Ballistic Magnon Transport and Phonon Scattering in the Antiferromagnet Nd_2CuO_4

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The thermal conductivity of the antiferromagnet Nd_2CuO_4 was measured down to 50 mK. Using the spin-flop transition to switch on and off the acoustic Nd magnons, we can reliably separate the magnon and phonon contributions to heat transport. We find that magnons travel ballistically below 0.5 K, with a thermal conductivity growing as T^3, from which we extract their velocity. We show that the rate of scattering of acoustic magnons by phonons grows as T^3, and the scattering of phonons by magnons peaks at twice the average Nd magnon frequency.

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The collective excitations of an antiferromagnet are magnons. Like electrons and phonons, magnons can transport heat. Several groups have recently used heat transport to probe spin excitations in quantum magnets [1–6]. The results tend to be difficult to interpret because magnons interact strongly with phonons. However, as $T \rightarrow 0$, the two become decoupled and their respective mean free paths are expected to reach the size of the sample. This ballistic regime is easily reached for phonons, but it has rarely been demonstrated for magnons, because acoustic magnons are almost always gapped. If they were not gapped, their thermal conductivity would be hard to separate from that of acoustic phonons, as both are expected to exhibit a T^3 temperature dependence in the ballistic regime. In this context, the antiferromagnetic insulator Nd_2CuO_4 offers a rare opportunity. Jin *et al.* have recently shown that in this material magnon heat transport can be switched on and off with a magnetic field via a spin-flop transition [7].

In this Letter, we use this ''switch'' to investigate the physics of magnons. First, by cooling down to 50 mK, we show that the ballistic regime for magnon transport, characterized by a clear T^3 conductivity, sets in below 0.5 K. The magnitude gives us the magnon velocity, whose value agrees with a recent calculation [8]. Second, we show how above 0.5 K magnons are scattered by phonons at a rate proportional to T^3. Finally, we show that the scattering of phonons by magnons is strongly peaked at a temperature where the optimum phonon energy equals twice the average magnon energy, in line with a model of dominant one phonon–two magnon processes [9].

Nd_2CuO_4 is the insulating parent compound of the electron-doped high-T_c superconductor. The spins on the Cu^{2+} ions order antiferromagnetically below $T_N \approx 275$ K, with spins lying in the CuO_2 planes. As the temperature is lowered, the ordered spin structure undergoes a series of rearrangements, to reach a noncollinear state at low temperature ($T < 30$ K), with the spin direction changing by 90° in alternate planes along the c axis [8,10–12]. Because of the magnetic exchange interaction between Cu^{2+} and Nd^{3+}, moments on the Nd site also order in the same structure. The magnon spectrum of Nd_2CuO_4, studied by neutron scattering [11,13–15], is characterized by two sets of branches, associated, respectively, with spins on Cu and Nd sites. Cu magnons have energies above 5 meV [11,13], and four optical Nd magnon branches lie in the range 0.2 to 0.8 meV [14,15]. A recent model predicts that the Nd spin sublattice should also support an *acoustic* magnon mode, with a slope of 10 meVÅ and a very small gap (5 μeV) [8]. An in-plane magnetic field of 4.5 T causes a spin-flop transition from noncollinear to collinear state. This change of spin structure modifies the Nd magnon spectrum, as evidenced by the additional heat conduction that appears at the transition [7].

Single crystals of Nd_2CuO_4 were grown by a standard flux method. Our sample was annealed in helium for 10 h at 900 °C, and cut to a rectangular shape of dimensions $1.50 \times 0.77 \times 0.060$ mm^3 (length × length × thickness in the c direction). Contacts were made with silver epoxy, diffused at 500 °C for 1 h. The thermal conductivity $\kappa(T)$ was measured using a standard steady-state technique [16], in a dilution refrigerator (50 mK to 2 K, with RuO_2 thermometers), and in a ^{4}He cryostat (above 2 K, with Cernox thermometers). The magnetic field H was applied perpendicular to the heat current, both in-plane and along the c axis. Note that in a magnetic insulator, the thermal conductivity is the sum of phonon and magnon conductivities: $\kappa(T) = \kappa_p(T) + \kappa_m(T)$.

