Nonreciprocal reflection of infrared radiation from structures with antiferromagnets and dielectrics

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(Received 10 April 1996)

We calculate the reflectivity from structures involving both dielectric and antiferromagnetic films. In contrast to earlier results involving only antiferromagnetic films, we find that nonreciprocal reflection occurs even in the absence of any absorption. The calculations are done with a transfer matrix method and examples of nonreciprocal reflectivity are shown both with and without damping. In addition a general thermodynamic argument is presented which shows why nonreciprocity is allowed for the structure involving both dielectrics and antiferromagnets while it is forbidden for a simple antiferromagnetic film. The thermodynamic argument also gives some symmetry relations shown in our explicit calculations. [S0163-1829(96)04638-3]

I. INTRODUCTION

Recently a series of experiments[1–4] have examined the reflection of infrared radiation from antiferromagnets. A key feature in these experiments is the occurrence of nonreciprocal reflectivity. Here reversing the directions of the incident and reflected waves leads to a different reflection coefficient when there is an external magnetic field parallel to the surface of the antiferromagnet.

A number of papers have explored nonreciprocal reflection from antiferromagnets theoretically as well.[5–8] In the past it has been argued that for semi-infinite or thin film antiferromagnets nonreciprocity in the reflection intensities, which may, in principle, be enhanced by surface roughness,[5] could occur only when absorption was present.[6] This is a surprising result, but, in the general case of arbitrary angle between the applied field and the plane of incidence, mixing between s- and p-polarized waves can lead to nonreciprocity within a particular polarization.[7] However, in the Voigt geometry where the plane of incidence is perpendicular to the applied magnetic field and the field is parallel to the easy axis, no such mixing occurs and the reflection intensities are strictly reciprocal in the absence of damping.

In this paper we examine different geometries which also lead to nonreciprocal reflection. Our geometries are physically realistic and include an antiferromagnetic film on a substrate, a dielectric film on an antiferromagnetic film, and a dielectric overlayer on a semi-infinite antiferromagnetic. In contrast to earlier systems, the nonreciprocity explored here exists in the Voigt configuration and without any damping being present. For these geometries we compare the nonreciprocity which exists in the presence of damping to that which exists without any damping.

In addition to our explicit calculations for specific examples, we also present a general thermodynamic argument showing how the combination of a dielectric film and antiferromagnetic film allows nonreciprocal reflectivity where an antiferromagnetic film by itself does not. Our thermodynamic argument will also establish certain symmetry relations between the nonreciprocal reflectivity from the dielectric surface and the nonreciprocal reflectivity from the antiferromagnetic surface.

The paper is organized as follows. In Sec. II we present an outline of the theoretical calculations where we use a transfer matrix method to obtain reflectivity from a general dielectric/antiferromagnet structure. We then use this method to obtain specific results in Sec. III for particular geometries showing nonreciprocity both with and without damping. In Sec. IV we present the thermodynamic arguments which illustrate the general features of the reflectivity from dielectric/antiferromagnetic structures. Finally in Sec. V we present a summary and our conclusions.

II. THEORY

We consider layered systems in the Voigt geometry, with both the external field and the easy axis perpendicular to the plane of incidence. We take this direction to be the z direction; the y direction is normal to the layers. In this geometry, the only nonzero components of the magnetic permeability tensor of a simple uniaxial antiferromagnet at frequency ω are[9]

$$\mu_{xx}=\mu_{yy}=1+4\pi\gamma^2 H_A M_S(Y^+ + Y^-), \tag{1}$$

$$\mu_{xy}=-\mu_{yx}=i4\pi\gamma^2 H_A M_S(Y^+ - Y^-), \tag{2}$$

$$\mu_{zz}=1, \tag{3}$$

where

$$Y^{\pm}=[\omega_r^2-(\omega\pm\gamma H_0+i\Gamma)^2]^{-1}. \tag{4}$$

H_A is the anisotropy field, M_S the sublattice magnetization, γ the gyromagnetic ratio, H_0 the external field, and Γ the damping. The antiferromagnetic resonance frequency ω_r is given by

$$\omega_r = \gamma(2H_A H_E + H_A^2)^{1/2}, \tag{5}$$

where H_E is the exchange field.

