moment, and we see

$$m_{Az}^{(2)}(0) = -\frac{1}{M_0}[m_{Ax}^{(1)}(\omega) m_{Ax}^{(1)*}(\omega) + m_{Ay}^{(1)}(\omega) m_{Ay}^{(1)*}(\omega)] \tag{2-7a}$$

$$m_{Bz}^{(2)}(0) = \frac{1}{M_0}[m_{Bx}^{(1)}(\omega) m_{Bx}^{(1)*}(\omega) + m_{By}^{(1)}(\omega) m_{By}^{(1)*}(\omega)] \tag{2-7b}$$

These lead directly to the z component to be

$$m_z^{(2)}(0) = -\frac{1}{2M_0}(m_x^{(1)} n_x^{(1)*} + n_x^{(1)} m_x^{(1)*} + m_y^{(1)} n_y^{(1)*} + n_y^{(1)} m_y^{(1)*}) \tag{2-8}$$

Here we have used $m_i^{(2)}(0)$ and $n_i^{(2)}(0)$ directly to represent $m_i^{(2)}(0) + m_i^{(2)*}(0)$ and $n_i^{(2)}(0) + n_i^{(2)*}(0)$ for simplicity. Substituting the linear results into (2-6) and (2-8), and using

the definitions of $n_i^{(2)}(0)=\sum_{jk}N_{ijk}^{(2)}(0)H_jH_k^*$ and $m_i^{(2)}(0)=\sum_{jk}\chi_{ijk}^{(2)}(0)H_j(\omega)H_k^*(\omega)$, we find the corresponding nonzero elements

$$\chi_{xxz}^{(2)}(0)=\chi_{yyz}^{(2)}(0)=\chi_{yzy}^{(2)}(0)^*=\chi_{xzx}^{(2)}(0)^*=\omega_m(\omega_0\chi_{xx}^{(1)}-\omega_aN_{xx}^{(1)})/[M_0(\omega_r^2-\omega_0^2)] \quad (2\text{-}9a)$$

$$\chi_{xyz}^{(2)}(0)=\chi_{xzy}^{(2)}(0)^*=-\chi_{yxz}^{(2)}(0)=-\chi_{yzx}^{(2)}(0)^*=\omega_m(\omega_0\chi_{xy}^{(1)}-\omega_aN_{xy}^{(1)})/[M_0(\omega_r^2-\omega_0^2)] \quad (2\text{-}9b)$$

$$\chi_{zxx}^{(2)}(0)=\chi_{zyy}^{(2)}(0)=-(\chi_{xx}^{(1)}N_{xx}^{(1)*}+\chi_{xy}^{(1)}N_{xy}^{(1)*}+c.c)/4M_0 \quad (2\text{-}9c)$$

$$\chi_{zyx}^{(2)}(0)=-\chi_{zxy}^{(2)}(0)=(N_{xx}^{(1)}\chi_{xy}^{(1)*}+\chi_{xx}^{(1)}N_{xy}^{(1)*}-c.c.)/4M_0 \quad (2\text{-}9d)$$

$$\begin{aligned}&N_{xxz}^{(2)}(0)=N_{yyz}^{(2)}(0)=N_{xzx}^{(2)}(0)^*=N_{yzy}^{(2)}(0)^*\\&=\omega_m[\omega_0N_{xx}^{(1)}-(\omega_a+2\omega_e)\chi_{xx}^{(1)}]/[M_0(\omega_r^2-\omega_0^2)]\end{aligned} \quad (2\text{-}9e)$$

$$\begin{aligned}&N_{xyz}^{(2)}(0)=N_{xzy}^{(2)}(0)^*=-N_{yxz}^{(2)}(0)=-N_{yzx}^{(2)}(0)^*\\&=\omega_m[\omega_0N_{xy}^{(1)}-(\omega_a+2\omega_e)\chi_{xy}^{(1)}]/[M_0(\omega_r^2-\omega_0^2)]\end{aligned} \quad (2\text{-}9f)$$

$$N_{zxx}^{(2)}(0)=N_{zyy}^{(2)}(0)=-(N_{xy}^{(1)}N_{xy}^{(1)*}+\chi_{xx}^{(1)}\chi_{xx}^{(1)*}+\chi_{xy}^{(1)}\chi_{xy}^{(1)*}+N_{xx}^{(1)}N_{xx}^{(1)*})/4M_0 \quad (2\text{-}9g)$$

$$N_{zyx}^{(2)}(0)=-N_{zxy}^{(2)}(0)=-(\chi_{xy}^{(1)}\chi_{xx}^{(1)*}+N_{xy}^{(1)}N_{xx}^{(1)*}-c.c.)/4M_0 \quad (2\text{-}9h)$$

Next, we are going to derive the second-harmonic (SH) magnetization and susceptibility. They will not be used only in the third-order susceptibility, but also be applied to describe the SH generation in various AF systems. In equations (2-2), reserving only the SH terms, we obtain the following equations

$$-2i\omega m_x^{(2)}(2\omega)=\omega_0m_y^{(2)}(2\omega)+\omega_a''n_y^{(2)}(2\omega)+\omega_mh_zm_y^{(1)}/M_0 \quad (2\text{-}10a)$$

$$-2i\omega n_x^{(2)}(2\omega)=\omega_0n_y^{(2)}(2\omega)+(\omega_a''+2\omega_e)m_y^{(2)}(2\omega)+\omega_mh_zn_y^{(1)}/M_0 \quad (2\text{-}10b)$$

$$-2i\omega m_y^{(2)}(2\omega)=-\omega_0m_x^{(2)}(2\omega)-\omega_a''n_x^{(2)}(2\omega)-\omega_mh_zm_x^{(1)}/M_0 \quad (2\text{-}10c)$$

$$-2i\omega n_y^{(2)}(2\omega)=-\omega_0n_x^{(2)}(2\omega)-(\omega_a''+2\omega_e)m_x^{(2)}(2\omega)-\omega_mh_zn_x^{(1)}/M_0 \quad (2\text{-}10d)$$

with $\omega_a''=\omega_a+2i\omega\tau$. Meanwhile the conservation of each sulattice magnetic moment results in

$$m_z^{(2)}(2\omega)=-\frac{1}{2M_0}[m_x^{(1)}n_x^{(1)}+m_y^{(1)}n_y^{(1)}] \quad (2\text{-}10e)$$

Applying the expressions of the first-order components and the expressions

$$m_i^{(2)}(2\omega) = \sum_{jk} \chi_{ijk}^{(2)}(2\omega) H_j H_k \tag{2-11a}$$

$$n_i^{(2)}(2\omega) = \sum_{jk} N_{ijk}^{(2)}(2\omega) H_j H_k \tag{2-11b}$$

one finds

$$\begin{aligned}\chi_{xxz}^{(2)}(2\omega) = \chi_{xzx}^{(2)}(2\omega) = \chi_{yyz}^{(2)}(2\omega) = \chi_{yzy}^{(2)}(2\omega) = AB\omega_m^2\omega_0\{\omega_a' Z_{--}(\omega) \\ Z_{-+}(2\omega) + \omega_a'' Z_{-+}(\omega) Z_{--}(2\omega) + 4\omega^2[\omega_a' Z_{+-}(2\omega) + \omega_a'' Z_{+-}(\omega)]\} / M_0\end{aligned} \tag{2-12a}$$

$$\begin{aligned}\chi_{xyz}^{(2)}(2\omega) = \chi_{xzy}^{(2)}(2\omega) = -\chi_{yxz}^{(2)}(2\omega) = -\chi_{yzx}^{(2)}(2\omega) = -iAB\omega\omega_m^2\{2\omega_0^2[\omega_a' \\ Z_{-+}(2\omega) + 2\omega_a'' Z_{-+}(\omega)] + \omega_a'' Z_{+-}(\omega) Z_{--}(2\omega) + 2\omega_a' Z_{--}(\omega) Z_{+-}(2\omega)\} / M_0\end{aligned} \tag{2-12b}$$

$$\chi_{zxx}^{(2)}(2\omega) = \chi_{zyy}^{(2)}(2\omega) = 2A\omega_m^2\omega_a'\omega_0 / M_0 \tag{2-12c}$$

$$\begin{aligned}N_{xxz}^{(2)}(2\omega) = N_{xzx}^{(2)}(2\omega) = N_{yyz}^{(2)}(2\omega) = N_{yzy}^{(2)}(2\omega) = -AB\omega_m^2\{\omega_a'(\omega_a'' + 2\omega_e) \\ [Z_{--}(2\omega) Z_{--}(\omega) + 8\omega_0^2\omega^2] + \omega_0^2 Z_{-+}(2\omega) Z_{-+}(\omega) + 2\omega^2 Z_{+-}(2\omega) Z_{+-}(\omega)\} / M_0\end{aligned} \tag{2-12d}$$

$$\begin{aligned}N_{xyz}^{(2)}(2\omega) = N_{xzy}^{(2)}(2\omega) = -N_{yxz}^{(2)}(2\omega) = -N_{yzx}^{(2)}(2\omega) = iAB\omega\omega_0\omega_m^2\{2\omega_a'(\omega_a'' + 2\omega_e) \\ [Z_{--}(2\omega) + 2Z_{--}(\omega)] + 2Z_{+-}(2\omega) Z_{-+}(\omega) + Z_{-+}(2\omega) Z_{+-}(\omega)\} / M_0\end{aligned} \tag{2-12e}$$

$$N_{zxx}^{(2)}(2\omega) = N_{zyy}^{(2)}(2\omega) = \frac{A^2\omega_m^2}{M_0}[\omega^2 Z_{+-}(\omega)^2 - \omega_a'^2 Z_{--}(\omega)^2 - \omega_0^2 Z_{-+}(\omega)^2 + 4\omega_a'^2\omega^2\omega_0^2] \tag{2-12f}$$

where $B = 1/[\omega_r''^2 - (2\omega + \omega_0)^2][\omega_r''^2 - (2\omega - \omega_0)^2]$.

2.3 The third-order approximation

The third-order magnetization also contains two part, or one varies with time according to $\exp(-i\omega t)$ and the orther is the third-harmonic part with $\exp(-3i\omega t)$. Because we do not consern with the third-harmonic (TH) generation, so the first-order, and second-order and TH terms in equations (2-2) all are ignored. Thus we have

$$-i\omega m_x^{(3)}(\omega) = \omega_0 m_y^{(3)}(\omega) + \omega_a' n_y^{(3)}(\omega) + \eta_x \tag{2-13a}$$

$$-i\omega n_x^{(3)}(\omega) = \omega_0 n_y^{(3)}(\omega) + (\omega_a' + 2\omega_e) m_y^{(3)}(\omega) + \eta_x' \tag{2-13b}$$

$$-i\omega m_y^{(3)}(\omega) = -\omega_0 m_x^{(3)}(\omega) - \omega_a' n_x^{(3)}(\omega) + \eta_y \tag{2-13c}$$

$$-i\omega n_y^{(3)}(\omega) = -\omega_0 n_x^{(3)}(\omega) - (\omega_a' + 2\omega_e) m_x^{(3)}(\omega) + \eta_y' \tag{2-13d}$$

$$
\begin{aligned}
&-i\omega m_z^{(3)}(\omega)=\frac{\omega_m}{M_0}[m_x^{(2)}(0)H_y+m_x^{(2)}(2\omega)H_y^*-m_y^{(2)}(0)H_x\\
&-m_y^{(2)}(2\omega)H_x^*]+\frac{\tau}{M_0}[-2i\omega m_{Ax}^{(1)*}(\omega)\\
&m_{Ay}^{(2)}(2\omega)-i\omega m_{Ax}^{(2)}(0)m_{Ay}^{(1)}(\omega)+i\omega m_{Ax}^{(2)}(2\omega)m_{Ay}^{(1)*}(\omega)\\
&+2i\omega m_{Ay}^{(1)*}(\omega)m_{Ax}^{(2)}(2\omega)+i\omega m_{Ay}^{(2)}(0)m_{Ax}^{(1)}(\omega)\\
&-i\omega m_{Ay}^{(2)}(2\omega)m_{Ax}^{(1)*}(\omega)]-2i\omega m_{Bx}^{(1)*}(\omega)m_{By}^{(2)}(2\omega)\\
&-i\omega m_{Bx}^{(2)}(0)m_{By}^{(1)}(\omega)+i\omega m_{Bx}^{(2)}(2\omega)m_{By}^{(1)*}(\omega)\\
&+2i\omega m_{By}^{(1)*}(\omega)m_{Bx}^{(2)}(2\omega)+i\omega m_{By}^{(2)}(0)m_{Bx}^{(1)}(\omega)\\
&-i\omega m_{By}^{(2)}(2\omega)m_{Bx}^{(1)*}(\omega)]
\end{aligned}
\tag{2-13e}
$$

where

$$
\begin{aligned}
&\eta_x=\eta_{Ax}+\eta_{Bx}=\frac{\omega_m}{M_0}[m_y^{(2)}(0)H_z+m_y^{(2)}(2\omega)H_z^*-m_z^{(2)}(0)H_y\\
&-m_z^{(2)}(2\omega)H_y^*]+\frac{\omega_a}{M_0}[m_{Ay}^{(1)}(\omega)m_{Az}^{(2)}(0)\\
&+m_{Ay}^{(1)*}(\omega)m_{Az}^{(2)}(2\omega)+m_{By}^{(1)}(\omega)m_{Bz}^{(2)}(0)+m_{By}^{(1)*}(\omega)m_{Bz}^{(2)}(2\omega)]\\
&+\frac{\tau}{M_0}[-2i\omega m_{Ay}^{(1)*}(\omega)m_{Az}^{(2)}(2\omega)+i\omega m_{Az}^{(2)}(0)\\
&m_{Ay}^{(1)}(\omega)-i\omega m_{Az}^{(2)}(2\omega)m_{Ay}^{(1)*}(\omega)-2i\omega m_{By}^{(1)*}(\omega)m_{Bz}^{(2)}(2\omega)\\
&+i\omega m_{Bz}^{(2)}(0)m_{By}^{(1)}(\omega)-i\omega m_{Bz}^{(2)}(2\omega)m_{By}^{(1)*}(\omega)]
\end{aligned}
\tag{2-14a}
$$

$$
\begin{aligned}
&\eta_y=\eta_{Ay}+\eta_{By}=\frac{\omega_m}{M_0}[m_z^{(2)}(0)H_x+m_z^{(2)}(2\omega)H_x^*-m_x^{(2)}(0)H_z\\
&-m_x^{(2)}(2\omega)H_z^*]-\frac{\omega_a}{M_0}[m_{Az}^{(2)}(0)m_{Ax}^{(1)}(\omega)\\
&+m_{Az}^{(2)}(2\omega)m_{Ax}^{(1)*}(\omega)+m_{Bz}^{(2)}(0)m_{Bx}^{(1)}(\omega)+m_{Bz}^{(2)}(2\omega)m_{Bx}^{(1)*}(\omega)]\\
&+\frac{\tau}{M_0}[-i\omega m_{Az}^{(2)}(0)m_{Ax}^{(1)}(\omega)+i\omega m_{Az}^{(2)}(2\omega)\\
&m_{Ax}^{(1)*}(\omega)+2i\omega m_{Ax}^{(1)*}(\omega)m_{Az}^{(2)}(\omega)-i\omega m_{Bz}^{(2)}(0)m_{Bx}^{(1)}(\omega)\\
&+i\omega m_{Bz}^{(2)}(2\omega)m_{Bx}^{(1)*}(\omega)+2i\omega m_{Bx}^{(1)}(\omega)m_{Bz}^{(2)}(2\omega)]
\end{aligned}
\tag{2-14b}
$$

$$
\begin{aligned}
&\eta_x'=\eta_{Ax}-\eta_{Bx}=\frac{\omega_m}{M_0}[n_y^{(2)}(0)H_z+n_y^{(2)}(2\omega)H_z^*-n_z^{(2)}(0)H_y\\
&-n_z^{(2)}(2\omega)H_y^*]+\frac{\omega_a}{M_0}[m_{Ay}^{(1)}(\omega)m_{Az}^{(2)}(0)
\end{aligned}
$$

$$
\begin{aligned}
&+m_{Ay}^{(1)*}(\omega)m_{Az}^{(2)}(2\omega)-m_{By}^{(1)}(\omega)m_{Bz}^{(2)}(0)-m_{By}^{(1)*}(\omega)m_{Bz}^{(2)}(2\omega)]\\
&+\frac{2\omega_e}{M_0}[m_{By}^{(1)}(\omega)m_{Az}^{(2)}(0)+m_{By}^{(1)*}(\omega)m_{Az}^{(2)}(2\omega)\\
&-m_{Ay}^{(1)}(\omega)m_{Bz}^{(2)}(0)-m_{Ay}^{(1)*}(\omega)m_{Bz}^{(2)}(2\omega)]\\
&+\frac{\tau}{M_0}[-2i\omega m_{Ay}^{(1)*}(\omega)m_{Az}^{(2)}(2\omega)+i\omega m_{Az}^{(2)}(0)m_{Ay}^{(1)}(\omega)\\
&-i\omega m_{Az}^{(2)}(2\omega)m_{Ay}^{(1)*}(\omega)+2i\omega m_{By}^{(1)*}(\omega)m_{Bz}^{(2)}(2\omega)\\
&-i\omega m_{Bz}^{(2)}(0)m_{By}^{(1)}(\omega)+i\omega m_{Bz}^{(2)}(2\omega)m_{By}^{(1)*}(\omega)]
\end{aligned}
\tag{2-14c}
$$

$$
\begin{aligned}
&\eta_y'=\eta_{Ay}-\eta_{By}=\frac{\omega_m}{M_0}[n_z^{(2)}(0)H_x+n_z^{(2)}(2\omega)H_x^*-n_x^{(2)}(0)H_z\\
&-n_x^{(2)}(2\omega)H_z^*]-\frac{\omega_a}{M_0}[m_{Az}^{(2)}(0)m_{Ax}^{(1)}(\omega)+\\
&m_{Az}^{(2)}(2\omega)m_{Ax}^{(1)*}(\omega)-m_{Bz}^{(2)}(0)m_{Bx}^{(1)}(\omega)-m_{Bz}^{(2)}(2\omega)m_{Bx}^{(1)*}(\omega)]\\
&+\frac{2\omega_e}{M_0}[m_{Bz}^{(2)}(0)m_{Ax}^{(1)}(\omega)+m_{Bz}^{(2)}(2\omega)m_{Ax}^{(1)*}(\omega)\\
&-m_{Az}^{(2)}(0)m_{Bx}^{(1)}(\omega)-m_{Az}^{(2)}(2\omega)m_{Bx}^{(1)*}(\omega)]+\frac{\tau}{M_0}\\
&[-i\omega m_{Az}^{(2)}(0)m_{Ax}^{(1)}(\omega)+i\omega m_{Az}^{(2)}(2\omega)m_{Ax}^{(1)*}(\omega)\\
&+2i\omega m_{Ax}^{(1)*}(\omega)m_{Az}^{(2)}(\omega)+i\omega m_{Bz}^{(2)}(0)m_{Bx}^{(1)}(\omega)\\
&-i\omega m_{Bz}^{(2)}(2\omega)m_{Bx}^{(1)*}(\omega)-2i\omega m_{Bx}^{(1)}(\omega)m_{Bz}^{(2)}(2\omega)]
\end{aligned}
\tag{2-14d}
$$

