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# Nonlinear infrared transmission through and reflection off antiferromagnetic films

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**Abstract**
We present a theory of nonlinear infrared transmission through and reflection off an antiferromagnetic film with uniaxial anisotropy. In the geometry with the anisotropy axis parallel to the film surface and normal to the plane of incidence, using a linearly polarized wave (TE wave) obliquely incident upon the film, we obtain the transmission and reflection spectra. For an incident intensity available in experiments, the obvious nonlinear effects are seen in the vicinity of the resonant frequency. The nonlinear reflection and transmission can be much stronger or weaker than the linear ones, depending on frequencies or incident angles. The nonlinear interaction also dramatically increases or decreases the absorption of electromagnetic energy in the film.

## 1. Introduction

Nonlinear magnetic phenomena were found in ferromagnets many years ago, but nonlinear properties of antiferromagnets (AFs) were not given great attention until the 1990s. AF resonances are usually in the millimetre or far infrared region, for which the experimental methods are optical or quasi-optical ones, so the nonlinear optical properties of AFs should be interesting.

The nonlinear properties of common nonlinear optical materials result from their nonlinear resonance to the electric field of electromagnetic waves, or their electric polarization $\vec{P}^{\mathrm{NL}} = \overset{\leftrightarrow}{\chi}^{(1)}\vec{E} + \overset{\leftrightarrow}{\chi}^{(2)}\vec{E}\vec{E} + \overset{\leftrightarrow}{\chi}^{(3)}\vec{E}\vec{E}\vec{E} + \cdots$. However, magnetically nonlinear characteristics of materials come from their nonlinear resonance to the magnetic field component of electromagnetic waves, the magnetic polarization $\vec{m}^{\mathrm{NL}} = \overset{\leftrightarrow}{\chi}^{(1)}\vec{h} + \overset{\leftrightarrow}{\chi}^{(2)}\vec{h}\vec{h} + \overset{\leftrightarrow}{\chi}^{(3)}\vec{h}\vec{h}\vec{h} + \cdots$. The magnetic nonlinear effects for common materials are weak, since the magnetic component of electromagnetic waves is small in numerical value, but the nonlinear magnetic susceptibilities for ferromagnets, ferrimagnets and antiferromagnets may be very large as the wave frequency

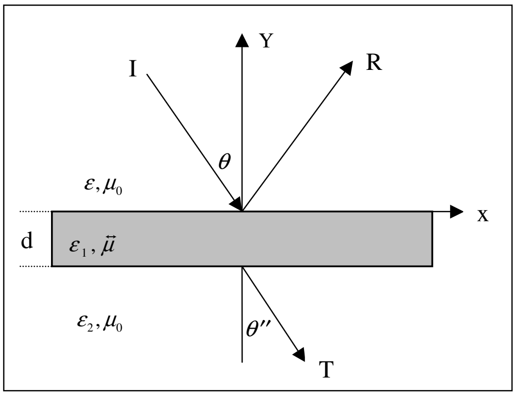


**Figure 1.** Geometry and coordinate system for nonlinear reflection and transmission of a AF film with thickness $d$. The anisotropy axis is along the $z$-axis and the incident plane parallel to the $x$–$y$ plane. $I$ is the obliquely incident radiation (TE wave), $R$ the reflection off and $T$ the transmission through the film.

is in the range of their resonant frequency. Thus magnetic nonlinear phenomena can also be observed in these materials. In addition, high power laser can help scientists to study magnetically nonlinear properties.

In discussions on relevant subjects, the nonlinear magnetic susceptibilities comprise a base for investigating the nonlinear properties. From the nonlinear equations satisfied by alternating magnetizations in AFs, the nonlinear magnetic susceptibilities were obtained in different forms for various purposes [1–4]. Almeida and Mills [1] dealt with the bi-stability and multi-stability of transmission for circularly polarized infrared radiation normally incident upon AF films with the uniaxial anisotropy normal to the film surfaces, and they discussed the same subject for AF superlattices in another paper [2]. All the nonlinear susceptibilities within the third-order approximation were obtained with a circularly polarized magnetic field in the cylindrical coordinate system [3]. However, for general applications the susceptibility expressions in the $xyz$ coordinate system may be more convenient. Wang, Fu and Li [4, 5] derived these expressions and investigated nonlinear surface and bulk magnetic polaritons in AF superlattices. The other properties, for example the nonlinear propagation of electromagnetic waves at the interface between two AFs and the surface of an AF, as well as AF solitons were discussed in [6–8, 15]. In addition, in the case of linearity the transmission through an AF film and superlattice were also discussed for normally incident radiation [9–11].

