Spin waves (SWs) are collective oscillations of spins that are also known as the Damon–Eshbach (DE) mode, the backward-volume magnetostatic spin waves (BVMSWs), and the forward-volume magnetostatic spin waves (FVMSWs).\cite{3,13–16} The MSSWs are nonreciprocal and exhibit different characteristics at different interfaces of magnetic films.\cite{3,14} For both MSSWs and FVMSWs, the slopes of the dispersion relations are positive for positive frequencies and wavevectors so that both phase and group velocities are positive but not equal in general.\cite{3,14} The BVMSWs have positive phase velocities and negative group velocities. This backward propagation is useful in velocity-related applications such as the inverse Doppler effect.\cite{3,17,18} One interesting question is whether a negative group velocity can happen to MSSWs in a hybridized system. Such backward MSSWs should be very interesting and important because surface SWs are in general useful in SW-based devices such as SW filters, SW beamsplitters, and SW emitters.\cite{1,19–27}

Conventional nonreciprocal MSSWs are unidirectional and propagate along \( \mathbf{m} \times \mathbf{n} \). Here \( \mathbf{m} \) is the magnetization direction and \( \mathbf{n} \) is the normal direction of the surface that points outward of the magnetic film. Thus \( \mathbf{n} \) is up for the upper surface and down for the bottom surface.\cite{13} When two magnetic layers are stacked together, there are pairs of left- and right-propagating waves localized at the top and bottom surfaces of two layers. The two counterpropagating waves are strongly coupled. As shown by previous works on very thin bilayers, this coupling can induce nonreciprocity for the interface MSSWs\cite{28–32} yet still with positive group velocities. Recently, there are indications of backward MSSWs in YIG/YIG/GaAs multilayers.\cite{13} However, only few data points with a very long wavelength \( (k < 5 \text{ rad m}^{-1}) \) were provided, probably due to the small saturation magnetization of YIG as well as the similarity of two YIG layers. According to the previous theoretical studies,\cite{29} we expect that backward MSSWs are possible when two counterpropagating MSSWs are sufficiently different from each other (can be realized in materials with very different saturation magnetizations) and their coupling is strong enough. The reason that backward MSSWs were not found before is either because the film thickness is too small so that the coupling of MSSWs on different surfaces is important or because two MSSWs are too similar. Thus, we use thicker bilayers (Co/FeNi) in this study. In addition to the usual nonreciprocal property of MSSWs from the magnetostatic interactions, the Dzyaloshinskii–Moriya interaction (DMI) is chiral in nature and can also lead to an asymmetric dispersion relation that, in turn, results in the nonreciprocal behavior of MSSWs.

Spin waves (SWs) in magnetic films have attracted much attention in recent years because of great potential applications in spintronic devices.\cite{1–7} SWs are collective oscillations of spins at the gigahertz frequency in typical ferromagnetic materials.\cite{8,9} Their wavelengths are of orders of magnitude shorter than that of electromagnetic waves at the same frequency so that SWs can be used in micro- or nanosize SW devices.\cite{2,3,8–10} SWs can be classified by the dominating interactions as exchange types and magnetostatic ones.\cite{11,12} It is well known that there are three different kinds of magnetostatic SWs in thin ferromagnetic films. They are the magnetostatic surface spin waves (MSSWs) that are also known as the Damon–Eshbach (DE) mode, the magnetic volume spin waves (MVSWs) also called the FVMSWs, and the forward-volume magnetostatic spin waves (FVMSWs).\cite{11,13–16} The MSSWs are nonreciprocal and exhibit different characteristics at different interfaces of magnetic films.\cite{3,14} For both MSSWs and FVMSWs, the slopes of the dispersion relations are positive for positive frequencies and wavevectors so that both phase and group velocities are positive but not equal in general.\cite{3,14} The BVMSWs have positive phase velocities and negative group velocities. This backward propagation is useful in velocity-related applications such as the inverse Doppler effect.\cite{3,17,18} One interesting question is whether a negative group velocity can happen to MSSWs in a hybridized system. Such backward MSSWs should be very interesting and important because surface SWs are in general useful in SW-based devices such as SW filters, SW beamsplitters, and SW emitters.\cite{1,19–27}
In this Letter, we use Brillouin light scattering (BLS) to obtain dispersion relations of surface SWs of the Co/FeNi bilayer system. These two materials are chosen because of their distinct saturation magnetizations and other magnetic properties. The MSSWs and perpendicular standing spin waves (PSSWs) were observed. The dispersion relations of MSSWs obtained from the anti-Stokes measurements are backward MSSWs whereas that obtained from Stokes measurements has the usual MSSW spectrum. The experimental results agree well with the numerical calculations when true material parameters are used.