Ballistic regime.—Figure 1 shows the temperature dependence of κ, in $H = 0$, 10 T $\perp c$, and 10 T $\parallel c$ [17]. The in-plane magnetic field causes a huge enhancement of κ, as discovered by Jin *et al.* [7] above 2 K [18]. The advantage of going lower in temperature by a factor of 40 is that one can then unambiguously decouple κ_p and κ_m. In Fig. 2 we plot the zero-field data (upper panel) as $\kappa(0)$ vs $T^{2.6}$, and the difference between in-field data and zero-field data (lower panel) as $\Delta\kappa = \kappa(10\ \mathrm{T} \perp c) - \kappa(0)$ vs T^3. In both cases, the data follow a straight line below about 0.5 K, which defines the ballistic regime for both types of

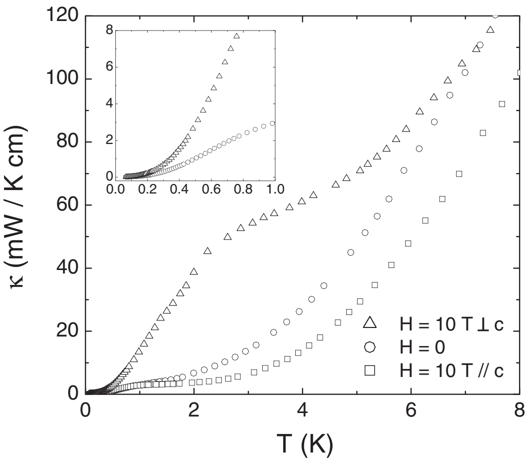

FIG. 1. Temperature dependence of the in-plane thermal conductivity κ of Nd_2CuO_4, in zero magnetic field (circles: $H = 0$) and in a field of 10 T applied either in the basal plane (triangles: $H \perp c$) or along the c axis (squares: $H \parallel c$). Inset: Enlargement below 1 K.

carriers, wherein the mean free path is limited by sample boundaries.

(a) *Phonons* ($T < 0.5$ K).—As $T \to 0$, the phonon mean free path in insulators becomes limited only by the physical dimensions of the sample. In this ballistic regime, the phonon conductivity is given by [19]

$$\kappa_p = \frac{2}{15}\pi^2 k_B (k_B T/\hbar)^3 \langle v_p^{-2}\rangle l_0, \tag{1}$$

where $\langle v_p^{-2}\rangle$ is the inverse square of the sound velocity averaged over the three acoustic branches in all **q** directions, and l_0 is the temperature-independent mean free path, given by the cross-sectional area A of the sample: $l_0 = 2\sqrt{A/\pi}$. For our sample, $l_0 = 0.24$ mm.

For samples with smooth surfaces, a fraction of phonons undergo specular reflection and the mean free path exceeds l_0, estimated for diffuse scattering. Because specular reflection depends on the ratio of phonon wavelength to surface roughness, the mean free path depends on temperature and κ_p no longer grows strictly as T^3 [19]. The general result is that $\kappa_p \propto T^\alpha$, with $\alpha < 3$ [20]. Typical values of α are 2.6–2.8 (see [16] and references therein). For instance, $\kappa_p \propto T^{2.7}$ in the insulator La_2CuO_4 [21] (where all magnons are gapped). In Fig. 2, the zero-field conductivity of our Nd_2CuO_4 crystal can be fitted to $\kappa_p = bT^\alpha$ below 0.4 K, with $\alpha = 2.6$. This is a first indication that $\kappa(0)$ is mostly due to phonons, with a ballistic regime below ~0.5 K.

