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Nonreciprocal propagation of surface acoustic wave in Ni/$LiNbO_3$

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We investigated surface acoustic wave propagation in a Ni/$LiNbO_3$ hybrid device. We found that the absorption and phase velocity are dependent on the sign of the wave vector, which indicates that the surface acoustic wave propagation has nonreciprocal characteristics induced by simultaneous breaking of time-reversal and spatial inversion symmetries. The nonreciprocity was reversed by 180° rotation of the magnetic field. The origin of the nonreciprocity is ascribed to interference of shear-type and longitudinal-type magnetoelastic couplings.

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Breaking of spatial inversion symmetry (SIS) and time-reversal symmetry (TRS) has a large influence on the spin-dependent energy band dispersion of electrons, $\epsilon(\boldsymbol{k},\sigma)$, where $\sigma = \uparrow,\downarrow$ is the electron spin state, and $\boldsymbol{k}$ is the wave vector. When both the symmetries are preserved, $\epsilon(\boldsymbol{k},\uparrow) = \epsilon(\boldsymbol{k},\downarrow) = \epsilon(-\boldsymbol{k},\uparrow) = \epsilon(-\boldsymbol{k},\downarrow)$. If only TRS is broken, $\epsilon(\boldsymbol{k},\uparrow) = \epsilon(-\boldsymbol{k},\uparrow)$ but $\epsilon(\boldsymbol{k},\uparrow) \neq \epsilon(-\boldsymbol{k},\downarrow)$. The energy difference corresponds to spin splitting in ferromagnetic metals. For systems with TRS but without SIS, $\epsilon(\boldsymbol{k},\uparrow) = \epsilon(-\boldsymbol{k},\downarrow)$ but $\epsilon(\boldsymbol{k},\uparrow) \neq \epsilon(-\boldsymbol{k},\uparrow)$ when $\boldsymbol{k}$ is along some specific crystal axis. The Rashba effect [1] and the Dresselhaus effect [2] are examples of this type of spin splitting. The degeneracy of $+\boldsymbol{k}$ and $-\boldsymbol{k}$ electronic states is completely lifted by the breaking of both the symmetries.

The symmetry dependence of energy dispersion should be common to other elementary excitations in solids, such as photons, magnons, phonons, etc. For the photonic case, the circular polarization corresponds to the spin state, and the slope of the energy dispersion is the inverse of refractive index. In this sense, natural optical activity and Faraday rotation correspond to the spin splitting in SIS-broken and TRS-broken electronic systems, respectively. Recently, nonreciprocal directional dichroism observed in systems without TRS or SIS has been investigated extensively [3–5]. This phenomenon is caused by the difference of the refractive indices between $+k$ and $-k$ photons irrespective of the polarization direction, which originates from the photonic energy dispersion asymmetry in TRS-broken and SIS-broken systems. Quite recently, similar nonreciprocal propagation of magnons has been observed in SIS-broken ferromagnetic films [6,7] and chiral ferromagnets [8,9] originating from the Dzyaloshinskii-Moriya interaction.

A few papers have reported the nonreciprocal propagation of acoustic waves. Heil *et al.* observed nonreciprocal acoustic waves on an Al surface at 4 K, caused by coupling between the lattice deformation and long-lived cyclotron motion of conduction electrons at low temperature [10]. On the other hand, Fleury *et al.* fabricated a macroscopic nonreciprocal acoustic wave circulator utilizing the air blown by commercial CPU fans [11]. Here, we study the nonreciprocal propagation of surface acoustic waves in ferromagnetic Ni/$LiNbO_3$ hybrid devices. The advantages of ferromagnetic nonreciprocal acoustic wave devices over the previously studied nonreciprocal devices are that nonreciprocity can be realized even at room temperature, and that devices can be microfabricated. A surface acoustic wave (SAW) is an acoustic wave propagating on the surface (Rayleigh wave). The amplitude exponentially decays in the interior of the material. It has circular polarization, and the polarization direction is reversed upon reversal of the wave vector direction. SAWs can be efficiently excited and detected by electromagnetic waves on piezoelectric materials such as $LiNbO_3$ with the use of interdigital transducers (IDTs) [12]. Recently, Weiler and co-workers have reported that ferromagnetic resonance can be excited by SAWs in a ferromagnetic Ni/$LiNbO_3$ hybrid device [13,14]. It should be noted that the surface is obviously an SIS-broken system, and TRS is broken by the ferromagnet, and therefore, nonreciprocal propagation of the SAWs is expected in such a device. We investigated SAW propagation in a similar Ni/$LiNbO_3$ hybrid device and show that the SAW propagation was certainly nonreciprocal. We inferred that interference between shear-type and longitudinal-type magnetoelastic couplings plays an important role in the nonreciprocal propagation of SAWs.