## 2. Nonlinear reflection and transmission

As we know, linear results or expressions are considered as bases on which one can discuss nonlinear properties, but the linear results can be found from the linear approximation of nonlinear results, so the useful linear expressions for our subject can be obtained from our nonlinear expressions. Thus we shall directionally derive the nonlinear expressions for the reflection and transmission.

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 figure 1, 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. The wave electric field can be described as $E_z = (E_I e^{-ik_y y} + E_R e^{ik_y y})e^{i(k_x x-\omega t)}$ above the film and as

$E_z = E_T e^{-ik''_y y} e^{i(k_x x - \omega t)}$ below. In these two regions, the magnetic field components are

$$h_x = (i\omega\mu_0)^{-1}\frac{\partial E_z}{\partial y}, \qquad h_y = -(i\mu_0\omega)^{-1}\frac{\partial E_z}{\partial x}. \tag{1}$$

In the film, the wave solution is no longer linear, but fortunately we found its expression in the process of discussing nonlinear polaritons in AF superlattices [5]. Its magnetic field is written as

$$\vec{h}^{NL} = \frac{k'_y}{\mu_0\mu_1\omega}\Bigg\{[(A'e^{ik'_y y} - B'e^{-ik'_y y}) + A'\eta_1(y)]\vec{e}_x - \frac{k_x}{k'_y}[(A'e^{ik'_y y} + B'e^{-ik'_y y}) + A'\eta_2(y)]\vec{e}_y\Bigg\}e^{i(k_x x - \omega t)}, \tag{2}$$

and electric field as

$$E_z^{NL} = \{[A'e^{ik'_y y} + B'e^{-ik'_y y} + A'\eta_3(y)]\}e^{i(k_x x - \omega t)}, \tag{3}$$

where $\eta_j(y)$ represents the nonlinear terms, and in the linear case $\eta_j(y) = 0$. The propagation constants are $k_x = (\omega/c)\sqrt{\varepsilon}\sin\theta$, $k_y = (\omega/c)\sqrt{\varepsilon}\cos\theta$, $k' = \sqrt{\varepsilon_1\mu_1(\omega/c)^2 - k_x^2}$ with $\mu_1$ presented in the appendix, and $k'' = \sqrt{\varepsilon_2(\omega/c)^2 - k_x^2}$. The nonlinear terms have been given for an AF superlattice in [5]. With slight corrections we can find the terms available to an AF film, or

$$\eta_1(y) = L_1 e^{ik'_y y} + L_2 e^{-ik'_y y} + L_3 e^{3ik'_y y} + L_4 e^{-3ik'_y y}, \tag{4a}$$

$$\eta_2(y) = L'_1 e^{ik'_y y} + L'_2 e^{-ik'_y y} + L'_3 e^{3ik'_y y} + L'_4 e^{-3ik'_y y}, \tag{4b}$$

and

$$\eta_3(y) = \frac{c^2}{\mu_1\varepsilon_1\omega^2}\left[k_x^2\eta_2(y) - ik'_y\frac{\partial\eta_1(y)}{\partial y}\right], \tag{4c}$$

where the nonlinear coefficients are written as

$$L_1 = -P\left|\alpha A'\right|^2 ik'_y y\left(\chi_+ + \frac{k_x}{ik'_y}\delta_+\right), \qquad L_2 = -P\alpha\left|A'\right|^2 ik'_y y\left(\chi_- + \frac{k_x}{ik'_y}\delta_-\right), \tag{5a}$$

$$L_3 = -P\left|A'\right|^2\alpha^*\left(\chi_- - \frac{3k_x}{ik'_y}\delta_-\right)\Bigg/4, \qquad L_4 = P\alpha^2\left|A'\right|^2\left(\chi_+ - \frac{3k_x}{ik'_y}\delta_+\right)\Bigg/4, \tag{5b}$$

$$L'_1 = -P\left|\alpha A'\right|^2\left[\chi_+(1 + ik'_y y) - \frac{\delta_+}{ik_x k'_y}(2k'^2_y + k_x^2 - ik'_y y k_x^2)\right], \tag{5c}$$

$$L'_2 = -\alpha P\left|A'\right|^2\left[\chi_-(1 - ik'_y y) - \frac{\delta_-}{ik_x k'_y}(2k'^2_y + k_x^2 + ik_x^2 k'_y y)\right], \tag{5d}$$

$$L'_3 = -P\left|A'\right|^2\alpha^*\left[3\chi_- - \frac{\delta_-}{ik_x k'_y}(k_x^2 - 8k'^2_y)\right]\Bigg/4, \tag{5e}$$