Our Co(30)/FeNi(t) bilayer films were deposited on single-crystal Si (111) substrates by radio frequency (RF) magnetron sputtering. The numbers in parentheses are the film thicknesses in nanometers. The FeNi layer thickness t varied from 30 to 50 nm that was controlled by varying the sputtering time (with 10% uncertainty in thickness). The base pressure of the sputtering chamber was about $5 \times 10^{-5}$ Pa. The pressure inside the chamber was 0.3 Pa and the RF power was 50 W during the sputtering process. The static magnetization of Co(30)/FeNi(t) bilayer was measured by a vibrating sample magnetometer (VSM).

Figure 1 shows the in-plane magnetic hysteresis loops of bilayer samples at room temperature when the field is applied along the easy magnetization axis (a) and along the hard magnetization axis (b). As shown in Figure 1, the magnetic hysteresis loops are smooth in both cases. This means that the bilayer is coupled like a single layer. Ferromagnetic resonance (FMR) measurements and relevant experimental parameters can be found in Supporting Information.

BLS measurements were carried out for the Co/FeNi bilayer at room temperature. Wavenumber-selective BLS is very effective in obtaining the dispersion relation of surface waves. The scattering process can be described by inelastic scattering and is known as the 180° backscattering geometry (see the inset of Figure 2a). In experiments, the external magnetic field H is in plane and perpendicular to the MSSW wave vector. The incident plane of the laser light is perpendicular to the external field, as shown in the inset of Figure 2a. The wave vector of the MSSWs is $k_l = \frac{4\pi \sin \theta}{\lambda}$, where $\theta$ is the incident angle of the light and $\lambda$ is the laser wavelength (532 nm). The laser power incident on the surface of the sample is about 30 mW in the experiments. In our work, the range of $k_l$ varies from $-16.53$ to $16.53 \mathrm{rad} \mu \mathrm{m}^{-1}$ with a step size of $1.18 \mathrm{rad} \mu \mathrm{m}^{-1}$ by varying the laser light incident angle. In our experiment, the distance between the mirrors is 5 mm. The free spectral range of the interferometer is $\pm 34.94 \mathrm{GHz}$ along with 1024 channels in the analog–digital conversion. The resolution of the scanning frequency is 0.068 GHz. The typical BLS spectra for the FeNi(30) single layer and Co(30)/FeNi(30) bilayer are shown in Figure 2 with the absolute value of the wave vector as $k_l = 16.53 \mathrm{rad} \mu \mathrm{m}^{-1}$ and external magnetic field as $H = 500 \mathrm{Oe}$. In the light-scattering process, the wave vector change in the anti-Stokes scattering (positive frequency) is defined as the positive and that in the Stokes scattering (negative frequency) is defined as the negative. From Figure 2, both the FeNi(30) single layer and the Co(30)/FeNi(30) bilayer spectrum display four different peaks. The two peaks of the larger signal are for MSSWs and the other two peaks with weaker signals for PSSWs. For the FeNi(30) single layer, MSSWs show one Stokes peak $f(-k)$ with a negative frequency shift ($-12.87 \mathrm{GHz}$) and one anti-Stokes peak $f(k)$ with a positive frequency shift ($12.82 \mathrm{GHz}$). This suggests that the two MSSWs propagate in opposite directions and with the same absolute value of frequency within experimental error. However, for the Co(30)/FeNi(30) bilayer, the MSSWs of the Co(30)/FeNi(30) bilayer show one Stokes peak $f(-k)$ with a negative frequency shift ($-13.28 \mathrm{GHz}$) and one anti-Stokes peak $f(k)$ with a positive frequency shift ($9.67 \mathrm{GHz}$). This suggests that the two MSSWs propagate in opposite directions and have a distinct frequency difference. In contrast, for the PSSW mode, the frequencies of both FeNi(30) single layer and Co(30)/FeNi(30) bilayer are almost the same.

