Specific heat and thermal conductivity of ferromagnetic magnons in Yttrium Iron Garnet

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received 9 March 2013; accepted in final form 26 July 2013; published online 23 August 2013

PACS 75.30.Ds – Spin waves
PACS 75.40.Cx – Static properties (order parameter, static susceptibility, heat capacities, critical exponents, etc.)
PACS 75.40.Gb – Dynamic properties (dynamic susceptibility, spin waves, spin diffusion, dynamic scaling, etc.)

Abstract – The specific heat and thermal conductivity of an insulating ferrimagnet Y₃Fe₅O₁₂ (Yttrium Iron Garnet, YIG) single crystal were measured down to 50 mK. The ferromagnetic magnon specific heat $C_m$ shows a characteristic $T^{1.5}$-dependence down to 0.77 K. Below 0.77 K, a downward deviation is observed, which is attributed to the magnetic dipole-dipole interaction with typical magnitude of $10^{-4}$ eV. The ferromagnetic magnon thermal conductivity $\kappa_m$ does not show the characteristic $T^2$-dependence below 0.8 K. To fit the $\kappa_m$ data, both magnetic defect scattering effect and dipole-dipole interaction are taken into account. These results provide a complete picture of the thermodynamic and thermal transport properties of the low-lying ferromagnetic magnons.

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Introduction. – Recently, the ferrimagnetic insulator Y₃Fe₅O₁₂ (Yttrium Iron Garnet, YIG) has drawn great attention due to long-range transport by its spin current [1,2]. In these experiments, an electronic signal can be transferred by the angular momentum of the spin waves in insulating YIG, via the spin Hall and inverse spin Hall effects, which provides a new method to transfer information purely by spin waves [1,2]. Furthermore, it was discovered that in YIG films the quanta of spin waves, namely magnons, undergo Bose-Einstein condensation under microwave pumping in a magnetic field [3-6]. In this context, it is important to understand the thermodynamic and transport properties of the spin waves.

In spin wave theory for magnets, the quanta of spin waves are antiferromagnetic (AFM) or ferromagnetic (FM) magnons. They have totally different dispersion relations, and distinct thermodynamic and transport properties. The AFM magnons have a linear dispersion relation, so both their specific heat and boundary-limited thermal conductivity at low temperature obey the $T^3$-dependence [7,8]. For FM magnons, the situation is more complex due to the existence of the magnetic dipole-dipole interaction [9,10].

At not very low temperature, the dispersion relation $E = Dk^2$ is a good approximation, and the specific heat and boundary-limited thermal conductivity of FM magnons show the characteristic $T^{1.5}$- and $T^2$-dependence, respectively [11–13]. The equations are

$$C_m(T) = \frac{15\zeta(5/2) k_B^5 T^{1.5}}{32\pi^{1.5} D^{1.5}} \quad (1)$$

and

$$\kappa_m(T) = \frac{\zeta(3) k_B^3 L T^2}{\pi^2 h D}, \quad (2)$$

where $\zeta$ is the Riemann function, and $L$ is the boundary-limited mean free path [7,11–13]. However, at sub-kelvin temperature range, the dipole interaction

$$H_{d-d} = \mu_B^2 \sum_{i \neq j} \frac{r_{ij}^2 (S_i S_j) - 3(r_i S_i)(r_j S_j)}{r_{ij}^3}, \quad (3)$$

with typical order of $10^{-4}$ eV, has to be considered for FM magnons [14]. It significantly modifies the dispersion relation of FM magnons below 1 K, and makes the approximate form $E = Dk^2$ no longer valid [15].

The dipole interaction is long range and anisotropic. It is a basic interaction in magnets and critical for many
phenomena such as the demagnetization factor and the formation of domain walls in ferromagnets, spin ice behavior in Ising pyrochlore magnets, and spin anisotropy in ferromagnetic films [16–18]. Theoretically, when the effect of dipole interaction was taken into account for FM magnons, both the $T^{1.5}$-dependence of $C_m$ and the $T^2$-dependence of $\kappa_m$ changed [9,10,14,19]. Yet so far experimental verification of this effect on FM magnons is still lacking.

YIG is an archetypal ferrimagnetic insulator. Its low-energy magnetic excitations are FM magnons because at low temperature the important spin-wave branch in a ferrite has the same form as in a ferromagnet [20,21]. The study of this interesting and complex compound started in the late 1950s, and it has become indispensable for investigating the properties of magnets since then [14]. In fact, YIG is the first material in which thermodynamic and transport properties of FM magnons.