To excite a high-frequency SAW that couples to ferromagnetic resonance, we prepared a SAW device composed of two microfabricated IDTs and a ferromagnetic Ni thin film evaporated on $LiNbO_3$ substrates, as shown in Fig. 1(a). We fabricated the IDTs by a photolithography technique and by electron beam evaporation of aluminum. The finger length and width of the IDTs were 1 mm and 5 μm, respectively. The space between fingers was also 5 μm. The number of fingers in the IDTs was 30. A Y-cut $LiNbO_3$ substrate was used, and the SAW propagation direction was along the Z direction of $LiNbO_3$. The fundamental frequency of the SAWs was estimated to be $\simeq$170 MHz [12]. A ferromagnetic nickel film with a thickness of 30 nm was sputtered onto the $LiNbO_3$ substrate between the two IDTs.

Figure 1(b) shows the microwave transmission spectrum between the two IDTs, measured by a vector network analyzer (Agilent E5071C). We observed a sharp peak around 170 MHz, which is consistent with the designed fundamental frequency of the IDTs. In addition, the 5th, 9th, and 13th harmonic peaks were also discerned. In this study, we used the 13th harmonic resonance ($f \simeq 2.24$ GHz), which was the highest observed in our device and was comparable with the ferromagnetic resonance frequency of the Ni film, as discussed later.

There was some direct electromagnetic crosstalk between the IDTs, in addition to the coupling via SAW propagation. In order to separate the SAW signal from the electromagnetic crosstalk, we performed a time-domain measurement using pulsed microwaves. Figures 1(c) and 1(d) display demodulated

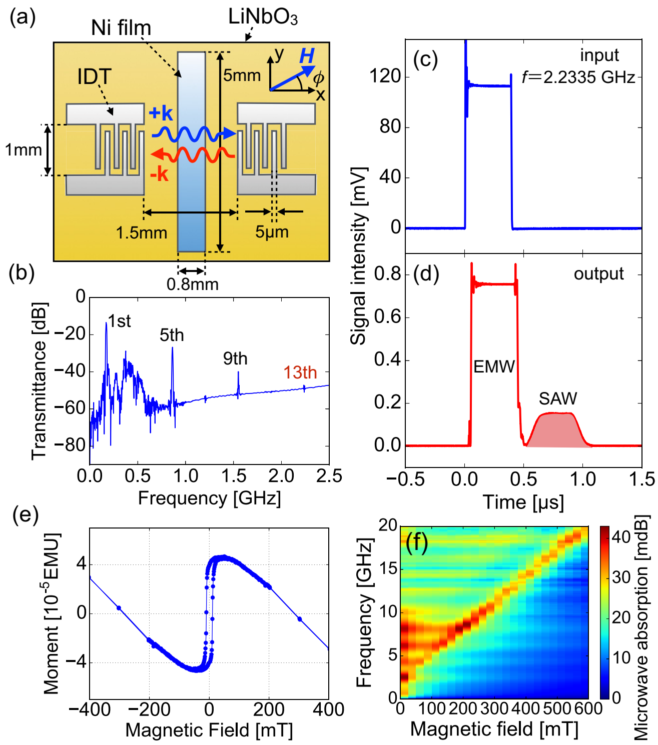

FIG. 1. (a) A schematic diagram of a Ni/$LiNbO_3$ hybrid SAW device used in this study. A rectangular ferromagnetic Ni film was evaporated on device. (b) Typical transmission spectrum. The 13th harmonic of approximately 2.23 GHz was used in this study. (c),(d) Amplitude oscillograms for (c) input and (d) output signals at $f = 2.2335$ GHz. Note that SAW signal and EMW noise were successfully separated in the time domain using a microwave homodyne circuit. (e) Magnetization curve of Ni film. In the higher magnetic field region, the magnetization is decreased because of a diamagnetic signal from the $LiNbO_3$ substrate. (f) Microwave absorption of Ni film at 280 K.

