Direct Measurement of Magnon Temperature: New Insight into Magnon-Phonon Coupling in Magnetic Insulators

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We present spatially resolved measurements of the magnon temperature in a magnetic insulator subject to a thermal gradient. Our data reveal an unexpectedly close correspondence between the spatial dependencies of the exchange magnon and phonon temperatures. These results indicate that if—as is currently thought—the transverse spin Seebeck effect is caused by a temperature difference between the magnon and phonon baths, it must be the case that the magnon temperature is spectrally nonuniform and that the effect is driven by the sparsely populated dipolar region of the magnon spectrum.

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Spin-caloric phenomena–which allow spin currents to be produced from thermal gradients in magnetic materials–present tantalizing possibilities for manipulating spin information using heat in future spin-caloritronic devices [1–10]. The key to unraveling their underlying physics is to understand spin-lattice interactions, i.e., the coupling between magnons (the quanta of excitations of the magnetic structure of the material) and phonons (the quanta of lattice vibrations). Current understanding of a range of thermoelectric magnetic phenomena, including the spin Seebeck and magnon-drag [9,11] effects, hinges on the assumption that the coupling between the magnon and phonon systems in the materials in which they have been observed is sufficiently weak that they can establish dynamic thermal equilibria at distinct temperatures. In such a case, when a thermal (phonon) gradient ∇T is applied along a sample, it will produce a magnon density gradient in the same direction. The motion of magnons down this gradient leads to the magnon temperature T_m being lifted up above the phonon temperature T_p in the cooler regions [12–14] and pulled down below it in the hotter ones. This difference in T_m and T_p generates a spin current which may be measured (using, for example, the inverse spin Hall effect [2]). However, until now, no experiment has been demonstrated capable of directly measuring and comparing the magnon and phonon temperatures. Here, we report a new technique for measuring the spatial distribution of phonon and magnon temperatures in a magnetic insulator subject to a lateral thermal gradient. From a series of measurements, we determine that the contribution of exchange magnons to the transverse spin Seebeck effect [2,3] in magnetic insulators must be negligible.

Magnons are bosonic quasiparticles with angular momentum $\hbar$ in the exchange limit. The population of magnons of frequency ω at temperature T_m is described by Bose-Einstein statistics and given by $[\exp(\hbar\omega/k_B T_m) - 1]^{-1}$, where k_B is the Boltzmann constant [15]. Since each thermal exchange magnon reduces the total magnetization of a magnetic system by one Bohr magneton, the local magnetization is a measure of the local magnon population, and hence the magnon temperature. It follows that, in principle, spatial variations in the magnon temperature of a magnetic system can be determined through spatially resolved measurements of its magnetization. However, the changes in magnetization which must be detected are very small, making the measurement task a challenging one. Here, we demonstrate that the technique of Brillouin light scattering (BLS) spectroscopy offers an elegant solution. Although BLS is best known as a tool for the spatially resolved investigation of dipolar magnons (which typically have characteristic wavelengths between several microns to millimeters [16]), it can also be used to detect highly spatially localized thermal exchange magnons (wavelengths of order of hundreds of nanometers). The frequency of these exchange magnons depends very strongly on the local magnetization and can thus be used to measure it. The technique we have developed enables the study of thermally induced magnetization gradients via BLS measurements of the local frequency of a thermal exchange magnon mode of known fixed wave number [17].

