Magnons and Phonons Optically Driven out of Local Equilibrium in a Magnetic Insulator

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The emerging field of spin caloritronics has been stimulated by a number of recent discoveries, such as large magnon thermal conductivity [1,2], spin Seebeck effect [3–10], spin Peltier effect [11,12], magneto-Seebeck effect [13], and thermal spin transfer torque (STT) [14,15]. These discoveries hold promise for new technologies based on thermally driven spin transport phenomena. For example, thermal STT improves upon current STT-based memory devices by reducing the threshold current for spin transport phenomena.

The coupling and possible nonequilibrium between magnons and other energy carriers have been used to explain several recently discovered thermally driven spin transport and energy conversion phenomena. Here, we report experiments in which local nonequilibrium between magnons and phonons in a single crystalline bulk magnetic insulator, \( \text{Y}_3\text{Fe}_5\text{O}_{12} \), has been created optically within a focused laser spot and probed directly via micro-Brillouin light scattering. Through analyzing the deviation in the magnon number density from the local equilibrium value, we obtain the diffusion length of thermal magnons. By explicitly establishing and observing local nonequilibrium between magnons and phonons, our studies represent an important step toward a quantitative understanding of various spin-heat coupling phenomena.

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The emerging field of spin caloritronics has been stimulated by a number of recent discoveries, such as large magnon thermal conductivity [1,2], spin Seebeck effect [3–10], spin Peltier effect [11,12], magneto-Seebeck effect [13], and thermal spin transfer torque (STT) [14,15]. These discoveries hold promise for new technologies based on thermally driven spin transport phenomena. For example, thermal STT improves upon current STT-based memory devices by reducing the threshold current for STT-induced magnetic switching [16]. In addition, the large magnon thermal conductivity observed in some cuprate crystals can find potential applications for thermal management [1]. Moreover, the spin Seebeck and spin Peltier effects are being explored for applications in novel thermoelectric energy conversion devices.

The coupling and nonequilibrium between different energy carriers, namely magnons, phonons, and electrons, play an important role in the current theories of spin caloritronic phenomena. For example, in the magnon-mediated transverse SSE model [4,17], it is speculated that the magnon population is out of local equilibrium with the phonon bath. However, such local nonequilibrium has not been directly observed [18]. Indeed, to drive and observe nonequilibrium between magnons and phonons, a temperature gradient must be established within a length scale smaller than the distance over which magnons relax toward complete thermodynamic equilibrium with the phonon bath. Experimental studies on creating and detecting local magnon-phonon nonequilibrium, and probing its associated fundamental length scale, can help to further advance the field of spin caloritronics.

In this Letter, we demonstrate that magnons and phonons can be driven out of local equilibrium in a bulk crystal of the magnetic insulator \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) (yttrium iron garnet, or YIG) that is irradiated by a focused laser beam to obtain a temperature gradient on the order of \( 10^6 \text{ K m}^{-1} \), 2 orders of magnitude larger than those achieved in previous experiments [18,19]. Using the Brillouin light scattering (BLS) technique, we are able to directly probe both the phonon temperature and magnon number density at the same location. Our measurements show that the magnon number density in the laser spot is apparently lower than the local equilibrium value. The fact that one can drive magnons and phonons out of local equilibrium by localized heating within a few microns suggests that the characteristic coupling length between thermal magnons and phonons is comparable to or longer than a few microns in YIG. The measured deviation in magnon number density from equilibrium also allows us to obtain a thermal magnon diffusion length of about 3 \( \text{μm} \) based on a diffusion model. These findings are essential for reaching a microscopic understanding of a host of spin caloritronic phenomena and exploring their potential device applications.