$$L'_4 = P\alpha^2\left|A'\right|^2\left[3\chi_+ - \frac{\delta_+}{ik_x k'_y}(k_x^2 - 8k'^2_y)\right]\Bigg/4, \tag{5f}$$

for real $k'_y$, or as

$$L_1 = -P\alpha \left|A'\right|^2 \mathrm{i}k'_y y \left(\chi_+ + \frac{k_x}{\mathrm{i}k'_y}\delta_+\right), \qquad L_2 = -P \left|\alpha A'\right|^2 \mathrm{i}k'_y y \left(\chi_- + \frac{k_x}{\mathrm{i}k'_y}\delta_-\right), \tag{6a}$$

$$L_3 = -P \left|A'\right|^2 \left(\chi_- - \frac{3k_x}{\mathrm{i}k'_y}\delta_-\right) \Big/ 4, \qquad L_4 = P\alpha \left|\alpha A'\right|^2 \left(\chi_+ - \frac{3k_x}{\mathrm{i}k'_y}\delta_+\right) \Big/ 4, \tag{6b}$$

$$L'_1 = -P\alpha \left|A'\right|^2 \left[\chi_+(1 + \mathrm{i}k'_y y) - \frac{\delta_+}{\mathrm{i}k_x k'_y}(2k'^2_y + k_x^2 - \mathrm{i}k_x^2 k'_y y)\right], \tag{6c}$$

$$L'_2 = P \left|\alpha A'\right|^2 \left[\chi_-(1 - \mathrm{i}k'_y y) - \frac{\delta_-}{\mathrm{i}k_x k'_y}(2k'^2_y + k_x^2 + \mathrm{i}k_x^2 k'_y y)\right], \tag{6d}$$

$$L'_3 = -P \left|A'\right|^2 \left[3\chi_- - \frac{\delta_-}{\mathrm{i}k_x k'_y}(k_x^2 - 8k'^2_y)\right] \Big/ 4, \tag{6e}$$

$$L'_4 = P\alpha \left|\alpha A'\right|^2 \left[3\chi_+ - \frac{\delta_+}{\mathrm{i}k'_y k_x}(k_x^2 - 8k'^2_y)\right] \Big/ 4, \tag{6f}$$

for imaginary $k'_y$, where $\chi_\pm = -k'_y\chi^{(3)}_{xxxy} \pm k_x\chi^{(3)}_{xyyx}$, $\delta_\pm = \mathrm{i}k_x\chi^{(3)}_{xxxy} \pm \mathrm{i}k'_y\chi^{(3)}_{xyyx}$, $P = k_x/\mu_0^2\mu_1^3\omega^2$ and $\alpha = -B'/A'$. Considering the relations between electric amplitude $A'$ and magnetic amplitude $A_x$ and $B_x$ in the case of linearity, $A_x = \frac{k'_y}{\mu_0\mu_1\omega}A'$ and $B_x = -\frac{k'_y}{\mu_0\mu_1\omega}A'$, as well as the factor $M_0^{-2}$ included in $\chi^{(3)}_{ijkl}$, $\eta_j(y)$ is a second-order quantity in $h/M_0$.

According to the known field components, the transmission and reflection coefficients in the case of nonlinearity can be found from the boundary conditions of $h_x$ and $E_z$ continuous at $y = 0$ and $-d$. We find four equations

$$E_\mathrm{I} + E_\mathrm{R} = [A' + A'\eta_3(0) + B'], \tag{7a}$$

$$\frac{k_y}{\omega\mu_0}(-E_\mathrm{I} + E_\mathrm{R}) = \frac{k'_y}{\mu_0\mu_1\omega}[A' - B' + A'\eta_1(0)], \tag{7b}$$

$$A'\mathrm{e}^{-\mathrm{i}k'_y d} + B'\mathrm{e}^{\mathrm{i}k'_y d} + A'\eta_3(-d) = E_\mathrm{T}\mathrm{e}^{\mathrm{i}k''_y d}, \tag{7c}$$

$$\frac{k'_y}{\mu_0\mu_1\omega}\left[A'\mathrm{e}^{-\mathrm{i}k'_y d} - B'\mathrm{e}^{\mathrm{i}k'_y d} + A'\eta_1(-d)\right] = -\frac{k''_y}{\mu_0\omega}E_\mathrm{T}\mathrm{e}^{\mathrm{i}k''_y d}. \tag{7d}$$

