Anomalous increasing of the intensity of field dependence optical mode ferromagnetic resonance in the exchange coupled bilayer system

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Acoustic and optical ferromagnetic resonance (FMR) in the interlayer exchange coupled Fe_{20}Ni_{80}/Co bilayer have been investigated. In the optical mode, unexpected increasing tendencies of absorption intensity at the resonance frequency, under an increasing magnetic field, has been observed. We presented analytical calculations where the exchange coupling between Co and Fe_{20}Ni_{80} layers, the magnetization and the in-plane uniaxial anisotropy are taken into account, to interpret the increasing of the maximum values of the optical permeability. The results of both experimental measurements and theoretical calculation show that such tendencies are dependent on the layer thickness \( t \), and that there is a critical field above which the absorption intensity begins to decrease. These results might help us to understand the mechanism of interlayer exchange coupling induced optical FMR and might enlighten us to find new possibility of high frequency applications of magnetic materials.

I. INTRODUCTION

Ferromagnetic resonance (FMR) is the one of the most ubiquitous technique to investigate the micro wave magnetic properties of interlayer exchange coupled magnetic multi-layers.\(^{1-5}\) By measuring the permeability spectra as a function of exchange coupling strength, magnetization, anisotropy, damping factor, magnetic layer thickness and even applied magnetic field, one can obtain a lot of basic magnetic properties of the multi-layer systems, and can test theories that describe the mechanisms of these coupling structures.

The existence of exchange coupling between magnetic moments consisting of two magnetic layers or sandwiched films have been studied by several groups. In these multilayered systems, two modes of FMR, known as acoustic and optical FMR, respectively, can be observed in experiment.\(^{6,7}\) The energy of the interaction are described by two exchange coupling parameters, the bilinear coupling \( J_1 \) and the biquadratic coupling \( J_2 \). Generally, \( J_1 \) is dominant in the films, where moments of the two layers are parallel\(^{11-13}\) or antiparallel\(^{16-18}\) to each other to meet the demand of energy minimization. While some authors have shown that the biquadratic coupling can also become dominating\(^{19,20}\), leading to a 90°-type coupling, in which the moments of the two ferromagnetic layers are vertical to each other. It was found that the biquadratic exchange coupling arise from spatial fluctuation of the interlayer thickness in a sandwiched structure.\(^{9}\)

For the \( J_1 \) exchange coupling (ferromagnetic or antiferromagnetic corresponding to \( J_1 > 0 \) or \( J_1 < 0 \)) dominated cases, the exchange energy per unit area at the interface can be written as,

\[
E_{ex} = -J \frac{M_1 \cdot M_2}{M_1 M_2} .
\]

where \( J \) is the bilinear exchange coupling coefficient with the unit of erg/cm\(^2\), \( M_1 \) and \( M_2 \) are the saturation magnetizations of the individual layers. Note that we neglected the subscript of \( J \) here for simplicity. It is found that the exchange coupling strength depend dramatically on the thicknesses of the magnetic films,\(^{9,12}\) as well as of the interlayer.\(^{10}\) Heinrich and coworkers found out that the magnetic coupling in epitaxial bcc Fe(001)/Cu(001) /Fe(001) trilayers changes from ferromagnetic to antiferromagnetic as the Cu(001) interlayer thickness changes. The exchange energy can be derived from frequency difference of the acoustic and optical modes.\(^{21,22}\) For the magnetic coupling, the resonance frequency of optical mode is higher than that of acoustic mode, while for the antiferromagnetic coupling, the optical resonance frequency is lower than that of acoustic mode. The dispersive relations and dependence of resonance intensities on exchange coupling field, saturation magnetization and anisotropic field can be obtained from solving Landau-Lifshitz-Gilbert equation.\(^{12,13,23-25}\) Note that the resonance intensity mentioned here is defined as relating to the area under FMR absorption line.