Substituting the definitions of $m_i^{(1)}$, $m_i^{(2)}$, $n_i^{(1)}$ and $n_i^{(2)}$ into equations (2-13,2-14), and after some complicated algebra, we finally obtain

$$\chi_{xxxx}^{(3)}(\omega)=A[\omega_0 Z_{-+}(\omega)f-\omega_a' Z_{--}(\omega)f'-i\omega Z_{+-}(\omega)a+2i\omega\omega_0\omega_a' a'] \tag{2-15a}$$

$$\chi_{xyyx}^{(3)}(\omega)=A[\omega_0 Z_{-+}(\omega)e-\omega_a' Z_{--}(\omega)e'-i\omega Z_{+-}(\omega)b+2i\omega\omega_0\omega_a' b'] \tag{2-15b}$$

$$\chi_{xzzx}^{(3)}(\omega)=A[\omega_0 Z_{-+}(\omega)g-\omega_a' Z_{--}(\omega)g'-i\omega Z_{+-}(\omega)c+2i\omega\omega_0\omega_a' c'] \tag{2-15c}$$

$$\chi_{xxyx}^{(3)}(\omega)=\frac{A}{2}[-\omega_0 Z_{-+}(\omega)h+\omega_a' Z_{--}(\omega)h'-i\omega Z_{+-}(\omega)d+2i\omega\omega_0\omega_a' d'] \tag{2-15d}$$

$$\chi_{xxxy}^{(3)}(\omega)=A[-\omega_0 Z_{-+}(\omega)b+\omega_a' Z_{--}(\omega)b'-i\omega Z_{+-}(\omega)e+2i\omega\omega_0\omega_a' e'] \tag{2-15e}$$

$$\chi_{xyyy}^{(3)}(\omega)=A[-\omega_0 Z_{-+}(\omega)a+\omega_a' Z_{--}(\omega)a'-i\omega Z_{+-}(\omega)f+2i\omega\omega_0\omega_a' f'] \tag{2-15f}$$

$$\chi_{xzzy}^{(3)}(\omega)=A[-\omega_0 Z_{-+}(\omega)c+\omega_a' Z_{--}(\omega)c'-i\omega Z_{+-}(\omega)g+2i\omega\omega_0\omega_a' g'] \tag{2-15g}$$

$$\chi^{(3)}_{xxyy}(\omega)=\frac{A}{2}[\omega_0 Z_{-+}(\omega)d-\omega'_a Z_{--}(\omega)d'-i\omega Z_{+-}(\omega)h+2i\omega\omega_0\omega'_a h'] \tag{2-15h}$$

$$\chi^{(3)}_{xxzz}(\omega)=\frac{A}{2}[\omega_0 Z_{-+}(\omega)p-\omega'_a Z_{--}(\omega)p'-i\omega Z_{+-}(\omega)l+2i\omega\omega_0\omega'_a l'] \tag{2-15i}$$

$$\chi^{(3)}_{xyzz}(\omega)=\frac{A}{2}[-\omega_0 Z_{-+}(\omega)l+\omega'_a Z_{--}(\omega)l'-i\omega Z_{+-}(\omega)p+2i\omega\omega_0\omega'_a p'] \tag{2-15j}$$

$$\begin{aligned}\chi^{(3)}_{zxzx}(\omega)=&\frac{\tau}{4M_0}[6\chi^{(1)*}_{xy}\chi^{(2)}_{xxz}(2\omega)-6\chi^{(1)*}_{xx}\chi^{(2)}_{xyz}(2\omega)\\&+3N^{(1)*}_{xy}N^{(2)}_{xxz}(2\omega)-3N^{(1)*}_{xx}N^{(2)}_{xyz}(2\omega)+\\&\chi^{(1)}_{xx}\chi^{(2)}_{xzy}(0)-\chi^{(1)}_{xy}\chi^{(2)}_{xzx}(0)+N^{(1)}_{xx}N^{(2)}_{xzy}(0)-N^{(1)}_{xy}N^{(2)}_{xzx}(0)]\\&+\frac{i\omega_m}{2\omega M_0}[\chi^{(2)}_{xzy}(0)+2\chi^{(2)}_{xyz}(2\omega)]\end{aligned} \tag{2-15k}$$

$$\begin{aligned}\chi^{(3)}_{zyzx}(\omega)=&\frac{\tau}{4M_0}[6\chi^{(1)*}_{xy}\chi^{(2)}_{xyz}(2\omega)+6\chi^{(1)*}_{xx}\chi^{(2)}_{xxz}(2\omega)\\&+3N^{(1)*}_{xy}N^{(2)}_{xyz}(2\omega)+3N^{(1)*}_{xx}N^{(2)}_{xxz}(2\omega)\\&+\chi^{(1)}_{xx}\chi^{(2)}_{xzx}(0)+\chi^{(1)}_{xy}\chi^{(2)}_{xzy}(0)+N^{(1)}_{xx}N^{(2)}_{xzx}(0)\\&+N^{(1)}_{xy}N^{(2)}_{xzy}(0)]+\frac{i\omega_m}{2\omega M_0}[\chi^{(2)}_{xzx}(0)-2\chi^{(2)}_{xxz}(2\omega)]\end{aligned} \tag{2-15l}$$

$$\begin{aligned}\chi^{(3)}_{zxxz}(\omega)=&\frac{\tau}{2M_0}[\chi^{(1)}_{xx}\chi^{(2)}_{xyz}(0)+N^{(1)}_{xx}N^{(2)}_{xyz}(0)-N^{(1)}_{xy}N^{(2)}_{xxz}(0)\\&-\chi^{(1)}_{xy}\chi^{(2)}_{xxz}(0)]+\frac{i\omega_m}{\omega M_0}\chi^{(2)}_{xyz}(0)\end{aligned} \tag{2-15m}$$

$$\begin{aligned}\chi^{(3)}_{zyyz}(\omega)=&\frac{\tau}{2M_0}[\chi^{(1)}_{xx}\chi^{(2)}_{xyz}(0)+N^{(1)}_{xx}N^{(2)}_{xyz}(0)-N^{(1)}_{xy}N^{(2)}_{xxz}(0)\\&-\chi^{(1)}_{xy}\chi^{(2)}_{xxz}(0)]+\frac{i\omega_m}{\omega M_0}\chi^{(2)}_{xyz}(0)\end{aligned} \tag{2-15n}$$

$$\begin{aligned}\chi^{(3)}_{zxzy}(\omega)=&-\frac{\tau}{4M_0}[6\chi^{(1)*}_{xx}\chi^{(2)}_{xxz}(2\omega)+6\chi^{(1)*}_{xy}\chi^{(2)}_{xyz}(2\omega)\\&+3N^{(1)*}_{xx}N^{(2)}_{xxz}(2\omega)+3N^{(1)*}_{xy}N^{(2)}_{xyz}(2\omega)+\\&\chi^{(1)}_{xx}\chi^{(2)}_{xzx}(0)+\chi^{(1)}_{xy}\chi^{(2)}_{xzy}(0)+N^{(1)}_{xx}N^{(2)}_{xzx}(0)+N^{(1)}_{xy}N^{(2)}_{xzy}(0)]\\&+\frac{i\omega_m}{2\omega M_0}[2\chi^{(2)}_{xxz}(2\omega)-\chi^{(2)}_{xzx}(0)]\end{aligned} \tag{2-15o}$$

$$\chi^{(3)}_{zyzy}(\omega)=\frac{\tau}{4M_0}[6\chi^{(1)*}_{xy}\chi^{(2)}_{xxz}(2\omega)-6\chi^{(1)*}_{xx}\chi^{(2)}_{xyz}(2\omega)$$
$$+3N^{(1)*}_{xy}N^{(2)}_{xxz}(2\omega)-3N^{(1)*}_{xx}N^{(2)}_{xyz}(2\omega)+\chi^{(1)}_{xx}\chi^{(2)}_{xzy}(0)$$
$$-\chi^{(1)}_{xy}\chi^{(2)}_{xzx}(0)+N^{(1)}_{xx}N^{(2)}_{xzy}(0)-N^{(1)}_{xy}N^{(2)}_{xzx}(0)] \quad (2\text{-}15p)$$
$$+\frac{i\omega_m}{2\omega M_0}[\chi^{(2)}_{xzy}(0)+2\chi^{(2)}_{xyz}(2\omega)]$$

with the coefficients

$$a=\frac{i\omega\tau}{2M_0}[3\chi^{(2)}_{zxx}(2\omega)\chi^{(1)*}_{xy}+3N^{(2)}_{zxx}(2\omega)N^{(1)*}_{xy}$$
$$-\chi^{(1)}_{xy}\chi^{(2)}_{zxx}(0)-N^{(1)}_{xy}N^{(2)}_{zxx}(0)] \quad (2\text{-}16a)$$
$$-\frac{\omega_a}{2M_0}[\chi^{(1)}_{xy}\chi^{(2)}_{zxx}(0)+N^{(1)}_{xy}N^{(2)}_{zxx}(0)$$
$$+N^{(2)}_{zxx}(2\omega)N^{(1)*}_{xy}+\chi^{(2)}_{zxx}(2\omega)\chi^{(1)*}_{xy}]$$

$$b=\frac{i\omega\tau}{2M_0}[3\chi^{(2)}_{zxx}(2\omega)\chi^{(1)*}_{xy}+3N^{(2)}_{zxx}(2\omega)N^{(1)*}_{xy}$$
$$+\chi^{(1)}_{xx}\chi^{(2)}_{zyx}(0)+N^{(1)}_{xx}N^{(2)}_{zyx}(0)] \quad (2\text{-}16b)$$
$$+\frac{1}{2M_0}\{\omega_a[\chi^{(1)}_{xx}\chi^{(2)}_{zyx}(0)+N^{(1)}_{xx}N^{(2)}_{zyx}(0)-\chi^{(2)}_{zxx}(2\omega)\chi^{(1)*}_{xy}$$
$$-N^{(2)}_{zxx}(2\omega)N^{(1)*}_{xy}]-2\omega_m\chi^{(2)}_{zyx}(0)\}$$

$$c=-\frac{\omega_m}{M_0}\chi^{(2)}_{xzy}(0) \quad (2\text{-}16c)$$

$$d=\frac{i\omega\tau}{2M_0}[\chi^{(1)}_{xx}\chi^{(2)}_{zxx}(0)+N^{(1)}_{xx}N^{(2)}_{zxx}(0)$$
$$-\chi^{(1)}_{xy}\chi^{(2)}_{zyx}(0)-N^{(1)}_{xy}N^{(2)}_{zyx}(0)] \quad (2\text{-}16d)$$
$$+\frac{1}{2M_0}\{\omega_a[\chi^{(1)}_{xx}\chi^{(2)}_{zxx}(0)-\chi^{(1)}_{xy}\chi^{(2)}_{zyx}(0)+N^{(1)}_{xx}N^{(2)}_{zxx}(0)$$
$$-N^{(1)}_{xy}N^{(2)}_{zyx}(0)]-2\omega_m\chi^{(2)}_{zxx}(0)\}$$

$$e=\frac{i\omega\tau}{2M_0}[-3\chi^{(2)}_{zxx}(2\omega)\chi^{(1)*}_{xx}-3N^{(2)}_{zxx}(2\omega)N^{(1)*}_{xx}$$
$$+\chi^{(1)}_{xy}\chi^{(2)}_{zyx}(0)+N^{(1)}_{xy}N^{(2)}_{zyx}(0)]$$

$$+\frac{1}{2M_0}\{\omega_a[\chi^{(1)}_{xy}\chi^{(2)}_{zyx}(0)+N^{(1)}_{xy}N^{(2)}_{zyx}(0)+\chi^{(2)}_{zxx}(2\omega)\chi^{(1)*}_{xx}$$
$$+N^{(2)}_{zxx}(2\omega)N^{(1)*}_{xx}]-2\omega_m\chi^{(2)}_{zxx}(2\omega)\} \quad (2\text{-}16e)$$

$$f = \frac{i\omega\tau}{2M_0}[-3N_{zxx}^{(2)}(2\omega)N_{xx}^{(1)*} - 3\chi_{zxx}^{(2)}(2\omega)\chi_{xx}^{(1)*} + \chi_{xx}^{(1)}\chi_{zxx}^{(2)}(0) + N_{xx}^{(1)}N_{zxx}^{(2)}(0)] + \frac{1}{2M_0}\{\omega_a[\chi_{xx}^{(1)}\chi_{zxx}^{(2)}(0) + N_{xx}^{(1)}N_{zxx}^{(2)}(0) + N_{zxx}^{(2)}(2\omega)N_{xx}^{(1)*} + \chi_{zxx}^{(2)}(2\omega)\chi_{xx}^{(1)*}] - 2\omega_m[\chi_{zxx}^{(2)}(2\omega) + \chi_{zxx}^{(2)}(0)]\} \quad (2\text{-}16f)$$

$$g = \frac{\omega_m}{M_0}\chi_{xzx}^{(2)}(0) \quad (2\text{-}16g)$$

$$h = -\frac{i\omega\tau}{2M_0}[\chi_{xx}^{(1)}\chi_{zyx}^{(2)}(0) + N_{xx}^{(1)}N_{zyx}^{(2)}(0) + \chi_{xy}^{(1)}\chi_{zxx}^{(2)}(0) + N_{xy}^{(1)}N_{zxx}^{(2)}(0)] - \frac{1}{2M_0}\{\omega_a[\chi_{xx}^{(1)}\chi_{zyx}^{(2)}(0) + N_{xx}^{(1)}N_{zyx}^{(2)}(0) + \chi_{xy}^{(1)}\chi_{zxx}^{(2)}(0) + N_{xy}^{(1)}N_{zxx}^{(2)}(0)] - 2\omega_m\chi_{zyx}^{(2)}(0)\} \quad (2\text{-}16h)$$

$$l = -\frac{\omega_m}{M_0}[2\chi_{xyz}^{(2)}(2\omega) + \chi_{xyz}^{(2)}(0)] \quad (2\text{-}16i)$$

$$p = \frac{\omega_m}{M_0}[2\chi_{xxz}^{(2)}(2\omega) + \chi_{xxz}^{(2)}(0)] \quad (2\text{-}16j)$$

