Topical Review

Magnon and phonon thermometry with inelastic light scattering

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Abstract
Spin caloritronics investigates the interplay between the transport of spin and heat. In the spin Seebeck effect, a thermal gradient across a magnetic material generates a spin current. A temperature difference between the energy carriers of the spin and lattice subsystems, namely the magnons and phonons, is necessary for such thermal nonequilibrium generation of spin current. Inelastic light scattering is a powerful method that can resolve the individual temperatures of magnons and phonons. In this review, we discuss the thermometry capabilities of inelastic light scattering for measuring optical and acoustic phonons, as well as magnons. A scattering spectrum offers three temperature sensitive parameters: frequency shift, linewidth, and integrated intensity. We discuss the temperatures measured via each of these parameters for both phonon and magnons. Finally, we discuss inelastic light scattering experiments that have examined the magnon and phonon temperatures in thermal nonequilibrium which are particularly relevant to spin caloritronic phenomena.

Keywords: magnon, phonon, thermometry, Raman spectroscopy, Brillouin light scattering, thermal nonequilibrium, spin Seebeck effect

(Some figures may appear in colour only in the online journal)

1. Introduction
Thermal transport processes are driven by thermal nonequilibrium, i.e. heat flowing from high temperature to low temperature. However, temperature is only well defined for equilibrium systems. This issue is partially resolved by considering small portions of a system to be in local thermal equilibrium, where a temperature can be defined [1]. This approach fails when the system size falls below the mean free paths of the energy carriers and the carriers can no longer thermalize. With the advancements in technology, nanostructures with 10nm dimensions are routinely fabricated [2]. These length scales are well below the mean free path of phonons [3, 4]. When electrons, optical phonons, and acoustic phonons are driven out of equilibrium, drastically different thermal and transport properties are found in nanostructures from those in bulk materials [5–7].

A host of recently discovered spin caloritronic phenomena necessitate a nonequilibrium. The spin Seebeck effect (SSE) is an exemplary spin caloritronic effect [8]. When a temperature gradient is applied to a magnetic layer, a spin current is generated along the direction of the heat flow. Upon partial transmission through the interface between the magnetic layer and a HM, the spin current is converted to a voltage,
In measuring magnons and phonons with ILS, there are three distinct temperature dependent characteristics. The first is the measured frequency shift, which is proportional to the energy of the quasiparticle. The second is the linewidth, which is the measured frequency shift, which is proportional to the quasiparticle's lifetime (or damping) and determined by quasiparticle scattering. The final is the integrated intensity, which is proportional to the population of the quasiparticle. This review article will discuss the details and applications of these ILS characteristics for magnon and phonon thermometry. Then we will discuss the efforts using ILS to measure the thermal nonequilibrium between magnon and phonon.
Phonons are quantized lattice vibrations. Phonons behave as bosons, with their population described by the Bose–Einstein distribution. While phonon frequency can be derived with a simple harmonic model, the temperature dependence of the frequency can only be understood by taking into account anharmonic terms in the lattice potential, which allows for phonon–phonon interactions [53]. After describing temperature dependences of the frequency shift, linewidth, and integrated intensity of the ILS spectra of phonons, we discuss the similarities and differences between optical and acoustic phonons. Finally, the temperature sensing capacity of each parameter is evaluated.

2. ILS phonon thermometry

Phonons are quantized lattice vibrations. Phonons behave as bosons, with their population described by the Bose–Einstein distribution. While phonon frequency can be derived with a simple harmonic model, the temperature dependence of the frequency can only be understood by taking into account anharmonic terms in the lattice potential, which allows for phonon–phonon interactions [53]. After describing temperature dependences of the frequency shift, linewidth, and integrated intensity of the ILS spectra of phonons, we discuss the similarities and differences between optical and acoustic phonons. Finally, the temperature sensing capacity of each parameter is evaluated.

2.1. ILS frequency shifts of phonons

The optical phonon temperature dependence results from phonon–phonon scattering. This scattering may only occur if anharmonic terms of atomic displacements (e.g. cubic and/or quartic) are included in the interatomic potential energy [54]. Hart et al first measured the temperature dependence of optical phonons in silicon over the range of 20–770 K using Raman scattering [55]. The observed frequency shift can be described by a three phonon scattering model below 800 K [54]. To describe the behavior at even higher temperatures, additional 4-phonon scattering terms must be considered [56]. The expression for describing the anharmonic frequency shift can be greatly simplified if restricted to specific scattering events. For example, in the 3-phonon scattering process, one commonly assumes that a phonon at \( \omega_a \) decays into two phonons at half of its frequency, \( \omega_b = \omega_c = \omega_a/2 \). Similarly, for a 4-phonon scattering, one optical phonon at \( \omega_a \) is assumed to decay into three phonons of equal energy, \( \omega_b = \omega_c = \omega_d = \omega_a/3 \). The temperature dependence of these scattering events describes the Raman frequency shifts for temperatures higher than 800 K.