A second argument comes from the magnitude of b. Taking $l_0 = 0.24$ mm as a lower bound on the mean free path and applying Eq. (1) at $T = 0.4$ K, where $\kappa(0) = 0.61$ mW/K cm, we obtain $\bar{v}_p = (\langle v_p^{-2}\rangle)^{-1/2} = 3.2 \times 10^3$ m/s. A mean free path larger by a factor of 2 would give $\bar{v}_p = 4.5 \times 10^3$ m/s. These values are in good agreement with the measured sound velocities of

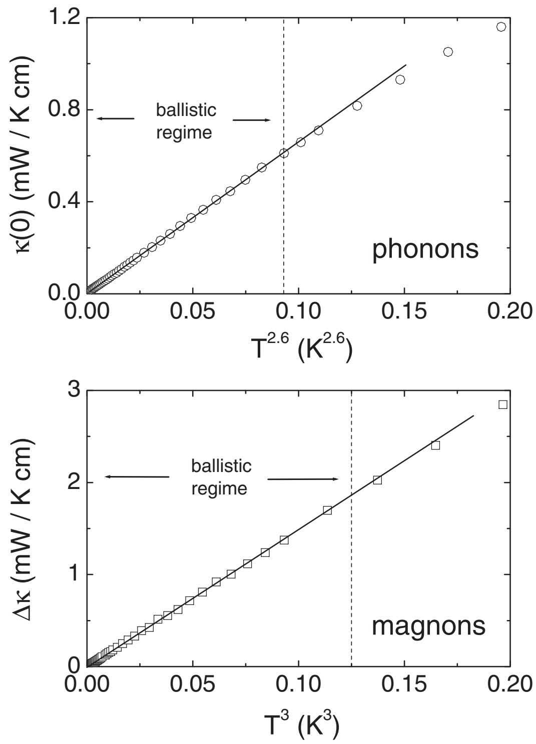

FIG. 2. Top: Zero-field thermal conductivity $\kappa(0)$ vs $T^{2.6}$. The heat at $H = 0$ is carried entirely by phonons: $\kappa_p \simeq \kappa(0)$ (see text). The line is a linear fit below 0.4 K. Bottom: The additional thermal conductivity induced by a 10 T in-plane field, $\Delta\kappa = \kappa(10\ \mathrm{T} \perp c) - \kappa(0)$, plotted as $\Delta\kappa$ vs T^3. The line is a linear fit below 0.5 K. The additional heat is carried entirely by acoustic magnons: $\kappa_m = \Delta\kappa$ (see text).

Nd_2CuO_4: $v_L = 6.05 \times 10^3$ m/s and $v_T = 2.46 \times 10^3$ m/s for longitudinal and transverse modes along [100] [22]. So it appears that phonons account for the full $\kappa(0)$. All the evidence suggests that magnon transport is negligible in the noncollinear state at any temperature in the range considered here (up to 20 K), so that $\kappa(0) \simeq \kappa_p$ at all T in that range.

(b) *Magnons* ($T < 0.5$ K).—Magnon transport is revealed by looking at the difference between the two curves in the inset of Fig. 1, $\Delta\kappa = \kappa(10\ \mathrm{T} \perp c) - \kappa(0)$, plotted in the bottom panel of Fig. 2. We get the striking result that $\Delta\kappa = dT^3$, below 0.5 K. Such a field-induced additional T^3 contribution to κ at low T can be attributed unambiguously to acoustic magnons. To our knowledge, this is the first observation of the characteristic T^3 dependence of magnon thermal conductivity [23]. As bosons with a linear energy dispersion at low q, acoustic magnons should contribute to heat transport in the same way as acoustic phonons, provided there is no gap in their spectrum. Hence for gapless acoustic magnons, Eq. (1) should apply in the ballistic regime, with $\langle v_m^{-2}\rangle$ replacing $\langle v_p^{-2}\rangle$ [24]. Note, however, that the prefactor will depend on the number of magnon branches. In the simplest case, one expects two degenerate magnon branches, with the degeneracy lifted by a magnetic field. As our data are in 10 T, we assume that only one branch lies at zero energy. Hence

$$\kappa_m = \frac{1}{3} \times \frac{2}{15} \pi^2 k_B (k_B T/\hbar)^3 \langle v_m^{-2} \rangle l_0. \qquad (2)$$