In order to calculate the reflectivity of a layered system it is useful to employ a transfer matrix method. The procedure adopted here is basically a variation of that employed for layered semiconductors in previous studies.[10] Here we summarize the basic method. We restrict the discussion to s polarization because only s-polarized radiation interacts with the magnetic system for the Voigt geometry.

In the medium of incidence (layer 1, vacuum in all the present examples), the in-plane component of the wave vector q_x is determined by the angle of incidence θ:

$$q_x = \varepsilon_1^{1/2}(\omega/c)\sin\theta, \tag{6}$$

where ε_1 is the dielectric constant for the incident layer. The quantity q_x is continuous throughout the layered structure. The out-of-plane component q_{ny} (n is a layer index), however, varies from layer to layer. In s polarization it is given by

$$q_{ny} = [\varepsilon_n \mu_{vn}(\omega/c)^2 - q_x^2]^{1/2}, \tag{7}$$

where μ_{vn} is the Voigt permeability of the nth layer and is given by

$$\mu_{vn} = \mu_{1n} + \mu_{2n}^2/\mu_{1n}, \tag{8}$$

where μ_{1n} and μ_{2n} represent μ_{xx} and μ_{xy}, respectively, for the nth layer. For nonmagnetic layers μ_{vn} is equal to 1.

We now consider the variation of the electric fields, which are directed along the z axis, within each layer. At the top of each layer we can divide the field into two components $a_{n\mathrm{T}}$ and $b_{n\mathrm{T}}$. The former represents the total field associated with propagation in the $-y$ (downward) direction and the latter represents that associated with propagation in the $+y$ (upward) direction. We can also define equivalent fields $a_{n\mathrm{B}}$ and $b_{n\mathrm{B}}$ at the bottom of the layer. If the layer has thickness d_n, then the fields at its top and bottom may be related by the matrix equation

$$\begin{pmatrix} a_{n\mathrm{T}} \\ b_{n\mathrm{T}} \end{pmatrix} = \begin{pmatrix} \exp(-iq_{ny}d_n) & 0 \\ 0 & \exp(iq_{ny}d_n) \end{pmatrix} \begin{pmatrix} a_{n\mathrm{B}} \\ b_{n\mathrm{B}} \end{pmatrix}. \tag{9}$$

We may also use the boundary conditions at any chosen interface to relate the fields on each of its sides. At the interface between the nth and $(n+1)$th layers we use the continuity of E_z and H_x. The resulting relationship is of the form

$$\begin{pmatrix} a_{n\mathrm{B}} \\ b_{n\mathrm{B}} \end{pmatrix} = \frac{1}{X_n^- - X_n^+} \begin{pmatrix} X_{n+1}^- - X_n^+ & X_{n+1}^+ - X_n^+ \\ X_n^- - X_{n+1}^- & X_n^- - X_{n+1}^+ \end{pmatrix} \begin{pmatrix} a_{(n+1)\mathrm{T}} \\ b_{(n+1)\mathrm{T}} \end{pmatrix}, \tag{10}$$

where

$$X_n^{\pm} = [(\mu_{2n}/\mu_{1n})q_x \pm q_{ny}]/\mu_{vn}. \tag{11}$$

The matrices of the two types represented by Eqs. (9) and (10) may then be multiplied together over the whole structure so as to relate the fields in the top layer (layer 1) to those in the bottom one (layer N). Since there is no upward wave in layer N, which is semi-infinite, we may put $b_{N\mathrm{T}} = 0$, so that the overall matrix equation may be expressed in the form

$$\begin{pmatrix} a_{1\mathrm{B}} \\ b_{1\mathrm{B}} \end{pmatrix} = R \begin{pmatrix} a_{N\mathrm{T}} \\ 0 \end{pmatrix}, \tag{12}$$

where R is the 2×2 matrix resulting from the multiplication of the various transfer matrices across the layers and interfaces. The ratio $b_{1\mathrm{B}}/a_{1\mathrm{B}}$ may thus be obtained, and hence the reflectivity.