The dispersion relation of FeNi(30) single layer and Co(30)/FeNi(t) bilayers with various FeNi thicknesses of $t = 30$, 40, and 50 nm was measured by BLS. Figure 3 shows the density plot of BLS intensity in the frequency–wave vector plane. The left side of the vertical black dotted line corresponds to negative wave vector (Stokes) and the right side of the vertical black dotted line corresponds to positive wave vector (anti-Stokes). In the experiments, a constant magnetic field of $H = 500 \mathrm{Oe}$ is along the x direction and $k_l$ varies from $-16.53$ to $16.53 \mathrm{rad} \mu \mathrm{m}^{-1}$ along the y direction. As shown in Figure 3a, the lower frequency mode is MSSWs and the higher frequency mode is PSSWs. For the lower frequency mode, the frequency increases with the wave vector. For the higher frequency mode, the frequency does not change with the wave vector. As shown in Figure 3b, c, d, the lower frequency mode of the Co(30)/FeNi(t) bilayers is

![Figure 1](image1.png)  
**Figure 1.** The normalized in-plane hysteresis loops of Co/FeNi bilayer. a) The hysteresis loops of FeNi layer of various thicknesses. The field is along the easy magnetization axis. b) The hysteresis loops of FeNi layer of various thicknesses. The field is along the hard magnetization axis.
MSSWs from the FeNi layer (no signal from Co due to the penetration depth of the laser light). The higher frequency mode is a mixture of PSSWs and MSSWs, dominated by PSSWs (pure PSSW modes are drawn in the white dashed line). For the lower frequency mode, \( f(k^-) \) increases with the increase in wave vector \( k \), which behaves like the common MSSWs. However, \( f(k^+) \) decreases with the increase in wave vector \( k \), a feature of backward MSSWs. The data of the negative magnetic field (see Supporting Information) further prove the results of backward MSSWs.

To further substantiate our results, we compute the MSSW spectrum of Co/FeNi bilayer. The sample is schematically shown in the inset of Figure 3d. The sample is assumed to be in the \( \omega-k \) plane for FeNi(30) single layer, and for Co/FeNi bilayer with various FeNi layer thicknesses of 30 nm. The two sides of the vertical black dotted line correspond to different wave vector coordinates, respectively. The red dashed curves are of the calculated MSSW spectrum. The white dashed curves are of the PSSW spectrum. The fixed external field \( H = 500 \) Oe is applied along the x direction. Inset: Schematic diagram of the bilayer system.

\[ \frac{\partial \mathbf{M}_i}{\partial t} = -\gamma \mathbf{M}_i \times \left( \frac{A_i}{\mu_0 M_s^2} \nabla^2 \mathbf{M}_i + \mathbf{M}_i \right) + \frac{\alpha_i}{M_s^2} \mathbf{M}_i \times \frac{\partial \mathbf{M}_i}{\partial t} \] (3)

where \( A_i \) are the intralayer exchange constants in FeNi and Co, and \( \alpha_i \) is the damping coefficient. If we do not consider surface effects such as surface anisotropy or surface spin transfer torque,\(^{[42]}\) the LLG equation requires \( \frac{\partial \mathbf{M}_i}{\partial z} = 0 \) at boundaries 01 and 23, and the condition at boundary 12 is\(^{[44]}\)