$$a' = \frac{i\omega\tau}{2M_0}[3\chi_{xy}^{(1)*}N_{zxx}^{(2)}(2\omega) + 3N_{xy}^{(1)*}\chi_{zxx}^{(2)}(2\omega) - \chi_{xy}^{(1)}N_{zxx}^{(2)}(0) - N_{xy}^{(1)}\chi_{zxx}^{(2)}(0)] - \frac{1}{2M_0}\{2\omega_e[\chi_{xy}^{(1)}N_{zxx}^{(2)}(0) + \chi_{xy}^{(1)*}N_{zxx}^{(2)}(2\omega) - N_{xy}^{(1)}\chi_{zxx}^{(2)}(0) - N_{xy}^{(1)*}\chi_{zxx}^{(2)}(2\omega)] + \omega_a[\chi_{xy}^{(1)}N_{zxx}^{(2)}(0) + \chi_{xy}^{(1)*}N_{zxx}^{(2)}(2\omega) + N_{xy}^{(1)*}\chi_{zxx}^{(2)}(2\omega) + N_{xy}^{(1)}\chi_{zxx}^{(2)}(0)]\} \quad (2\text{-}17a)$$

$$b' = \frac{i\omega\tau}{2M_0}[3\chi_{xy}^{(1)*}N_{zxx}^{(2)}(2\omega) + 3N_{xy}^{(1)*}\chi_{zxx}^{(2)}(2\omega) + N_{xx}^{(1)}\chi_{zyx}^{(2)}(0) + \chi_{xx}^{(1)}N_{zyx}^{(2)}(0)] + \frac{1}{2M_0}\{2\omega_e[\chi_{xx}^{(1)}N_{zyx}^{(2)}(0) - \chi_{xy}^{(1)*}N_{zxx}^{(2)}(2\omega) - N_{xx}^{(1)}\chi_{zyx}^{(2)}(0) + N_{xy}^{(1)*}\chi_{zxx}^{(2)}(2\omega)] + \omega_a[\chi_{xx}^{(1)}N_{zyx}^{(2)}(0) - \chi_{xy}^{(1)*}N_{zxx}^{(2)}(2\omega) + N_{xx}^{(1)}\chi_{zyx}^{(2)}(0) - N_{xy}^{(1)*}\chi_{zxx}^{(2)}(2\omega)] - 2\omega_m N_{zyx}^{(2)}(0)\} \quad (2\text{-}17b)$$

$$c' = -\frac{\omega_m}{M_0} N_{xzy}^{(2)}(0) \tag{2-17c}$$

$$\begin{aligned} d' = {} & \frac{i\omega\tau}{2M_0}[\chi_{xx}^{(1)} N_{zxx}^{(2)}(0) - \chi_{xy}^{(1)} N_{zyx}^{(2)}(0) + N_{xx}^{(1)} \chi_{zxx}^{(2)}(0) \\ & - N_{xy}^{(1)} \chi_{zyx}^{(2)}(0)] + \frac{1}{2M_0}\{2\omega_e[\chi_{xx}^{(1)} N_{zxx}^{(2)}(0) \\ & - \chi_{xy}^{(1)} N_{zyx}^{(2)}(0) - N_{xx}^{(1)} \chi_{zxx}^{(2)}(0) + N_{xy}^{(1)} \chi_{zyx}^{(2)}(0)] + \omega_a[\chi_{xx}^{(1)} N_{zxx}^{(2)}(0) \\ & - \chi_{xy}^{(1)} N_{zyx}^{(2)}(0) - N_{xy}^{(1)} \chi_{zyx}^{(2)}(0) \\ & + N_{xx}^{(1)} \chi_{zxx}^{(2)}(0)] - 2\omega_m N_{zxx}^{(2)}(0)\} \end{aligned} \tag{2-17d}$$

$$\begin{aligned} e' = {} & \frac{i\omega\tau}{2M_0}[-3\chi_{xx}^{(1)*} N_{zxx}^{(2)}(2\omega) - 3N_{xx}^{(1)*} \chi_{zxx}^{(2)}(2\omega) + \chi_{xy}^{(1)} N_{zyx}^{(2)}(0) \\ & + N_{xy}^{(1)} \chi_{zyx}^{(2)}(0)] + \frac{1}{2M_0}\{2\omega_e[\chi_{xx}^{(1)*} N_{zxx}^{(2)}(2\omega) \\ & + \chi_{xy}^{(1)} N_{zyx}^{(2)}(0) - N_{xx}^{(1)*} \chi_{zxx}^{(2)}(2\omega) - N_{xy}^{(1)} \chi_{zyx}^{(2)}(0)] + \omega_a[\chi_{xx}^{(1)*} N_{zxx}^{(2)}(2\omega) \\ & + \chi_{xy}^{(1)} N_{zyx}^{(2)}(0) + N_{xx}^{(1)*} \chi_{zxx}^{(2)}(2\omega) \\ & + N_{xy}^{(1)} \chi_{zyx}^{(2)}(0)] - 2\omega_m N_{zxx}^{(2)}(2\omega)\} \end{aligned} \tag{2-17e}$$

$$\begin{aligned} f' = {} & -\frac{i\omega\tau}{2M_0}[3\chi_{xx}^{(1)*} N_{zxx}^{(2)}(2\omega) + 3N_{xx}^{(1)*} \chi_{zxx}^{(2)}(2\omega) - \chi_{xx}^{(1)} N_{zxx}^{(2)}(0) \\ & - N_{xx}^{(1)} \chi_{zxx}^{(2)}(0)] + \frac{1}{2M_0}\{2\omega_e[\chi_{xx}^{(1)} N_{zxx}^{(2)}(0) \\ & + \chi_{xx}^{(1)*} N_{zxx}^{(2)}(2\omega) - N_{xx}^{(1)} \chi_{zxx}^{(2)}(0) - N_{xx}^{(1)*} \chi_{zxx}^{(2)}(2\omega)] + \omega_a[\chi_{xx}^{(1)} N_{zxx}^{(2)}(0) \\ & + \chi_{xx}^{(1)*} N_{zxx}^{(2)}(2\omega) + N_{xx}^{(1)} \chi_{zxx}^{(2)}(0) \\ & + N_{xx}^{(1)*} \chi_{zxx}^{(2)}(2\omega)] - 2\omega_m (N_{zxx}^{(2)}(0) + N_{zxx}^{(2)}(2\omega))]\} \end{aligned} \tag{2-17f}$$

$$g' = \frac{\omega_m}{M_0} N_{xzx}^{(2)}(0) \tag{2-17g}$$

$$\begin{aligned} h' = {} & -\frac{i\omega\tau}{2M_0}[\chi_{xx}^{(1)} N_{zyx}^{(2)}(0) + \chi_{xy}^{(1)} N_{zxx}^{(2)}(0) + N_{xx}^{(1)} \chi_{zyx}^{(2)}(0) \\ & + N_{xy}^{(1)} \chi_{zxx}^{(2)}(0)] - \frac{1}{2M_0}\{2\omega_e[\chi_{xx}^{(1)} N_{zyx}^{(2)}(0) \\ & + \chi_{xy}^{(1)} N_{zxx}^{(2)}(0) - N_{xx}^{(1)} \chi_{zyx}^{(2)}(0) - N_{xy}^{(1)} \chi_{zxx}^{(2)}(0)] + \omega_a[\chi_{xx}^{(1)} N_{zyx}^{(2)}(0) \\ & + \chi_{xy}^{(1)} N_{zxx}^{(2)}(0) + N_{xx}^{(1)} \chi_{zyx}^{(2)}(0) \\ & + N_{xy}^{(1)} \chi_{zxx}^{(2)}(0)] - 2\omega_m N_{zyx}^{(2)}(0)\} \end{aligned} \tag{2-17h}$$

$$l' = -\frac{\omega_m}{M_0}[2N_{xyz}^{(2)}(2\omega) + N_{xyz}^{(2)}(0)] \tag{2-17i}$$

$$p' = \frac{\omega_m}{M_0}[2N_{xxz}^{(2)}(2\omega) + N_{xxz}^{(2)}(0)] \tag{2-17j}$$

The symmetry relations among the third-order elements are found to be

$\chi_{xxxx}^{(3)}(\omega) = \chi_{yyyy}^{(3)}(\omega)$, $\chi_{xyyx}^{(3)}(\omega) = \chi_{yxxy}^{(3)}(\omega)$

$\chi_{xzzx}^{(3)}(\omega) = \chi_{yzzy}^{(3)}(\omega)$, $\chi_{xxxy}^{(3)}(\omega) = -\chi_{yyyx}^{(3)}(\omega)$, $\chi_{xyyy}^{(3)}(\omega) = -\chi_{yxxx}^{(3)}(\omega)$

$\chi_{xxyx}^{(3)}(\omega) = \chi_{xyxx}^{(3)}(\omega) = -\chi_{yxyy}^{(3)}(\omega) = -\chi_{yyxy}^{(3)}(\omega)$, $\chi_{xzzy}^{(3)}(\omega) = -\chi_{yzzx}^{(3)}(\omega)$, $\chi_{zxzy}^{(3)}(\omega) = \chi_{zzxy}^{(3)}(\omega)$,

$\chi_{zxzx}^{(3)}(\omega) = \chi_{zzxx}^{(3)}(\omega)$, $\chi_{xxyy}^{(3)}(\omega) = \chi_{xyxy}^{(3)}(\omega) = \chi_{yxyx}^{(3)}(\omega) = \chi_{yyxx}^{(3)}(\omega)$, $\chi_{zyzx}^{(3)}(\omega) = \chi_{zzyx}^{(3)}(\omega)$,

$\chi_{xxzz}^{(3)}(\omega) = \chi_{xzxz}^{(3)}(\omega) = \chi_{yyzz}^{(3)}(\omega) = \chi_{yzyz}^{(3)}(\omega)$, $\chi_{zyzy}^{(3)}(\omega) = \chi_{zzyy}^{(3)}(\omega)$,

$\chi_{xyzz}^{(3)}(\omega) = \chi_{xzyz}^{(3)}(\omega) = -\chi_{yxzz}^{(3)}(\omega) = -\chi_{yzxz}^{(3)}(\omega)$.

Although there are 81 elements of the third-order susceptibility tensor and their expressions are very complicated, but many among them may not be applied due to the plane or line polarization of used electromagnetic waves. for example when the magnetic field $\vec{H}$ is in the x-y plane, the third-order elements with only subscripts x and y, such as $\chi_{xxxx}^{(3)}(\omega)$, $\chi_{xxyx}^{(3)}(\omega)$, $\chi_{xyyx}^{(3)}(\omega)$ and $\chi_{xyyy}^{(3)}(\omega)$ *et. al.*, are usefull. In addition, if the external magnetic field H_0 is removed, many the first- second- and third-order elements will disappear, or become 0. In the following sections, when one discusses AF polaritons the damping is neglected, but when investigating transmission and reflection the damping is considered.

3. Linear polaritons in antiferromagnetic systems

The linear AF polaritons of AF systems (AF bulk, AF films and superlattices) are eigen modes of electromagnetic waves propagating in the systems. The features of these modes can predicate many optical and electromagnetic properties of the systems. There are two kinds of the AF polaritons, the surface modes and bulk modes. The surface modes propagate along a surface of the systems and exponentially attenuate with the increase of distance to this surface. For these AF systems, an optical technology was applied to measure the AF polariton spectra (Jensen, 1995). The experimental results are completely consistent with the theoretical predications. In this section, we take the Voigt geometry usually used in the experiment and theoretical works, where the waves propagate in the plane normal to the AF anisotropy axis and the external magnetic field is pointed along this anisotropy axis.

3.1 Polaritons in AF bulk and film

Bulk AF polaritons can be directly described by the wave equation of EMWs in an AF crystal,

$$\nabla(\nabla \cdot \vec{H}) - \nabla^2 \vec{H} - \varepsilon_a \omega^2 \vec{\mu} \cdot \vec{H} = 0 \tag{3-1}$$

where ε_a is the AF dielectric constant and $\vec{\mu}$ is the magnetic permeability tensor. It is interesting that the magnetic field of AF polaritons vibrates in the x-y plane since the field

does not couple with the AF magnetization for it along the z axis. We take the magnetic field as $\vec{H} = \bar{A}\exp(i\vec{k}\cdot\vec{r} - i\omega t)$ with the amplitude $\bar{A}$. Thus applying equation (3-1) we find directly the dispersion relation of bulk polaritons

$$k_x^2 + k_y^2 = \varepsilon_a \mu_\nu \omega^2 \tag{3-2}$$

with $\mu_\nu = [\mu_1^2 - \mu_2^2]/\mu_1$ the AF effective permeability. Equation (3-2) determines the continuums of AF polaritons in the $k-\omega$ figure (see Fig.2).

The best and simplest example available to describe the surface AF polariton is a semi-infinite AF. We assume the semi-infinite AF occupies the lower semi-space and the upper semi-space is of vacuum. The y axis is normal to the surface. The surface polariton moves along the x axis. The wave field in different spaces can be shown by

$$\vec{H} = \begin{cases} \bar{A}_0 \exp(-\alpha_0 y + ik_x x - i\omega t), & (\text{in the vaccum}) \\ \bar{A}\exp(\alpha y + ik_x x - i\omega t), & (\text{in the AF}) \end{cases} \tag{3-3}$$

where α_0 and α are positive attenuation factors . From the magnetic field (3-3) and the Maxwell equation $\nabla \times \vec{H} = \partial\vec{D}/\partial t$, we find the corresponding electric field

$$\vec{E} = \vec{e}_z \begin{cases} \dfrac{i}{\varepsilon_0\omega}[ik_x A_{0y} + \alpha_0 A_{0x}]\exp(-\alpha_0 y + ik_x x - i\omega t) \\ \dfrac{i}{\varepsilon_a\omega}[ik_x A_y - \alpha A_x]\exp(\alpha y + ik_x x - i\omega t), \end{cases} \tag{3-4}$$

Here there are 4 amplitude components, but we know from equation $\nabla\cdot(\vec{\mu}\cdot\vec{H}) = 0$ that only two are independent. This bounding equation leads to

$$A_{0y} = ik_x A_{0x}/\alpha_0 \, , \, A_y = i(k_x\mu_1 - \alpha\mu_2)A_x/(k_x\mu_2 - \alpha\mu_1) \tag{3-5}$$

The wave equation (3-1) shows that

$$\alpha_0^2 = k_x^2 - (\omega/c)^2 \, , \, \alpha^2 = k_x^2 - \mu_\nu(\omega/c)^2 \tag{3-6}$$

determining the two attenuation constants. The boundary conditions of H_x and E_z continuous at the interface (y=0) lead to the dispersion relation

$$\mu_1(\alpha_0\mu_v + \varepsilon_a\alpha) = \varepsilon_a\mu_2 k_x \tag{3-7}$$

where the permeability components and dielectric constants all are their relative values. Equation (3-7) describes the surface AF polariton under the condition that the attenuation factors both are positive. In practice, Eq.(3-6) also shows the dispersion relation of bulk modes as that attenuation factor is vanishing.

We illustrate the features of surface and bulk AF polaritons in Fig.2. There are three bulk continua where electromagnetic waves can propagate. Outside these regions, one sees the surface modes, or the surface polariton. The surface polariton is non-reciprocal, or the polariton exhibits completely different properties as it moves in two mutually opposite directions, respectively. This non-reciprocity is attributed to the applied external field that

breaks the magnetic symmetry of the AF. If we take an AF film as example to discuss this subject, we are easy to see that the surface mode is changed only in quantity, but the bulk modes become so-called guided modes, which no longer form continua and are some separated modes (Cao & Caillé, 1982).

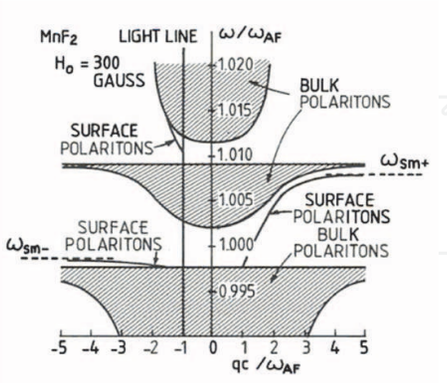


Fig. 2. Surface polariton dispersion curves and bulk continua on the MnF_2 in the geometry with an applied external field. After Camley & Mills,1982

3.2 Polaritons in antiferromagnetic multilayers and superlattices

There have been many works on the magnetic polaritons in AF multilayers or superlattices. This AF structure is the one-dimension stack, commonly composed of alternative AF layers and dielectric (DE) layers, as illustrated in Fig.3.

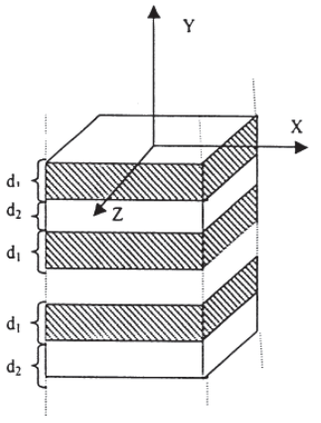


Fig. 3. The structure of AF superlattice and selected coordinate system.

