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Phonon-magnon interaction in low dimensional quantum magnets observed by dynamic heat transport measurements

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1. Fitting FFM traces to Parker's formula

FFM temperature traces have been fitted to Parker's formula to extract the effective thermal conductivity shown in Fig. 1 (b) of the Letter. Parker's formula [1] is a solution of the one-dimensional homogeneous heat diffusion problem

$$C \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial x^2} \quad (1)$$

solved for a sample with surfaces at $x=0$ and $x=L$ (L is the sample length), where thermal insulation boundary conditions

$$\left. \frac{\partial T}{\partial x} \right|_{x=0,L} = 0 \quad (2)$$

apply. At $t=0$ the optical pulse homogeneously heats a layer of thickness μ from the back surface ($x=0$):

$$T(x, 0) = \Delta T \theta(x - \mu) \quad (3)$$

Here $\theta(x)$ is the Heavyside theta used to describes the step-like heating profile. The temperature rise at equilibrium can be written as a function of the absorbed heating power density Q and the sample specific heat C :

$$\Delta T = \frac{Q}{C} \quad (4)$$

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Using Fourier techniques to integrate the heat equation the time evolution of the temperature at the front surface ($x=L$) is obtained:

$$T(x = L, t) = \frac{\mu}{L} \Delta T \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-d q^2 t} \right] \quad (5)$$

Here $d = k/C$ is the sample diffusivity and $q = \pi/n$.

2. Two temperature model: analytical solution for the FFM experiment

To fit the FFM data we solved the two temperature model

$$C_l \frac{\partial T_l}{\partial t} = k_l \frac{\partial^2 T_l}{\partial x^2} - g(T_l - T_m) \quad (6.1)$$

$$C_m \frac{\partial T_m}{\partial t} = k_m \frac{\partial^2 T_m}{\partial x^2} + g(T_l - T_m) \quad (6.2)$$

For a homogeneous system between $x=0$ and $x=L$ using, in accordance with Parker's single temperature diffusion model, thermal insulation boundary conditions:

$$\left. \frac{\partial T_{l,m}}{\partial x} \right|_{x=0,L} = 0 \quad (7)$$

and as initial conditions:

$$T_l(x, 0) = \Delta T_l \theta(x - \mu) \quad (8.1)$$

$$T_m(x, 0) = \Delta T_m \theta(x - \mu) \quad (8.2)$$

Using the Fourier method for the spatial coordinate, the solution of the model (6) for the lattice temperature is

$$\begin{aligned} T_l(L, t) = & \frac{\mu \Delta T_m + \alpha/\beta \Delta T_l}{L} - \frac{\mu \Delta T_m - \Delta T_l}{L} \frac{1 + \alpha/\beta}{1 + \alpha/\beta} e^{-(\alpha+\beta)t} \\ & + \frac{\mu}{L} \sum_{n=1}^{\infty} (-1)^n \cos \frac{\pi \mu n}{L} \operatorname{sinc} \frac{\pi \mu n}{L} \left\{ \left[\left(1 - \frac{G_n}{\phi_n} \right) \Delta T_l \right. \right. \\ & \left. \left. - \frac{2\beta}{\phi_n} \Delta T_m \right] e^{-(F_n + \phi_n) \frac{t}{2}} \right. \\ & \left. + \left[\left(1 + \frac{G_n}{\phi_n} \right) \Delta T_l + \frac{2\beta}{\phi_n} \Delta T_m \right] e^{-(F_n - \phi_n) \frac{t}{2}} \right\} \end{aligned} \quad (9)$$

Where

$$F_n = \alpha + \beta + (\delta + \gamma)q^2 \quad (10.1)$$

$$G_n = \alpha - \beta - (\delta - \gamma)q^2 \quad (10.2)$$

$$\phi_n = \sqrt{\alpha^2 + 2\alpha[\beta + (\gamma - \delta)q^2] + [\beta - (\gamma - \delta)q^2]^2} \quad (10.3)$$

This is the function used in the fitting of data showed in Fig. 2 of the paper. We have used here the reduced parameters of the model:

$$\alpha = \frac{g}{C_m} \quad \beta = \frac{g}{C_l} \quad \gamma = \frac{k_m}{C_m} \quad \delta = \frac{k_l}{C_l} \quad (11)$$

In the limit $g \rightarrow 0$ the solution shown above reduces to Parker's formula [1] for the lattice alone,

$$\lim_{g \rightarrow 0} T_l(L, t) = \frac{\mu}{L} \Delta T_l \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-\delta q^2 t} \right] \quad (12)$$

and for the case $g \rightarrow \infty$ it reduces to a Parker's formula for the joint (magnon + phonon) system

$$\lim_{g \rightarrow \infty} T_l(L, t) = \frac{\mu}{L} \Delta T_{\text{eff}} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-\delta_{\text{eff}} q^2 t} \right] \quad (13)$$

where $\Delta T_{\text{eff}} = \frac{C_m \Delta T_m + C_l \Delta T_l}{C_m + C_l}$ is the effective temperature rise and $\delta_{\text{eff}} = \frac{k_m + k_l}{C_m + C_l}$ is the effective diffusivity of the joined system.