Though the temperature dependence of the optical phonon frequency results from anharmonic effects, the Raman frequency shift can act as a temperature sensor by simply calibrating the frequency shift as a function of temperature. With this simple calibration process, the Raman frequency shift has become a popular method for measuring local thermal conductivity, such as in polycrystalline diamond [57, 58]. Other examples include thermal conductivity measurements in 2D materials like graphene and MoS2 [59–61]. Figure 4 shows an example of thermal conductivity measurements in MoS2 using Raman scattering. After calibrating the temperature dependence of the frequency shift, a laser was used to induced local heating, and the corresponding laser power dependent Raman spectra are displayed in figure 4(a). The power dependence is then correlated with the temperature through the frequency shift and the thermal conductivity can be calculated with a heat diffusion equation.
Acoustic phonons measured via BLS can serve as temperature sensors as well. ILS of acoustic phonons is not as well developed for temperature sensing because BLS measurements are more technologically challenging. Because of the linear dispersion of acoustic phonons, the leading temperature dependence for acoustic phonons results from this dispersion rather than the anharmonic effects which determine the optical phonon temperature dependence. The frequency of an acoustic phonon is given by

$$\omega (q_a) = v_a q_a,$$

where \(v_a\) is the group velocity of acoustic phonon branch \(a\). Acoustic phonon frequency measured in backscattering BLS is given by

$$\omega = 2 \pi n k_0 \sin \theta,$$

where \(n\) is the refractive index, \(k_0\) is the wavevector of the incident light, and \(\theta\) is between the incident and scattered light within the material [62]. The two main temperature dependent quantities are the phonon group velocity and the refractive index. The incident angle is also temperature dependent since it depends on the refractive index. This dependence is smaller than the direct contribution from the refractive index, so that the incident angle is assumed constant in our analysis.

Both phonon group velocity and refractive index are material specific parameters. The group velocity is proportional to the elastic stiffness and density of the material. The temperature dependence of both can be related to the Grüneisen parameter [54, 63, 64]. In the anharmonic lattice potential, the phonon frequencies become dependent on the equilibrium lattice volume [53]. The Grüneisen parameter describes how the phonon frequency changes with volume. As the temperature changes, the volume will change due to thermal expansion. The Grüneisen parameter is typically positive, which, based on its definition, indicates a decrease of frequency with an increase in volume [53, 65]. Thus, the temperature dependence of the group velocity is due to the volume change from thermal expansion and is expected to decrease with increasing temperature.

Figure 5 shows the temperature dependent BLS frequency shift of the longitudinal acoustic phonon in silicon propagating along the [110] crystal direction [66]. The raw frequency shift is opposite to that expected from decreasing group velocity with increasing temperature. The temperature dependence of the refractive index, shown in the figure inset, is the reason for the observed opposite frequency shift. Electronic properties determine the behavior of the refractive index. In general for semiconductors (e.g. Si) the band gap lies in the optical wavelength range [67]. Variations of the band gap energy with temperature consequently influence the refractive index near that energy [68, 69]. After removing the refractive index effects, the measured BLS frequency shift follows the temperature dependence expected from the group velocity change,
as shown in figure 5. For insulators, the refractive index may not be as sensitive to temperature as semiconductors. When the wavelength of interest is well below the band gap, the temperature dependence of the band gap has less of an influence on the refractive index. This was observed in yttrium iron garnet (YIG), which has a band gap near 2.8 eV [70, 71]. The BLS frequency shift of the longitudinal acoustic phonon was observed to decrease with temperature in YIG, when probed with a 2.33 eV laser. Observing the temperature dependence of the BLS frequency for acoustic phonons requires the refractive index to be relatively constant with temperature as compared to the phonon group velocity.

Based on the discussion above, we conclude that frequency shifts in both Raman and BLS spectra measure an average phonon temperature. The Raman frequency shift measures the average temperature of the phonons involved in scattering with the probed optical phonon. The BLS frequency shift results from the temperature dependence of the Grüneisen parameter, which is a parameter that averages the behavior of all phonon modes. Thus, the BLS frequency shift measures the average temperature of all phonon modes. In thermal equilibrium, it is not necessary to make this distinction, as all phonons will be at the same temperature. In the case of thermal nonequilibrium, different phonon modes may be at different temperatures and thus need to be measured separately.