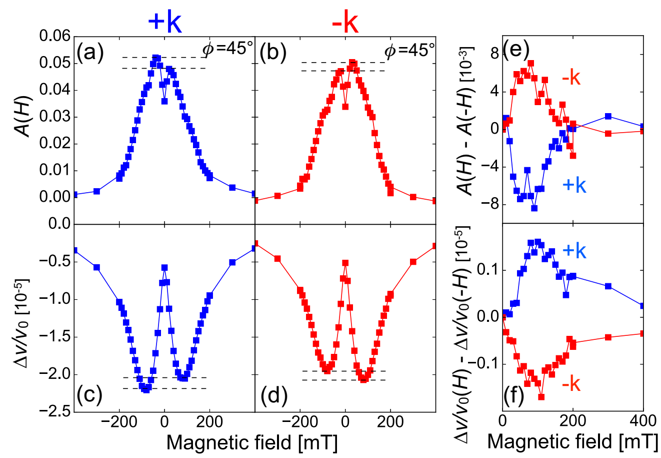

FIG. 2. (a),(b) Absorption, $A(H)$, of SAWs propagating along (a) "$+k$" and (b) "$-k$" directions shown in Fig. 1(b) as a function of external magnetic field along $\phi = 45^\circ$. (c),(d) Relative change in phase velocity of SAW $\Delta v/v_0$ propagating along (c) "$+k$" and (d) "$-k$" directions as a function of magnetic field along $\phi = 45^\circ$. The dashed lines emphasize the asymmetry of magnetic field dependence of absorption and the relative change in phase velocity. (e),(f) The nonreciprocities of (e) the absorption, $A(H) - A(-H)$, and (f) relative change in phase velocity, $\Delta v/v_0(H) - \Delta v/v_0(-H)$, for $+k$ and $-k$.

microwave input and output signals through the SAW device, respectively. Using a large difference in group velocity between the SAW and the electromagnetic waves (EMWs), we successfully separated them in the time domain. Our measurement system was based on a microwave homodyne circuit [15], which allowed simultaneous and highly accurate measurement of both the amplitude and phase for the transmitted SAW signal. We performed the SAW propagation measurement at 280 K in a split-type superconducting magnet (Spectromag, Oxford instrument) equipped with a rotation stage. With this experimental setup, the external magnetic field could be applied in any direction in the SAW device plane. The in-plane field direction was specified by the angle ϕ, as shown in Fig. 1(a).

Figures 2(a) and 2(c) show the SAW absorption $A(H)$ and the relative decrease of phase velocity $\Delta v/v_0(H)$ as a function of external magnetic field along $\phi = 45^\circ$. Here, $A(H)$ is defined by the decrease in transmittance normalized by the transmittance at high enough field (the absorption due to magnetoelastic coupling is minimal at high field, as discussed below). The decrease in phase velocity is also normalized by the high-field value. The presented data are averages of those taken by field increasing measurement and field decreasing measurement. With decreasing magnetic field from +400 mT, the absorption increased considerably. It showed a maximum accompanying a small dip structure around zero magnetic field. Then, the absorption decreased again toward high negative magnetic field. Corresponding to the enhancement of the absorption, the phase velocity decreased in the low-magnetic-field region. Around zero magnetic field, it showed a sharp peak corresponding to the dip structure of the absorption. We plot in Fig. 1(e) the magnetization curve of Ni film with the thickness of 40 nm sputtered on $LiNbO_3$ substrate, which is measured by a magnetic property measurement system (Quantum Design). The magnetization reversal process was restricted to the low-field region below 30 mT.