Before looking for the nonlinear reflection and transmission, the amplitude $A'$ and ratio $\alpha$ should be obtained for a given incident field $E_\mathrm{I}$ since they are included in the nonlinear terms. In the third-order approximation it is enough to know their linear expressions, so ignoring the nonlinear terms in equation (7), we see that

$$A' = \frac{1}{2}\left[1 + r + \frac{\mu_1 k_y}{k'_y}(r - 1)\right]E_\mathrm{I}, \tag{8a}$$

with

$$r = \frac{E_\mathrm{R}}{E_\mathrm{I}} = \frac{\mu_1 k'_y(k_y - k''_y)\cos k'_y d + (k'^2_y - \mu_1^2 k_y k''_y)\mathrm{i}\sin k'_y d}{\mu_1 k'_y(k_y + k''_y)\cos k'_y d - (k'^2_y + \mu_1^2 k_y k''_y)\mathrm{i}\sin k'_y d}, \tag{8b}$$

and

$$\alpha = \frac{\mu_1 k''_y + k'_y}{\mu_1 k''_y - k'_y}\mathrm{e}^{-2\mathrm{i}k'_y d}. \tag{9}$$

Note that $\eta_1(y)$ and $\eta_3(y)$ in equation (7) are both of second order and neglect the terms higher than second order, the nonlinear refection and transmission coefficients are obtained, or

$$r = \frac{E_R}{E_I} = \frac{\mu_1(k_y - k_y'')\cos k_y'd + (k_y' - \mu_1^2 k_y k_y''/k_y')\mathrm{i}\sin k_y'd - \mathrm{NL}_+}{\mu_1(k_y + k_y'')\cos k_y'd - (k_y' + \mu_1^2 k_y k_y''/k_y')\mathrm{i}\sin k_y'd - \mathrm{NL}_-}, \quad (10a)$$

$$t = \frac{E_T}{E_I} = \frac{\mu_1 k_y[2 - \eta_1(0) - \eta_3(0)]\mathrm{e}^{\mathrm{i}k_y''d}}{\mu_1(k_y + k_y'')\cos k_y'd - (k_y' + \mu_1^2 k_y k_y''/k_y')\mathrm{i}\sin k_y'd - \mathrm{NL}_-}, \quad (10b)$$

in which the nonlinear terms $\mathrm{NL}_\pm$ are shown with

$$\begin{aligned}\mathrm{NL}_\pm = \tfrac{1}{2}\{&\pm(k_y' - \mu_1 k_y'')[\cos(k_y'd) \pm \mathrm{i}\mu_1 k_y \sin(k_y'd)/k_y']\eta_1(-d)\mathrm{e}^{\mathrm{i}k_y'd} + (\mp k_y' + \mu_1 k_y)\\ &\times [\cos(k_y'd) - \mathrm{i}\mu_1 k_y'' \sin(k_y'd)/k_y']\eta_1(0)\\ &+ (k_y' - \mu_1 k_y'')[\pm \mathrm{i}\sin(k_y'd) + \mu_1 k_y \cos(k_y'd)/k_y']\\ &\times \mathrm{e}^{\mathrm{i}k_y'd}\eta_3(-d) + (\pm k_y' - \mu_1 k_y)[\mathrm{i}\sin(k_y'd) - \mu_1 k_y'' \cos(k_y'd)/k_y']\eta_3(0)\}. \end{aligned} \quad (11)$$

Finally the reflectivity and transmissivity are defined as $R = |r|^2$ and $T = |t|^2$ for $\varepsilon = \varepsilon_2$, but $T = |t|^2\, \varepsilon_2 \cos\theta''/\varepsilon\cos\theta$ for $\varepsilon \neq \varepsilon_2$ [12]. Here we should discuss three special situations: the cases of normal incidence ($k_x = 0$), $k_y' = 0$ and imaginary $k_y''$.

In the first situation ($k_x = 0$), from the expressions of $L_1$ to $L_4$ and $L_1'$ to $L_4'$, we find $\eta_1(y) = \eta_3(y) = 0$. It is quite obvious that one finds no nonlinear effects on the reflection and transmission in the case of normal incidence. In the second case ($k_y' = 0$), the magnetic amplitudes $A = B = 0$ and then the magnetic field component $h_x$ vanishes in the AF film, which also means there are no nonlinear effects in this case. Opposite to that, when $\omega \to \sqrt{\omega_r^2 + 2\omega_a\omega_m}$, $\mu_1$ becomes a very small complex quantity. This means that the wave magnetic field is very intense in the film; in turn, nonlinear effects also should be strong. For $\varepsilon > \varepsilon_2$, $k_y''$ becomes imaginary as the incident angle $\theta$ exceeds a special value, then the transmission vanishes. The nonlinear effect can be seen only from the reflection. These judgments are proven in the next section.

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 equation (10).