2.2. ILS phonon linewidth

Another temperature dependent quantity in light scattering spectra is the linewidth, which is proportional to the decay rate (inversely proportional to the lifetime) of the measured quasiparticle. These decay rates are determined by the particle scattering processes. In the Raman study of silicon by Hart et al., the linewidth is shown to be dependent on 3-phonon scattering and can be expressed as

\[ \Gamma(T) = \Gamma(0) \left( 1 + \frac{2}{\pi T} \right), \]  

where \( \Gamma(0) \) is the linewidth at zero temperature, and \( x = \frac{\omega_0}{T} \). This commonly used Klemens model assumes that a higher energy phonon decays into two phonons with equal energy [72, 73]. As with the optical phonon frequency, the 3-phonon scattering is approximated to only depend on scattering with phonons of half the energy of the probed phonon. Another model proposed by Menéndez and Cardona considers the case of two lower energy phonons with different energies by examining the phonon density of states and yielded better agreement for both silicon and germanium [73]. Balkanski et al. showed that for larger temperatures (~800 K), 4-phonon scattering terms must be considered in the linewidth.

The scattering of acoustic phonons is similar to that of optical phonons. In [66], the temperature dependence of the [110] longitudinal acoustic phonon of silicon was measured with BLS. The results show the linewidth is linearly dependent on temperature, as it would be if the acoustic phonon linewidth followed the Klemens’ model in the temperature range measured. However, a comparison to the Klemens’ model requires linewidth measurements at low temperature, approximately 100 K or below, which was not performed in this previous study.

The phonon linewidth temperature dependence results from phonon–phonon scattering processes. Consequently, the temperature measured by the linewidth is the temperature of the phonons that dominantly scatter with the probed phonon. For optical phonons, the temperature measured by linewidth is the temperature of the two phonons that the probed phonon decays into. For the low wavevector acoustic phonons measured in BLS, the dominant scattering processes are not well established. Whether the combination of two phonons into a higher energy phonon or the splitting of one phonon into two lower energy phonons is the dominant scattering process is not well known. These scattering processes are often used in theoretical calculations of the acoustic phonon lifetimes [74–76]. The calculations consider sums of anharmonic interactions over phonon modes of all wavevectors. As a result, the dominant contributions to acoustic modes at specific wavevectors do not follow a simple rule and depend on the particular material.

The above discussion has focused on phonon–phonon scattering as the only contribution to the phonon linewidth, which is not always the case. Other factors, such as electron–phonon scattering and boundary scattering, also make contributions to the total lifetime. This total lifetime can be calculated with Matthiessen’s rule [1]. Sample dependent properties: size, boundary roughness, and impurities will contribute temperature independent terms to the lifetime, and thus the linewidth. Electron–phonon scattering and scattering with quasiparticles, excitons or magnons, may contribute temperature dependent terms to the phonon linewidth. The degree of the contribution from interparticle scattering will depend on the material and the excitation conditions. For example, with above band gap excitation, bulk silicon, germanium, and alpha phase tin have negligible electron–phonon scattering contributions to the linewidth [55, 56, 73]. In graphene and other more complex materials, such halide perovskites, phonon linewidths may be dominated by electron–phonon scattering [77–79]. The main caveat of temperature sensing based on linewidth is that the dominant contributions to the temperature dependence must be known in order to identify which material the temperature measurement.

2.3. ILS integrated intensity of phonons

The integrated intensity of ILS by phonons reflects the population of the phonon being probed [80]. Thus, the intensity can be used as a mode specific temperature sensor. Taking the ratio of the anti-Stokes and Stokes intensities removes the temperature dependence of material parameters. This ratio is given by

\[ I_{AS}^{\omega} = C_{\exp} \left( \frac{\omega_0}{\omega} \right)^n \exp \left( -\frac{n\omega}{\omega_0 - \omega} \right), \]  

where there are only three parameters that influence the ratio. The first parameter is \( C_{\exp} \), which captures the difference in experimental collection efficiency between light at anti-Stokes
and Stokes frequencies. For example, in the Raman scattering spectra of silicon measured with a 532 nm probing laser, the anti-Stokes peak is at 518 nm and the Stokes peak is at 547 nm. Instrument response may not be the same at these different wavelengths. $C_{\text{exp}}$ captures this effect. The next parameter is the frequency of the phonon being probed, $\omega$. The final parameter is the phonon temperature $T$. Once $C_{\text{exp}}$ and $\omega$ are taken into account, the scattering ratio can be used as a mode specific phonon temperature sensor.