Therefore the dip structure of the SAW absorption and the sharp peak of the phase velocity were caused by the magnetization reversal process. On the other hand, the magnetic field dependencies of SAW absorption and phase velocity in the higher magnetic field region were induced by the change in the difference in frequency between of the SAWs and ferromagnetic resonance (FMR). In Fig. 1(f), we show the microwave absorption of Ni film with the thickness of 40 nm sputtered on $LiNbO_3$ substrate at 280 K as functions of frequency and magnetic field, which is measured in the measurement system shown in literature [8]. The absorption peak frequency due to FMR was observed to be increased linearly with the magnetic field in the high-field region, while SAW frequency did not show any magnetic field dependence. If the linear magnetic field dependence is extended to zero magnetic field, the zero field frequency is around 2 GHz, which is comparable with the SAW frequency used in our measurement. Thus the magnetic field effectively tuned the energy difference correlated to the magnetoelastic coupling.

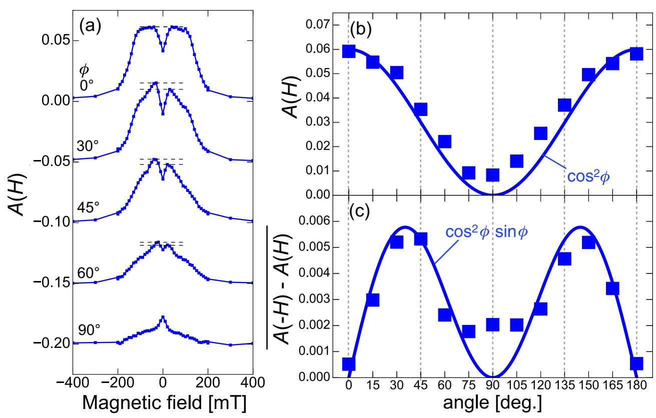

FIG. 3. (a) Magnetic field dependence of SAW absorption $A(H)$ in several magnetic field directions. The dashed lines emphasize the asymmetry of the magnetic field dependence. (b) The SAW absorption $A(H)$ at $\mu_0 H = 100$ mT as a function of angle ϕ. (c) Averaged nonreciprocity of absorption $\overline{A(-H) - A(H)}$ as a function of angle ϕ. We averaged $A(-H) - A(H)$ in the field region of 40 mT $\leqslant |\mu_0 H| \leqslant$ 150 mT. Solid lines are theoretical angle dependence (see text).

The magnetic field dependence of SAW absorption reflected the field dependent magnetoelastic coupling.

Importantly, we observed a difference in absorption between negative and positive magnetic fields, as emphasized by the dashed lines in Fig. 2. Correspondingly, the phase velocity also showed asymmetric field dependence. These asymmetries cannot be ascribed to the magnetization process because the magnetic moment saturated above 30 mT. Therefore, the asymmetries were likely caused by the intrinsic difference of SAW absorption between positive and negative magnetic fields, denoted as nonreciprocal SAW propagation. The SAW absorption $A(k,H)$ for wave vector k at magnetic field H should satisfy the relation $A(k,H) = A(-k,-H)$. To check this relation, we observed the absorption and the phase velocity for SAWs with opposite wave vector [Figs. 2(b) and 2(d)]. As is clearly shown in this figure, the field asymmetries of absorption and phase velocity became reversed when the SAWs propagated along the opposite direction. Figures 2(e) and 2(d) show the differences in absorption and the relative change in phase velocity between positive and negative magnetic fields. The differences were completely reversed by reversal of propagation direction.