We measured the temperature-dependent magnon and phonon spectra using the micro-BLS technique as shown in Fig. 1 [19–21]. A green laser with a wavelength of \( \lambda = 532 \text{ nm} \) and power of 8 mW was used as the probing laser in all BLS measurements. An additional red laser with \( \lambda = 660 \text{ nm} \) was used to create local heating in some of the measurements. The power of the heating laser was varied from 0 to 19.1 mW. The beam sizes for the probing and heating lasers were obtained by fitting the beam intensity with a Gaussian function and were found to have an effective radius of \( w_g = 0.8 \text{ μm} \) and \( w_r = 1.3 \text{ μm} \), respectively. The YIG sample was oriented with the [110] direction normal to the surface, and an external magnetic
field of 49.5 mT was applied along the [110] in-plane direction of the sample in all measurements.

We first discuss the magnon and phonon modes probed in the BLS spectra shown in Fig. 2. The dominant phonon and magnon modes probed by BLS have the same wave vector ($q$) determined by the change in the photon momentum, as required by momentum conservation, $q = k_s - k_i$, where $k_s$ and $k_i$ are the scattered and incident wave vectors of the laser averaged over the light cone, respectively. In our backscattering geometry, $k_s$ and $k_i$ are nearly antiparallel; thus, $q = 4\pi n/\lambda = 5.53 \times 10^7$ m$^{-1}$, where $n = 2.34$ is the index of refraction for YIG at $\lambda = 532$ nm [22]. This wave vector translates to a wavelength of 113 nm for both the magnon and phonon modes probed. The calculated frequencies of the probed magnon and phonon modes agree well with our experimental observations (see the Supplemental Material [23] for details).

We first uniformly heated the YIG sample with an external heating stage while recording the temperature-dependent BLS spectra, as shown in Fig. 2. While the phonon and magnon populations increase with temperature according to the Bose-Einstein distribution, the intensity, linewidth, and frequency of the BLS peak show complicated temperature dependence [45,46]. In Fig. 2(a), the peak frequency in the magnon BLS spectra shifts downwards in frequency by 0.25 GHz when the temperature is increased from 302 to 345 K. This shift arises from the reduction in the saturation magnetization with increasing temperature. As each magnon reduces the magnetic moment by the same amount, the measured magnon frequency shift can be used to probe the change in the local magnon number density. We note, however, that probing the magnon number density is insufficient to determine the magnon temperature when local nonequilibrium exists between magnons and the lattice within a length scale shorter than the magnon diffusion length. Within this length scale, a nonzero magnon chemical potential can produce an approximately constant magnon number density as the magnon temperature is changed [47,48].

The phonon BLS spectra exhibit a downward frequency shift of 0.14 GHz as the temperature increases from 302 to 345 K, as shown in Fig. 2(b). Because the phonon frequency shift is caused by bond softening and anharmonic coupling among phonon modes, the phonon frequency shift is influenced by the occupancy of all other phonon modes that are coupled to the long-wavelength phonon mode probed by BLS [45]. As such, the phonon peak shift can be used to probe the average temperature of the broad spectrum of thermally excited phonons instead of only the temperature corresponding to the long-wavelength mode directly probed by BLS.

In order to create and probe local nonequilibrium between magnons and phonons, we measured the phonon and magnon BLS spectra with the addition of a red heating laser while maintaining the sample stage at room temperature. Both the magnon and phonon frequencies shift down with increasing heating laser power [Fig. 3(b)] similar to the stage heating condition [Fig. 3(a)]. Figure 3(c) shows the equivalent stage temperature rise for the stage heating condition that yields the same phonon frequency shift as that in the red laser heating experiment at each laser power. The difference in strain between the stage and laser heating cases was taken into account in Figs. 3(c) and 3(d). (See the Supplemental Material [23] for detailed calculations of the strain effects and BLS measurements under hydrostatic pressure.)