The anti-Stokes to Stokes ratio has been measured in Raman experiments and demonstrated to follow the expected temperature dependence [55, 56]. It has been used to investigate numerous systems which includes microscale thermal mapping of AlGaN/GaN HFETs and electrothermal MEMs devices, as well as the sample size effect on graphene thermal conductivity [81–83]. Raman spectra of graphene are shown in figure 6, where the difference between the anti-Stokes and Stokes intensities can be seen. All of these measurements assume local thermal equilibrium, where the measured phonon temperature is the equilibrium temperature. One recent study of graphene used the Raman intensity ratio to measure the temperature of specific phonon modes under local laser heating. These temperatures were different from the equilibrium temperature measured by Raman frequency shift, showing that the different phonon modes could be optically driven out of equilibrium with respect to each other [84].

The anti-Stokes to Stokes ratio applies equally to phonon measured using BLS as it does in Raman. However, in practice, the BLS anti-Stokes to Stokes intensity ratio cannot measure temperatures accurately. Figure 7 shows why this is the case. BLS measures low frequency excitations, where the exponential term in the anti-Stokes to Stokes ratio becomes very small, effectively quenching any temperature dependence. The BLS integrated intensity can still act as a mode specific sensor, as the intensity is proportional to the probed phonon’s population. The full expression for the BLS intensity is

$$I = I_0 \left| \frac{\varepsilon}{2\varepsilon_0} \frac{\omega}{\omega_0} \right|^2 e^{i\mathbf{\hat{e}}_t \cdot \mathbf{\hat{e}}_s} \left(\mathbf{P} \cdot \hat{e}_t\right)^2 \left\{ \frac{n+1}{n} \right\}$$  (4)

where $\varepsilon$ is the permittivity of the material, $\varepsilon_0$ is the permittivity of free space, $V$ is the scattering volume, $\omega_0$ is the scattered frequency (either $\omega_0 + \omega$ or $\omega_0 - \omega$), $c$ is the speed of light in vacuum, $g$ is the density of the material, $v$ is the group velocity of the phonon, $\mathbf{\hat{e}}_t$ is the polarization of the scattered (incident) light, $\hat{P}$ is the photoelastic tensor contracted with the unit vectors of the phonon propagation and phonon polarization, $n$ is the phonon occupation number where $n+1$ or $n$ corresponds to Stokes or anti-Stokes scattering, and $I_0$ is the intensity of the incident light [80]. The BLS intensity of acoustic phonons in silicon was shown to follow a linear temperature dependence [66]. This dependence is in agreement with that predicted by the Bose–Einstein distribution, suggesting the BLS intensity depends on the temperature of the probed phonon. The other parameters have negligible temperature dependence in silicon, though this is not true for all materials in general. For example, perovskites can have strongly temperature dependent photoelastic properties [85]. Thus, for the BLS intensity to act as an acoustic phonon mode specific temperature sensor, the material property temperature dependences must first be removed.

3. ILS of magnons

Spin waves are collective excitations that deviate from the saturated magnetization direction. Magnons are quanta of spin waves. These two terms are often used interchangeably. Magnons, as phonons, behave as bosons and are described by the Bose–Einstein distribution. Temperature sensing with ILS of magnons is not as well developed as it is for phonons. Raman scattering has certainly been used to study magnons,
though mostly on two-magnon scattering in antiferromagnets. Two-magnon scattering cross sections are larger compared to those from one-magnon scattering for antiferromagnets [86, 87]. Figure 8 shows the Raman spectrum for antiferromagnetic nickel oxide, where the 2-magnon peak is the dominant feature of the spectrum [88]. Though the SSE has been observed in antiferromagnets, ferromagnets are still the material of focus for studying the SSE [89, 90]. Therefore, in this section we will focus on how BLS can be used to measure magnon temperatures in ferromagnets.

3.1. BLS frequency shift of magnons

To describe magnon dispersions in ferromagnets, several methods have been used including the Bloch and Holstein–Primakoff methods, Maxwell’s equations, and the Landau–Lifshitz equation [91, 92]. The most basic and commonly used spin wave dispersion is the Kittel equation for FM resonance in a planar film

$$\omega_K = \gamma \sqrt{H(H + 4\pi M)}$$  \hspace{1cm} (5)

where $\gamma$ is the gyromagnetic ratio, $H$ is the external field applied along the film plane, and $M$ is the saturation magnetization [67]. In the case of different geometries and/or in the presence of anisotropy, the proper demagnetization and anisotropy fields should be incorporated into the effective magnetization [91, 93]. Though the Kittel equation describes uniform magnetic precession in a planar film, the equation serves as a good approximation of magnons with small wavevectors probed by light scattering experiments. Table 1 lists the magnon dispersion equations for select types of magnon modes. Low wavevector or long wavelength magnons are referred to as magnetostatic waves, or dipolar magnons.