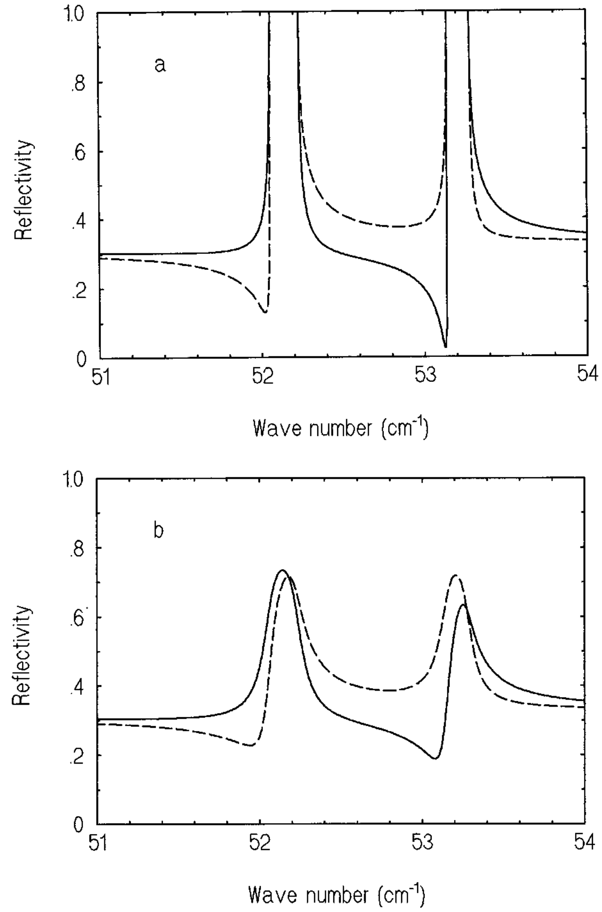

FIG. 1. Reflectivity spectra for a 3 μm dielectric layer deposited on a semi-infinite antiferromagnetic substrate, in the presence of a 0.5 T external field. The parameters for the antiferromagnet and the dielectric are those observed for FeF_2 at 4.2 K and for Si, respectively. (a) $\Gamma=0$; (b) $\Gamma=0.05$ cm^{-1}. Solid curves: $\theta=+45°$; dashed curves: $\theta=-45°$.

III. RESULTS

We now apply the above analysis to specific examples. All results are for s polarization with the easy axis and the external field normal to the plane of incidence.

In Fig. 1 we show the oblique incidence reflectivity off a dielectric film deposited on a semi-infinite antiferromagnetic substrate. The parameters chosen for the antiferromagnet are those for FeF_2 at 4.2 K, as used in interpreting experimental far infrared reflectivity and attenuated total reflection results;[2–4] $M_S=0.056$ T, $H_A=19.745$ T, $H_E=53.313$ T, $\omega_r=52.45$ cm^{-1}, $\gamma=1.05$ cm^{-1}/T, and $\epsilon=5.5$. The film is

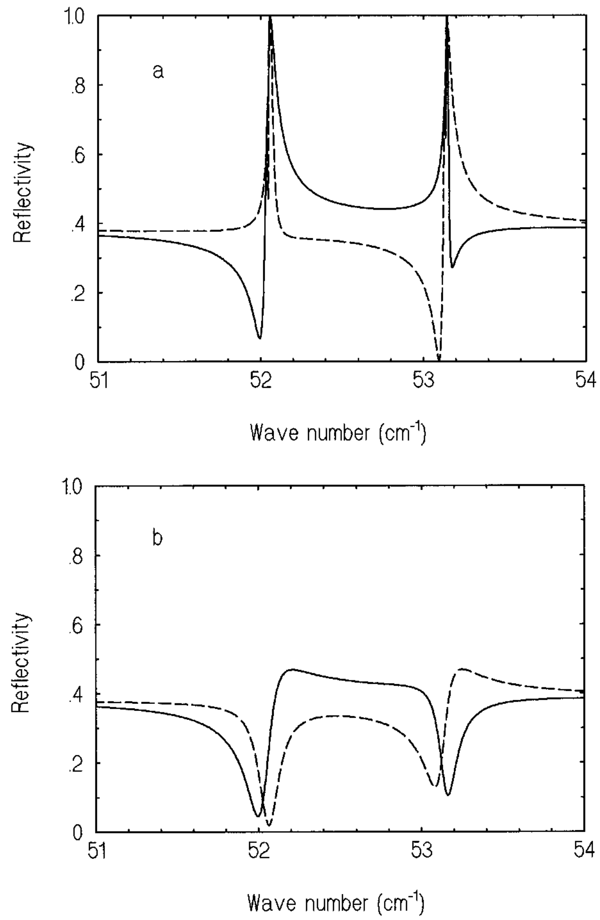

FIG. 2. Reflectivity spectra for a 6 μm antiferromagnet layer deposited on a semi-infinite dielectric substrate, in the presence of a 0.5 T external field. The parameters for the two materials are as for Fig. 1. (a) $\Gamma=0$; (b) $\Gamma=0.05$ cm^{-1}. Solid curves: $\theta=+45°$; dashed curves: $\theta=-45°$.