These results demonstrate nonreciprocal SAW propagation in this Ni/$LiNbO_3$ hybrid device. To study the nature of nonreciprocal SAW propagation, we investigated the magnetic field angle dependence of SAW absorption. Figure 3(a) shows the SAW absorption at various ϕ. The absorption was large at $\phi = 0^\circ$ and decreased with increasing ϕ. Figure 3(b) shows the absorption at $\mu_0 H = 100$ mT as a function of ϕ. The absorption seems almost proportional to $\cos^2\phi$. The nonreciprocity also showed a large angle dependence. While the nonreciprocity was negligible at $\phi = 0^\circ$, the absorption at $\phi = 30^\circ$ clearly showed the nonreciprocal behavior. The nonreciprocity decreased with increasing ϕ from 30°. Figure 3(c) shows the average nonreciprocity $\overline{A(-H) - A(H)}$ as a function of ϕ. Here, $\overline{A(-H) - A(H)}$ is defined as the average difference of normalized absorptions between positive and negative magnetic fields in the field region of 40 mT $\leqslant |\mu_0 H| \leqslant$ 150 mT. The nonreciprocity steeply increased with ϕ in the low-ϕ region. After showing the maximum around $\phi = 30^\circ$, it gradually decreased in the larger ϕ region.

To discuss the microscopic origin of these experimental observations, let us theoretically discuss the SAW excitation of FMR. In general, the lowest order term of magnetoelastic coupling energy density F_{coupling} that does not vanish under the time-reversal symmetry is written with components of magnetization vector m_i, strain tensors ϵ_{ij}, and four rank tensors of magnetoelastic coupling constant $B_{i,j,k,l}$;

$$F_{\text{coupling}} = \sum_{i,j,k,l=x,y,z} B_{i,j,k,l} m_i m_j \epsilon_{kl}, \tag{1}$$

where $B_{i,j,k,l}$ is symmetrical with respect to the pairs of subscripts i,j and k,l. In a polycrystalline thin film, F_{coupling} is unchanged under any rotation around the normal axis of the film and any mirror operations with respect to the planes including the normal axis.

Considering this symmetry, magnetoelastic coupling energy has the following form:

$$\begin{aligned} F_{\text{coupling}} = {} & m_x^2(b_1\epsilon_{xx} + b_2\epsilon_{yy} + b_3\epsilon_{zz}) \\ & + m_y^2(b_2\epsilon_{xx} + b_1\epsilon_{yy} + b_3\epsilon_{zz}) \\ & + m_z^2(b_4(\epsilon_{xx} + \epsilon_{yy}) + b_5\epsilon_{zz}) \\ & + 2m_x m_y(b_1 - b_2)\epsilon_{xy} + 4m_y m_z b_6 \epsilon_{yz} \\ & + 4m_z m_x b_6 \epsilon_{zx}, \end{aligned} \tag{2}$$

where $b_1 = B_{x,x,x,x}$, $b_2 = B_{x,x,y,y}$, $b_3 = B_{x,x,z,z}$, $b_4 = B_{z,z,x,x}$, $b_5 = B_{z,z,z,z}$, $b_6 = B_{x,z,x,z}$. Here, we assume that the film is perpendicular to the z axis. When the SAW propagates along the x axis, ϵ_{xx}, ϵ_{zz}, and ϵ_{zx} oscillate as follows [16]:

$$\epsilon_{xx} = \epsilon_{xx0} \exp[i(kx - \omega t)], \tag{3}$$

$$\epsilon_{zz} = \epsilon_{zz0} \exp[i(kx - \omega t)], \tag{4}$$

$$\epsilon_{zx} = i\epsilon_{zx0} \exp[i(kx - \omega t)], \tag{5}$$

where ω is the SAW frequency, and ϵ_{xx0}, ϵ_{zz0}, and ϵ_{zx0} are real and decrease exponentially along the z direction. The phases of longitudinal strains ϵ_{xx}, ϵ_{zz}, and shear strain ϵ_{zx} differ by $\pi/2$, forming the ellipsoidal polarization of the elastic oscillation. When k is reversed, the ellipsoidal direction becomes reversed, and ϵ_{zx0} shows a different sign. We also assumed that the static magnetization was parallel to the device plane:

$$\begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix} = \begin{pmatrix} m_0 \cos\phi \\ m_0 \sin\phi \\ 0 \end{pmatrix}. \tag{6}$$