![Figure 7. Temperature dependence the exponential factor that appears in the anti-Stokes to Stokes ILS intensity ratio. For BLS frequencies, the factor varies only slight (~2%) over the 200 K temperature range. For Raman frequencies, the factor varies by a factor of 5. This difference in temperature dependence of the exponential factor suggests that the Raman intensity ratio can be used as a temperature sensor while the same approach does not lead to a sensitive temperature sensor based on BLS intensity ratio.](image)

![Figure 8. Raman spectra of nickel oxide at various temperatures. 2LO refers to the 2-phonon peak for the longitudinal optical phonons. 2M refers to the 2-magnon peak. At room temperature the 2M peak is the dominant feature due to the strong 2-magnon scattering cross section in antiferromagnets. As the temperature increase, the magnetization decreases leading to a change in frequency and intensity for the 2M peak. Reprinted with permission from [88]. Copyright 2017, AIP Publishing LLC.](image)

| Magnon dispersion equation                                                                 | Magnon type                          |
|------------------------------------------------------------------------------------------|--------------------------------------|
| $\omega_K = \gamma \sqrt{H(H + 4\pi M)}$                                                  | Uniform precession                   |
| $\omega_{MS} = \frac{\theta_q}{\gamma} \sqrt{H(H + 4\pi M \sin^2 \theta_q)}$            | Magnetostatic waves                  |
| $\omega_{EMS} = \gamma \sqrt{(H + Dq^2)(H + Dq^2 + 4\pi M \sin^2 \theta_q)}$            | Exchange magnetostatic waves         |
| $\omega_{BV} = \sqrt{H(H + 4\pi M \left(1 - \frac{e^{-q l}}{q l}\right))}$                | Backward-volume magnetostatic waves  |
| $\omega_{Physics} = \gamma \sqrt{(H + \frac{4\pi M}{3})^2 - \left(\frac{4\pi M}{3} e^{-q l}\right)^2}$ | Perpendicular standing spin waves   |

Table 1. Frequency dispersion relations for select magnon modes. $D$ is the exchange constant and $l$ is the thin film thickness. FM resonance corresponds to uniform spin precession ($q = 0$). Magnetostatic waves corresponds to bulk spin waves for an isotropic sphere. Exchange magnetostatic waves include the exchanges interaction term. Additional anisotropy terms can also be included as needed. Backward volume and surface magnetostatic spin waves are thin film modes that respectively propagate parallel and perpendicular to the magnetization direction. Perpendicular standing spin waves have wavevectors quantized in the thickness direction. For further details, see [67, 91–95].
The dipole–dipole interaction dominates for longer wavelengths \[91\]. At short wavelengths, the exchange interaction dominates, thus high wavevector magnons are referred to as exchange magnons \[94\]. Magnons probed with BLS fall between these two limits, where dipole–dipole and exchange interactions must both be considered.

It is important to note that all these dispersions depend on the saturation magnetization of the material. Figure 9 shows the dispersion and temperature dependence of a few select magnon modes. The saturation magnetization is the main contribution to the temperature dependence of magnon frequencies. The saturation magnetization is related to magnon number by

\[ M(T) = M(0) \left( 1 - \frac{1}{N_S} \sum q n(q) \right) \]  (6)

where \( M(T) \) is the saturation magnetization at temperature \( T \), \( M(0) \) is the zero temperature magnetization, \( N \) is the total number of spins, \( S \) is the mean spin, and \( n(q) \) is the Bose–Einstein distribution for a magnon with frequency \( \omega(q) \) [53]. As the temperature increases, the magnon population increases. This results in a further deviation from the ground state spin alignment, and thus a reduction of the saturation magnetization at higher temperatures.

The BLS frequency of magnons has been used to measure local temperatures. The temperature profile for permalloy (Ni80Fe20) and YIG thin films under thermal gradients have been mapped with BLS frequency shifts \[95, 96\]. Another study used BLS frequency shifts to measure the local laser heating of magnons \[70\]. The magnon frequency shifts in these measurements result from the temperature dependence of the magnetization. Therefore, temperatures measured with BLS frequency shift of magnons in these studies represent an average temperature of all magnons.

### 3.2. BLS linewidth of magnons

Similar to phonons, magnon linewidths are determined by the decay rates, or relaxation time, of the probed particle. The relaxation of magnetic excitations is often characterized by Gilbert damping \[91, 92\]. This damping parameter is typically measured with FM resonance, which corresponds to spatially uniform magnetic oscillations with zero wavevector \[97–99\]. The BLS linewidth is determined by the relaxation or decay of a magnon with a finite wavevector. As a result, the relaxation measured with BLS linewidth may differ from that measured by Gilbert damping.