taken to have a dielectric constant of 11.6, the value for silicon. We show results both without damping and with the experimental value[2–4] of $\Gamma=0.05$ cm^{-1}. We consider a $+0.5$ T field in the z direction, with the angle of incidence either $+45°$ or $-45°$, corresponding to positive or negative values of q_x, respectively. We could equivalently have held the angle of incidence at $+45°$ and reversed the direction of the field.

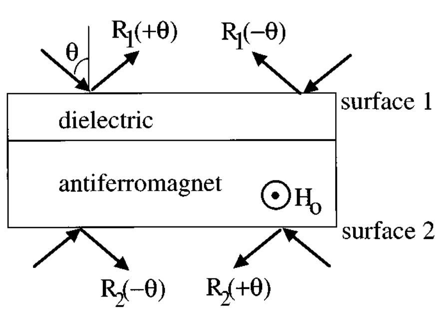

FIG. 3. The four types of reflectivity illustrated in Fig. 4 for a free-standing dielectric/antiferromagnet bilayer structure. The external field is coming out of the page, in the $+z$ direction.

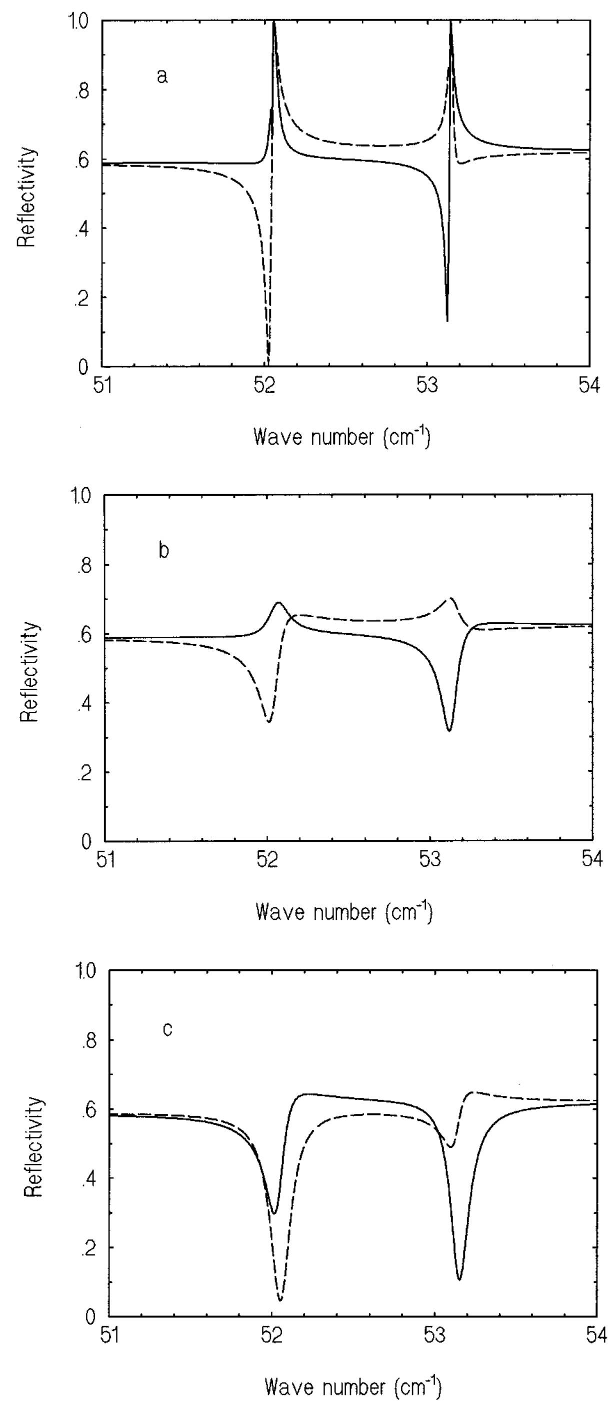

FIG. 4. Reflectivity spectra for a free-standing dielectric/antiferromagnet bilayer structure, in the presence of a 0.5 T external field. The dielectric layer is 3 μm thick and the antiferromagnet layer is 6 μm thick. The parameters for the two layers are as for Fig. 1. (a) $\Gamma=0$. Solid curve: $R_1(+45°)$ or $R_2(-45°)$; dashed curve: $R_1(-45°)$ or $R_2(+45°)$. (b) $\Gamma=0.05$ cm^{-1}, reflectivity from dielectric surface. Solid curve: $R_1(+45°)$; dashed curve: $R_1(-45°)$. (c) $\Gamma=0.05$ cm^{-1}, reflectivity from antiferromagnet surface. Solid curve: $R_2(+45°)$; dashed curve: $R_2(-45°)$.

As can be seen from the figure, the reflectivity R is nonreciprocal [i.e., $R(+45°)\neq R(-45°)$], regardless of whether damping is present or not. This contrasts with the case for reflection off a semi-infinite antiferromagnet or the symmetric case of a free-standing antiferromagnetic film; in either of these two cases damping is necessary for nonreciprocal reflection to occur.[6] In the particular example illustrated here, the undamped spectrum actually shows more nonreciprocity than the damped one.

The details of the nonreciprocity in Fig. 1 are very sensitive to the thickness of the dielectric overlayer. For very thin films (less than 3000 Å) there is essentially no nonreciprocity in the absence of damping. For the case shown in Fig. 1, a 3 μm dielectric film, the nonreciprocity is most evident at frequencies just below those of the reststrahl peaks. If the thickness is increased to 9 μm, the nonreciprocity becomes strongest at frequencies just above those of the reststrahl peaks. At a thickness of about 14 