In this paper, we implemented a theoretical calculation and experimental measurements of permeability for a exchange coupled bilayer system consisting of two different ferromagnetic layers in intimate contact. Both numerical calculation and FMR results for the permeability of the bilayer are presented. Differences of acoustic and optical resonance behaviors under a increasing external magnetic field \( H \) are studied with several parameters are taken into account, including layer thickness \( t \), magnetization \( M \), in-plane uniaxial anisotropy (IPUMA) \( H_k \), damping factor \( \alpha \), and exchange strenght \( J \).

II. THEORETICAL MODEL

The schematic of the bilayer structure is shown in Fig.\(^\Dagger\) We consider that the bilayer, which is composed
The total energy $E$ consists of the Zeeman energy (interaction of the magnetizations and the magnetic field), the in-plane uniaxial anisotropy\cite{24,26} and the dipolar energy\cite{27} of FM1 and FM2 layers, as well as the exchange coupling energy between FM1 and FM2.

At equilibrium, the first derivatives of $E$ with respect to $x_i$, $y_i$ and $z_i$ must be equal to zero. It is apparent that when $x_1 = 1$ and $y_1 = z_1 = 0$, the system reach to equilibrium condition, at which the magnetizations, $\mathbf{M}_1$ and $\mathbf{M}_2$, are all lie in the $x$ direction in the films plane. Note that in the antiferromagnetic situation, however, $\mathbf{M}_1$ and $\mathbf{M}_2$ are antiparallel, i.e., $x_1 \times x_2 = -1$.

Now we consider the dynamic behaviors of $M_1$ and $M_2$ in a weak microwave magnetic field $h$. It is described by the Landau-Lifshitz equation with the Gilbert damping term

$$\frac{d\mathbf{M}_i}{dt} = \gamma \mathbf{M}_i \times \left[ \nabla M_i \left( \frac{E}{t_i} \right) + \frac{\alpha}{\gamma M_i} \frac{d\mathbf{M}_i}{dt} - h_i e^{j \omega t} \right].$$

Here $\gamma$ is the gyromagnetic ratio, $\alpha$ is the damping factor and $\omega$ is the angular frequency of $h$. Considering that the magnetization vectors excited by $h$ oscillate about the equilibrium position, the Eq. (4) can be linearized by expanding the free energy $E$ in Taylor series up to second order. The motion of the moments then can be written in matrix form as

$$\begin{pmatrix}
\gamma E_{yy} - j \alpha \omega M_1 \\
\gamma E_{yz} - j \alpha \omega M_1 \\
\gamma E_{zy} + j \alpha \omega M_1 \\
\gamma E_{aa} + j \alpha \omega M_1 \\
\gamma E_{bb} + j \alpha \omega M_1 \\
\gamma E_{ab} - j \alpha \omega M_1 \\
\gamma E_{ba} + j \alpha \omega M_1
\end{pmatrix} \times \begin{pmatrix}
\Delta y \\
\Delta z \\
\Delta a \\
\Delta b
\end{pmatrix} = \begin{pmatrix}
\gamma M_1 h_y \\
0 \\
0 \\
0
\end{pmatrix},$$

where $E_{ij} = \frac{\partial^2 E}{\partial i \partial j}$ are the second partial derivative of energy with respect to $i$ and $j$ ($i, j = y, z, a, b$) at the equilibrium position. Note that, in Eq. (5), we let $y_1 = y$, $z_1 = z$, $y_2 = a$ and $z_2 = b$ to make the calculation simple, and $\Delta y$, $\Delta z$, $\Delta a$ and $\Delta b$ denote the small variations of $y$, $z$, $a$ and $b$, respectively. By substituting Eq. (2) into Eq. (5) and simplifying the matrix equation, we can obtain
\[
\begin{pmatrix}
\Omega_{k1} - j \omega & -j \omega & 0 \\
-j \omega & \Omega_{m1} + j \omega & -\omega_{j2} \\
-\omega_{j1} & 0 & \Omega_{k2} - j \omega \\
0 & -\omega_{j1} & \Omega_{m2} + j \omega
\end{pmatrix}
\times
\begin{pmatrix}
\Delta m_{1y} \\
\Delta m_{1z} \\
\Delta m_{2y} \\
\Delta m_{2z}
\end{pmatrix}
= 
\begin{pmatrix}
\omega_{m1} h_y \\
\omega_{m2} h_y \\
0 \\
0
\end{pmatrix}
\]