In FM insulators, the relevant scattering processes are magnon–magnon and magnon–phonon. In FM metals, magnon–electron scattering is also important. While two-magnon scattering depends on sample specific properties including surface roughness, grain boundaries, and impurity distribution, the temperature dependence of two-magnon scattering is only due to the magnetization. Dipole–dipole interactions lead to 3-magnon scattering, and the exchange interaction leads to 4-magnon scattering \[91\]. For high quality, bulk FM insulators, two magnon scattering should be negligible compared to 3- and 4-magnon scattering. BLS linewidths measure 3-magnon and 4-magnon scattering rates since both dipole–dipole and exchange interactions are relevant for magnons probed with ILS. Parametric microwave pumping of magnons in YIG found that the 4-magnon scattering is the dominant scattering process with some contribution from 3-magnon scattering.
The wavevector dependent FM resonance linewidths were found to be well described by only these two processes, suggesting magnon–phonon interactions are negligible in the magnon linewidth of bulk YIG samples [91].

Temperatures measured by BLS linewidth of magnons are then the temperatures of the particles which scatter with the probed magnon. For materials like YIG, one can conjecture that the dominant modes in the 3- and 4-magnon scattering will be ~GHz range magnons, since scattering rates decrease as the difference in energy of the particles increases. Thus, the BLS linewidth of magnons measures the averages temperatures of GHz range magnons. For other materials, it may be the case that magnon–phonon or magnon–electron interactions dominate. Then, the BLS linewidth would be measuring the average temperature of the particles interacting with the probed magnon.

### 3.3. BLS integrated intensity of magnons

The magnon BLS integrated intensity depends on the probed magnon population and thus its temperature. Other temperature dependent quantities also influence the BLS intensity. These temperature dependences must be identified and accounted for. The BLS intensity for magnons is described by

$$ I = I_0 \frac{\hbar \gamma M_{A}^{4}}{2 \pi e^{2}} |A^{\mp}|^{2} \left\{ \begin{array}{c} n + 1 \\ n \end{array} \right\} $$  \hspace{0.5cm} (7)

where $A^{\mp}$ is a tensor coefficient relating electric susceptibility to the magnetization, which depends on the crystal’s structure, magnetization direction, and the polarization of the incident and scattered light [101]. The superscript indicates the coefficient can be different for Stokes (−) and anti-Stokes (+) scattering. In addition to the magnon population, the magnetization and the tensor coefficient are temperature dependent. The temperature dependence of the magnetization is well known; thus, the temperature dependence of tensor coefficient must be determined to use BLS intensity as a magnon mode specific temperature sensor.

The temperature dependence of the BLS intensity has been studied in YIG and permalloy, shown in figures 10(a) and (c) [102]. Immediately it can be seen that the BLS intensities for these two samples have opposite temperature dependences. The YIG BLS intensity decreases with increasing temperature, whereas the permalloy BLS intensity increases. To see the temperature dependence of the tensor coefficient, the BLS intensity is compared to the Bose–Einstein distribution of the probed magnon after the magnetization is removed. The BLS intensity and Bose–Einstein distribution function are both normalized to their values at 300 K for comparison, as shown in figures 10(b) and (d).

For YIG, the magnetization corrected BLS intensity follows the Bose–Einstein distribution, suggesting the tensor coefficients are relatively temperature insensitive. For permalloy, there is a large difference between them. There are two possible reasons for the observed difference. One possibility is that the tensor coefficients are temperature sensitive, which one might expect since permalloy is metallic, therefore having a more complex response at optical frequencies. The other possibility is that, due to heating from the probe laser, the probed magnon in permalloy is out of thermal equilibrium, and thus its population is not well described by the Bose–Einstein distribution. Regardless, this study showed that the BLS intensity can be used to measure a specific magnon mode’s temperature in thermal equilibrium once the temperature dependence of the magnetization and tensor coefficients are removed.

### 4. Magnon–phonon thermal nonequilibrium measured with ILS

The ability of ILS to measure magnon and phonon temperatures has been outlined in the previous sections. In this section, we discuss the efforts on measuring magnons and phonons in thermal nonequilibrium with ILS.