Our experiments were performed using a monocrystalline yttrium iron garnet (YIG) film (3 mm × 10 mm, thickness 6.7 μm) grown by liquid phase epitaxy on a 0.5 mm thick gallium gadolinium garnet (GGG) substrate and magnetized along the long axis by an in-plane magnetic field $B = 250\,\mathrm{mT}$. A schematic diagram of the experimental setup is shown in Fig. 1(a). Two Peltier elements parallel to the short edges of the film and separated by a distance of 3.2 mm were used to create a thermal gradient along its long axis. The thermal gradient was measured using an infrared (IR) camera with a temperature resolution of 0.1 K and a

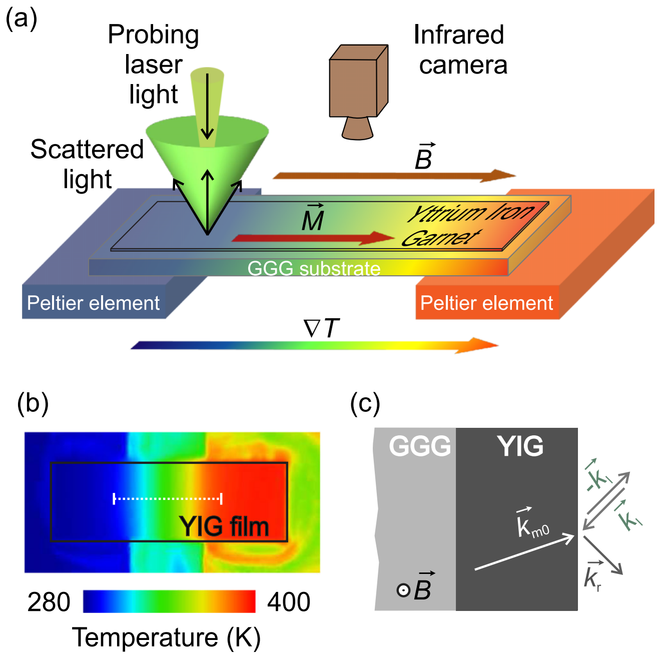

FIG. 1 (color online). (a) In our experiments, two opposite ends of a YIG film (on a GGG substrate) were placed on Peltier elements to create a lateral thermal gradient ∇T. The film was magnetized in plane with an externally applied magnetic field $B = 250$ mT parallel to ∇T, and the magnon temperature was measured using BLS. An IR camera was used to obtain thermal images of the system. (b) Infrared image of the YIG film shown in (a). The dashed white line indicates the path along which the laser was scanned in order to perform the magnon temperature measurements. (c) The orientation of the incident (probing) ($\vec{k}_l$), reflected ($\vec{k}_r$), and backscattered (signal) ($-\vec{k}_l$) photon wave vectors relative to the YIG film surface. The magnon wave vector $\vec{k}_{m0}$ satisfies the momentum conservation condition $|\vec{k}_{m0}| = 2n|\vec{k}_l|$, where $n = 2.36$ is the refractive index of YIG.

spatial resolution of 40 μm. Prior to the experiments, the BLS laser-spot position (i.e., the magnetization measurement point) was identified visually in the infrared image. For these purposes, a relatively high laser power (40 mW) was used. A much smaller power (7 mW) was used for the magnetization measurements so as to minimize local heating (< 0.1 K) and the formation of thermal gradients around the laser spot. During the measurement process, any thermal drifts were controlled, and the laser-spot temperature was kept stable to within ± 0.3 K. All BLS measurements were performed in the second free spectral range of a multipass (3 + 3 pass) tandem Fabry-Perot interferometer [20] to improve the frequency resolution.

Typically, in YIG films having thicknesses in the micron range, a broad spectrum of magnons is thermally excited at room temperature [15,21]. Different magnon modes can be probed using BLS by changing the direction of the incident-photon wave vector (i.e., the angle of the optical probing beam) relative to the magnetization of the film [16]. Owing to the weak spin-orbit coupling in YIG, the efficiency of inelastic photon scattering, and therefore the intensity of the BLS signal for a given thermal magnon density, is small. However, a particular thermal magnon mode $\vec{k}_{m0}$ always exists, which travels along the probing light direction inside the film and satisfies $|\vec{k}_{m0}| = 2n|\vec{k}_l|$, where n (= 2.36 for YIG [22]) is the refractive index of the film, and $\vec{k}_l$ is the photon wave vector ($|\vec{k}_l| = 1.18 \times 10^5$ rad cm^{-1} for green laser light of wavelength 532 nm). Conservation of momentum implies that photons scattered by thermal magnons satisfying $\vec{k} = \vec{k}_{m0}$ propagate “back” along their original path (i.e., the scattering angle is 180°). The position of the $\vec{k}_{m0}$ mode is always well defined in the magnon spectrum; in what follows, we shall refer to it as the backscattering magnon (BSM) mode. Under the conditions of our experiments, the BSM mode lies in the region of exchange-dominated magnons with a wave number $|\vec{k}_{m0}| = k_{m0} = 5.67 \times 10^5$ rad cm^{-1} and a wavelength of 110 nm.