with

\[
\begin{align*}
\Omega_{k1} &= \omega_0 + \omega_{k1} + \omega_{j1}, \quad \Omega_{m1} = \omega_0 + \omega_{k1} + \omega_{m1} + \omega_{j1} \\
\Omega_{k2} &= \omega_0 + \omega_{k2} + \omega_{j2}, \quad \Omega_{m2} = \omega_0 + \omega_{k2} + \omega_{m2} + \omega_{j2}
\end{align*}
\]

where \(\omega_0 = \gamma H, \quad \omega_{k1} = \frac{2 K_1}{M_1}, \quad \omega_{j1} = \frac{\gamma J}{d_1 M_1}\) and \(\omega_{m1} = 4\pi \gamma M_i(i = 1, 2)\). On solving this equation with the help of computer, one can get the real and imaginary permeability of both acoustic and optical modes for the bilayer systems numerically.

III. EXPERIMENTAL DETAILS

A set of Fe\(_{20}\)Ni\(_{80}\)/Co bilayer films with different layer thicknesses are fabricated and studied in this paper. The samples were grown by radio frequency (rf) magnetron sputter deposition on 0.43 mm thick Si (111) substrates, which were attached to oblique sample holders with oblique angle of 30° to induce the in-plane uniaxial anisotropy, in a ultrahigh vacuum chamber. The base pressure of the chamber prior to sputtering was pumped to approximately \(6 \times 10^{-5}\) Pa. The deposition pressure during the fabrication was maintained at 0.3 Pa at Ar ambient with gas rate flow of 10 SCCM (cubic centimetre per minute at STP). The sputter targets of Co metal and Fe\(_{20}\)Ni\(_{80}\) alloy are 3 inches in diameter. The rf power of 50 W was used to deposit the films. The thickness of the films was controlled by controlling the deposition time for each layer. The film structure are shown in Fig. 1 where FM1 represents Co layer and FM2 is Fe\(_{20}\)Ni\(_{80}\) layer in this case. For simplicity, the Co layer thickness \(d_1\) was fixed to be 28.6 nm, while the Fe\(_{20}\)Ni\(_{80}\) layer have varying thicknesses, \(d_2 = 29.2, 58.6\) and 68.4 nm.

The FMR measurements of the films were performed via vector network analyser (VNA, Agilent E8363B, USA) with a home made shorted-circuited microstrip line (MSL) jig connected to it through a subminiature assembly coaxial connector. The resistance of the MSL is 50 Ω to meet impedance matching of VNA's test port. During the measurements, the micro magnetic field \(h\) was perpendicular to the easy axis (EA) of the samples. For each sample, an increasing planer magnetic field \(H\) along EA was applied.