3. Interplay between defects and phonon-magnon interaction time

The scope of this section to show that: i) the interplay of a long magnon-phonon thermalization time AND the presence of defects in the magnetic structure lowers the measured conductivity with respect to the one of the homogeneous sample; ii) this effect is greater for the dynamic FFM than the SSM; and iii) that in our case, to the end of interpreting experimental data, the inhomogeneous heat conduction problem can justifiably be mapped into the homogeneous one, with the magnetic thermal conductivity suitably renormalized from its intrinsic value.

Point i) and ii) explain, in a qualitative way, the spin ladder anomaly measured in the FFM experiment as a direct consequence and a first, most evident proof of the long magnon-phonon thermalization time and the presence of defects in the one-dimensional magnetic structure. Point iii) justifies the fixing of the k_m parameter in the homogeneous 2T model to approximately half of the intrinsic value in order to fit the spin ladder data.

To model the defected magnetic structure, two pieces of the LDQM under consideration (spin chain or ladder), each of length L' , are joined together through a thin nonmagnetic layer of thickness l , in which the heat is carried only by phonons. (see Figure 1). Thermal insulation boundary conditions are applied for the magnetic subsystem at both external boundaries ($x=0$ and $x=L$) and interfaces ($x=L'$ and $x=L'+l$), whereas the phonon subsystem is taken as continuous and homogeneous throughout the whole sample. The parameter $\chi=l/L$ mimics in the model the defect concentration of the magnetic structure.

Figure 1 upper part: Broken LDQM sample used to simulate the magnetic defects compared with the homogeneous one (lower part). In red the phonon-only breaking layer.

3.1 Modeling the SSM measurements

Following the method outlined by Sanders and Walton [2], the effective thermal conductivity measured by the SSM on the homogeneous sample is

$$k_{SSM} = \frac{k_T}{\epsilon_0} \quad (14)$$

where

$$\epsilon_0 = 1 + \frac{k_m}{k_l} \frac{\tanh(L/2\xi)}{L/2\xi} \quad (15)$$

is the thickness renormalization factor of the total conductivity $k_T = k_l + k_m$. Here, the *healing length* ξ can be defined as:

$$\left(\frac{L}{\xi}\right)^2 = gL^2 \left(\frac{1}{k_l} + \frac{1}{k_m}\right) = \left(\frac{c_l}{c_l + c_m} \tau_m + \frac{c_m}{c_l + c_m} \tau_l\right) \frac{1}{\tau_{mp}} \quad (16)$$

Here the diffusion times for the magnetic and lattice part τ_m and τ_l have been introduced in place of the conductivities, as well as the magnon phonon thermalization time τ in place of the coupling g . The healing length can be seen as the width of the spatial region surrounding a phonon heating source in which the two fluids remain not thermalized.

The corresponding formula for the broken sample can be found as

$$k_{SSM}^{\text{cut}} = \frac{L}{\frac{2L'\epsilon'}{k_T} + \frac{l}{k_l}} = \frac{k_T}{\epsilon'} \frac{1}{1 + \frac{l}{L'} \frac{k_T}{k_l 2\epsilon'}} \approx \frac{k_T}{\epsilon'} \left(1 - \chi \frac{k_T}{k_l \epsilon'}\right) \quad (17)$$

where

$$\epsilon' = 1 + \frac{k_m}{k_l} \frac{\tanh(L'/2\xi)}{L'/2\xi} \quad (18)$$

is the thickness renormalization factor for the broken sample. It can be seen that the value of k_{SSM} is reduced from the value *relative to the homogeneous part of the broken sample* (length L') by a factor linear in the (small) thickness ratio χ and in the second order in the (small) parameter $1/\epsilon'$. This indicates that, to the first order, the spurious thickness dependence induced by the sample breaking in addition to the renormalized homogeneous case can be neglected.