We now compare magnon frequency shifts in the two heating configurations. Figure 3(d) shows the magnon
frequency shift as a function of the strain-corrected phonon frequency shift based on Figs. 3(a) and 3(b). Because the measured phonon frequency shift is determined by the local phonon temperature rise when the strain effect is accounted for, the same x-axis value in Fig. 3(d) represents the same average local phonon temperature rise in the probe laser spot in two different heating experiments. It is apparent that the magnon frequency shift is smaller for the laser heating case than for the uniform stage heating condition. In addition, Figs. 3(e) and 3(f) show the phonon and magnon spectra, respectively, obtained under 30 K stage heating and 13.2 mW laser heating. While the phonon spectral shapes are nearly identical [Fig. 3(e)], the magnon spectra are clearly shifted [Fig. 3(f)]. As the magnon frequency shift is directly related to the magnon number density, this result clearly shows that the local magnon number density within the heating laser spot is not relaxed to the equilibrium value at the local phonon temperature. In comparison, two prior BLS measurements have been conducted in an attempt to directly verify the existence of local nonequilibrium in either magnetic insulators [18] or metals [19]. However, the \( \sim 10^4 \) K m\(^{-1} \) temperature gradients applied over millimeter scale were not sufficient to produce observable nonequilibrium between magnons and phonons.

To describe the established nonequilibrium between magnons and phonons quantitatively, we adopt the following steady state magnon diffusion equation, after using an averaged diffusion length \( l_r \) for the broad spectrum of thermal magnons [49]:

\[
\nabla^2 n = \frac{n - n_0}{l_r^2},
\]

(1)

where \( n \) is the magnon number density and \( n_0 \) is the magnon number density in local equilibrium with the phonon temperature \( T_p(r) \), which is calculated using the Bose-Einstein distribution \( n_0 = \sum_k \left[ \exp(\hbar \omega(k)/k_B T_p(r)) - 1 \right]^{-1} \), where \( h \) and \( k_B \) are the reduced Planck’s constant and Boltzmann constant, respectively. \( \hbar \omega(k) \) is the spectroscopic energy of the magnon mode with wave vector \( k \), and the summation is over all magnon modes. Each magnon decreases the magnetic moment by \( g \mu_B \), where \( g \) and \( \mu_B \) are the Landé \( g \) factor and the Bohr magneton, respectively. Therefore, the deviation from local equilibrium, \( \delta n = n - n_0 \), leads to a deviation in the local magnetization, \( \delta M = -g \mu_B \delta n \). This deviation further gives rise to a deviation in the magnon peak frequency, \( \delta f_m \approx (\partial f_m / \partial T_p) H \delta T_p \). Under the experimental conditions, we find that \( \nabla^2 n_0 \approx (\partial n_0 / \partial T_p)_H \nabla^2 T_p \) is an accurate approximation [23]. Hence, Eq. (1) can be rewritten as

\[
\nabla^2 T_p + \nabla^2 \delta \theta_m = \frac{\delta \theta_m}{l_r^2},
\]

(2)

where

\[
\delta \theta_m = \frac{\delta f_m}{(\partial f_m / \partial T_p)_H}.
\]

Here, \( (\partial f_m / \partial T_p)_H \) depends on the local phonon temperature, \( T_p(r, z) \), and was obtained from the measurements of \( f_m \) at different stage temperatures while accounting for the heating from the green probe laser [23]. Equation (2) can be used to solve for \( \delta \theta_m(r, z) \), and subsequently \( \delta f_m(r, z) \).

The phonon temperature profile is determined by the steady state energy equation,
The weighted average value in the probe laser spot, deviation profile between the two heating conditions, of \( \kappa_m \) and \( \kappa_p \) are the magnon and phonon contributions to the total thermal conductivity, \( \kappa = \kappa_p + \kappa_m \), and \( Q \) is the power density of the absorbed laser. Here, \( T_m \) is the magnon temperature. It has previously been found that \( \kappa_m \) is much smaller than \( \kappa_p \) in YIG near room temperature [50,51]. In addition, \( \nabla T_p \) is greater than \( |\nabla (T_p - T_m)| \). Hence, the energy equation can be approximated as (see the Supplemental Material [23] for details of the energy equation)

\[
\nabla \cdot (\kappa_p \nabla T_p) + \nabla \cdot (\kappa_m \nabla T_m) + Q = 0,
\]

where \( \kappa_m \) and \( \kappa_p \) are the magnon and phonon contributions to the total thermal conductivity, \( \kappa = \kappa_p + \kappa_m \), and \( Q \) is the power density of the absorbed laser. Here, \( T_m \) is the magnon temperature. It has previously been found that \( \kappa_m \) is much smaller than \( \kappa_p \) in YIG near room temperature [50,51]. In addition, \( \nabla T_p \) is greater than \( |\nabla (T_p - T_m)| \). Hence, the energy equation can be approximated as (see the Supplemental Material [23] for details of the energy equation)

\[
\nabla \cdot (\kappa \nabla T_p) + Q \approx 0.
\]