In the limit case of small stack period, the effective-medium method was developed (Oliveros, et. al., 1992; Camley, 1992; Raj & Tilley, 1987; Almeida & Tilley, 1990; Cao & Caillé, 1982; Almeida & Mills,1988; Dumelow & Tilley,1993; Elmzughi, 1995a, 1995b). According to this method, one can consider these structures as some homogeneous films or bulk media with effective magnetic permeability and dielectric constant. This method and its results are very simple in mathematics. Of course, this is an approximate method. The other method is called as the transfer-matrix method (Born & Wolf, 1964; Raj & Tilley, 1989), where the electromagnetic boundary conditions at one interface set up a matrix relation between field amplitudes in the two adjacent layers, or adjacent media. Thus amplitudes in any layer can be related to those in another layer by the product of a series of matrixes. For

an infinite AF superlattice, the Bloch's theorem is available and can give an additional relation between the corresponding amplitudes in two adjacent periods. Using these matrix relations, bulk AF polaritons in the superlattices can be determined. For one semi-finite structure with one surface, the surface mode can exist and also will be discussed with the method.

3.2.1 The limit case of short period, effective-medium method

Now we introduce the effective-medium method, with the condition of the wavelength λ much longer than the stack period $D = d_1 + d_2$ (d_1 and d_2 are the AF and DE thicknesses). The main idea of this method is as follows. We assume that there are an effective relation $\vec{B} = \ddot{\mu}_{eff} \cdot \vec{H}$ between effective magnetic induction and magnetic field, and an effective relation $\vec{D} = \ddot{\varepsilon}_{eff} \cdot \vec{E}$ between effective electric field and displacement, where these fields are considered as the wave fields in the structures. But $\vec{b} = \ddot{\mu} \cdot \vec{h}$ and $\vec{d} = \varepsilon \vec{e}$ in any layer, where $\ddot{\mu}$ is given in section 2 for AF layers and $\ddot{\mu} = 1$ for DE layers. These fields are local fields in the layers. For the components of magnetic induction and field continuous at the interface, one assumes

$$H_x = h_{1x} = h_{2x}\,,\ H_z = h_{1z} = h_{2z}\,,\ B_y = b_{1y} = b_{2y} \tag{3-8a}$$

and for those components discontinuous at the interface, one assumes

$$B_x = f_1 b_{1x} + f_2 b_{2x}\,,\ B_z = f_1 b_{1z} + f_2 b_{2z}\,,\ H_y = f_1 H_{1y} + f_2 H_{2y} \tag{3-8b}$$

where the AF ratio $f_1 = d_1 / (d_1 + d_2)$ and the DE ratio $f_2 = 1 - f_1$. Thus the effective magnetic permeability is obtained from equations (3-8) and its definition $\vec{B} = \ddot{\mu}_{eff} \cdot \vec{H}$,

$$\ddot{\mu}_{eff} = \begin{pmatrix} \mu_{xx}^e & i\mu_{xy}^e & 0 \\ -i\mu_{xy}^e & \mu_{yy}^e & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{3-9}$$

with the elements

$$\mu_{xx}^e = f_1\mu_1 + f_2 - \frac{f_1 f_2 \mu_2^2}{f_1 + f_2\mu_1}\,,\ \mu_{yy}^e = \frac{\mu_1}{f_1 + f_2\mu_1}\,,\ \mu_{xy}^e = \frac{f_1\mu_2}{f_1 + f_2\mu_1} \tag{3-10}$$

On the similar principle, we can find that the effective dielectric permittivity tensor is diagonal and its elements are

$$\varepsilon_{xx}^e = \varepsilon_{zz}^e = f_1\varepsilon_1 + f_2\varepsilon_2\,,\ \varepsilon_{yy}^e = \varepsilon_1\varepsilon_2 / (f_1\varepsilon_2 + f_2\varepsilon_1) \tag{3-11}$$

On the base of these effective permeability and permittivity, one can consider the AF multilayers or superlattices as homogeneous and anisotropical AF films or bulk media, so the same theory as that in section 3.1 can be used. Magnetic polaritons of AF multilayers (Oliveros, et.al., 1992; Raj & Tilley, 1987), AF superlattices with parallel or transverse surfaces (Camley, et. al., 1992; Barnas, 1988) and one-dimension AF photonic crystals (Song, et.al., 2009; Ta, et. al.,2010) have been discussed with this method.

3.2.2 Polaritons and transmission of AF multilayers: transfer-matrix method

If the wavelength is comparable to the stack period, the effective-medium method is no longer available so that a strict method is necessary. The transfer-matrix method is such a method. In this subsection, we shall present magnetic polaritons of AF multilayers or superlattices with this method. We introduce the wave magnetic field in two layers in the *l*th stack period as follows.

$$\vec{H} = \mathrm{e}^{ik_x x - i\omega t}\begin{cases}(\vec{A}_+^l\,\mathrm{e}^{ik_1 y} + \vec{A}_-^l\,\mathrm{e}^{-ik_1 y}) & (\text{in the AF layer})\\ (\vec{B}_+^l\,\mathrm{e}^{ik_2 y} + \vec{B}_-^l\,\mathrm{e}^{-ik_2 y}) & (\text{in the DE layer})\end{cases} \tag{3-12}$$

where k_1 and k_2 are determined with $k_1^2 + k_x^2 = \varepsilon_1\mu_v\omega^2$ and $k_2^2 + k_x^2 = \varepsilon_2\mu_0\omega^2$. Similar to Eq. (3-4) in subsection 3.1, the corresponding electric field in this period is written as

$$\vec{E} = \vec{e}_z e^{ik_x x - i\omega t}\begin{cases}\dfrac{i}{\varepsilon_1\omega}[(ik_x A_{+y}^l - ik_1 A_{+x}^l)\mathrm{e}^{ik_1 y} + (ik_x A_{-y}^l + ik_1 A_{-x}^l)\mathrm{e}^{-ik_1 y}]\\ \dfrac{i}{\varepsilon_2\omega}[(ik_x B_{+y}^l - ik_2 B_{+x}^l)\mathrm{e}^{ik_2 y} + (ik_x B_{-y}^l + ik_2 B_{-x}^l)\mathrm{e}^{-ik_2 y}]\end{cases} \tag{3-13}$$

Here there is a relation between per pair of amplitude components, or

$$A_{\pm y}^l = i(k_x\mu_1 \mp ik_1\mu_2)A_{\pm x}^l / (k_x\mu_2 \mp ik_1\mu_1) = \lambda_\pm A_{\pm x}^l,\ B_{\pm y}^l = \mp k_x B_{\pm x}^l / k_2 \tag{3-14}$$

As a result, we can take $A_{\pm x}^l$ and $B_{\pm x}^l$ as 4 independent amplitude components. Next, according to the continuity of electromagnetic fields at that interface in the period, we find

$$A_{+x}^l e^{ik_1 d_1} + A_{-x}^l e^{-ik_1 d_1} = B_{+x}^l + B_{-x}^l \tag{3-15a}$$

$$\frac{1}{\varepsilon_1}[(-k_x A_{+y}^l + k_1 A_{+x}^l)\mathrm{e}^{ik_1 d_1} - (k_x A_{-y}^l + k_1 A_{-x}^l)\mathrm{e}^{-ik_1 d_1}] = \frac{\omega\mu_0}{k_2}(B_{+x}^l - B_{-x}^l) \tag{3-15b}$$

At the interface between the *l*th and *l+1*th periods, one see

$$(A_{+x}^{l+1} + A_{-x}^{l+1}) = B_{+x}^l e^{ik_2 d_2} + B_{-x}^l e^{-ik_2 d_2} \tag{3-15c}$$

$$\frac{1}{\varepsilon_1}[(k_1 A_{+x}^{l+1} - k_x A_{+y}^{l+1}) - (k_1 A_{-x}^{l+1} + k_x A_{-y}^{l+1})] = \frac{\omega\mu_0}{k_2}(B_{+x}^l\,\mathrm{e}^{ik_2 d_2} - B_{-x}^l\,\mathrm{e}^{-ik_2 d_2}) \tag{3-15d}$$

Thus the matrix relation between the amplitude components in the same period is introduced as

$$\begin{pmatrix} B_{+x}^l \\ B_{-x}^l \end{pmatrix} = \begin{pmatrix} \Gamma_{11} & \Gamma_{12} \\ \Gamma_{21} & \Gamma_{22} \end{pmatrix}\begin{pmatrix} A_{+x}^l \\ A_{-x}^l \end{pmatrix} \tag{3-16}$$

where the matrix elements are given by

$$\Gamma_{11} = \frac{e^{ik_1 d_1}}{2}(1+\Delta_+),\ \Gamma_{12} = \frac{e^{-ik_1 d_1}}{2}(1-\Delta_-),\ \Gamma_{21} = \frac{e^{ik_1 d_1}}{2}(1-\Delta_+),\ \Gamma_{22} = \frac{e^{-ik_1 d_1}}{2}(1+\Delta_-) \tag{3-17}$$

with $\Delta_{\pm} = k_2(k_1 \mp \lambda_{\pm} k_x) / \omega\mu_0\varepsilon_1$. From (3-15), the other relation also is obtained, or

$$\begin{pmatrix} B_{+x}^{l} \\ B_{-x}^{l} \end{pmatrix} = \begin{pmatrix} \Lambda_{11} & \Lambda_{12} \\ \Lambda_{21} & \Lambda_{22} \end{pmatrix} \begin{pmatrix} A_{+x}^{l+1} \\ A_{-x}^{l+1} \end{pmatrix} \tag{3-18}$$

with

$$\Lambda_{11} = \frac{e^{-ik_2 d_2}}{2}(1+\Delta_{+}) \,,\, \Lambda_{12} = \frac{e^{-ik_2 d_2}}{2}(1-\Delta_{-}) \,,\, \Lambda_{21} = \frac{e^{ik_2 d_2}}{2}(1-\Delta_{+}) \,,\, \Lambda_{22} = \frac{e^{ik_2 d_2}}{2}(1+\Delta_{-}) \tag{3-19}$$

Commonly, the matrix relation between the amplitude components in the *l*th and *l+1*th periods is written as

$$\begin{pmatrix} A_{+x}^{l} \\ A_{-x}^{l} \end{pmatrix} = \Gamma^{-1}\Lambda \begin{pmatrix} A_{+x}^{l+1} \\ A_{-x}^{l+1} \end{pmatrix} = T \begin{pmatrix} A_{+x}^{l+1} \\ A_{-x}^{l+1} \end{pmatrix} \tag{3-20}$$

In order to discuss bulk AF polaritons, an infinite AF superlattice should be considered. Then the Bloch's theorem is available so that $A_{\pm x}^{l+1} = gA_{\pm x}^{l}$ with $g = \exp(-iQD)$, and then the dispersion relation of bulk magnetic polaritons just is

$$\cos(QD) = \frac{1}{2}(T_{11} + T_{22}) \tag{3-21}$$

It can be reduced into a more clearly formula, or

$$\cos(QD) = \cos(k_1 d_1)\cos(k_2 d_2) - \frac{k_1^2 + k_2^2\mu_v^2 - k_x^2\mu_2^2 / \mu_1^2}{2k_1 k_2 \mu_v}\sin(k_1 d_1)\sin(k_2 d_2) \tag{3-22}$$

When one wants to discuss the surface polariton, the semi-infinite system is the best and simplest example. In this situation, the Bloch's theorem is not available and the polariton wave attenuates with the distance to the surface, according to $\exp(-\alpha lD)$, where lD is the distance and α is the attenuation coefficient and positive. As a result,

$$\cosh(\alpha D) = \frac{1}{2}(T_{11} + T_{22}) \tag{3-23}$$

It should remind that equation (3-23) cannot independently determine the dispersion of the surface polariton since the attenuation coefficient is unknown, so an additional equation is necessary. We take the wave function outside this semi-infinite structure as $\vec{H} = \vec{A}_0 \exp(-\alpha_0 y + ik_x x - i\omega t)$ with α_0 the vacuum attenuation constant. The two components of the amplitude vector are related with $A_{0y} = ik_x A_{0x} / \alpha_0$ and $k_x^2 - \alpha_0^2 = (\omega / c)^2$. The corresponding electric field is $E_z = -(i\omega\mu_0 / \alpha_0)H_x$. The boundary conditions of field components H_x and E_z continuous at the surface lead to

$$A_{0x} = A_{+x} + A_{-x} \tag{3-24a}$$

$$-(i\omega\mu_0\varepsilon_1/\alpha_0)A_{0x} = (k_1A_{+x} - k_xA_{+y}) - (k_1A_{-x} + k_xA_{-y}) \tag{3-24b}$$

$$A_{+x} = g'(T_{11}A_{+x} + T_{12}A_{-x}) \tag{3-24c}$$

with $g' = \exp(-\alpha D)$. These equations result in another relation,

$$g'T_{12}(k_x\lambda_+ - k_1 - \Delta_0) + (1 - g'T_{11})(k_x\lambda_- + k_1 - \Delta_0) = 0 \tag{3-25}$$

Eqs. (3-23) and (3-25) jointly determine the dispersion properties of the surface polariton under the conditions of $\alpha, \alpha_0 > 0$.

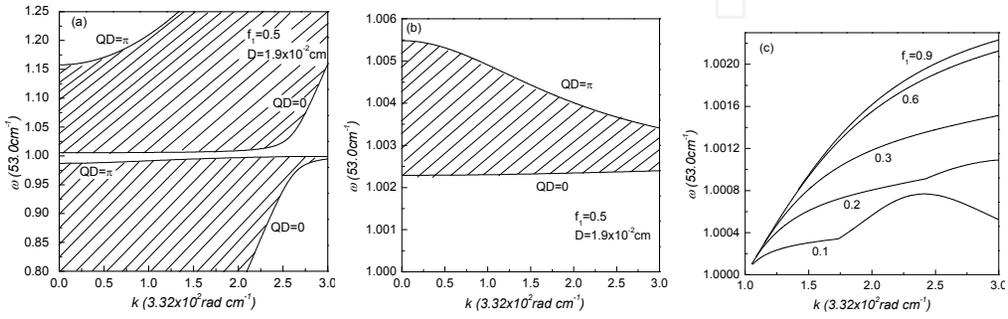


Fig. 4. Frequency spectrum of the polaritons of the FeF_2/ZnF_2 superalttice. (a) shows the top and bottom bands, and (b) presents the middle band. The surface mode is illustrated in (c). f_1 denotes the ratio of the FeF_2 in one period of the superlattice. After Wang & Li, 2005.

We present a figure example to show features of bulk and surface polaritons, as shown in Fig.4. Because of the symmetry of dispersion curves with respective to *k=0,* we present only the dispersion pattern in the range of *k>0.* The bulk polaritons form several separated continuums, and the surface mode exists in the bulk-polariton stop-bands. The bulk polaritons are symmetrical in the propagation direction, or possess the reciprocity, but is not the surface mode. These properties also can be found from the dispersion relations. For the bulk polaritons, the wave vector appears in dispersion equation (3-22) in its k_x^2 style, but for the surface mode, k_x and k_x^2 both are included dispersion equation (3-25).

3.2.3 Transmission of AF multilayers

In practice, infinite AF superlattices do not exist, so the conclusions from them are approximate results. For example, if the incident-wave frequency falls in a bulk-polariton stop-band of infinite AF superlattice, the transmission of the corresponding AF multilayer must be very weak, but not vanishing. Of course, it is more intensive in the case of frequency in a bulk-polariton continuum. Based on the above results, we derive the transmission ratio of an AF multilayer, where this structure has two surfaces, the upper surface and lower surface. We take a TE wave as the incident wave, with its electric component normal to the incident plane (the x-y plane) and along the z axis. The incident wave illuminates the upper surface and the transmission wave comes out from the lower surface. We set up the wave function above and below the multilayer as

$$\vec{H} = [\vec{I}_0\exp(-ik_0y) + \vec{R}_0\exp(ik_0y)]\exp(ik_xx)\text{ ,(above the system)} \tag{3-26a}$$

$$\vec{H} = \vec{T}_0 \exp(-ik_0 y + ik_x x) \text{ (below the system)} \tag{3-26b}$$

The wave function in the multilayer has been given by (3-12) and (3-13). By the mathematical process similar to that in subsection 3.2.2, we can obtain the transmission and reflection of the multilayer with N periods from the following matrix relation,

$$\begin{pmatrix} I_0 \\ R_0 \end{pmatrix} = \Lambda_0 T^{N-1} \Gamma^{-1} \Lambda_1 \begin{pmatrix} T_0 \\ T_0 \end{pmatrix} \tag{3-27}$$

in which two new matrixes are shown with

$$\Lambda_0 = \begin{pmatrix} 1+\Delta'_+ & 1+\Delta'_- \\ 1-\Delta'_+ & 1-\Delta'_- \end{pmatrix}, \Lambda_1 = \begin{pmatrix} 1-k_2/k_0 & 0 \\ 0 & 1+k_2/k_0 \end{pmatrix} \tag{3-28}$$

with $k_0 = [(\omega/c)^2 - k_x^2]^{1/2}$ and $\Delta'_\pm = k_0(k_x \lambda_\pm \mp k_1)/\omega\mu_0\varepsilon_1$. Thus the reflection and transmission are determined with equation (3-27). In numerical calculations, the damping in the permeability cannot is ignored since it implies the existence of absorption. We have obtained the numerical results on the AF multilayer, and transmission spectra are consistent with the polariton spectra (Wang, J. J. et. al, 1999), as illustrated in Fig.5.