Figure 2(a) shows the position of the BSM mode on the theoretical magnon-dispersion curves [23] corresponding to an in-plane magnetized YIG film (subject to bias magnetic field $B = 250$ mT) at 300 K (upper curve) and 400 K

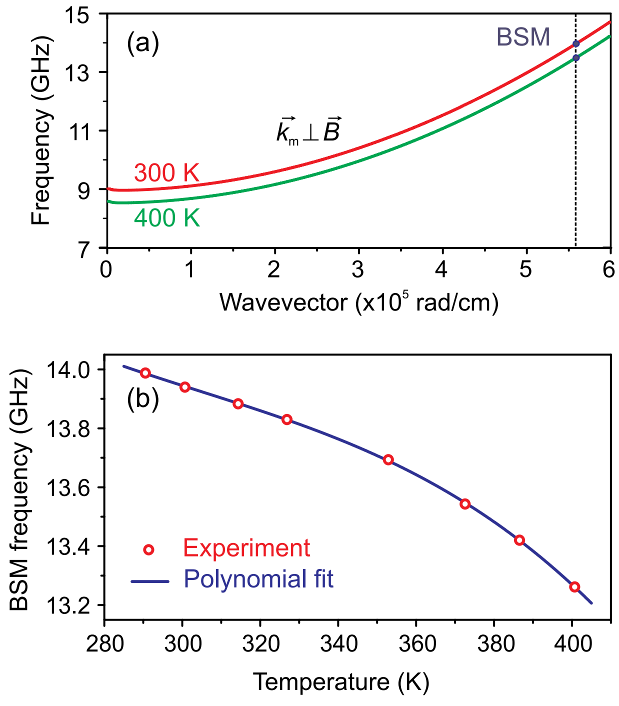

FIG. 2 (color online). (a) The dispersion relations for magnons propagating perpendicular to the magnetization at temperatures of 300 and 400 K in a 6.7 μm thick YIG film subject to a bias magnetic field $B = 250$ mT. Bullets show the positions of the BSM mode at $k_m = 5.67 \times 10^5$ rad/cm. (b) Measured (open symbols) and polynomially fitted (solid line) BSM frequency as a function of temperature of the YIG film. The frequency decreases monotonically with temperature as a result of the increasing magnon population.

(lower curve) [24]. Owing to its short wavelength, good coupling to the laser measurement beam, and extreme sensitivity to its local magnetic environment, the BSM mode provides the ideal means to measure the spatial dependence of the magnetization—and hence magnon temperature—of a magnetic system. In our experiments, we probed and measured the BSM mode frequency using an incident laser beam at an angle of 45° to the film surface [Fig. 1(c)]. This configuration allowed us to position the IR camera normal to the film surface so as to simultaneously measure the spatial distribution of the phonon temperature T_p without parallax error.

In a first reference experiment, BLS was carried out on a uniformly heated sample so as to measure the thermal dependence of the BSM mode. In this configuration, with the Peltier elements maintained at the same temperature, the phonon temperature was found to be uniform along the length of the YIG strip in the gap between them. The measurements show a monotonic decrease in the magnon frequency as the temperature of the YIG film increases as a result of the rising thermal magnon population which reduces the magnetization of the system [see Fig. 2(b)]. (Note that the magnon approximation for the magnetization is known to be valid for this temperature span [26].) Although the monotonic decrease in the magnon frequency cannot be described theoretically by considering the magnetization the only temperature-dependent quantity, the dominating contribution comes from the magnetization [27].