## 3. Numerical examples

We take a FeF2 film as an example for numerical calculations, with the physical parameters [13, 14]: the relative dielectric constant $\varepsilon_1 = 5.5$, the sublattice magnetization $M_0 = 0.56$ kG and the damping coefficient $\tau = 1.0 \times 10^{-3}$ are the same as that used in [13]; the anisotropy field $H_a = 197.0$ kG, the exchange field $H_e = 540.0$ kG and the gyromagnetic ratio $\gamma = 1.97 \times 10^{10}$ rad s$^{-1}$ kG$^{-1}$, leading to the resonant frequency $\omega_r/2\pi c = 52.5$ cm$^{-1}$. 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 $\eta_j(y)$, is fixed at $S_I = 4.7$ MW cm$^{-2}$, corresponding to a magnetic amplitude of 16 G 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 in the two geometries. In the first geometry, the AF film is put in a vacuum, and in the next it is located between two dielectrics with $\varepsilon = 2.3$ and $\varepsilon_2 = 1.0$, respectively.

In the first geometry, the transmission and reflection versus frequency $\omega$ are illustrated in figure 2 for the incident angle $\theta = 30°$ and are shown in figure 3 versus incident angle $\theta$ for $\omega/2\pi c = 52.8$ cm$^{-1}$. First, the nonlinear modification is more evident in reflection for frequencies higher than $\omega_r$. We see a very obvious discontinuity on the nonlinear $R$ and $T$

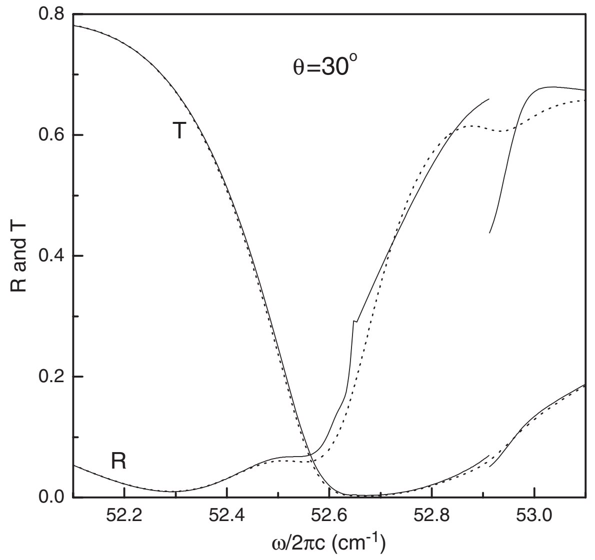


**Figure 2.** Reflectivity and transmissivity versus frequency for a fixed angle of incidence of 30°, where the dotted lines represent the linear results. The film is put into free space and the intensity of incident radiation $S_I = 4.7$ kW cm$^{-2}$.

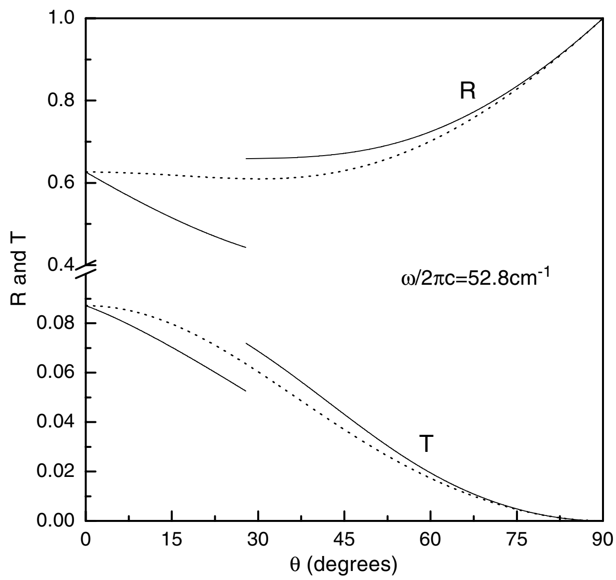


**Figure 3.** Reflectivity and transmissivity versus angle of incidence for the frequency fixed at 52.8 cm$^{-1}$ and the intensity of incident radiation $S_I = 4.7$ kW cm$^{-2}$. The dotted lines show the linear results.

curves at $\omega/2\pi c = 52.9$ cm$^{-1}$, corresponding to the smallest value of $\mu_1$ 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. At a special frequency corresponding to the point of intersection of the linear and nonlinear curves