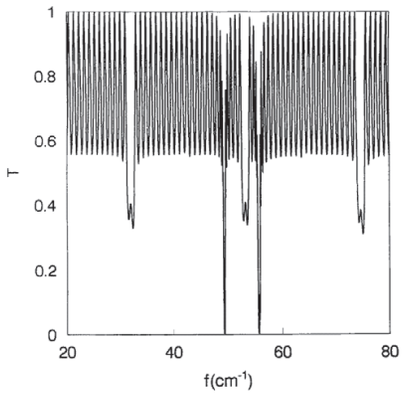


Fig. 5. Transmission curve for FeF_2 multilayer in Voigt geometry. After Wang, J. J. et. al, 1999.

4. Nonlinear surface and bulk polaritons in AF superlattices

In the previous section, we have discussed the linear propagation of electromagnetic waves in various AF systems, including the transmission and reflection of finite thickness multilayer. The results are available to the situation of lower intensity of electromagnetic waves. If the intensity is very high, the nonlinear response of magnetzation in AF media to the magnetic component of electromagnetic waves cannot be neglected. Under the present laser technology, this case is practical. Because we have found the second- and third-order magnetic susceptibilities of AF media, we can directly derive and solve nonlinear dispersion equations of electromagnetic waves in various AF systems. There also are two situations to be discussed. First，if the wavelenght λ is much longer than the superlattice period L ($\lambda \gg L$), the superlattice behaves like an anisotropic bulk medium(Almeida & Mills,1988; Raj & Tilley,1987), and the effective-medium approch is reasonable. We have introduced a

nonlinear effective-medium theory(Wang & Fu, 2004), to solve effective susceptibilities of magnetic superlattices or multilayers. This method has a key point that the effective second- and third-order magnetizations come from the contribution of AF layers or $\vec{m}_e^{(2)} = f_1 \vec{m}^{(2)}$ and $\vec{m}_e^{(3)} = f_1 \vec{m}^{(3)}$.

4.1 Polaritons in AF superlattice

In this section we shall use a stricter method to deal with nonlinear propagation of AF polaritons in AF superlattices. In section 2, we have obtained various nonlinear susceptibilities of AF media, which means that one has obtained the expressions of $\vec{m}^{(2)}$ and $\vec{m}^{(3)}$. In AF layers, the polariton wave equation is

$$\nabla(\nabla \cdot \vec{H}^{NL}) - \nabla^2 \vec{H}^{NL} - k_0^2 \vec{\mu} \cdot \vec{H}^{NL} = k_0^2 \vec{m}^{(3)}, \qquad k_0^2 = \varepsilon_1 (\omega / c)^2, \tag{4-1}$$

where $\vec{\mu}$ is the linear permeability of antiferromagnetic layers given in section 2, and the nonzeroelements $\mu_{yy} = \mu_{xx} = \mu$, $\mu_{zz} = 1$. The third-order magnetization is indicated by $m_i^{(3)} = \sum_{jkl} \chi_{ijkl}^{(3)} H_j H_k H_l^*$ with the nonlinear susceptibility elements presened in section 2. As an approximation, we consider the field components H_i in $m_i^{(3)}$ as linear ones to find the nonlinear solution of $\vec{H}^{NL}$ included in wave equanion (4-1). For the linear surface wave propagating along the x-axis and the linear bulk waves moving in the x-y plane, $\partial / \partial z = 0$. Thus the wave equation is rewritten as

$$ik_x \frac{\partial}{\partial y} H_y^{NL} - \frac{\partial^2}{\partial y^2} H_x^{NL} - \varepsilon_1 \omega^2 \mu_1 H_x^{NL} = \varepsilon_1 \omega^2 \Gamma(y)(\chi_{xxxy}^{(3)} H_x^{NL} - \chi_{xyyx}^{(3)} H_y^{NL}) \tag{4-2a}$$

$$ik_x \frac{\partial}{\partial y} H_x^{NL} + (k_x^2 - \varepsilon_1 \omega^2 \mu) H_y^{NL} = \varepsilon_1 \omega^2 \Gamma(y)(\chi_{xxxy}^{(3)} H_y^{NL} - \chi_{xyyx}^{(3)} H_x^{NL}) \tag{4-2b}$$

$$(k^2 - \frac{\partial^2}{\partial y^2} - \varepsilon_1 \omega^2) H_z^{NL} = \varepsilon_1 \omega^2 m_z^{(3)} \tag{4-2c}$$

with $\Gamma(y) = (H_x H_y^* - H_x^* H_y)$. Eq.(4-2c) implies that H_z is a third-order small quantity and equal to zero in the circumstance of linearity (TM waves). We begin from the linear wave solution that has been given section 3 to look for the nonlinear wave solution in AF layers. In the case of linearity, the relations among the wave amplitudes, $A_{\pm y} = \mp i k_x A_{\pm x} / \alpha_1$ with $\alpha_1 = [k_x^2 - \varepsilon_1 \mu (\omega / c)^2]^{1/2}$. The nonlinear terms in equations (4-2) should contain a factor $F(m) = \exp(-mn\beta D)$ with $m = 3$ and β is defined as the attenuation constant for the surface modes, and m=1 and $\beta = iQ$ with Q the Bloch's wavwnumber for the bulk modes. $A_1 \sim D_1$ and $A_2 \sim D_2$ are nonlinear coefficients. After solving the derivation of equation (4-2b) with respect to y, substituting it into (4-2a) leads to the wave solutions

$$\begin{aligned} H_x = {} & A_{+x} e^{i(k_x x - \omega t)} e^{-n\beta D} \{ e^{\alpha_1 (y+nD)} + \alpha' e^{-\alpha_1 (y+nD)} + f_n [(y+nD)\alpha_1 L_1 e^{\alpha_1 (y+nD)} \\ & + \alpha (y+nD)\alpha_1 L_2 e^{-\alpha_1 (y+nD)} + L_3 e^{3\alpha_1 (y+nD)} + L_4 e^{-3\alpha_1 (y+nD)}] \} \end{aligned} \tag{4-3a}$$

and

$$H_y = -\frac{ik_x}{\alpha_1} A_{+x} e^{-\beta nD} e^{i(k_x x-\omega t)} \{e^{\alpha_1 (y+nD)} - \alpha' e^{-\alpha_1 (y+nD)} + f_n[((y+nD)\alpha_1 L_1 + S) \\ e^{\alpha_1 (y+nD)} + \alpha(-(y+nD)\alpha_1 L_2 + T) e^{-\alpha_1 (y+nD)} + L_3' e^{3\alpha_1 (y+nD)} + L_4' e^{-3\alpha_1 (y+nD)}]\} \quad (4\text{-}3b)$$

in which $f_n = 1$ for the bulk modes and $f_n = \exp(-2n\beta D)$ for the surface modes. The expressions of coefficients in Eqs.(4-3a) and (4-3b) are presented as follows:

1. When α_1 is a real number, the coefficients in Eq.(4-3a) are

$$A_1 = 2\alpha k_x A_m \chi_+ , B_1 = -2|\alpha|^2 k_x A_m \chi_- , C_1 = 2k_x A_m \chi_- , D_1 = -2\alpha|\alpha|^2 k_x A_m \chi_+ \quad (4\text{-}4a)$$

$$A_2 = 2i\alpha k_x A_m \delta_+ , \; B_2 = 2ik_x |\alpha|^2 A_m \delta_- , C_2 = -2ik_x A_m \delta_- , D_2 = -2ik_x \alpha |\alpha|^2 A_m \delta_+ \quad (4\text{-}4b)$$

where $\chi_\pm = i\alpha_1 \chi^{(3)}_{xxxy} \pm k_x \chi^{(3)}_{xyyx}$, $\delta_\pm = ik_x \chi^{(3)}_{xxxy} \pm \alpha_1 \chi^{(3)}_{xyyx}$, $A_m = \varepsilon_1 \omega^2 |A|^2 / [k_x^2 + |\alpha_1|^2]$. The field strength $|A|^2 = |A_x|^2 + |A_y|^2 = [|\alpha_1|^2 + k_x^2] |A_x|^2 / |\alpha_1|^2$. From the boundary conditions of the linear field, one also can easily prove that α included in the formulae is

$$\alpha = A_{-x} / A_{+x} = (\alpha_0 \mu + \alpha_1) / (\alpha_0 \mu - \alpha_1) \quad (4\text{-}5a)$$

for the surface modes and

$$\alpha = \frac{e^{-\alpha_1 d_1} [\alpha_1 \cosh(\alpha_2 d_2) - \mu\alpha_2 \sinh(\alpha_2 d_2)] - \alpha_1 e^{-iQD}}{\alpha_1 e^{-iQD} - e^{\alpha_1 d_1} [\alpha_1 \cosh(\alpha_2 d_2) + \mu\alpha_2 \sinh(\alpha_2 d_2)]} \quad (4\text{-}5b)$$

for the bulk modes. The coefficients in Eq.(4-3) can written as

$$L_1 = \frac{1}{2\varepsilon_1 \omega^2 \mu} (A_1 - \frac{ik_x}{\alpha_1} A_2) = \frac{A_m k_x \alpha}{\mu \varepsilon_1 \omega^2} (\chi_+ + \frac{k_x}{\alpha_1} \delta_+) \\ L_2 = -\frac{1}{2\varepsilon_1 \omega^2 \mu\alpha} (B_1 + \frac{ik_x}{\alpha_1} B_2) = \frac{A_m \alpha^* k_x}{\mu \varepsilon_1 \omega^2} (\chi_- + \frac{k_x}{\alpha_1} \delta_-) \quad (4\text{-}6a)$$

$$L_3 = \frac{1}{8\varepsilon_1 \omega^2 \mu} (C_1 - \frac{3ik_x}{\alpha_1} C_2) = \frac{A_m k_x}{4\mu \varepsilon_1 \omega^2} (\chi_- - \frac{3k_x}{\alpha_1} \delta_-) \\ L_4 = \frac{1}{8\varepsilon_1 \omega^2 \mu} (D_1 + \frac{3ik_x}{\alpha_1} D_2) = -\frac{A_m \alpha |\alpha|^2 k_x}{4\mu \varepsilon_1 \omega^2} (\chi_+ - \frac{3k_x}{\alpha_1} \delta_+) \quad (4\text{-}6b)$$

$$L_3' = 3L_3 + \frac{i}{\alpha_1 k_x} C_2 = \frac{A_m}{4\mu \varepsilon_1 \omega^2} [3k_x \chi_- - \frac{\delta_-}{\alpha_1} (k_x^2 + 8\alpha_1^2)] \\ L_4' = -3L_4 + \frac{i}{\alpha_1 k_x} D_2 = \frac{A_m \alpha |\alpha|^2}{4\mu \varepsilon_1 \omega^2} [3k_x \chi_+ - \frac{\delta_+}{\alpha_1} (k_x^2 + 8\alpha_1^2)] \quad (4\text{-}6c)$$

$$S = L_1 + \frac{i}{\alpha_1 k_x} A_2 = \frac{A_m \alpha}{\mu \varepsilon_1 \omega^2}[k_x \chi_+ + \frac{\delta_+}{\alpha_1}(2\alpha_1^2 - k_x^2)]$$
$$T = k_x L_2 + \frac{i}{\alpha_1 \alpha k_x} B_2 = \frac{k_x A_m \alpha^*}{\mu \varepsilon_1 \omega^2}[k_x \chi_- + \frac{\delta_-}{\alpha_1}(2\alpha_1^2 - k_x^2)] \quad (4\text{-}6d)$$

2. If α_1 is imaginary, i.e. $\alpha_1 = i\lambda$, these coefficients should be changed into

$$A_1 = -2|\alpha|^2 k_x A_m \chi_+ , B_1 = 2\alpha k_x A_m \chi_- , C_1 = -2k_x \alpha^* A_m \chi_- , D_1 = 2k_x \alpha^2 A_m \chi_+ \quad (4\text{-}7a)$$

$$A_2 = -2ik_x |\alpha|^2 A_m \delta_+ , B_2 = -2ik_x \alpha A_m \delta_- , C_2 = 2ik_x \alpha^* A_m \delta_- , D_2 = 2ik_x \alpha^2 A_m \delta_+ \quad (4\text{-}7b)$$

$$L_1 = -\frac{A_m |\alpha|^2 k_x}{\varepsilon_1 \omega^2 \mu}(\chi_+ + \frac{k_x}{\alpha_1}\delta_+) , L_2 = -\frac{A_m k_x}{\varepsilon_1 \omega^2 \mu}(\chi_- + \frac{k_x}{\alpha_1}\delta_-) \quad (4\text{-}8a)$$

$$L_3 = -\frac{A_m \alpha^* k_x}{4\varepsilon_1 \omega^2 \mu}(\chi_- - \frac{3k_x}{\alpha_1}\delta_-) , L_4 = \frac{A_m \alpha^2 k_x}{4\varepsilon_1 \omega^2 \mu}(\chi_+ - \frac{3k_x}{\alpha_1}\delta_+) \quad (4\text{-}8b)$$

$$L_3' = -\frac{A_m \alpha^*}{4\varepsilon_1 \omega^2 \mu}[3k_x \chi_- - \frac{\delta_-}{\alpha_1}(k_x^2 + 8\alpha_1^2)] , L_4' = -\frac{A_m \alpha^2}{4\varepsilon_1 \omega^2 \mu}[3k_x \chi_+ - \frac{\delta_+}{\alpha_1}(k_x^2 + 8\alpha_1^2)] \quad (4\text{-}8c)$$

$$S = -\frac{A_m |\alpha|^2}{\varepsilon_1 \omega^2 \mu}[k_x \chi_+ + \frac{\delta_+}{\alpha_1}(2\alpha_1^2 - k_x^2)] , T = -\frac{A_m}{\varepsilon_1 \omega^2 \mu}[k_x \chi_- + \frac{\delta_-}{\alpha_1}(2\alpha_1^2 - k_x^2)] \quad (4\text{-}8d)$$

Note that all these coefficients contain implicitly the factor $\Delta = |A|^2 / 4\pi M_0^2$, so we say that they are of the second-order. For simplicity in the process of deriving dispersion equations, we introduce three second-order quantities,

$$\eta_1(y + nD) = (y + nD)\alpha_1 L_1 e^{\alpha_1 (y+nD)} + \alpha(y + nD)\alpha_1 L_2 e^{-\alpha_1 (y+nD)} + L_3 e^{3\alpha_1 (y+nD)} + L_4 e^{-3\alpha_1 (y+nD)} \quad (4\text{-}9a)$$

$$\eta_2(y + nD) = [(y + nD)\alpha_1 L_1' + S]e^{\alpha_1 (y+nD)} + \alpha[-(y + nD)\alpha_1 L_2 + T]e^{-\alpha_1 (y+nD)} + L_3' e^{3\alpha_1 (y+nD)} + L_4' e^{-3\alpha_1 (y+nD)} \quad (4\text{-}9b)$$

and

$$\theta(y + nD) = \frac{i\alpha_1}{\varepsilon_1 \omega^2 k}[A_2 e^{\alpha_1 (y+nD)} + B_2 e^{-\alpha_1 (y+nD)} + C_2 e^{3\alpha_1 (y+nD)} + D_2 e^{-3\alpha_1 (y+nD)}] \quad (4\text{-}9c)$$

Thus the nonlinear magnetic field can be rewritten as

$$\vec{H} = A_x\{[e^{\alpha_1 (y+nD)} + \alpha' e^{-\alpha_1 (y+nD)} + \eta_1(y + nD) f_n)]\vec{e}_x - \frac{ik}{\alpha_1}[e^{\alpha_1 (y+nD)} - \alpha' e^{-\alpha_1 (y+nD)} + \eta_2(y + nD) f_n]\vec{e}_y\} e^{-\beta nD} e^{i(kx - \omega t)} \quad (4\text{-}10a)$$

and the third-order magnetization is equal to

$$m_y^{(3)} = -\frac{ik}{\alpha_1} A_x \theta(y+nD) f_n e^{i(kx-\omega t)} e^{-n\beta D} \tag{4-10b}$$

The two formulae will be applied for solving the dispersion equations of the nonlinear surface and bulk polaritons from the boundary conditions satisfied by the wave fields.