3.2 Modeling the FFM measurements

The analytical SSM results can be compared with the numerical FFM results. By solving the reduced version of the two temperature model

$$\frac{\partial T_l}{\partial t} = \frac{1}{\tau_l} \frac{\partial^2 T_l}{\partial y^2} - \frac{c_m}{c_m + c_l \tau_{mp}} \frac{1}{\tau_{mp}} (T^l - T^m) \quad (19.1)$$

$$\frac{\partial T_m}{\partial t} = \frac{1}{\tau_m} \frac{\partial^2 T_m}{\partial y^2} + \frac{c_l}{c_m + c_l \tau_{mp}} \frac{1}{\tau_{mp}} (T^l - T^m) \quad (19.2)$$

Here the conductivities are included in the diffusion times

$$\tau_{l,m} = \frac{L^2 c_{l,m}}{k_{l,m}} \quad (20)$$

And $y=x/L$ is the reduced spatial coordinate. The time-dependent traces of the temperature at the back sample surface are s and the value of the conductivity can be obtained by fitting to Parker's formula to the back surface temperature trace. In this way, the measurement on the 'broken' spin chain and the 'broken' spin ladder material can be simulated. Results are shown in Figure 1. The material parameters employed here are shown in Table 1 in the main paper.

Figure 2 : Simulated temperature traces for the homogeneous and broken spin chain (fast thermalization time) and ladder (slow thermalization time). Here $x=0.01$. The half rise-time is shown here for the homogeneous (t_0) and cut (t_0^{cut}) samples. For the ladder case the sample with two breaks is shown.

The broken spin ladder (Fig. 2 left) shows a marked reduction ($k_{\text{FFM}}^{\text{cut}} \approx 0.5k_{\text{FFM}}^0$) in the thermal conductivity with respect to the homogeneous case due to its magnon-phonon thermalization time. For the spin chain, (Fig. 2 right) characterized by a faster thermalization, the reduction is much smaller. It is worth noting that the dependence on the layer thickness is small in both cases. This is an indication that, also in the FFM case, the breaks induces negligible additional thickness dependence with respect to the renormalized homogeneous case. This allows us, in order to fit the ladder data, to treat the effect of the defects in the magnetic structure by suitably renormalizing the magnetic conductivity of the homogenous solution of the 2T model, as done in the Letter.

A comparison between the SSM and FFM results on the homogeneous and broken sample is presented in Table 1 as the magnon-phonon thermalization time τ_{mp} is varied with respect to the magnon diffusion time τ_m . In particular, it can be seen that both the SSM and FFM give a smaller thermal conductivity in the broken sample than in the homogeneous one, and that this ‘anomaly’ is between 4 and 1.5 times larger in the FFM (8th column) than in the SSM (5th column). As the thermalization time is reduced, the healing length becomes small with respect to the sample length, and the broken sample results converge towards the homogeneous ones for both method. Crucially, however, there is a thermalization time regime in which the SSM gives essentially the homogeneous value (within the 10-15% reproducibility of the method), while the FFM shows still a measurable anomaly in which the broken conductivity is about 50-70% of the homogeneous (and therefore intrinsic) one.

Although quantitative agreement is beyond the reach of this simple model, these qualitative results point out that: i) the FFM is generally more sensitive to the presence of defects in the magnetic structure than the SSM; ii) The same sample can give rather different conductivities with static and dynamic measurement methods.

τ_{mp}/τ_m	ξ/L	ϵ_0	ϵ'	$\kappa_{\text{SSM}}^{\text{cut}}/\kappa_{\text{SSM}}^0$	t_0	t_0^{cut}	$\kappa_{\text{FFM}}^{\text{cut}}/\kappa_{\text{FFM}}^0$
0.2	0.05	9.15	17.3	0.53	6.0	36	0.17
0.1	0.036	6.76	12.5	0.54	4.4	26	0.17
0.01	0.011	2.82	4.64	0.6	3.2	10	0.32
10^{-3}	0.0036	1.57	2.15	0.73	3.0	5.5	0.55
10^{-4}	0.00114	1.18	1.36	0.87	3.0	4.0	0.75
10^{-5}	0.00036	1.06	1.11	0.95			
10^{-6}	0.000114	1.02	1.03	0.98			

Table 1: Results of the numerical FFM and SSM experiments for different values of the magnon-phonon thermalization time τ for the homogeneous and the broken (cut) sample. The corresponding healing length ξ is also shown. Third-fifth column: SSM results. The normalization factors for the homogeneous ϵ_0 and broken sample ϵ' are compared, as well as the ratio of the two resulting conductivities. Sixth-eight column: FFM results. The half-rise times of the temperature time trace for the homogeneous (t_0) and cut (t_0^{cut}) samples (see Figure 2) and the resulting conductivity ratios are shown. Model parameters are derived from the parameters used to fit the data presented in the paper: $c_l=30$, $c_m=1$, $\tau_l=1600$ $\tau_m=1$, $l=0.01$ and $L=1$.

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