IV. RESULTS AND DISCUSSIONS

Fig. 2 shows the permeability spectra of the Fe\(_{20}\)Ni\(_{80}\)/Co bilayer measured without external dc magnetic field, with the thicknesses of Co and Fe\(_{20}\)Ni\(_{80}\) layer are \(t_1 = 28.6\) nm and \(t_2 = 58.6\) nm, respectively. Two modes of FMR are observed, the one is the acoustic mode \((\omega_{ac} = 1.99\) GHz\)) , the other one is the optical mode \((\omega_{op} = 6.31\) GHz\)). The numerical results of zero-field permeability of the Fe\(_{20}\)Ni\(_{80}\)/Co bilayer according to Eq. (6) are presented in Fig. 2 (b). The parameters used in simulations are \(4\pi M_1 = 15.17\) kG, \(H_{k1} = 48.3\) Oe, \(\alpha_1 = 0.02\) for Co layer; \(4\pi M_2 = 11.58\) kG, \(H_{k2} = 27.6\) Oe, \(\alpha_2 = 0.012\) for Fe\(_{20}\)Ni\(_{80}\) layer, and the interlayer exchange coupling strength is \(J = 0.61\) erg/cm\(^2\). The theoretical calculation are in well consistent with those of experiments as shown in Fig. 2 (a) and (b). A large resonance frequency of \(\omega_{op} = 6.31\) GHz is obtained in the optical mode owing to the presence of the interlayer exchange coupling. Fig. 3 shows the experimental measured zero-field imaginary permeability of the bilayer[Fig. 3(a)] as a comparison to the theoretical permeability of the Co and Fe\(_{20}\)Ni\(_{80}\) layer, respectively, of which the two layers are exchange coupled to each other[Fig. 3(b)]. The resonance peak of the optical mode is significantly lower.
than that of the acoustic one, as shown in Fig. 3 (a). This has already been explained by several authors that in the case of strong ferromagnetic coupling, the out-of-phase moment’s precession in optical mode [see the insert of Fig. 3 (b)] causes an offset of FMR between Co and Fe$_{20}$Ni$_{80}$ layers in the bilayer. One can see that the magnetic moments precession of the two layers in optical mode is significantly different from that in acoustic mode. For the acoustic FMR, the magnetic moments precess in-phase, while the moments precess out-of-phase for the optical mode, leading to a negative imaginary permeability of the Fe$_{20}$Ni$_{80}$ layer, as shown in Fig. 3 (b). Hence the absorption intensity of the optical mode of the bilayer are vastly crippled.

The experimental results of dc magnetic field dependent imaginary permeability are presented in Fig. 4 with the applied field lies in the film plane and along the easy directions of the bilayer. A evident downward trend of the acoustic mode was observed, however, the absorption peak of the optical mode increased unusually with the increase of the applied field, which is completely opposite to that of the acoustic mode. The difference of peak values of the two modes decreased from 1098 to 80 when $H$ increases from 0 to 225 Oe. This increasing might have a bearing on the out-of-phase precession of magnetic moments. Fig. 5 shows the external field dependent of resonance frequencies and peak values of the acoustic and optical modes. At zero field, the resonance frequency of the optical mode is 4.32 GHz higher than that of the acoustic mode, as shown in Fig. 5 (a). For the acoustic mode, the radio frequency (rf) components of the magnetization vectors of Co and Fe$_{20}$Ni$_{80}$ layers precess in phase and the dispersive relation of acoustic mode degenerate with that of single layer because that the interlayer exchange coupling produces no dynamic contributions to the resonance. For the optical mode, however, the rf component of the two layers precess out of phase and therefore the coupling produces effect exchange fields of $H_{c1} = J/d_1 M_1 = 157$ Oe and $H_{c2} = J/d_2 M_2 = 130$ Oe in Co and Fe$_{20}$Ni$_{80}$ layers, respectively. The frequencies of both acoustic and optical modes increase the dc field $H$ increased, the black and red line in Fig. 5 (a) denote the calculated dispersive relations of acoustic and optical FMR.

The variation of peak values of the acoustic and optical imaginary permeability as functions of the applied field $H$ are presented in Fig. 5 (b). Compared to that of the acoustic mode, the peak value of the optical is significantly small. Due to the presence of uniaxial anisotropy $H_k$ and applied field $H$, when applying a transverse microwave magnetic field $h$, the magnetization vectors in each layer precess about the equilibrium position either in-phase or out-of-phase corresponding to acoustic and optical mode, respectively. The transverse (along the $h$ directions) rf components of the magnetization of Co and Fe$_{20}$Ni$_{80}$ layers are parallel to each other, and it produces a relative large microwave absorption. On the contrary, the rf components in the optical mode are always antiparallel, resulting in a small absorption in optical resonance.