Seeking the dispersion relations of AF polaritons should begin from the boundary conditions of the magnetic field H_x and magnetic induction field B_y continuous at the interfaces and surface ($y = -nD, -nD - d_1$ and 0). The results (4-3a) and (4-3b) related to the nth AF layer, as well as the solutions in the vacuum $\vec{H} = \vec{A}_0 e^{-\alpha_0 y} e^{i(kx-\omega t)}$ and in the nth NM layer $\vec{H} = [\vec{C} e^{\alpha_2 (y+jD+d_1)} + \vec{D} e^{-\alpha_2 (y+jD+d_1)}] e^{-\beta jD} e^{i(kx-\omega t)}$ will be used to determine the dispersion relations. In the following several paragraphs, we shall calculate the dispersion relations of the surface and bulk modes, respectively.

3. Bulk dispersion equation

For the bulk polaritons, there are 6 amplitude coefficients in the wave solutions, $A_x, \alpha', C_x, C_y, D_x$ and D_y . The magnetic induction .. in AF layers and $B_y = H_y$ in NM layers. The boundary conditions of B_y and H_x continuous at the interfaces ($y = -nD$ and $-nD - d_1$) imply four equations, and $\nabla \cdot \vec{H} = 0$ in a NM layer leads to two additional relations $C_y = -ikC_x / \alpha_2$ and $D_y = ikD_x / \alpha_2$. Thus we have

$$A_x[1 + \alpha' + \eta_1(0) f_n] = (C_x e^{-\alpha_2 d_2} + D_x e^{\alpha_2 d_2}) e^{iQD} \tag{4-11a}$$

$$\frac{A_x}{\alpha_1}[\mu(1 - \alpha' + \eta_2(0) f_n) + \theta(0) f_n] = \frac{1}{\alpha_2}(C_x e^{-\alpha_2 d_2} - D_x e^{\alpha_2 d_2}) e^{iQD} \tag{4-11b}$$

$$A_x[e^{-\alpha_1 d_1} + \alpha' e^{\alpha_1 d_1} + \eta_1(-d_1) f_n] = C_x + D_x \tag{4-11c}$$

$$\frac{A_x}{\alpha_1}[\mu(e^{-\alpha_1 d_1} - \alpha' e^{\alpha_1 d_1} + \eta_2(-d_1) f_n) + \theta(-d_1) f_n] = \frac{1}{\alpha_2}(C_x - D_x) \tag{4-11d}$$

From these four equations, we find the dispersion relation of the nonlinear bulk polaritons,

$$\cos(QD) - \cosh(\alpha_1 d_1)\cosh(\alpha_2 d_2) - \frac{\alpha_1^2 + \alpha_2^2 \mu^2}{2\alpha_1 \alpha_2 \mu} \sinh(\alpha_1 d_1)\sinh(\alpha_2 d_2) = \frac{1}{4} N \tag{4-12}$$

with the nonlinear factor N described by

$$\begin{aligned} N = {} & \eta_1(0)[-e^{-iQD} + \cosh(\alpha_2 d_2) e^{\alpha_1 d_1} + (\alpha_1 / \mu\alpha_2)\sinh(\alpha_2 d_2) e^{\alpha_1 d_1}] \\ & + [\eta_2(0) + \theta(0)/\mu][-e^{-iQD} + \cosh(\alpha_2 d_2) e^{\alpha_1 d_1} + (\alpha_2 \mu / \alpha_1)\sinh(\alpha_2 d_2) e^{\alpha_1 d_1}] \\ & + \eta_1(-d_1)[-e^{\alpha_1 d_1} e^{iQD} + \cosh(\alpha_2 d_2) - (\alpha_1 / \mu\alpha_2)\sinh(\alpha_2 d_2)] + [\eta_2(-d_1) \\ & + \theta(-d_1)/\mu][-e^{\alpha_1 d_1} e^{iQD} + \cosh(\alpha_2 d_2) - (\alpha_2 \mu / \alpha_1)\sinh(\alpha_2 d_2)] \end{aligned} \tag{4-13}$$

Due to the nonlinear interaction, the nonlinear term $N/4$ appears in the dispersion equation of the polaritons and is directly proportional to Δ. This term is a second-order quantity and makes a small correct to the dispersion properties of the linear bulk polaritons. Generally speaking, this nonlinear dispersion equation is a complex relation. However in some special circumstances it may be a real one. Here we illustrate it with an example. If $Q=0$, the bulk wave moves along the x-axis and the dispersion equation is a real equation for real α_1. For such a dispersion equation, ω has a real solution, otherwise the solution of ω is a complex number with the real part ω^{NL}, so-called the nonlinear mode frequency, and the imaginary part $\Delta\tau$, the attenuation or gain coefficient. In addition, it is very interesting that the unreciprocity of the bulk modes, $\omega(\vec{k})\neq\omega(-\vec{k})$ with $\vec{k}=(k,Q,0)$, is seen, due to the existence of $\exp(-iQD)$ in the nonlinear term $N/4$ as a function of QD with the period 2π.

4. Surface dispersion relations

For the surface modes, note $f_n=\exp(-2n\beta D)$ and take the transformation $iQ\to\beta$ in equations (4-10), we can find

$$\cosh(\beta D)-\cosh(\alpha_1 d_1)\cosh(\alpha_2 d_2)-\frac{\alpha_1^2+\alpha_2^2\mu^2}{2\alpha_1\alpha_2\mu}\sinh(\alpha_1 d_1)\sinh(\alpha_2 d_2)=\frac{1}{4}N'e^{-2\beta nD} \quad (4\text{-}14)$$

in which N' can be obtained directly from Eq.(4-13) with the same transformation. This nonlinear term is directly proportional to the multiply of Δ and $\exp(-2n\beta D)$, so in the same condition the nonlinearity makes larger contribution to the bulk modes than the surface modes. We can use the linear expression of $\exp(-\beta D)$ to reduce the nonlinear term on the right-hand of Eq.(4-14), but have to derive its nonlinear expression to describe $\cosh(\beta D)$ on the left-hand, since its nonlinear part may has the same numerical order as that of $N'\exp(-2n\beta D)/4$. So we need another equation to determine it. Applying the boundary conditions at the surface, $y=0$ and $n=0$, we can find

$$\alpha_1[1+\alpha'+\eta_1(0)]=-\{\mu[1-\alpha'+\eta_2(0)]+\theta(0)\}\alpha_0 \quad (4\text{-}15)$$

Combining this with Eqs.(4-11a-c), the equation determining β is found,

$$\begin{aligned}e^{\beta D}=\{&(1+\alpha'+\eta_1(0)f_n)\cosh(\alpha_2 \mathrm{d}_2)+\alpha_2\mu[1-\alpha'+(\eta_2(0)+\theta(0)/\mu)f_{\mathrm{n}}]\\ &\sinh(\alpha_2 \mathrm{d}_2)/\alpha_1\}/[\mathrm{e}^{-\alpha_1 d_1}+\alpha' e^{\alpha_1 d_1}+\eta(-d_1)f_n]\end{aligned} \quad (4\text{-}16)$$

with

$$\alpha'=\frac{1}{\alpha_0\mu-\alpha_1}\{\alpha_0\mu+\alpha_1+\alpha_1\eta_1(0)+\alpha_0[\mu\eta_2(0)+\theta(0)]\} \quad (4\text{-}17)$$

$f_n=\exp(-2n\beta D)$ in Eq.(4-16) also can be considered as an linear quantity since it always appears in the multiply of it and Δ. We also should note that there is a series of nonlinear surface eigen-modes as *n* can be any integer value equal to or larger than 1. Actually the nonlinear contribution decreases rapidly as *n* is increased, so only for small *n*, the nonlinear

effect is important. In addition, increasing Δ and decreasing n have a similar effect in numerical calculation.

Because the nonlinear terms in Eqs.(4-12) and (4-14) all contain $\chi^{(3)}_{ijkl}$ directly proportional to $1/(\omega_r^2-\omega^2)^4$, the nonlinear effects may be too strong for us to use the third-order approximation for the nonlinear magnetization when ω is near to ω_r. In this situation we will take a smaller value of Δ to assure of the availability of this approximation.

We take the FeF_2/ZnF_2 superlattice as an example for numerical calculations, the physical parameters of FeF_2 are given in table 1. While the relative dielectric constant of ZnF_2 are $\varepsilon_2 = 8.0$. We apply the SL period $D = 1.9\times10^{-2} cm$, and take $n = 1$ for the surface modes. The nonlinear factor $\Delta = \left|A/(4\pi M_0)\right|^2$ is the relative strength of the wave field. The nonlinear shift in frequency is defined as $\Delta\omega = (\omega^{NL}-\omega)/\omega_r$, where the nonlinear frequency ω^{NL} and attenuation or gain coefficient $\Delta\tau$ as the real and imagine parts of the frequency solution from the nonlinear dispersion equations both are solved numerically. ω is determined by the linear dispersion relations.

	H_a	H_e	$4\pi M_0$	ε	γ
FeF_2	$197kG$	$533kG$	$7.04\ kG$	5.5	1.97×10^{10} rad s^{-1} kG
MnF_2	$7.87kG$	$550kG$	$5.65\ kG$	5.5	1.97×10^{10} rad s^{-1} kG

Table 1. Physical parameters for FeF_2 and MnF_2.

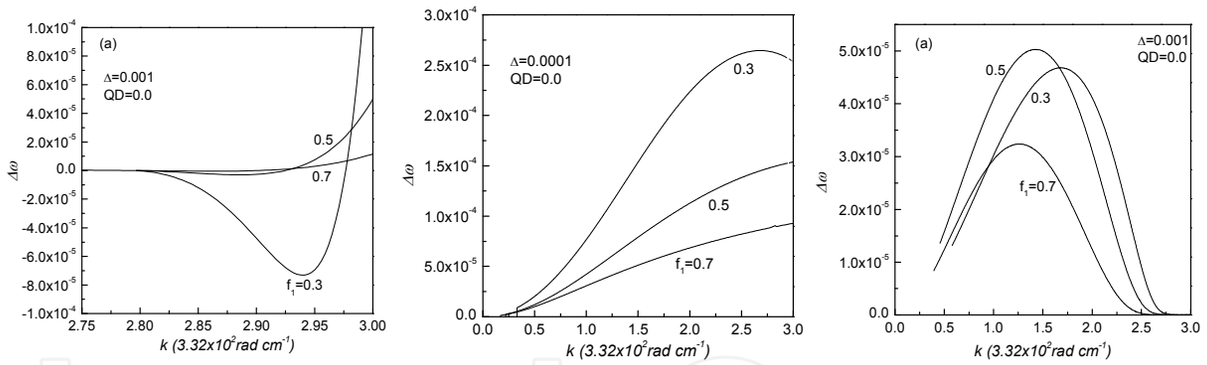


Fig. 6. Nonlinear shift in frequency(a) in the bottom band(b) in the middle band and (c) in the top band. After Wang & Li 2005.

We illustrate the nonlinear shift in frequency as function of the component of wave vector k in the three bulk-mode bands seperately in Fig. 6. is offered to illustrate the bottom band. (a) and ((b) show for $Q = 0$ and π/D respectively. As shown in Fig.6(a), in the bottom band, when $f_1 = 0.3 and 0.5$, the nonlinear shift is downward or negative in the region of smaller k, but becomes positive from negative with the increase of k. For a SL with thicker AF layers, for example $f_1 = 0.7$, the shift is always positive. For the top bulk band, $\Delta\omega$ always is positive and possesses its maximum. In the middle band, Fig. 6(b) shows the positive frequency shift that increases basically with k. In terms of the shape of a band, the second-order derivative of linear frequency with respect to k, $\partial^2\omega/\partial k^2$ for a mode in it can be roughly estimated to be positive or negative. According to the Lighthill criterion $\Delta\omega\cdot\partial^2\omega/\partial k^2 \le 0$ for the existence of solitons(Lighthill,1965). One confirms from the figures

that $\partial^2\omega/\partial k^2>0$ for modes in the top band, $\partial^2\omega/\partial k^2<0$ in the bottom band, but $\partial^2\omega/\partial k^2<0$ or $\partial^2\omega/\partial k^2>0$ in the middle band, depending on k. The soliton solution may be found since the Lighthill criterion can be fulfilled in the two bands. In the middle bulk band, the mode attenuation is vanishing, the nonlinearity is very evident and the nonlinear shift is positive.

We examine the surface magnetic polariton in the case of nonlinearity, which is shown in Fig.7. Similar to those in the middle bulk band, the surface-mode frequency also is very closed to ω_r, as a result, the nonlinear effect also is stronger. The attenuation $\Delta\tau=0$ as the dispersion equations are real. The shift $\Delta\omega$ is negative for $f_1\geq 0.3$, but positive for f_1=0.1. For f_1=0.2, it is positive and increases with k in the range of small k, but negative in the range of large k and its absolute value decreases as k is increased. Although there can be a series of surface eigen-modes in the nonlinear situation, the obvious nonlinear effect can be seen only for $n=1$, so that we present only the corresponding mode. One should note that the Lighthill criterion is satisfied for f_1=0.1 and 0.2, as a result, the surface soliton may form from the surface magnetic polariton.

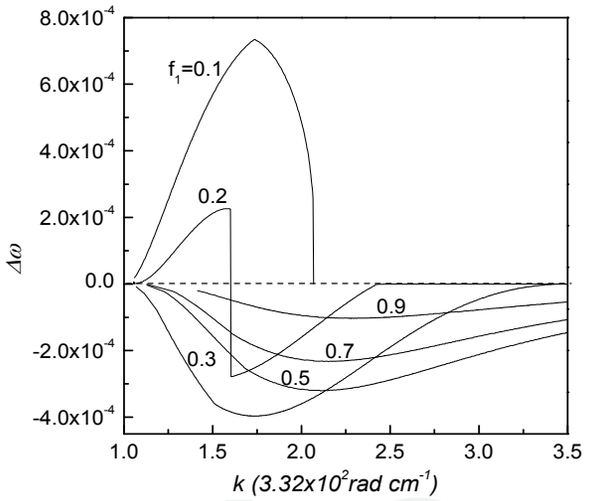


Fig. 7. Nonlinear frequency shift $\Delta\omega$ of the surface mode versus k for $\Delta=1.0\times10^{-4}$ and various values of f_1. After Wang & Li 2005.

4.2 Nonlinear infrared ransmission through and reflection off AF films

Finally, we discuss nonlinear transmission through the AF film. We assume that the media above and below the nonlinear AF film are both linear, but the film is nonlinear. Our geometry is shown in Fig. 8, where the anisotropy axis (the z axis) is parallel to the film surfaces and normal to the incident plane (the x–y plane). A linearly polarized radiation (TE wave) is obliquely incident on the upper surface.

Because we have known the nonlinear wave solution in the AF film and those above and below the film, to solve nonlinear transmission and reflection is a simple algebraical precess. Thus we directly present the finall results, the nonlinear refection and transmission coefficients

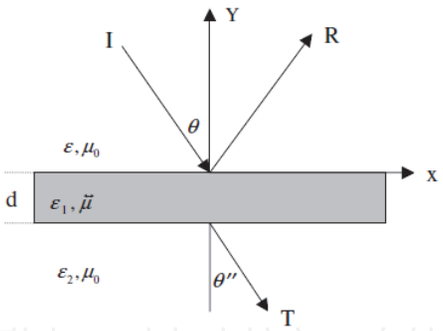


Fig. 8. Geometry and coordinate system for nonlinear reflection and transmission of an AF film with thickness d. R the reflection off and T the transmission through the film.

$$r=\frac{E_R}{E_I}=\frac{\mu_1\left(k_y-k_y''\right)\cos k_y'd+\left(k_y'-\mu_1^2k_y\,k_y''/k_y'\right)i\sin k_y'd-NL_+}{\mu_1\left(k_y+k_y''\right)\cos k_y'd-\left(k_y'+\mu_1^2k_y\,k_y''/k_y'\right)i\sin k_y'd-NL_-} \tag{4-18a}$$

$$t=\frac{E_T}{E_I}=\frac{\mu_1k_y\left[2-\eta_1(0)-\theta(0)\right]e^{ik_y''d}}{\mu_1\left(k_y+k_y''\right)\cos k_y'd-\left(k_y'+\mu_1^2k_y\,k_y''/k_y'\right)i\sin k_y'd-NL_-} \tag{4-18b}$$

in which the nonlinear terms $NL_\pm$ are shown with

$$\begin{aligned}NL_\pm=&\frac{1}{2}\{\pm\left(k_y-\mu_1k_y''\right)\left[\cos\left(k_yd\right)\pm i\mu_1k_y'\sin\left(k_yd\right)/k_y\right]\eta_1(-d)e^{ik_yd}\\&+\left(\mp k_y+\mu_1k_y'\right)\left[\cos\left(k_yd\right)-i\mu_1k_y''\sin\left(k_yd\right)/k_y\right]\eta_1(0)\\&+\left(k_y-\mu_1k_y''\right)\left[\pm i\sin\left(k_yd\right)+\mu_1k_y'\cos\left(k_yd\right)/k_y\right]e^{ik_yd}\theta(-d)\\&+\left(\pm k_y-\mu_1k_y'\right)\left[i\sin\left(k_yd\right)-\mu_1k''\cos\left(k_yd\right)/k_y\right]\theta(0)\}\end{aligned} \tag{4-19}$$

Finally the reflectivity and transmissivity are defined as $R=|r|^2$ and $T=|t|^2$ (Klingshirn, C. F. ,1997). Here we should discuss a special situation. In the situation $(k_x=0)$, from the expressions of L_1 to L_4 and L_1' to L_4', we find $\eta_1(y)=\theta(y)=0$. It is quite obvious that one finds no nonlinear effects on the reflection and transmission in the case of normal incidence. For $\varepsilon>\varepsilon_2$, k_y'' becomes imaginary as the incident angle θ exceeds a special value, then the transmission vanishes. The nonlinear effect can be seen only from the reflection. Due to the complicated expressions for the reflection and transmission coefficients, more properties of R and T can be obtained only by numerical calculation of Eq. (4-18).