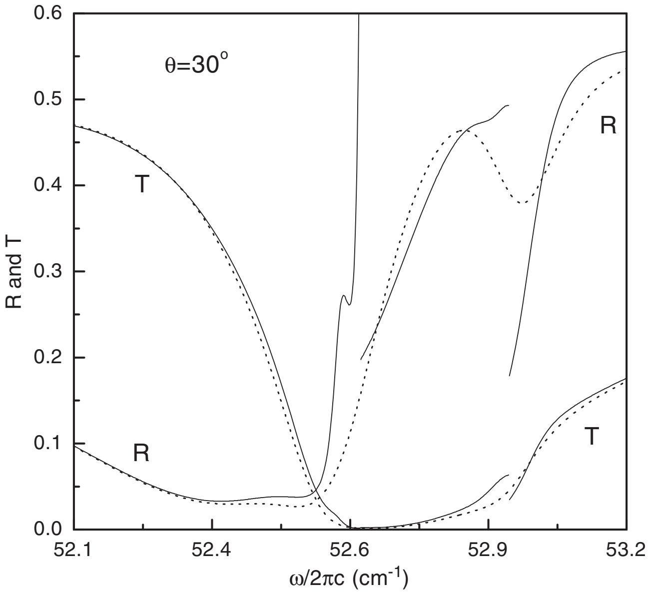


**Figure 4.** Reflection and transmission versus frequency for an angle of incidence of 30° in such a geometry where the dielectric with $\varepsilon = 2.3$ is above the film and the space below is a vacuum. The intensity of incident radiation $S_I = 4.7$ kW cm$^{-2}$ and the dotted lines show the linear results.

$k'_y = 0$, and as we know from the above section the modification disappears. Secondly, $R$ and $T$ versus $\theta$ for a fixed frequency are shown in figure 3. Here the discontinuity is also seen since the magnetic amplitude and the nonlinear terms vary with the wave vector $\vec{k}'$. It is more interesting that when the incident angle $\theta \leqslant 27.5°$ the reflection and transmission are both lower than the linear ones, implying that the absorption is reinforced. However, in the range of $\theta \geqslant 27.5°$ they both are higher than the linear ones, and as a result the absorption is evidently restrained. The nonlinear influence disappears for normal incidence.

In the second geometry, the reflection and transmission spectra are illustrated by figures 4–6. It is worth noting that if $k''_y = \sqrt{\varepsilon_2(\omega/c)^2 - k_x^2}$ is an imaginary value, $T$ vanishes in the linear and nonlinear cases and the complete reflection is seen, so that we find a critical angle $\theta_c = \arcsin(\sqrt{\varepsilon_2/\varepsilon}) = 41.3°$. $R$ and $T$ versus frequency (see figure 4) are different in two points from figure 2. First, two discontinuities are found on the $R$ spectrum, or one is at $\omega/2\pi c = 52.93$ cm$^{-1}$ and for the same reason as in the first geometry, but the other is at 52.55 cm$^{-1}$ and may come from the divergence of the third-order magnetic susceptibilities. Although this discontinuity does not appear in figure 2, we see the relevant signs of discontinuity there. Second, the nonlinear effects are more obvious than in the first geometry. In a small frequency region near to the resonant frequency, the nonlinear reflection becomes 15 times as large as the linear one, but only 1/2 of the linear one in the vicinity of $\omega/2\pi c = 52.93$ cm$^{-1}$. We take two examples for $R$ and $T$ as a function of incident angle, as illustrated in figures 5 and 6. One is related to $\omega/2\pi c = 52.4$ cm$^{-1}$ lower than the resonant frequency and the other to $\omega/2\pi c = 52.8$ cm$^{-1}$ higher than the resonant frequency. The first is completely different from figure 3. In the vicinity of $\theta \leqslant \theta_c$, both $T$ and $R$ are obviously higher than the linear values. For $\theta \geqslant \theta_c$, the transmission vanishes and only the reflection is seen, but the absorption is very large for angles near to 60°. At about $\theta = 43°$, the nonlinear interaction evidently constrains the absorption, then $R$ approaches three times the linear value.

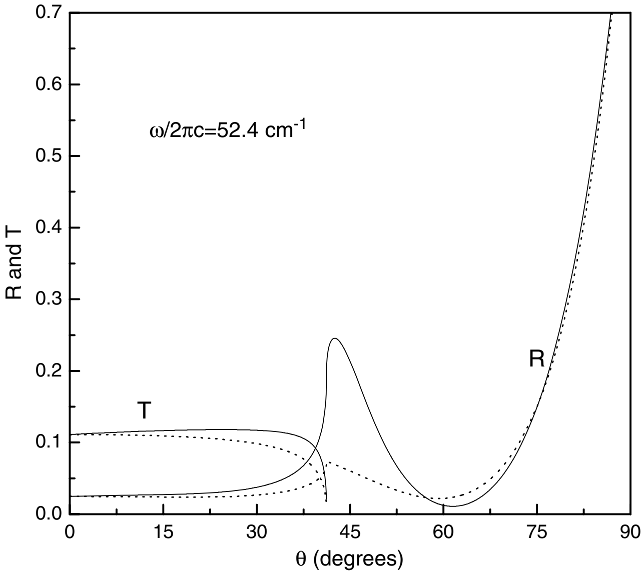


**Figure 5.** Reflection and transmission spectra with respect to the angle of incidence for a fixed frequency larger than the resonant frequency in the same geometry and intensity as those in figure 4.