μm, the nonreciprocity disappears almost completely, only to emerge again for even thicker overlayers. This behavior suggests that the nonreciprocity is a function of the phase in the dielectric film. The exact mechanism will be described in more detail in a later paper. As an indication of the wavelength scales involved, the free-space wavelength is about 190 μm near the magnon frequency, and 14 μm would correspond to a quarter wavelength in the film.

In Fig. 2 we show the result for an antiferromagnetic film deposited on a semi-infinite dielectric. Once again, the reflectivity is nonreciprocal, irrespective of damping. In this case it is also seen that damping qualitatively affects the overall shape of the $\theta = -45°$ spectrum; this is not an unusual phenomenon in spectra of layered structures.[11,12]

The thickness dependence of the nonreciprocity for a thin antiferromagnetic film on a dielectric substrate is quite different from the inverse problem explored in Fig. 1. In the absence of damping, the nonreciprocity persists even for very thin antiferromagnetic films of a few hundred angstroms. For such films, the nonreciprocity disappears as damping is increased. The reason for this sensitivity for very thin antiferromagnetic films is that, for small damping, the effective wavelength in the antiferromagnet can be very short because of the resonance in the permeability. When the amplitude of the permeability is reduced through an increase in damping, the effective wavelength in the antiferromagnet becomes much larger than the film thickness and the nonreciprocity disappears.

We now consider the reflectivity off a free-standing dielectric/antiferromagnet bilayer. In this case we can reflect off either side of the sample, equivalent to inversion of the sample. We therefore consider reflectivity for the incident wave in each of the four directions indicated in Fig. 3. In Fig. 4 we show the results for each of the corresponding reflectivities, both with and without damping. Nonreciprocal reflectivity is once again observed, and when damping is present all four spectra are different. With no damping present, however, we observe the additional result that

$$R_1(+\theta) = R_2(-\theta), \tag{13}$$

$$R_1(-\theta) = R_2(+\theta). \tag{14}$$

In the following section we use thermodynamic arguments to explain these results.