We take a FeF_2 film as an example for numerical calculations. with the physical parameters given in Table 1. The film thickness is fixed at $d=30.0\mu m$ and the incident wave intensity $S_I=\sqrt{\varepsilon_0/\mu_0}\,E_I^2/2$, implicitly included in the nonlinear coefficients, is fixed at $S_I=4.7MWcm^{-2}$, corresponding to a magnetic amplitude of 16G in the incident wave. In the figures for numerical results, we use dotted lines to show linear results and solid lines to show nonlinear results. We shall discuss transmission and reflection of the AF film put in a vacuum. The transmission and reflection versus frequency ω are illustrated in Fig.9 for the incident angle $\theta=30°$ and are shown in Fig.10 versus incident angle for $\omega/2\pi C=52.8cm^{-1}$

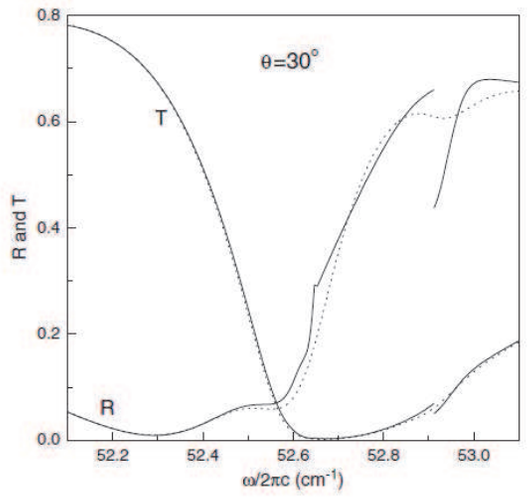


Fig. 9. Reflectivity and transmissivity versus frequency for a fixed angle of incidence of $30°$. After Bai, et. al. 2007.

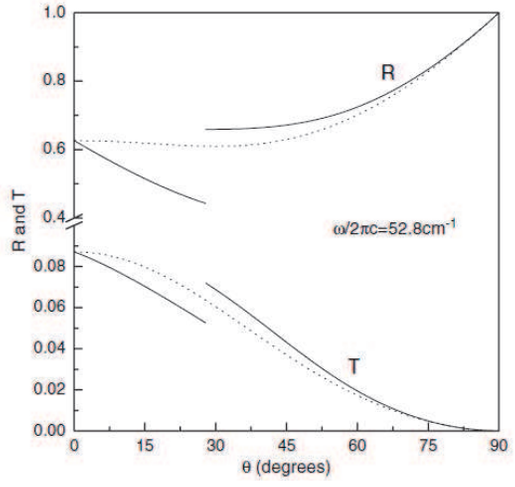


Fig. 10. Reflectivity and transmissivity versus angle of incidence for the frequency fixed at 52.8 cm^{-1}. After Bai, et. al. 2007.

First, the nonlinear modification is more evident in reflection for frequencies higher than ω_r. We see a very obvious discontinuity on the nonlinear R and T curves at $\omega/2\pi C = 52.9cm^{-1}$, corresponding to the smallest value of μ_1 whose real part changes in sign as the frequency moves cross this point. This causes the jump and obvious nonlinear modification, as the wave magnetic field is intense in the vicinity of this point. Secondly, R and T versus θ for a fixed frequency are shown in Fig.10. Here the discontinuity is also seen since the magnetic amplitude and the nonlinear terms vary with the wave vector $\bar{k}'$. It is more interesting that when the incident angle $\theta \le 27.5°$ the reflection and transmission are both lower than the linear ones, implying that the absorption is reinforced. However, in the range of $\theta \ge 27.5°$ they both are higher than the linear ones, and as a result the absorption is evidently restrained. The nonlinear influence disappears for normal incidence. we see the discontinuities on the reflection and transmission curves and the nonlinear effect is very obvious in the regions near to the jump points. The discontinuities are related to the

bi-stable states. The nonlinear interaction also play an important role in decreasing or increasing the absorption in the AF film.

5. Second harmonic generation in antiferromagnetic films

In this section, the most fundamental nonlinear effect, second harmonic generation (SHG) of an AF film between two dielectrics (Zhou & Wang, 2008) and in one-dimensional photonic crystals (Zhou, et. al., 2009) have been analyzed based on the second-harmonic tensor elements obtained in section 2. We know from the expression of SH magnetization that if $H_0 = 0$ the SH magnetization is vanishing, as a result the SHG is absent. So the external magnetic field is necessary for the SHG. We take such an AF structure as example to describe the SHG theory, where the AF film is put two different dielectrics. In the coordinate system selected in Fig.11, the AF anisotropic field and dc magnetic field both parallel to the z-axis and the x-y plane as the incident plane. *I* is the incident wave, *R* the reflection wave and *T* the transmission wave, related to incident angle θ, reflection angle $-\theta$ and transmission angle θ', respectively. If a subscript *s* is added to the above quantities, they represent the corresponding quantities of second harmonic (SH) waves. The pump wave in the film is not indicated in this figure. The dielectric constants and magnetic permeabilities are shown in corresponding spaces.

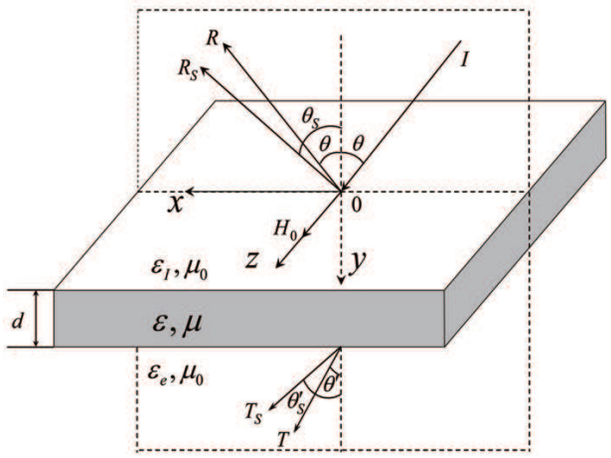


Fig. 11. Geometry and coordinate system.

Although we have obtained all elements of the SH susceptibility in section 2, but only two will be used in this geometry. It is because that a plane EMW of incidence can be decomposed into two waves, or a TE wave with the electric field normal to the incident plane and a TM wave with the magnetic field transverse to this plane. Due to no coupling between magnetic moments in the film and the TM wave (Lim, 2002, 2006; Wang & Li, 2005; Bai, et. al., 2007), the incident TM wave does not excite the linear and SH magnetizations, so can be ignored. Thus we take the TE wave as the incident wave *I* which produces the TE pump wave $\vec{H} = (H_x, H_y, 0)$ in the film. In this case, only one component of the SH magnetization can be found easily

$$m_z^{(2)}(\omega_s) = \chi_{xx}^{(2)}(\omega_s)(H_x H_x + H_y H_y) \tag{5-1}$$

with $\omega_s = 2\omega$ and the susceptibility elements

$$\chi_{xx}^{(2)}(2\omega) = \chi_{yy}^{(2)}(2\omega) = \frac{i\omega_m[\chi_{xx}^{(1)}\omega_0(\omega_r'^2 + \omega^2 - \omega_0^2) - \chi_{xy}^{(1)}\omega(\omega_r'^2 - \omega^2 + \omega_0^2)]}{M_0[\omega_r'^2 - (\omega - \omega_0)^2][\omega_r'^2 - (\omega + \omega_0)^2]} \tag{5-2}$$

The SHG magnetization arises as a source term in the harmonic wave equation and is excited by the pump wave, and in turn the pumping wave is induced by the incident wave. When the energy-flux density of the excited SH wave is much less than that of the incident wave, the assumption that the depletion of pump waves can be neglected(Shen, 1984) is commonly accepted. This assumption allows us to solve the pump wave in the film within the linear electromagnetic theory or with the linear optical method.

Based on the above assumption, to solve the pump wave is a linear problem. The method is well-known and just one usual optical process, so we give a simpler description for solving the pump wave in the film. Because the pump wave is a TE wave, we take its electric field to be

$$\vec{E} = \vec{e}_z[A_+ \exp(ik_y y) + A_- \exp(-ik_y y)]\exp(ik_x x - i\omega t) \tag{5-3}$$

where A_+ and A_- show the amplitudes of the forward and backward waves in the film, respectively. The electric fields above and below the film are

$$\vec{E}_a = \vec{e}_z[E_0 \exp(ik_{0y} y) + R_0 \exp(-ik_{0y} y)]\exp(ik_{0x} x - i\omega t) \tag{5-4}$$

$$\vec{E}_b = \vec{e}_z T_0 \exp(ik'_{0y} y)\exp(ik'_{0x} x - i\omega t) \tag{5-5}$$

The corresponding magnetic fields in different spaces are written as

$$\begin{aligned}\vec{H} = \frac{\exp(ik_x x - i\omega t)}{\omega\mu_0\mu_v}\{&\vec{e}_x k_y[(1+\delta)A_+ \exp(ik_y y) + (1-\delta)A_- \exp(-ik_y y)] \\ &+ \vec{e}_y k_x[(\delta'-1)A_+ \exp(ik_y y) - (\delta'+1)A_- \exp(-ik_y y)]\}\end{aligned} \tag{5-6a}$$

$$\begin{aligned}\vec{H}_a = \frac{\exp(ik_{0x} x - i\omega t)}{\omega\mu_0}\{&\vec{e}_x k_{0y}[E_0 \exp(ik_{0y} y) - R_0 \exp(-ik_{0y} y)] \\ &- \vec{e}_y k_{0x}[E_0 \exp(ik_{0y} y) + R_0 \exp(-ik_{0y} y)]\end{aligned} \tag{5-6b}$$

$$\vec{H}_b = \frac{T_0}{\omega\mu_0}(\vec{e}_x k'_{0y} - \vec{e}_y k'_{0x})\exp(ik'_{0y} y + ik'_{0x} x - i\omega t) \tag{5-6c}$$

where is $\mu_v = (\mu_1^2 - \mu_2^2)/\mu_1^2$ and μ_0 the vacuum magnetic permeability. $k_{0y} = k_0 \cos\theta$ and $k_0 = \varepsilon_1^{1/2}\omega/c$ is the wave number in the above space, and $k'_{0y} = k'_0 \cos\theta'$ and $k'_0 = \varepsilon_2^{1/2}\omega/c$ is the wave number below the film, but $k_y = [\varepsilon\mu_v(\omega/c)^2 - k_x^2]^{1/2}$. Here c is the vacuum velocity of light. $\delta = k_x\mu_2 / \mu_1 k_y$ and $\delta' = \mu_2 k_y / \mu_1 k_x$. The boundary conditions of the fields at the surfaces first require that $k_{0x} = k'_{0x} = k_x = k_0 \sin\theta$, and these wave-number components should be real since we assume that dielectric constants and magnetic permeabilities in nonmagnetic media all are real values. In addition, using the boundary conditions, we also find the pump-field amplitudes $A_\pm = E_0 f_\pm$ with E_0 the electric amplitude of I, and

$$f_{\pm}=\frac{[\Delta(1\mp\delta)\pm 1]\exp(-ik_y d)}{\cos k_y d(\Delta+\Delta')+i\delta(\Delta-\Delta')\sin k_y d-i[1+\Delta'\Delta(1-\delta^2)]\sin k_y d} \tag{5-7}$$

where d is the film thickness, $\Delta=k_y/\mu_\nu k_{0y}$ and $\Delta'=k_y/\mu_\nu k'_{0y}$. The wave amplitudes R_0 and T_0 of R and T are not necessary for seeking the SHG, so they are given up here.To solve the output amplitudes of SHG, R_s and T_s, we should look for the solution of the SH wave equation in the film. In fact, there are three component equations, but only one contains a source term and this equation is

$$-(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2})H_{sz}(\omega_s)-\varepsilon(\omega_s/c)^2 H_{sz}(\omega_s)=\varepsilon(\omega_s/c)^2 m_z^{(2)}(\omega_s) \tag{5-8}$$

The other two are homogeneous and do not contain the field component $H_{sz}(\omega_s)$. In addition, the other SH components cannot emerge voluntarily without source terms, so it is evident that the SH wave is a TM wave. Because the SH magnetization and pump field in the film both have been given, to find the solution of equation (5-8) is easy. Let

$$\begin{aligned}H_{sz}(\omega_s)=[&A_s\exp(ik_{sy}y)+B_s\exp(-ik_{sy}y)+a\exp(2ik_y y)\\&+b\exp(-2ik_y y)+c]\exp(2ik_x x-i\omega_s t)\end{aligned} \tag{5-9}$$

with $k_{sy}=[\varepsilon(\omega_s/c)^2-4k_x^2]^{1/2}$. Substituting SH solution (5-9), expression (5-1) and solution (5-6a) into equation (5-8), we find the nonlinear amplitudes

$$a=E_0^2\frac{\varepsilon_0\chi_{zxx}^{(2)}(\omega_s)f_+^2}{\mu_0(\mu_\nu-1)(\omega\mu_\nu/c)^2}[k_y^2(1+\delta)^2+k_x^2(1-\delta')^2] \tag{5-10a}$$

$$b=E_0^2\frac{\varepsilon_0\chi_{zxx}^{(2)}(\omega_s)f_-^2}{\mu_0(\mu_\nu-1)(\omega\mu_\nu/c)^2}[k_y^2(1-\delta)^2+k_x^2(1+\delta')^2] \tag{5-10b}$$

$$c=-E_0^2\frac{2\varepsilon_0\varepsilon(\omega_s/c)^2\chi_{zxx}^{(2)}(\omega_s)f_+f_-}{\mu_0[4k_x^2-\varepsilon(\omega_s/c)^2](\omega\mu_\nu/c)^2}[k_y^2(1-\delta^2)+k_x^2(\delta'^2-1)] \tag{5-10c}$$

Solution (5-9) shows that the SH wave in the film also propagates in the incident plane and it will radiate out from the film. We use

$$H_{sz}^a=R_s\exp[i(k_{sx}x-k_s y-\omega_s t)] \tag{5-11a}$$

to indicate the magnetic field of SH wave generated above the film and

$$H_{sz}^b=T_s\exp[i(k_{sx}x+k'_s y-\omega_s t) \tag{5-11b}$$

to represent the SH field below, with k_s and k'_s determined by $k_s^2+k_{sx}^2=\varepsilon_1(\omega_s/c)^2$ and $k_s'^2+k_{sx}^2=\varepsilon_2(\omega_s/c)^2$. The SH electric field in different spaces are found from to be

$$\vec{E}_s^a=\frac{R_s\exp[i(2k_x x-k_s y-\omega_s t)]}{\omega_s\varepsilon_0\varepsilon_1}[k_s\vec{e}_x+k_{sx}\vec{e}_y] \tag{5-12a}$$

$$\vec{E}_s = -\frac{\exp[i(2k_x x - \omega_s t)]}{\omega_s \varepsilon_0 \varepsilon}\{\vec{e}_x[k_{sy}(A_s \exp(ik_{sy}y) - B_s \exp(-ik_{sy}y) + 2k_y a \exp(2ik_y y) - 2k_y b \exp(-2ik_y y)] - 2k_x \vec{e}_y[a \exp(2ik_y y) + b \exp(-2ik_y y) + c]\} \quad (5\text{-}12b)$$

$$\vec{E}_s^b = \frac{T_s}{\omega_s \varepsilon_0 \varepsilon_2}(-k_s' \vec{e}_x + k_{sx} \vec{e}_y) \exp[i(2k_x x + k_s' y - \omega_s t)] \quad (5\text{-}12c)$$

Considering the boundary conditions of these fields continuous at the surfaces, there must be $k_{sx} = k_{sx}' = 2k_x$ and the these wave-number components all are real quantities, meaning the propagation angles of the SH outputs from the film

$$\theta_s = -\theta \quad (5\text{-}13a)$$

$$\theta_s' = arc \sin(\sqrt{\varepsilon_1 / \varepsilon_2} \sin\theta) \quad (5\text{-}13b)$$

It is proven that the SH wave outputs R_s and T_s have the same propagation direction as reflection wave *R* and transmission wave *T*, respectively.