#### 4.1. Nonequilibrium considerations for magnons and phonons

Temperature is only well defined in thermal equilibrium, leading to the question: can temperature be defined in nonequilibrium cases? Often, the answer to this question is yes, but only under the assumption that local thermal equilibrium is established for subsystems. The subsystems can be considered as a particular type of particle or quasiparticle. For example, in laser heating metals, electrons and phonons are considered subsystems capable of establishing thermal equilibrium among themselves described by an electron temperature and a phonon temperature [103]. These definitions are only possible if one assumes that scattering events among electrons and phonons are more frequent and effective than those between...

\[ \text{Figure 10. Temperature dependence of raw BLS intensity of magnons in (a) YIG and (c) permalloy. BLS intensity corrected for magnetization is shown in (b) for YIG and (d) for permalloy. The corrected intensities are normalized to their value at 300 K, and compared to normalized Bose–Einstein distributions for the respective magnon modes. Reprinted figure with permission from [102]. Copyright 2017 by the American Physical Society.} \]
electrons and phonons. The local temperature is also only established within a small and local volume, dependent on the particle scattering length. For the case of interest to this article, the magnons and phonons are considered subsystems in local thermal equilibrium, where magnon and phonon temperatures can be defined.

In nonequilibrium systems, the chemical potential for these quasiparticles must also be addressed. Phonon and magnon numbers are not conserved in thermal equilibrium, leading to zero chemical potential. In a driven system, a steady state can exist where the average number of quasiparticles is conserved. In this case, the quasiparticle can have a non-zero chemical potential. For example, chemical potential is solicited to describe magnon Bose–Einstein condensates. For these studies, parametric microwave pumping is used to excite magnons. The excited magnons relax to the lowest energy magnon, which occurs at a finite wavevector. The decay of the magnons into this specific magnon mode establishes a quasi-equilibrium for that mode, allowing it to establish a chemical potential. Increasing the parametric pumping power increases the magnon chemical potential, and above a threshold power, a magnon Bose–Einstein condensate is established. Similarly, this quasi-equilibrium and Bose–Einstein condensation has been seen for phonons using ultrafast laser pumping. In these cases, thermometry methods that rely on measuring quasiparticle population cannot uniquely measure temperature, as a change in population could arise from a change in either temperature or chemical potential.
The conclusion of the study was that their measurement is sensitive to short wavelength magnons, and the magnon-phonon nonequilibrium, important to the transverse SSE, must develop between the phonon and longer wavelength magnons. Nonequilibrium, important to the transverse SSE, must develop between the phonon and longer wavelength magnons. The acoustic phonons are driven out of equilibrium in a driven, nonequilibrium system measured by Agrawal et al. For this system, Schreier et al found a 20 μK difference in the magnon and phonon temperatures at the boundary of the YIG sample. These results by Schreier et al and Agrawal et al were in agreement as 20 μK was beyond the temperature sensitivity of the measurement.

An et al used local laser heating in bulk YIG to generate a large temperature gradient in an attempt to measure a difference in magnon and phonon temperatures [70]. The BLS frequency shift was employed to measure both the magnon and phonon temperatures as the sample was uniformly heated in the calibration step. A heating laser was then used to create a driven, nonequilibrium system on a few μm length scale. Figures 13(a) and (b) show the BLS spectra of phonons and magnons, comparing a spectrum at 30 K of uniform stage heating to that using 13.2 mW laser heating. The phonon spectra for uniform and laser heating overlapped, where the magnon spectra for uniform and laser heating clearly differed. Without relying on any model, these measured spectra clearly indicate that the magnons and phonons are driven out of equilibrium in the laser heating case. Instead of using the language of magnon-phonon nonequilibrium, the BLS frequency shift was correlated to magnon population changes. A two-temperature model was adapted with a magnon diffusion model, and the magnon diffusion length is calculated to be a few micrometers. This value is similar to the magnon diffusion length extract from non-local transport measurements [113, 114].

4.2. Measurements of magnon-phonon nonequilibrium

The first BLS study of probing magnon and phonon temperatures in a nonequilibrium system was performed by Agrawal et al [96]. In this study, a lateral temperature gradient was applied to a 6.7 μm thick YIG film, shown in figure 11(a). The magnon temperature along the temperature gradient was measured via BLS frequency shift while phonon temperature was measured using an infrared camera. As shown in figure 11(c), no difference between the phonon and magnon temperatures can be measured beyond the error bars. Using a 1D, two-temperature model, Agrawal et al calculated an upper bound of the magnon diffusion length of a few hundred micrometers. The conclusion of the study was that their measurement is sensitive to short wavelength magnons, and the magnon-phonon nonequilibrium, important to the transverse SSE, must develop between the phonon and longer wavelength magnons.