\[ \frac{1}{2} A_{12} \left( \mathbf{M}_1 \times \mathbf{M}_2 - \mathbf{M}_2 \times \mathbf{M}_1 \right) + \frac{A_{12}}{M_s^2} \left( \mathbf{M}_1 \times \frac{\partial \mathbf{M}_1}{\partial z} \right) = 0 \] (4)

\[ \frac{1}{2} A_{12} \left( \mathbf{M}_2 \times \mathbf{M}_1 + \mathbf{M}_1 \times \mathbf{M}_2 \right) + \frac{A_{12}}{M_s^2} \left( \mathbf{M}_2 \times \frac{\partial \mathbf{M}_2}{\partial z} \right) = 0 \] (5)

where \( A_{12} \) is the interfacial exchange constant in units of J/m². We expand \( \mathbf{M}_i \) and \( \mathbf{H}_i \) around their equilibrium values

\[ \mathbf{M}_{1,2} = \left( \mathbf{M}_{1,2}, 0, 0 \right) + \mathbf{m}_{1,2} \] (6)

\[ \mathbf{H}_i = \left( H, 0, 0 \right) + \mathbf{h}_i \] (7)

and keep only linear terms in the small quantities, \( \mathbf{m}_{1,2} \) and \( \mathbf{h}_i \). By assuming a harmonic form \( \mathbf{m}_{1,2}, \mathbf{h}_i \sim e^{i(\omega\cdot k)} \) and applying...
the boundary conditions, we obtain a secular equation whose solution is the dispersion relation. The two lower frequency branches are MSSWs shown in Figure 3, as the red dashed curves that agree well with the experiment (bright green). The parameters are $M_{s1} = 0.77 \times 10^6$ A m$^{-1}$, $M_{s2} = 1.42 \times 10^6$ A m$^{-1}$, and $\gamma = 28$ GHz T$^{-1}$, obtained from FMR experiments. The exchange constants as fitting parameters are $A_1 = 10$ pJ m$^{-1}$, $A_2 = 11$ pJ m$^{-1}$, and $A_{12} = 20$ mJ m$^{-2}$ that are reasonable. Indeed we also include the anisotropy field; see Supporting Information. The white dashed lines in Figure 3 indicate the PSSW frequency that decreases with the thickness of FeNi. Although the two layers are coupled, PSSWs can only be detected in the upper layer as the total thickness of the bilayer is larger than the laser light penetration depth.

**Figure 4** shows the theoretical group velocity as a function of the positive wave vector with various model parameters. For different bilayer thicknesses, the phase velocities $v_p = \omega/k$ are always positive, as shown in Figure 3. While the group velocities $v_g = \partial \omega/\partial k$ are negative for most wave vectors, as shown in Figure 4. For the FeNi layers of $t = 30, 40,$ and 50 nm, group velocities change sign at the wave vectors of $k_1 = 3.3, 2.45,$ and 1.95 rad $\mu$m$^{-1}$, respectively. The regions with opposite signs of phase velocities and group velocities are the backward SWs. It occurs when two magnetic layers have very different saturation magnetizations. It is the magnetostatic interaction between the two layers that leads to the nonreciprocal behavior of SWs and the backward MSSWs.\[28\] The largest magnitudes of negative group velocities for FeNi layers of $t = 30, 40,$ and 50 nm, are respectively 0.52, 0.64, and 0.72 $\times 10^3$ m s$^{-1}$. It can be understood from the fact of increases in magnetostatic interaction and unchanging exchange interaction with the increases in layer thickness. As a result, the nonreciprocity becomes stronger,\[30\] and, eventually, MSSWs become backward.

In summary, the backward MSSWs were observed in the coupled Co/FeNi bilayer by BLS. The results were further confirmed by theoretical calculations. We revealed that the coupling of two counterpropagating MSSWs at the interface, through the magnetostatic interaction between two layers, is responsible for BMSSWs. Furthermore, the largest magnitudes of negative group velocities increase with the thickness of FeNi.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

backward surface spin waves, Brillouin light scattering, negative group velocities

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**Figure 4.** Theoretical calculations of group velocity as a function of positive wave vector for different bilayer thicknesses.
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