Then, the effective ac magnetic field induced by the SAWs propagating along the x axis is

$$-\nabla_{\boldsymbol{m}} F_{\text{coupling}} = \mu_0 \begin{pmatrix} h_x \\ h_y \\ h_z \end{pmatrix} = -\begin{pmatrix} 2b_1 m_x \epsilon_{xx} + 2b_3 m_x \epsilon_{zz} \\ 2b_2 m_y \epsilon_{xx} + 2b_3 m_y \epsilon_{zz} \\ 4b_6 m_x \epsilon_{zx} \end{pmatrix} = -\begin{pmatrix} (2b_1 m_0 \epsilon_{xx0}\cos\phi + 2b_3 m_0 \epsilon_{zz0}\cos\phi)\exp[i(kx-\omega t)] \\ (2b_2 m_0 \epsilon_{xx0}\sin\phi + 2b_3 m_0 \epsilon_{zz0}\sin\phi)\exp[i(kx-\omega t)] \\ 4ib_6 m_0 \epsilon_{zx0}\cos\phi \exp[i(kx-\omega t)] \end{pmatrix}. \quad (7)$$

We deduce the effective magnetic field induced by the SAWs acting on the Ni film, assuming that the characteristics of the SAWs are not affected by the magnetic excitation in the ferromagnetic Ni film. This is justified by the small SAW absorption (6%). Only the ac magnetic field perpendicular to the magnetization, $h_\perp$, can excite FMR. The in-plane and out-of-plane components of $h_\perp$ are $h_x \sin\phi - h_y \cos\phi$ and $-h_z$, respectively. In addition, the susceptibility of FMR for a right-handed circularly polarized field χ_+ is large, whereas that for a left-handed field χ_- is negligible [17]. Thus, the SAW absorption induced by magnetoelastic FMR is proportional to

$$\chi_+''|h_\perp^+|^2 \propto |h_x \sin\phi - h_y\cos\phi - ih_z|^2 \propto |(b_1 - b_2)\epsilon_{xx0}\cos\phi\sin\phi + 2b_6\epsilon_{zx0}\cos\phi|^2, \quad (8)$$

where χ_+'' is the imaginary part of the magnetic susceptibility for the right-handed circularly polarized field $h_\perp^+$. As mentioned above, the SAW absorption was roughly proportional to $\cos^2\phi$, which indicates that the shear-type magnetoelastic coupling (b_6) was dominant over the longitudinal-type magnetoelastic couplings (b_1 and b_2) for our device [18].

When the vector of the SAWs is reversed, the circular direction of the elastic wave polarization is reversed, which is reflected by the sign of ϵ_{zx0}. The nonreciprocity can be induced by interference of the longitudinal- and shear-type magnetoelastic couplings and proportional to

$$(b_1 - b_2)b_6\epsilon_{xx0}\epsilon_{zx0}\cos^2\phi\ \sin\phi. \quad (9)$$

It seems that the minor b_1,b_2 term was still active, so that the nonreciprocal SAW propagation was finite. The angle dependence of nonreciprocity roughly coincided with Eq. (9) as shown in Fig. 3(c), which ensures the correctness of our theoretical analysis.

In conclusion, we successfully observed the nonreciprocal propagation of surface acoustic waves in a Ni/$LiNbO_3$ hybrid device. We inferred that interference of different magnetoelastic couplings is crucial for the nonreciprocity.

If the ratio of magnetic constants b_1, b_2, and b_6 is optimally tuned, it should be possible to realize complete nonreciprocal propagation, according to Eq. (8); namely, SAWs are attenuated by magnetic resonance along one direction but are totally unaffected in the opposite propagation direction. While the phonon diode effect has been extensively studied in acoustic metamaterials, which consist of periodic arrays of purely elastic objects [19], our observations may pave a route to nonreciprocal microwave and phononic functionalities based on media without SIS and TRS.

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dependence implied that the function form of magnetoelastic free energy is device dependent. The shear-type magnetoelastic coupling [b_6 term in Eq. (8)] is dominant in the present work, while the longitudinal magnetoelastic coupling [b_1,b_2 terms in Eq. (8)] is dominant in Weiler *et al.* The origin of device dependence has not been clarified yet and should be elucidated because the fine tuning of magnetoelastic coupling is crucial in order to achieve large nonreciprocal propagation.

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