Generally, in magnetic systems where the phonon temperature T_p is uniform throughout, magnons eventually establish equilibrium with phonons via magnon-phonon interactions, so that the temperatures of both subsystems are equal [12]. Accordingly, by fitting the BSM frequency versus T_p curve with a third-order polynomial, as shown in Fig. 2(b), the local magnon frequency can be expressed as a function of the magnon temperature T_m [28].

Once the reference experiments were complete, a thermal gradient ∇T was created and maintained along the YIG strip by passing electric currents through the two Peltier elements in opposite directions [Fig. 1(b)]. The phonon temperature profile was observed to be almost linear between the hot and the cold edges of the heat reservoirs ($\Delta T_p = 85$ K). The frequency of the BSM mode along the film and the phonon temperature were measured simultaneously. To establish the repeatability of these measurements and to estimate the measurement errors, the experiment was repeated four times and statistics were accumulated.

In Fig. 3, the phonon temperature T_p and the magnon temperature T_m, calculated from the third-order polynomial fitting [28] of measured frequency of the BSM mode, are plotted as a function of position along the YIG film. It is evident that T_m follows the trend of T_p within the limit of experimental error. The difference between the two temperatures along the film is shown in the inset with a 95% confidence level. The maximum difference between T_p and T_m is only about 2.8% of ΔT_p and, in contrast to theoretical expectations [12], this difference does not change monotonically between the hot and the cold edges.

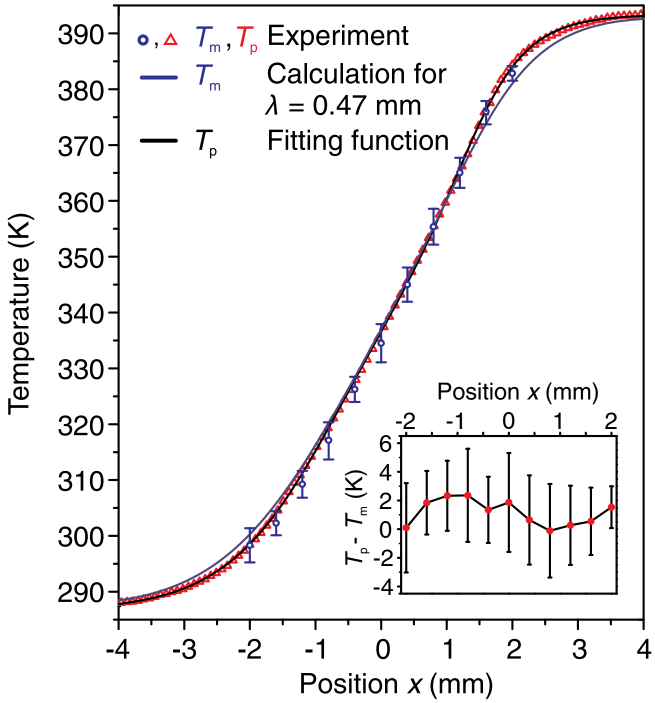

FIG. 3 (color online). Plotted are the measured phonon T_p (open triangles) and magnon T_m (open circles) temperatures along the BLS laser scan line indicated in Fig. 1(b). The T_p data are fitted with a Boltzmann sigmoid function. The profile of $T_m(x)$ is numerically calculated for the characteristic length parameter $\lambda = 0.47$ mm. The inset shows the difference between T_p and T_m with a 95% confidence level.