Schreier et al performed a theoretical analysis of magnon-phonon temperatures in YIG under different thermal gradients using a simple two temperature diffusion model [112]. First, they consider a 1D system consisting of a gadolinium gallium garnet (GGG) substrate, a 50 nm YIG layer, and a platinum layer, with a temperature gradient across the entire system. The calculated temperature differences between magnons and phonons at each interface were found to be small, approximately 0.2 K. A 3D version of the same structure was considered next, with a 7 nm platinum layer, and a temperature gradient induced by local laser heating incident on the platinum, shown in figure 12. Here, for 10 mW of laser power, the temperature difference between magnons and phonons was again found to be about 0.2 K. Finally, they considered the system measured by Agrawal et al. For this system, Schreier et al found a 20 μK difference in the magnon and phonon temperatures at the boundary of the YIG sample. These results by Schreier et al and Agrawal et al were in agreement as 20 μK was beyond the temperature sensitivity of the measurement.

An et al used local laser heating in bulk YIG to generate a large temperature gradient in an attempt to measure a difference in magnon and phonon temperatures [70]. The BLS frequency shift was employed to measure both the magnon and phonon temperatures as the sample was uniformly heated in the calibration step. A heating laser was then used to create a driven, nonequilibrium system on a few μm length scale. Figures 13(a) and (b) show the BLS spectra of phonons and magnons, comparing a spectrum at 30 K of uniform stage heating to that using 13.2 mW laser heating. The phonon spectra for uniform and laser heating overlapped, where the magnon spectra for uniform and laser heating clearly differed. Without relying on any model, these measured spectra clearly indicate that the magnons and phonons are driven out of equilibrium in the laser heating case. Instead of using the language of magnon-phonon nonequilibrium, the BLS frequency shift was correlated to magnon population changes. A two-temperature model was adapted with a magnon diffusion model, and the magnon diffusion length is calculated to be a few micrometers. This value is similar to the magnon diffusion length extract from non-local transport measurements [113, 114].
by the population of all magnon modes. Therefore, magnon frequency shifts measure an average magnon temperature.

Linewidths measure average temperatures of the quasiparticles that interact with the probed quasiparticle mode. For semiconductors, the optical phonon linewidth typically measures the temperature of the phonon at half the frequency of the probed phonon, following the Klemens model. For acoustic phonons in semiconductors, the temperature that the linewidth measures cannot be specifically assigned since the dominant phonon–phonon scattering is not well understood. However, the acoustic phonon linewidth likely measures the temperature of GHz range phonons as scattering efficiencies of particles are inversely proportional to the energy difference of the particles. For magnons in bulk FM insulators, the linewidth measures the temperatures of magnons involved with 3-magnon and 4-magnon scattering with the probed magnon. Depending on the materials, interparticle scattering, such as electron–phonon or magnon–phonon, can contribute additional temperature dependences to the linewidths, changing which temperature the linewidth measures.

The integrated intensity of ILS depends on the quasiparticle population, and thus the intensity measures the specific temperature of the probed quasiparticle. For optical phonons, their high energy allows the ratio of anti-Stokes to Stokes scattering intensity to have a strong temperature dependence, enabling this ratio to measure the temperature of the probed phonon mode. For acoustic phonons and FM magnons, the lower energy causes the intensity ratio to have a weak temperature dependence. Instead, the integrated intensity can act as a temperature sensor once other material dependent properties are corrected. The correction for acoustic phonons is the temperature dependence of the photoelastic coefficients. The corrections for FM magnons are the temperature dependence of the saturation magnetization and the magnetization dependent, electric susceptibility coefficients.

As the connection between magnon–phonon nonequilibrium and the SSE is not completely understood, we anticipate that ILS will prove useful in further studies on discovering and investigating new materials that optimize the SSE [18, 115–117]. Since ILS probes the region within the laser spot, special care must be used in selecting geometries to explore magnon–phonon nonequilibrium. In systems where the temperature gradient is perpendicular to the laser propagation direction, one can obtain a spatial mapping of the magnon and phonon temperatures. For example, in nanowires, ILS could be used to measure temperatures along the length of the wire [34]. The limiting length scales are the width and thickness of the nanowire. As long as a nanowire is wide enough to allow positioning the probe laser optically, and sufficient ILS signal can be detected, the temperature distribution along the length can possibly be measured. In systems where the temperature gradient is parallel to the laser propagation direction, ILS may be applied as long as the nonequilibrium region of interest is spatially constrained, such as in layered thin films or nanostructures such as nanopillars and magnetic tunnel junctions [35, 118, 119]. Thus, we expect ILS can be used to advance the understanding of magnon–phonon nonequilibrium and the SSE in many interesting systems.

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