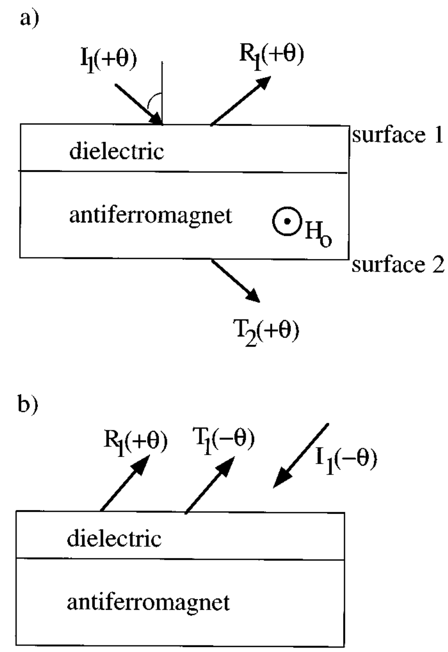

FIG. 5. Geometry for thermodynamic calculations. (a) illustration of conservation of energy process in Eq. (15); (b) illustration of detailed balance process of Eq. (21). Note that $T_1(-\theta)$ comes from an incident wave hitting surface 2 at an angle of $-\theta$.

IV. THERMODYNAMIC ARGUMENTS

In this section we show how nonreciprocal reflection is allowed for the antiferromagnet/dielectric structure but is not allowed for the simple case of a symmetric antiferromagnetic film. In addition we obtain Eqs. (13) and (14), found previously through direct calculations for a specific example, by a more general argument.

We neglect absorption so as to identify the nonreciprocity due to other effects. Also the arguments include only one polarization (s) so the results are limited to the Voigt configuration where there is no mixing between s- and p-polarizations in the reflection process.[7]

Consider the geometry illustrated in Fig. 5. A magnetic field is directed out of the page, and we consider waves incident on the structure from various directions. The angles are measured from the normal to the surface as usual. Incident angles are $+\theta$ when the light comes from the top left and $-\theta$ when the light comes from the top right. Similarly (obtainable by a rotation of 180° about the magnetic field) the incident angles are $+\theta$ for light from the bottom right and $-\theta$ for light from the bottom left.

A notation $I_1(+\theta)$ indicates the intensity of an incident wave hitting surface 1 at a positive incident angle. $R_1(+\theta)$ denotes the reflected intensity from the upper surface (1) coming from an incident wave with angle $+\theta$. Similarly $T_2(+\theta)$ corresponds to the transmitted wave leaving surface 2.

The discussion is based on two ideas: (1) Conservation of energy and, (2) detailed balance. Conservation of energy requires that the incident energy all goes into the transmitted and reflected waves. Thus considering the process shown in Fig. 5(a) we obtain

$$I_1(+\theta)=R_1(+\theta)+T_2(+\theta). \quad (15)$$

Similarly, reversing the angle of incidence, we obtain

$$I_1(-\theta)=R_1(-\theta)+T_2(-\theta). \quad (16)$$

Two additional equations can be obtained by considering the light incident from the bottom surface:

$$I_2(+\theta)=R_2(+\theta)+T_1(+\theta), \quad (17)$$

$$I_2(-\theta)=R_2(-\theta)+T_1(-\theta). \quad (18)$$

In thermal equilibrium, the incident intensities at a given angle must all be equal. Thus

$$I_1(+\theta)=I_1(-\theta)=I_2(+\theta)=I_2(-\theta). \quad (19)$$

Using this result, we may eliminate the intensities and obtain

$$R_1(+\theta)-R_1(-\theta)=T_2(-\theta)-T_2(+\theta). \quad (20)$$

There is also the condition of detailed balance—that the energy leaving a surface in a particular direction is equal to that incident going the opposite way. We consider as an example the energy leaving the top surface going to the upper right. There are two contributions to the intensity as illustrated in Fig. 5(b): (1) transmitted intensity, $T_1(-\theta)$, leaving surface 1 due to the incident wave on lower surface at angle $-\theta$ and (2) reflected intensity, $R_1(+\theta)$, due to the incident wave on upper surface at angle $+\theta$. So from detailed balance we get

$$I=T_1(-\theta)+R_1(+\theta). \quad (21)$$

Note that we have simply set the intensity to I since all the I's are the same in thermal equilibrium. Similarly considering all the other directions for exiting intensity we get