Finally we solve the amplitudes of the output SH wave. The continuity conditions of H_{sz} and E_{sx} at the interfaces lead to

$$R_s = A_s + B_s + a + b + c \quad (5\text{-}14a)$$

$$R_s = \frac{\varepsilon_1}{\varepsilon k_s}[k_{sy}(-A_s + B_s) + 2k_y(-a + b)] \quad (5\text{-}14b)$$

$$T_s \exp(ik_s' d) = A_s \exp(ik_{sy} d) + B_s \exp(-ik_{sy} d) + a \exp(2ik_y d) + b \exp(-2ik_y d) + c \quad (5\text{-}14c)$$

$$T_s \exp(ik_s' d) = \frac{\varepsilon_2}{\varepsilon k_s'}\{k_{sy}[A_s \exp(ik_{sy} d) - B_s \exp(-ik_{sy} d)] + 2k_y[a \exp(2ik_y d) - b \exp(-2ik_y d)]\} \quad (5\text{-}14d)$$

After eliminating A_s and B_s from the above equations, we find the magnetic field-amplitudes of the output SH waves,

$$R_s = \frac{1}{S}\{[(\Delta_2 - \Delta_0)\cos k_{sy} d + i(\Delta_2 \Delta_0 - 1)\sin k_{sy} d + \exp(2ik_y d)(-\Delta_2 + \Delta_0)]a + [(\Delta_2 + \Delta_0)\cos k_{sy} d - i(\Delta_2 \Delta_0 + 1)\sin k_{sy} d - (\Delta_2 + \Delta_0)\exp(-2ik_y d)]b + [\Delta_2(\cos k_s d - 1) - i \sin k_s d]c\} \quad (5\text{-}15a)$$

$$T_s = \frac{\exp(-ik_s' d)}{S}\{[(\Delta_1 + \Delta_0)(e^{2ik_y d}\cos k_{sy} d - 1) - i(1 + \Delta_0 \Delta_1)\exp(2ik_y d) \sin k_{sy} d]a + [(\Delta_1 - \Delta_0)(\exp(-2ik_y d)\cos k_{sy} d - 1) + i(\Delta_0 \Delta_1 - 1) \exp(-2ik_y d)\sin k_{sy} d]b + [\Delta_1(\cos k_{sy} d - 1) - i \sin k_{sy} d]c\} \quad (5\text{-}15b)$$

where

$$S = [(\Delta_2 + \Delta_1)\cos k_{sy}d - i(1 + \Delta_2\Delta_1)\sin k_{sy}d] \tag{5-15c}$$

$\Delta_0 = 2k_y / k_{sy}$, $\Delta_1 = \varepsilon k_s / k_{sy}\varepsilon_1$ and $\Delta_2 = k'_s\varepsilon / (k_{sy}\varepsilon_2)$. We see from the expressions of *a*, *b* and *c* that SH amplitudes R_s and T_s are directly proportional to E_0^2, the square of electric amplitude of incidence wave. According to the definition of electromagnetic energy-flux density, $S_I = (\varepsilon_0\varepsilon_1 / \mu_0)^{1/2}|E_0|^2 / 2$ is the incident density, but the SH output densities are expressed as $S_R = (\mu_0 / \varepsilon_0\varepsilon_1)^{1/2}|R_s|^2 / 2$ and $S_T = (\mu_0 / \varepsilon_0\varepsilon_2)^{1/2}|T_s|^2 / 2$. We can conclude that the output densities are directly proportional to the square of the input (incident) density, or say the conversion efficiency $\alpha = S_{R,T} / S_I$ is directly proportional to the input density. For a fixed incident density, if the SH outputs are intense, the conversion efficiency must be high. Then, we are going to seek for the cases or conditions in which the SH outputs are intense.

The numerical calculations are based on three examples, a single MnF_2 film, SiO_2/MnF_2/air and ZnF_2/MnF_2/air, in which the MnF_2 film is antiferromagnetic. The relative dielectric constants are 1.0 for air, 2.3 for SiO_2 and 8.0 for ZnF_2. The relative magnetic permeabilities of these media are 1.0. There are two resonance frequencies in the dc field of $1.0kG$, $\omega_1/2\pi c = 9.76cm^{-1}$ and $\omega_2/2\pi c = 9.83cm^{-1}$. We take the AF damping coefficient $\tau = 0.002$ and the film thickness $d = 255\mu m$. The incident density is fixed at $S_I = 1.0kW / cm^2$, which is much less than that in the previous papers (Almeida & Mills, 1987; Kahn, et. al., 1988; Costa, et. al., 1993; Wang & Li, 2005; Bai, et. al., 2007).

We first illustrate the output densities of a single film versus frequency ω and incident angle θ with Fig.12 (a) for S_R and (b) for S_T. Evidently in terms of their respective maxima, S_R is weaker than S_T by about ten times. Their maxima both are situated at the second resonant frequency ω_2 and correspond to the situation of normal incidence. The figure of S_R is more complicated than that of S_T since additional weaker peaks of S_R are seen at large incident angles.

Next we discuss the SH outputs of SiO_2/MnF_2/air shown in Fig.13. Incident wave *I* and reflective wave *R* are in the SiO_2 medium and transmission wave *T* in air. The maximum peak of S_R is between the two resonant frequencies and in the region of $\theta > \theta'_c = 41.3^o$. For the given parameters, this angle just satisfies $\sin\theta_c = \sqrt{\varepsilon_2 / \varepsilon_1}$ and is related to $k'_{0y} = 0$, so it can be called a critical angle. When $\theta > \theta'_c$, k'_{0y} is an imaginary number and transmission *T* vanishes. For $\theta < \theta'_c$, S_R is very weak and numerically similar to that of the single film. However, the maximum of S_T is about four times as large as that of S_R, and S_T decreases rapidly as the incident angle or frequency moves away from θ'_c or the resonant frequency region. We find that the maxima of S_R and S_T are in intensity higher than those shown in Fig.12 by about 40 and 13 times, respectively.

Finally we discuss the SH outputs of ZnF_2/MnF_2/air, with the dielectric constant of ZnF_2 larger than that of SiO_2. The spectrum of S_R is the most complicated and interesting, as shown in Fig.14 (a). First we see two special angles of incidence. The first angle has the same definition as θ'_c in the last paragraph and is equal to 20.1o. The second defined as θ_c corresponds to $k_y = 0$ and is equal to 55^o. For $\theta > \theta_c$, k_y becomes an imaginary number

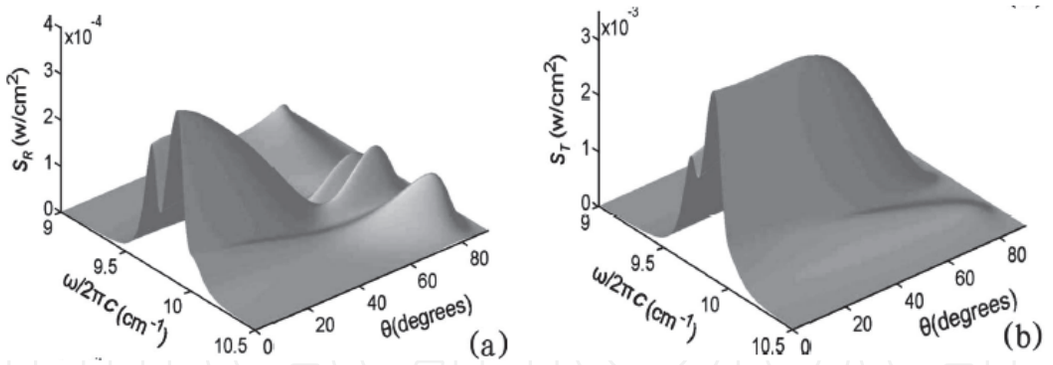


Fig. 12. SH outputs of a single AF film (MnF_2 film), S_R and S_T versus the incident angle and frequency. After Zhou & Wang, 2008.

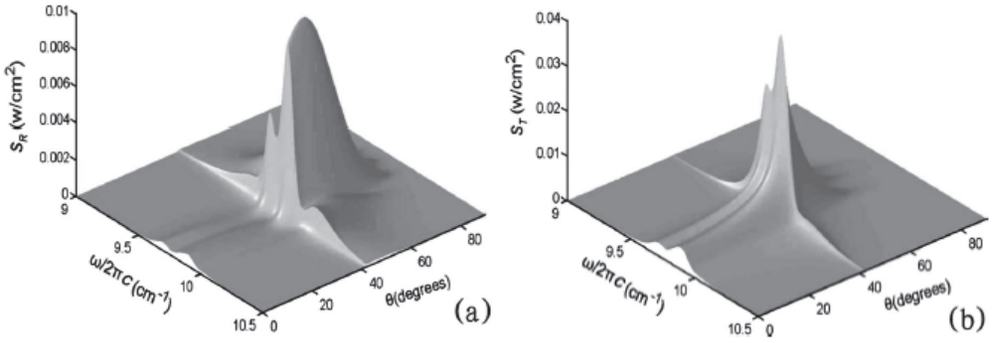


Fig. 13. SH outputs of SiO_2/MnF_2/air, S_R and S_T versus the incident angle and frequency. After Zhou & Wang, 2008.

and the incident wave *I* is completely reflected, so the SH wave is not excited. On this point, Fig.13(a) is completely different from Fig.12(a). More peaks of S_R appear between the two critical angles, but the highest peak stands between the two resonant frequencies and is near to θ_c. Outside of the region between θ_c and θ_c', we almost cannot see S_R. For S_T, the pattern is more simple, as shown in Fig.14 (b). Only one main peak is seen clearly, which arises at θ_c' and occupies a wider frequency range. Different from Fig.13, the maxima in Fig.14(a) and Fig.14(b) are about equal. Comparing Fig.14 with Fig.12, we find that the maximums of S_R and S_T are larger than those shown in Fig.12 by about 240 times and 20 times, respectively.

For the SH output peaks in Fig.13 and Fig.14, we present the explanations as follows. The pump wave in the film is composed of two parts, the forward and backward waves corresponding to the signs + and − in Eq.(5-3), respectively. The transmission (*T*) vanishes and the forward wave is completely reflected from the bottom surface of the film as k_{0y}' is equal to zero or an imaginary number. In this situation, the backward wave as the reflection wave is the most intense and equal in intensity to the forward wave. The interference of the two waves at the bottom surface makes the pump wave enlarged, and further leads to the appearance of the T_s-peak in the vicinity of the critical angle θ_c'. The intensity of R_s, however, depends on that of the pump wave at the upper surface. When the phase difference between the forward and backward waves satisfies $\phi = \pm 2k\pi$ (*k* is an integer) at

the surface, the interference results in the peaks of R_s. Thus the interference effect in the film plays an important role in the enhancement of the SHG.

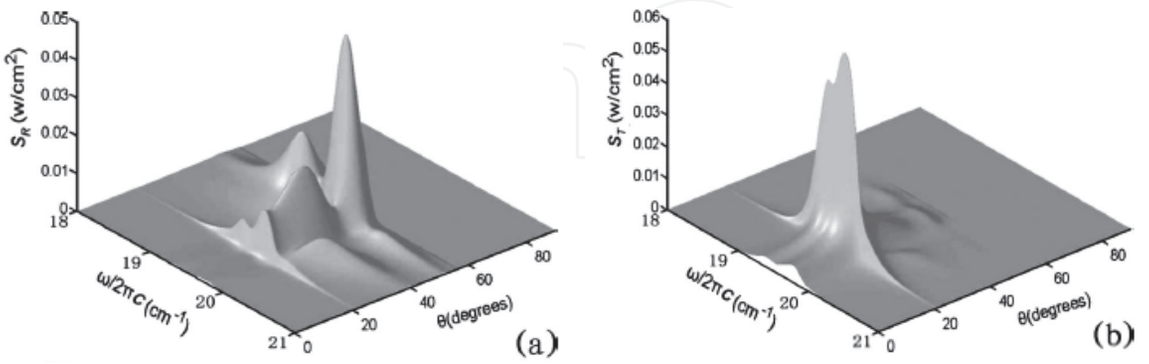


Fig. 14. SH outputs of ZnF_2/ MnF_2/air, S_R and S_T versus the incident angle and frequency. After Zhou & Wang, 2008.

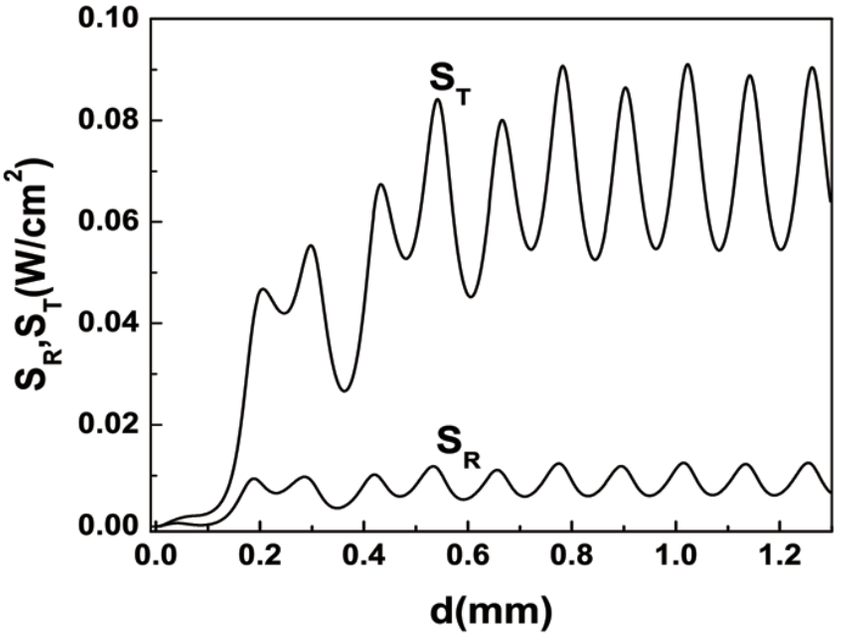


Fig. 15. SH outputs of SiO_2/MnF_2/air. S_R and S_T versus the film thickness for $\omega = 9.84cm^{-1}$ and $\theta = 41.3^\circ$. After Zhou & Wang, 2008.

It is also interesting for us to examine the SH outputs versus the film thickness. We take the SiO_2/MnF_2/air as an example and show the result in Fig.15. We think that the SH fringes result from the change of optical thickness of the film, and the SH outputs reache their individual saturation values about at $d = 800\mu m$, 0.09 W/cm², and 0.012 W/cm². If we enhance the incident wave density to 10.0kW/cm², the two output densities are increased by 100 times, to 9.0W/cm² and 1.2W/cm², or if we focus S_I on a smaller area, higher SH outputs are also obtained, so it is not difficult to observe the SH outputs.

If we put this AF film into one-dimension Photonic crystals (PCs), the SHG has a higher efficiency(Zhou, et. al., 2009). It is because that when some AF films as defect layers are introduced into a one-dimension PC, the defect modes may appear in the band gaps. Thus electromagnetic radiations corresponding to the defect modes can enter the PC and be greatly localized in the AF films. This localization effect has been applied to the SHG from a traditional nonlinear film embedded in one-dimension photonic crystals(Ren, et. Al., 2004 ; Si, et. al., 2001 ; Zhu, et.al., 2008, Wang, F., et. al. 2006), where a giant enhancement of the SHG was found.

6. Summary

In this chacter, we first presented various-order nonlinear magnetizations and magnetic susceptibilities of antiferromagnets within the perturbation theory in a special geometry, where the external magnetic field is pointed along the anisotropy axis. As a base of the nonlinear subject, linear magnetic polariton theory of AF systems were introduced, including the effective-medium method and transfer-matrix-method. Here nonlinear propagation of electromagnetic waves in the AF systems was composed of three subjects, nonlinear polaritons, nonlinear transmition and reflection, and second-harmonic generation. For each subject, we presented a theoretical method and gave main results. However, magnetically optical nonlinearity is a great field. For AF systems, due to their infrared and millimeter resonant-frequency feature, they may possess great potential applications in infrared and THz technology fields. Many subjects parallel to the those in the traditional nonlinear optics have not been discussed up to now. So the magnetically nonlinear optics is a opening field. We also hope that more experimental and theoretical works can appear in future.

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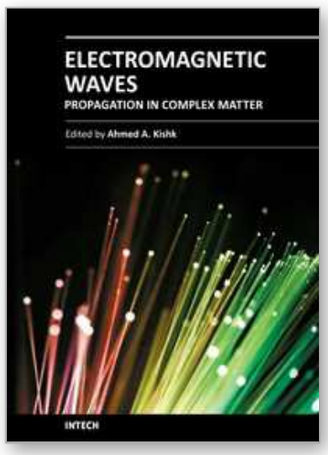
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