The previous lowest temperature for specific-heat measurement on YIG was 1 K [25,26]. The data on the specific heat $C_m(T)$ between 1 and 4 K showed the characteristic magnon $T^{1.5}$-dependence [25,26]. However, the magnon thermal conductivity $\kappa_m(T)$ between 0.23 and 1 K did not obey the characteristic $T^2$-dependence, which was explained by considering the effect of magnetic defect scattering [24,27,28].

In this paper, we present specific heat and thermal conductivity measurements of YIG single crystal down to 50 mK. The magnon specific-heat data deviate downward from the $T^{1.5}$-dependence below 0.77 K, which is attributed to the effect of dipole interaction. Below 0.2 K, the magnon thermal conductivity data cannot be fitted by only considering the boundary and magnetic defect scattering, and the effect of dipole interaction has to be taken into account. To our knowledge, these are the first experimental observations of the effect of dipole interaction on the FM magnon specific heat and thermal conductivity, giving complete understanding of the thermodynamic and transport properties of the low-lying FM magnons.

**Experimental details.** – A YIG single crystal was grown with an optical floating zone furnace [29–33]. The single crystal grew along the [332] crystallographic direction, as characterized by X-ray diffraction. Ultra-low temperature specific heat was measured by the two-tau method in a small dilution refrigerator adapted into a Physical Property Measurement System (PPMS, Quantum Design). The sample weights 45.55 mg, and is a disc with 3.30 mm in diameter and 0.92 mm in thickness.

Sample 1 (S1) was cut from the single crystal for thermal conductivity measurements. It is a rectangle with dimensions 2.08 x 0.84 mm$^2$ in the plane and 0.63 mm thick along the [332] direction (the sample growth direction). Sample 2 (S2) was obtained by thinning S1 to 0.23 mm. For both samples, the heat current was along the [110] direction.

Ultra-low temperature thermal conductivity was measured in a dilution refrigerator (Oxford Instruments), using a standard four-wire steady-state method with two RuO$_2$ chip thermometers, calibrated in situ against a reference RuO$_2$ thermometer. Four contacts were made on the sample surface by silver epoxy. Magnetic fields were applied parallelly to the heat current.

**Results and discussion.** – The quality of our YIG single crystal was characterized by X-ray diffraction (XRD), as shown in fig. 1. The main panel is the XRD pattern of the (332) plane. The inset shows the rocking scan curve of the (664) reflection, with two peaks from the Cu $Kα_1$ and $Kα_2$ radiations, respectively.

The specific heat of YIG single crystal below 2.5 K is shown in fig. 2. The sample coupling of the measurement is 98–100% above 0.3 K, and around 90% between 0.1–0.3 K, respectively, which indicates that our data are reliable. In the figure, $C/T^{1.5}$ was plotted as a function of $T^{1.5}$ in order to separate phonon and magnon contributions. Between 0.77 and 2.5 K, the total specific heat can be fitted by $C = aT^{1.5} + bT^3$, in which the first term is from the FM magnons and the second term is from the phonons. From the fitted coefficient $a = 6.7 \mu$J/cm$^3$K$^{2.5}$, we get $D = 5.2 \times 10^{-36}$ J/cm$^2$ according to eq. (1). This value is very close to Edmonds and Petersen’s result $D = 5.1 \times 10^{-36}$ J/cm$^2$ [25]. From the coefficient $b = 2.3 \mu$J/cm$^3$K$^4$, we get the Debye temperature $\Theta_D = 491$ K, in good agreement with previous results [34].

Below 0.77 K, however, there is an apparent deviation from the solid fitting line. As we have described, the dipole interaction will affect the specific heat of FM magnons...
be expressed as pressed by the field, as previously reported [22–24]. In Schottky anomaly from nuclear moments [38].

ure 4(a) shows the ultra-low temperature thermal conductivity below 1 K, by modifying the dispersion relation. The modified dispersion relation was proposed as the following form [15]:

\[ E(k) = \sqrt{(Dk^2 - N_z \hbar \omega_m)(Dk^2 - N_z \hbar \omega_m + \hbar \omega_m \sin^2 \theta_k)} \]

(4)

where \( \theta_k \) is the angle between the magnon wave vector and the magnetization direction, \( N_z \) is the z demagnetization factor, and \( \hbar \omega_m = g \mu_B 4\pi M \). Equation (4) is only valid within a single domain. For a single crystal of a single domain, \( N_z \) depends on the shape of the sample, while for a single crystal of a multidomain, \( N_z \) depends on the magnetization of neighboring domains [9]. The dispersion relation given in eq. (4) is plotted in fig. 3. The approximate dispersion relation \( E = Dk^2 \) is only valid for \( k_B T \gg \hbar \omega_m \). For YIG, \( 4\pi M = 2449 \text{Gs} \) [35,36], \( g = 2 \) [37], so \( \hbar \omega_m = g \mu_B 4\pi M = 0.32 k_B \). Therefore, the \( T^{1.5} \)-dependence of \( C_m \) should only hold for \( T \gg 0.32 K \). From our experimental data in fig. 2, \( C_m \) shows \( T^{1.5} \)-dependence above 0.77 K. The downward deviation below 0.77 K should come from the effect of dipole interaction. Note that the upturn below 0.38 K is the Schottky anomaly from nuclear moments [38].