When an magnetic field $H$ is applied along the easy axis, the peak value of acoustic mode decreases with the increase of $H$ owing to reduction of rf components of the two layers. While that of the optical mode increases as $H$ increases, this is related to the reduction speed of rf components of the magnetization vectors in two layers.
In order to understand the field-dependent behavior of the optical resonance, we calculated the optical imaginary permeability of the Fe$_{20}$Ni$_{80}$/Co bilayer and of the separated Co and Fe$_{20}$Ni$_{80}$ layers, respectively, as shown in Fig. 6 (a). The optical permeability of Fe$_{20}$Ni$_{80}$ layer is negative, which means that the moments in Co and Fe$_{20}$Ni$_{80}$ layers resonate out of phase. One can see that the absolute peak values of separated Co and Fe$_{20}$Ni$_{80}$ layers decrease in different speed with the increase of $H$, leading to an increase of peak value of the bilayer. The peak values of the Fe$_{20}$Ni$_{80}$/Co bilayer, see in Fig. 6 (b), equals numerically to the sum of those of separated Co and Fe$_{20}$Ni$_{80}$ layers, suggesting that the net rf component of the magnetization vectors in the bilayer comes from those of vector superpositions of the two sub layers.

In Fig. 7, we plot the calculated imaginary permeability of the Fe$_{20}$Ni$_{80}$/Co bilayer with $H$ ranges from 0 to 1100 Oe. The peak value dependence on the field $H$ of both acoustic and optical modes are plotted as the insertion. Apparently, the absorption peak value of the optical mode increases gradually when $H < 800$ Oe, while it turns to decrease when $H > 800$ Oe [Insert of Fig. 7]. This is due to the vector superposition effect result in an increase of optical mode at a small field. However, when $H$ is large enough, the magnetization vectors of both layers are saturated along the field direction leading to small precession angles, which means the decrease of optical resonance.

Fig. 8 shows the absorption peak values of the Fe$_{20}$Ni$_{80}$/Co bilayers as functions of $H$ with Fe$_{20}$Ni$_{80}$ layer thickness $t_2$ increasing from 29.2 to 98.6 nm. The thickness of Co layer remains unchanged. Fig. 8 (a), (b) and (c) denote the samples with Fe$_{20}$Ni$_{80}$ layer thickness $t_2$ = 29.2 nm, 58.6 nm, and 68.4 nm, respectively. Fig. 8 (a1), (b1) and (c1) are the measured absorption peak values of optical mode, same trend can be found in the numerical results, as shown in Fig. 8 (a2), (b2) and (c2), denoting that our model corresponds well to the experimental results. From Fig. 8 (a3), (b3) and (c3) one can see that the peak value of the optical mode depends on the Fe$_{20}$Ni$_{80}$ layer thicknesses. For the sample with $t_2$ = 29.2 nm, there is a critical field of about 800 Oe, above which the peak value turns to decrease with the increase of $H$. When $t_2$ increase to 68.4 nm, the peak value of the optical mode turns to decrease at $H = 200$ Oe, as shown in Fig. 8 (c3).

V. CONCLUSION

In summery, a theoretical model describing dynamic behaviors of the magnetic moments of an exchange coupled bilayer system under a microwave magnetic field was presented. A comparison of calculated permeability and the experimental data was investigated, and found that for a ferromagnetic coupled bilayer, the FMR frequency of optical mode is much higher than that of acoustic mode, which corresponding to an exchange strength of $J = 0.61$ erg/cm$^2$. The effect field of the exchange coupling for the Co and Fe$_{20}$Ni$_{80}$ layers are 157 and 130 Oe, respectively. The dispersive relations and the imaginary permeability dependence on external dc field $H$ were studied. The upward trend of absorption peak of the optical mode is much related to the out-of-phase precession of the magnetic moments. For the Fe$_{20}$Ni$_{80}$/Co bilayer with Fe$_{20}$Ni$_{80}$ layer thickness of 58.6 nm, the absorption peak value turns to decrease as $H > 800$ Oe. When the thickness of Fe$_{20}$Ni$_{80}$ layer rises from 28.6 to 68.4 nm, the critical field reduces to 200 Oe.

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FIG. 8. (a), (b) and (c) are peak values of the imaginary permeability the bilayers with Fe$_{20}$Ni$_{80}$ layer thickness corresponding to 29.2 nm, 58.6 nm and 68.4 nm, respectively, of which the (a1), (b1) and (c1) are derived from experiment data, and the rest are the calculated results.

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