Assuming that $\kappa(0) = \kappa_p$ and that κ_p is independent of magnetic field in the ballistic regime, we have $\kappa_m = \kappa - \kappa_p = \Delta\kappa$, so that $\kappa_m = dT^3$, with $d = 15$ mW K^{-4} cm^{-1}. Using Eq. (2) with $l_0 = 0.24$ mm, we get $\bar{v}_m = (\langle v_m^{-2} \rangle)^{-1/2} = 1.5 \times 10^3$ m/s.

No calculation of magnon modes has been reported for the *collinear* phase of Nd_2CuO_4. However, it is worth noting that our findings of an acoustic magnon mode with velocity 1500 m/s (10 meVÅ) and a gap smaller than 5 μeV (consistent with our lowest temperature of 50 mK) is in quantitative agreement with the theoretical calculation of Sachidanandam *et al.* for the *noncollinear* phase in zero magnetic field [8].

An intriguing difference between phonons and magnons in the ballistic regime is that only the former appear to undergo specular reflection. Indeed, the perfect T^3 dependence of κ_m implies that the fraction of magnons that are specularly reflected must be negligible, while it is not for phonons. We speculate that the strong anisotropy expected of the magnon velocity in Nd_2CuO_4, compared to the roughly isotropic phonon velocity, might account for this difference.

Inelastic regime.—Beyond the ballistic regime, the temperature interval up to 10 K or so is dominated by phonon-magnon scattering: (a) acoustic phonons are scattered strongly by magnons, and (b) acoustic magnons are scattered strongly by phonons.

(a) *Phonons* ($T > 0.5$ T).—A 10 T field along the c axis leaves the system in its noncollinear state, and, as seen in Fig. 1, it has negligible impact below 1 K. Above 1 K, however, it clearly suppresses κ, relative to $\kappa(0)$. Although the 10 T $\parallel c$ curve is more striking, both in-field and zero-field curves reveal that the rapid growth in phonon conductivity generally observed in insulators is almost entirely stopped between 1 and 3 K. An extremely effective inelastic scattering mechanism switches on near 1 K and off above 3 K. This range of energies is precisely that of Nd magnons in Nd_2CuO_4.

The theory of phonons scattered by magnons was given a thorough treatment by Dixon [9], who argues that the one phonon-two magnon process is dominant. Dixon gets two main results: (1) the scattering peaks at a temperature such that the optimum phonon frequency ($\sim 4k_BT$) is equal to the average energy of two magnons, and (2) a magnetic field enhances the effectiveness of magnon scattering, especially when the field is perpendicular to the spin direction. We attribute the suppression of κ induced by applying $H \parallel c$ to this enhanced effectiveness.

Phonons are scattered by magnons and several other processes (boundaries, defects, other phonons). However, because only magnon scattering depends on magnetic field, we can bring out the role of magnon scattering by focusing on the *change* in thermal *resistivity*, $w = 1/\kappa$, i.e., the thermal magnetoresistance $\Delta w \equiv w(10\ \mathrm{T} \parallel c) - w(0)$. In the top panel of Fig. 3, the normalized magnetoresistance, $\Delta w/w(0)$, is plotted vs T up to 20 K. Quite evidently, phonon transport emerges as a spectroscopy of magnons. (The same can be true of *electron* heat transport, as shown recently for $CeRhIn_5$ [25].) Indeed, the maximum scattering of phonons will occur when the peak phonon energy matches twice the average energy of optical magnons, $\langle E_{\mathrm{magnon}} \rangle$ [9]. At $T = T^*$, heat transport is dominated by thermal phonons with frequencies about $\omega^*_{\mathrm{phonon}} = 4k_BT^*/\hbar$. Therefore, in Nd_2CuO_4, where $\langle E_{\mathrm{magnon}} \rangle = 0.5$ meV is the average of the measured optical magnon energies (which range from 0.2 to 0.8 meV [14,15]), $w(T)$ is expected to peak when $\hbar\omega^*_{\mathrm{phonon}} = 2\langle E_{\mathrm{magnon}} \rangle \simeq 4k_BT^*$, i.e., $k_BT^* = 0.25$ meV, or $T^* \simeq 3$ K, precisely as observed.