Next we discuss the thermal conductivity results. Figure 4(a) shows the ultra-low temperature thermal conductivity of YIG in magnetic fields up to 11 T, plotted as \( \kappa/T \) vs. \( T \). One can see that \( \kappa \) is strongly suppressed by the field, as previously reported [22–24]. In the insulating YIG, the total thermal conductivity can be expressed as \( \kappa = \kappa_{ph} + \kappa_m \), in which \( \kappa_{ph} \) and \( \kappa_m \) are the phonon and magnon thermal conductivity, respectively. Here, the magnon-phonon scattering processes can be safely ignored for the following reasons. Firstly, the low-energy dispersion curves of magnon and phonon cross at a frequency equivalent to 9 K which is much higher than our temperature range, therefore, the first-order one-magnon–one-phonon scattering process should be ignored [24]. Secondly, the relaxation time of boundary scattering \( \tau_b = L/v_p = 1.5 \times 10^{-7} \) s for sample S1 is obtained, in which \( L = 2\sqrt{A/\pi} = 727 \mu \text{m} \) is the boundary-limited mean free path, and \( v_p \approx 5 \times 10^5 \text{cm/s} \) is the average phonon velocity [39]. In contrast, the relaxation time of higher-order magnon-phonon scattering \( \tau_{mp} \approx 1.5 \times 10^{-6} \) s was estimated at 2.5 K [39]. It will be even longer in our temperature range. Because \( \tau_{mp} \) is more than one order longer than \( \tau_b \), the higher-order magnon-phonon scattering should also be ignored.

Since \( \kappa_{ph} \) is usually not affected by the magnetic field, the rapid suppression of \( \kappa \) with the field in fig. 4(a) should come from the reduction of \( \kappa_m \). As we know, the external magnetic field \( H \) opens a gap \( \Delta = g \mu_B H \) in the magnon spectrum. When the external field is high enough to satisfy \( g \mu_B H \gg \kappa_B T \), there should be no magnon contribution. The saturated thermal conductivity from \( H = 4 \) to 11 T shown in fig. 4(a) suggests that there is only a phonon contribution below 0.8 K in \( H \geq 4 \) T, consistent with previous reports [23,24].

Therefore, the FM magnon thermal conductivity \( \kappa_m \) in zero field can be extracted by subtracting \( \kappa(4T) \) from \( \kappa(0T) \). In fig. 4(b), \( \kappa_m \) is plotted as \( \kappa_m/T \) vs. \( T \). For
AFM magnons, ballistic boundary-limited \( \kappa_m = aT^3 \) was observed below 0.5 K [8]. Apparently, for FM magnons in fig. 4(b), there is no such a simple power-law temperature dependence.

Theoretically, the thermal conductivity of magnons is calculated by the equation [19]

\[
\kappa = \frac{k_B}{24\pi^3\hbar} \int \left( \frac{E}{k_B T} \right)^2 \frac{e^{E/k_B T}}{(e^{E/k_B T} - 1)^2} (\nabla_k E) \, dk,
\]

where \( \ell \) is the mean free path of magnons. At not very low temperature, if the approximate dispersion relation \( E = Dk^2 \) of FM magnons is taken and only boundary scattering is considered, we get the \( T^2 \)-dependence of \( \kappa_m \) in eq. (2). Therefore, the anomalous temperature dependence of \( \kappa_m \) in fig. 4(b) must come from the dipole interaction or some other scattering mechanism. Previously, additional magnetic defect scattering was considered, and the data of \( \kappa_m \) between 0.23 and 1 K can be well fitted [24]. In this case, \( \ell^{-1} \) can be expressed as the sum of two terms

\[
\ell^{-1} = L^{-1} + \ell_D^{-1},
\]

where \( L^{-1} \) is from the boundary scattering and \( \ell_D^{-1} \) is from the magnetic defect scattering with \( \ell_D^{-1} = \alpha k^4 \).