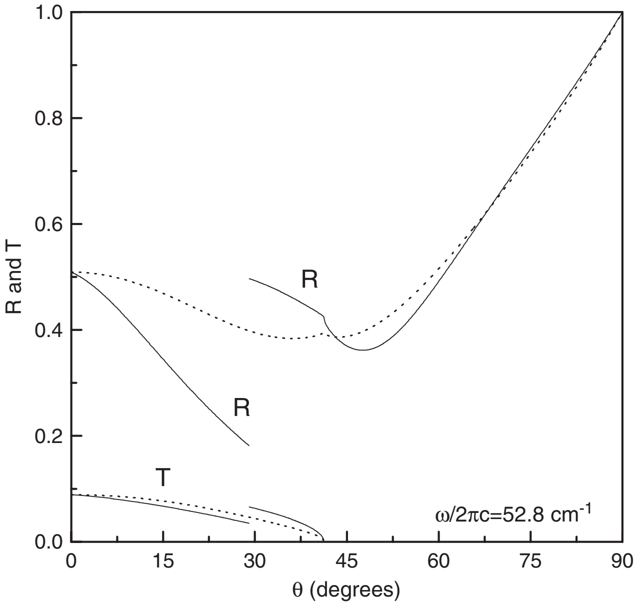


**Figure 6.** Reflection and transmission spectra with respect to the angle of incidence for a fixed frequency less than the resonant frequency in the same geometry and intensity as those in figure 4.

The second is similar to what is shown in figure 3, except for the absence of $T$ for angles of incidence larger than the critical value and stronger nonlinear modification.

## 4. Conclusions

We have discussed the reflection and transmission spectra of a nonlinearly antiferromagnetic film with uniaxial anisotropy in the vicinity of the resonant frequency, where the nonlinear interaction between the wave field and magnetic moments in the film are taken into account in the third-order approximation. For simplicity, a TE wave is taken as the incident radiation and the AF anisotropy axis is in the film surface and normal to the incident plane. In this geometry, we present a theoretical method for obtaining the expressions for the nonlinear reflection off and transmission through an AF film. The numerical results suggest some interesting properties of the reflection and transmission. The nonlinearity is weak in AFs, so one can observe the relevant properties only in special geometries and with higher power lasers, such as the free-electron and molecular gas lasers.

In the two geometries, 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 nonlinear interaction also play an important role in decreasing or increasing the absorption in the AF film. Generally speaking, the nonlinear effect is more evident in the second geometry for $\varepsilon > \varepsilon_2$ than in the first geometry.

## Acknowledgment

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## Appendix

Here we present the nonlinear susceptibilities of the uniaxial and bi-sublattice antiferromagnet used in our text. For the chosen geometry, as shown in figure 1, the antiferromagnetic sublattice magnetizations are

$$\vec{M}_{\mathrm{A}} = M_0\hat{z} + \vec{m}_{\mathrm{A}}, \qquad \vec{M}_{\mathrm{B}} = -M_0\hat{z} + \vec{m}_{\mathrm{B}} \tag{A.1}$$

and the magnetic field $\vec{h} = (h_x, h_y, 0)$. We should note that the alternating magnetizations are $\vec{m}_{\mathrm{A,B}} = \vec{m}^{(1)}_{\mathrm{A,B}} + \vec{m}^{(2)}_{\mathrm{A,B}} + \vec{m}^{(3)}_{\mathrm{A,B}}$ in the third-order approximation. $\vec{h}$ and $\vec{m}^{(1),(3)}$ include the factor $\mathrm{e}^{-\mathrm{i}\omega t}$, but $\vec{m}^{(2)}(2\omega)$ includes the factor $\mathrm{e}^{-2\mathrm{i}\omega t}$ and $\vec{m}^{(2)}(0)$ does not change with time. Similar to nonlinear optics, the complex conjugation $\vec{h}^*$ and $\vec{m}^{(1)*}$ should be considered in the third-order approximation. Thus, in the presence of a damping effect upon the motion of magnetizations, we have the equations of motion