The magnon and phonon temperatures T_m and T_p are coupled through the magnon-phonon interaction and, within the framework of a one-dimensional model as described in Ref. [12], obey

$$\frac{d^2 T_m(x)}{dx^2} + \frac{1}{\lambda^2}[T_p(x) - T_m(x)] = 0, \tag{1}$$

where λ is a characteristic length scale proportional to the square root of the magnon-phonon relaxation time. Equation (1) implies that in a magnetic system where the relaxation time, and hence λ, is large, the difference in $T_p(x)$ and $T_m(x)$ will be pronounced at the boundaries. However, until the work reported here, no experiment had been demonstrated capable of directly measuring the value of λ. Moreover, theoretical estimates for λ in YIG have varied significantly from 0.85 to 8.5 mm [13].

We have solved Eq. (1) to determine the value of λ in YIG under the conditions of our experiments. To obtain a continuous function describing the phonon temperature $T_p(x)$, the experimentally measured data were fitted with a

Boltzmann sigmoid function (Fig. 3). The fitted function was then substituted into Eq. (1) to obtain a second-order differential equation in $T_m(x)$ alone, and this equation was solved numerically for different values of λ. In our calculations, $dT_m(x)/dx$ was assumed to be zero at the sample boundaries (since, as the magnons are confined to the sample, heat can dissipate only through phonons at the edges). From our experimental data, we determined the maximum possible difference between $T_m(x)$ and $T_p(x)$ at the position $x = \pm 2$ mm and numerically calculated the value of $\lambda_{\max} \approx 0.47$ mm. This value is roughly 1 order of magnitude smaller than that estimated by Xiao *et al.* [13] for YIG using the experimental spin Seebeck data of Ref. [2].

Our findings have significant implications for the spin Seebeck effect. Current thinking attributes the spin Seebeck effect to the existence of a temperature difference between the magnon and phonon baths in a magnetic system subject to a lateral thermal gradient. The fact that our measurements indicate no such difference therefore presents a puzzle. However, a potential explanation presents itself: Our findings rule out the possibility that the temperature of short-wavelength thermal magnons $k_m \gtrsim 10^6$ rad cm^{-1}) that strongly dominate the room-temperature magnetization (magnon population $\propto k_m^2$ [15]) differs from the phonon temperature T_p. For such magnons, spin-lattice thermalization primarily occurs through three-particle (Cherenkov) processes [15,29]. The probability of these processes, which involve the creation or annihilation of a phonon by a magnon but do not change the total magnon population, is proportional to k_m^2. Accordingly, the relaxation channel they provide reduces rapidly with reducing k_m. In light of this, it is plausible that the magnon temperature is wave number dependent [i.e., $T_m = T_m(k_m)$] and that a difference between T_m and T_p capable of giving life to the spin Seebeck effect can be established in the longer-wavelength region of the magnon spectrum ($k_m < 10^5$ rad cm^{-1}) [30]. However, the measurement of $T_m(k_m)$ is currently beyond the experimental state of the art. It is interesting to note that, recently, spectral nonuniformity of the phonon temperature has also been predicted [31].

In summary, we have reported a means to determine the spatial distribution of the magnetization in a magnetic sample subject to a lateral thermal gradient by measuring the temperature-dependent frequency shift of a particular well-defined magnon mode. We have determined the upper limit of the length scale λ which characterizes the magnon-phonon interaction in the magnetic insulator YIG and found it to be significantly smaller than predicted [13]. This result reveals that the coupling of short-wavelength magnons which define the room-temperature magnetization of a magnetic system is too strong for them to contribute to the spin Seebeck effect (even in YIG, which is known to have significantly weaker magnon-phonon coupling than magnetic metals such as Permalloy in which the experimental observation of this effect was first reported). This suggests that, contrary to what was previously thought, the inequality between the magnon and phonon bath temperatures responsible for the spin Seebeck effect must be particular to the longer-wavelength region of the magnon spectrum. Our work not only sheds meaningful new light on the role of thermal magnons in the spin Seebeck effect but may lend valuable new direction to the development of spin caloritronics.

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Note added.—Recently, it came to our attention that our ideas to measure the magnon temperature have been taken up by Birt *et al.* [32] to measure the local magnon temperature in Permalloy films on glass substrates. The authors have correctly cited our manuscript on the arXiv server [33].

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