With this model, the magnon thermal conductivity is

\[
\kappa_m(T) = BT^2 \int_0^\infty \frac{x^3 \text{csch}^2 \left( \frac{x}{2} \right) dx}{1 + \beta T^2 x^2},
\]

where \( x = E/k_B T, \beta = \frac{\zeta(3)k_B^3 L}{2\pi^4 E}, \beta = \frac{\alpha k_B^3 \ell_D}{2}. \) By using this model, our \( \kappa_m \) data between 0.2 and 0.8 K can also be well fit, with the parameters \( B = 0.056 \text{ mW/K}^2 \text{cm} \) and \( \beta = 0.15 \text{ K}^{-2}. \) Using the obtained value of \( B, \) together with \( D = 5.2 \times 10^{-10} \text{ J/cm}^2, \) we get the boundary-limited mean free path \( L = 32.7 \text{ \mu m}, \) which we will discuss later. Here, we briefly compare the sizes of the two terms in eq. (6). The second term \( \ell_D^{-1} = \alpha k^4 = 4587(\frac{E}{k_B})^2 \) depends on the energy of magnons. It is equal to \( L^{-1} \) for magnons with energy \( E/k_B = 2.58 \text{ K}, \) and rapidly decreases with lowering the magnon energy. In our temperature range 0.07–0.8 K, most magnons have energy lower than 2.58k_B, thus \( \ell_D^{-1} < L^{-1}. \) Therefore, the term from boundary scattering is dominant, and the term from magnetic defect scattering is a correction.

However, the above model cannot fit the \( \kappa_m \) data below 0.2 K, as shown in fig. 4(b). We further consider the effect of the dipole interaction. Since the dispersion relation, eq. (4), is too complex to do a calculation easily, Ortenburger and Sparks chose the simplified dispersion relation [9]:

\[
E = Dk^2 + ck_B.
\]

Substituting this dispersion relation into eq. (5) results in

\[
\kappa_m = BT \int_0^\infty \frac{x^3 \text{csch}^2 \left( \frac{x}{2} \right)(Tx + c)}{1 + \beta (Tx + c)^2} dx.
\]

With the \( B \) and \( \beta \) values obtained above, the \( \kappa_m \) data can be fit well below 0.12 K, as shown in fig. 4(b). We want to emphasize that we tried to fit the \( \kappa_m \) data below 0.8 K with only boundary scattering and dipole interaction, without considering the magnetic defect scattering, but it did not work. Therefore, the magnetic defect scattering mechanism is necessary to explain the low-temperature \( \kappa_m \) data in YIG. Comparing the results of \( C_m \) and \( \kappa_m, \) the dipole interaction starts to affect thermal conductivity at a lower temperature than the specific heat. The reason may relate to the involvement of the magnetic defect scattering in thermal conductivity.

Finally, we discuss the boundary-limited mean free path \( L \) of FM magnons in YIG. From fig. 4(b), \( L = 32.7 \text{ \mu m} \) is obtained by the fitting. This value is one order smaller
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than the expected $L = 727 \mu m$ for sample S1, with $A$ being the cross-section area [40]. Such a phenomenon has been observed previously [22,23,41]. Friedberg and Harris speculated that it is due to the inner boundary of thin layers rich in Fe$^{2+}$ inside the sample [41]. To test this idea, we did the same thermal conductivity measurements on sample S2, obtained by thinning S1 from 0.63 to 0.23 mm. In fig. 5, the 0 and 4 T data of S2 are almost identical to those of S1, indicating the same $\kappa_m$ in S1 and S2. This result shows that $\kappa_m$ indeed does not change with the sample boundary, therefore the actual boundaries are inside the sample, likely the thin layers proposed by Friedberg and Harris [41]. These thin layers may form during the growing process.

**Summary.** – In summary, by extending the measurements of the specific heat and thermal conductivity down to 50 mK, we investigate the thermodynamic and transport properties of the low-lying ferromagnetic magnons in the YIG single crystal. The deviation of the magnon specific heat $C_m(T)$ from the characteristic $T^4$-dependence below 0.77 K is attributed to the effect of the magnetic dipole-dipole interaction. The magnon thermal conductivity $\kappa_m(T)$ is extracted by subtracting $\kappa_m(4T)$ from $\kappa_m(0T)$. Below 0.8 K, $\kappa_m(T)$ does not obey the characteristic $T^2$-dependence due to the magnetic defect scattering. With further decreasing temperature, the magnetic dipole-dipole interaction also has an effect on $\kappa_m(T)$ below 0.2 K. Our work provides a complete picture for the thermodynamic and transport properties of the low-lying ferromagnetic magnons.

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This work is supported by the Natural Science Foundation of China, the Ministry of Science and Technology of China (National Basic Research Program Nos. 2009CB929203 and 2012CB821402), Program for Professor of Special Appointment (Eastern Scholar) at Shanghai Institutions of Higher Learning.

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