Equations (2) and (4) were solved numerically using the commercial software COMSOL for both the red laser heating experiment at 13.2 mW and the stage heating condition with 33 K stage temperature rise, both of which yield the same strain-corrected phonon frequency shift in the probe laser spot according to Fig. 3(c). The calculations were based on independently measured temperature-dependent thermal conductivity and absorption coefficients, and the green probe laser present in both heating configurations was taken into account explicitly. Figure 4(a) shows the difference in the local phonon temperature rise in the two simulations, \( \Delta T_p(r, z) \), caused by the red heating laser. The weighted average value in the probe laser spot, \( \langle \Delta T_p(r, z) \rangle \), is 32 K, which agrees with the equivalent stage temperature rise of 33 ± 4 K shown in Fig. 3(c) for the experiment at 13.2 mW heating laser power. The agreement verifies the phonon temperature measured by the BLS method. The corresponding magnon frequency deviation profile between the two heating conditions, \( \delta f_m(r, z) \), is shown in Fig. 4(b) for \( l_r = 3.1 \) \( \mu m \). The weighted average value \( \langle \delta f_m(r, z) \rangle \) is plotted as a function of \( l_r \) in Fig. 4(c). The black solid line and gray shaded area are shown to indicate the mean of the measured \( \delta f_m \) and its uncertainty, respectively. From the intersection, we find \( l_r = 3.1 \pm 0.9 \) \( \mu m \). The diffusion length obtained in our measurements corresponds to the value at about 372 K, due to the heating by both lasers. Because of the relatively long spin diffusion length compared to the heating laser spot size, the calculated magnon number density in the red laser heating experiment is lower than that in the stage heating condition, as shown in Fig. 4(d), despite the same average phonon temperature in the probe laser spot. Details of the numerical calculations can be found in the Supplemental Material [23].

Our finding has important implications for spin caloritronics, where various relaxation processes of magnons play an important role. The scattering of magnons occurs by both spin-conserving and non-spin-conserving processes. Spin-conserving processes only relax energy and momentum. Such processes occur frequently and exhibit a long length scale of a few nanometers according to thermal conductivity measurements [50]. On the other hand, non-spin-conserving processes relax the magnon number density through spin-orbit interactions with the lattice bath and may exhibit a longer length scale [52,53]. A long magnon spin diffusion length is considered essential for SSE [7,17]. While it has been suggested that this length could be as long as millimeters based on the initial SSE measurements [3,9], a recent measurement of magnon transport in a transverse SSE geometry has reported a magnon diffusion length of ~8 \( \mu m \) for thermally excited magnons in YIG near room temperature [54]. However, it has been suggested that the interpretation of transverse SSE measurements can be complicated by the presence of unwanted longitudinal thermal gradients in addition to the intended transverse gradient in the experimental setup [55].

In summary, our experiments clearly demonstrate that magnons can be driven out of local equilibrium with the phonon bath in YIG when a large temperature gradient is generated on the scale of a few microns. Furthermore, our analysis shows that the observed local nonequilibrium is associated with a long thermal magnon spin diffusion.
length on the order of several microns. The optically based noncontact method for exciting and probing nonequilibrium magnon transport demonstrated here can be extended to various materials where such properties remain unknown.

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[23] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.117.107202, which includes Refs. [24–44], for the detailed sample growth process, detailed description of the BLS experiment, error analysis, calculations of the strain effects, the BLS measurement under hydrostatic pressure, the derivation of magnon diffusion and energy transport equations, and details of the numerical simulations.