(b) *Magnons* ($T > 0.5$ K).—We assume that magnons in 10 T $\parallel c$ scatter phonons just as well as in 10 T $\perp c$ [26], so that $\kappa_p(10\ \mathrm{T} \parallel c) \simeq \kappa_p(10\ \mathrm{T} \perp c)$, and $\kappa_m(10\ \mathrm{T} \perp c) \simeq \kappa(10\ \mathrm{T} \perp c) - \kappa(10\ \mathrm{T} \parallel c) \equiv \Delta\kappa'$ (which is equal to $\Delta\kappa$ below 1 K). In the bottom panel of Fig. 3, we plot $\Delta\kappa'$ vs T. The magnon conductivity κ_m peaks at 3 K,

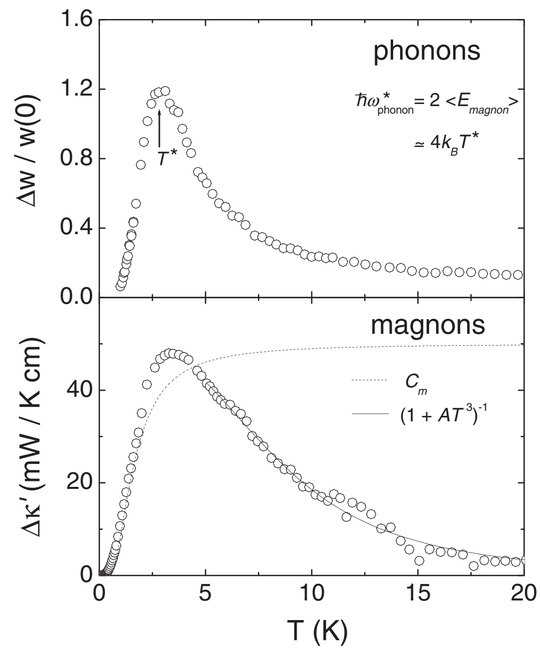

FIG. 3. Top: Normalized thermal magnetoresistance vs temperature, for $H \parallel c$. This is the field-dependent thermal resistivity of phonons [27]. The arrow indicates the temperature at which magnon scattering is maximum: $T^* = 3$ K. T^* is directly related to the magnon energy $\langle E_{\mathrm{magnon}} \rangle$ via the peak phonon frequency, $\omega^*_{\mathrm{phonon}}$ (see text). Bottom: Difference in thermal conductivity caused by rotating a 10 T field from in plane to c axis, $\Delta\kappa' = \kappa(10\ \mathrm{T} \perp c) - \kappa(10\ \mathrm{T} \parallel c)$, vs temperature. This is the conductivity of magnons. The dotted line is the estimated magnon specific heat (see text) and the solid line is a fit as indicated.