$$I=T_1(+\theta)+R_1(-\theta) \quad \text{(exit upper left)}, \quad (22)$$

$$I=T_2(+\theta)+R_2(-\theta) \quad \text{(exit lower right)}, \quad (23)$$

$$I=T_2(-\theta)+R_2(+\theta) \quad \text{(exit lower left)}. \quad (24)$$

By setting the last two equations equal one obtains

$$R_2(-\theta)-R_2(+\theta)=T_2(-\theta)-T_2(+\theta). \quad (25)$$

When we equate the right-hand sides of Eqs. (20) and (25) we obtain

$$R_1(+\theta)-R_1(-\theta)=R_2(-\theta)-R_2(+\theta). \quad (26)$$

This is an important result. This says any nonreciprocity in reflection from the top surface (surface 1) is equal to the nonreciprocity in reflection from the bottom surface (surface 2). Thus, nonreciprocity in reflection is now allowed. In contrast, there is no nonreciprocal reflection from an antiferromagnetic film without a dielectric. In that case symmetry requires

$$R_1(+\theta)=R_2(+\theta), \quad R_1(-\theta)=R_2(-\theta). \quad (27)$$

So Eq. (26) becomes

$$R_1(+\theta)-R_1(-\theta)=R_1(-\theta)-R_1(+\theta)$$
$$=-(R_1(+\theta)-R_1(-\theta)). \quad (28)$$

So we have $R_1(+\theta)-R_1(-\theta)$ equal to its negative and that only happens when $R_1(+\theta)=R_1(-\theta)$. So reciprocity in reflection is required for the case of an antiferromagnetic film and no damping.

Additional symmetry relations may be found through the thermodynamic results obtained above. For example by using Eqs. (15) and (23) we obtain Eq. (13) that $R_1(+\theta)=R_2(-\theta)$. Similarly by using Eqs. (16) and (24) we obtain Eq. (14). We can also obtain some results concerning the transmissivities. For example by combining Eqs. (15) and (21) we get

$$T_2(+\theta)=T_1(-\theta) \quad (29)$$

and by combining Eqs. (16) and (22) we get

$$T_2(-\theta)=T_1(+\theta). \quad (30)$$

We have checked Eqs. (13), (14), (29), and (30) by explicit numerical calculations and have found that they do indeed hold. These four equations provide the fundamental symmetry relations for transmissivities and reflectivities in the dielectric/antiferromagnetic structure.

The thermodynamic argument could, in principle, be extended to a nonsymmetric structure such as a vacuum/thin antiferromagnet/semi-infinite dielectric. The key idea in such an extension is that the the angles of propagation in the vacuum and in the dielectric would be different, but are connected by Snell's law. In addition, one has to be careful in the thermodynamic argument to use the definition of the transmittance as the ratio of the transmitted energy flux normal to the surface divided by the incident energy flux normal to the surface, with a similar definition of reflectance. If the angle of incidence in vacuum is denoted as θ_1 and the angle of the transmittance in the dielectric is θ_2, then Eq. (13) may be generalized to $R_1(+\theta_1)=R_2(-\theta_2)$. We have checked this result by an explicit numerical calculation. The other symmetry results of the thermodynamic argument can be similarly extended.

V. SUMMARY AND CONCLUSIONS

In this paper we have considered the reflection of electromagnetic radiation from uniaxial antiferromagnets in the common Voigt geometry. We have demonstrated, by both an explicit calculation and by general thermodynamic arguments, that one may obtain nonreciprocal reflection from structures involving both antiferromagnets and dielectrics even in the absence of damping mechanisms. This is in contrast to earlier works which showed that reflection in the Voigt geometry was reciprocal, both for semi-infinite antiferromagnets and for unsupported thin antiferromagnetic films.[6]

Our results are important for a number of reasons, both fundamental as well as practical. Future investigations of the properties of thin antiferromagnetic films and superlattices[13,14] will certainly involve fabrication on a dielectric substrate. Thus it is necessary to understand the influence of the substrate on the reflection spectrum. In terms of applications, antiferromagnets may be of interest for signal processing in the infrared. The previous work indicated that in the Voigt geometry nonreciprocal reflection was only possible with damping. However, significant damping will also degrade device performance. In this paper we show that it is in principle possible to have the best of both worlds, a system with minimal damping and which also shows significant nonreciprocity.

ACKNOWLEDGMENTS

The work of T.D. was supported by the Brazilian agency CAPES and that of R.E.C by U.S. ARO Grant No. DAAH04-94-G-0253 and by EPSRC.

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