$$\frac{\partial}{\partial t}\vec{M}_{\mathrm{A}} = \gamma\vec{M}_{\mathrm{A}} \times \vec{H}^{\mathrm{e}}_{\mathrm{A}} - \frac{\tau}{M_0}\vec{M}_{\mathrm{A}} \times \frac{\partial}{\partial t}\vec{M}_{\mathrm{A}}, \qquad \frac{\partial}{\partial t}\vec{M}_{\mathrm{B}} = \gamma\vec{M}_{\mathrm{B}} \times \vec{H}^{\mathrm{e}}_{\mathrm{B}} - \frac{\tau}{M_0}\vec{M}_{\mathrm{B}} \times \frac{\partial}{\partial t}\vec{M}_{\mathrm{B}} \tag{A.2}$$

where the effective fields on the two sublattices

$$\vec{H}^{\mathrm{e}}_{\mathrm{A}} = \vec{z}\frac{H_{\mathrm{a}}H^Z_{\mathrm{A}}}{M_0} - H_{\mathrm{e}}\frac{\vec{M}_{\mathrm{B}}}{M_0} + \vec{h}, \qquad \vec{H}^{\mathrm{e}}_{\mathrm{B}} = \vec{z}\frac{H_{\mathrm{a}}H^Z_{\mathrm{B}}}{M_0} - H_{\mathrm{e}}\frac{\vec{M}_{\mathrm{A}}}{M_0} + \vec{h}, \tag{A.3}$$

with the anisotropy field $H_{\mathrm{a}}$ and the exchange field $H_{\mathrm{e}}$. The last term on the right-hand side of every equation in (A.2) is the damping term with $\tau$ the damping coefficient.

In our previous papers [4, 5] we derived the nonlinear magnetic susceptibilities, without the damping term. Although we consider it here, the process of derivation is similar to the previous ones. Thus we directly present the necessary results. In the linear case (the first order),

$$\chi_{xx}^{(1)} = \chi_{yy}^{(1)} = \frac{2\omega_m \omega_{\mathrm{a}}'}{\omega_{\mathrm{r}}'^2 - \omega^2}, \tag{A.4}$$

with $\omega_{\mathrm{a}}' = \omega_{\mathrm{a}} - \mathrm{i}\tau\omega$ and $\omega_{\mathrm{r}}' = \sqrt{\omega_{\mathrm{a}}'(2\omega_{\mathrm{e}} + \omega_{\mathrm{a}}')}$ where $\omega_{\mathrm{a}} = \gamma H_{\mathrm{a}}$, $\omega_{\mathrm{e}} = \gamma H_{\mathrm{e}}$ and $\omega_m = 4\pi\gamma M_0$. The linear magnetic permeability $\mu_1 = 1 + \chi_{xx}^{(1)}$. The non-zero elements of the second-order susceptibilities are shown as

$$\chi_{zxy}^{(2)}(0) = -\chi_{zyx}^{(2)}(0) = \frac{4\mathrm{i}\omega\omega_{\mathrm{a}}\omega_m^2}{M_0|\omega_{\mathrm{r}}'^2 - \omega^2|^2}. \tag{A.5}$$

We note that harmonic generation cannot be seen in this geometry. The non-zero elements of the third-order magnetic susceptibility used in the text are indicated as

$$\chi_{xyxx}^{(3)} = \chi_{xxyx}^{(3)} = -\chi_{yxyy}^{(3)} = -\chi_{yyxy}^{(3)} = -\frac{1}{2}\chi_{xxxy}^{(3)} = \frac{1}{2}\chi_{yyyx}^{(3)} = \frac{4\mathrm{i}\omega^3\omega_{\mathrm{a}}'\omega_{\mathrm{a}}\omega_m^3}{M_0^2(\omega_{\mathrm{r}}'^2 - \omega^2)^2|\omega_{\mathrm{r}}'^2 - \omega^2|^2}, \tag{A.6}$$

$$\chi_{xyyx}^{(3)} = -2\chi_{xxyy}^{(3)} = -2\chi_{xyxy}^{(3)} = \chi_{yxxy}^{(3)} = -2\chi_{yyxx}^{(3)}$$
$$= -2\chi_{yxyx}^{(3)} = \frac{4\omega^2\omega_m^3\omega_{\mathrm{a}}(2\omega_{\mathrm{r}}'^2 - 3\omega_{\mathrm{a}}'^2 - \omega^2)}{M_0^2(\omega_{\mathrm{r}}'^2 - \omega^2)^2|\omega_{\mathrm{r}}'^2 - \omega^2|^2}. \tag{A.7}$$

The elements unused in the text are not derived in this appendix.

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