because the magnon heat capacity $C_m(T)$ saturates when T exceeds the maximum energy of the acoustic magnon branch, while the inelastic scattering due to phonons keeps increasing. That maximum energy is unknown, but we expect it to lie roughly in the range 0.2–0.8 meV. We estimate $C_m(T)$ using an Einstein model: $C_m = C_E = z^2 e^z (e^z - 1)^{-2}$ where $z = \hbar\omega_E / k_B T$, with $\hbar\omega_E =$ 0.5 meV. This is clearly inaccurate at very low T where $C_m \propto T^3$, but should be reasonable for temperatures comparable to and above the maximum energy, given roughly by $\hbar\omega_E$. As seen in Fig. 3, C_m is constant above 5 K or so and the temperature dependence of κ_m comes entirely from the scattering rate, which we model simply as the sum of boundary and phonon scattering: $\Gamma = \Gamma_0 + \Gamma_p$. As seen in Fig. 3, a good fit to $\kappa_m(T)$ is obtained with $\Gamma_p \propto T^3$ (and Γ_0 constant, as it should). This shows that acoustic phonons scatter acoustic magnons (as they do electrons) in proportion to their number, which increases as T^3. The magnon mean free path in Nd_2CuO_4 decreases by a factor of 15 between 5 and 20 K, and should continue to decrease up to roughly the Debye temperature. Although these data and analysis apply strictly only to the low-energy Nd magnon branch, they do suggest that magnon mean free paths in cuprates even at higher energies are unlikely to ever be independent of temperature in the range 20 to 200 K, as was assumed in the analysis of data on La_2CuO_4 [5]. Note that additional heat transport is expected to occur when the higher-energy Cu magnon modes become thermally excited as the temperature exceeds their gap, typically above 50 K or so.

In summary, by cooling to 50 mK, the characteristic T^3 heat transport expected of ballistic magnons has for the first time been observed, revealing the existence of an acoustic magnon branch (gapless or with a very small gap, with upper bound ~0.1 K) in the collinear phase of Nd_2CuO_4 ($H > 7$ T $\perp c$). Paradoxically, the properties of this mode agree well with recent calculations applied to the *noncollinear* phase ($H = 0$) [8], where our data (in $H = 0$ or $H \parallel c$) rule it out. A calculation specifically for the collinear phase in field is desirable, as well as a search for the acoustic branch via low-energy neutron scattering. Above the ballistic regime, phonons scatter magnons strongly, initially at a rate $\Gamma_p \propto T^3$, so that the magnon mean free path decreases rapidly with temperature, and should continue to do so up to roughly the Debye temperature. Phonons are themselves strongly scattered by the magnons, in the temperature range where one phonon–two magnon processes are most effective, namely, in the vicinity of $4k_B T = 2\langle E_{\mathrm{magnon}}\rangle$. In general, one can expect phonons in all cuprates (at any doping) to be scattered significantly by magnetic excitations (typically from Cu moments).

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[17] The magnitude of the in-plane magnetic field needed to induce the spin-flop transition is somewhat anisotropic [7]. For our sample, κ starts to increase with field above 1.5 T at 0.3 K, and saturates beyond 7 T. A field of 10 T was chosen to ensure that the sample has fully reached the collinear state at all relevant temperatures.
[18] Note that the pronounced anomaly at 5 K (sharp peak) in κ reported by Jin *et al.* [7] is entirely absent from our data.
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[23] T^3 is expected for 3D magnetic order, T^2 for 2D. With a c-axis correlation length in excess of 530 Å [12], Nd_2CuO_4 is clearly 3D.
[24] Note that in Nd_2CuO_4 $\langle v_m^{-2}\rangle$ is a 3D average over a strongly anisotropic spectrum.
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[26] This assumption is valid at temperatures where phonon energies are above twice the characteristic energy of the Nd magnon branches, since that characteristic energy is the same in both magnetic states, governed as it is by the crystal-field level splitting of the Nd rare earth ion, independently of the particular long-range order adopted in the crystal [8]. This translates to $T > 2$ K.
[27] The assumption that the difference data are predominantly due to a change in the thermal resistivity of phonons is supported by the fact that any contribution from magnon transport, most likely very small, would at any rate not be strongly dependent on field. For example, the magnon conductivity of the collinear state is observed not to depend on field (beyond the spin-flop field).