Size-dependent magnon thermal transport in a nanostructured quantum magnet

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SUMMARY

Magnons are fundamental excitations of spin waves in magnetic materials, and are able to carry both heat and spin information. Examining magnon thermal transport is essential for advancing energy-efficient spintronics and novel energy conversion technologies. Unlike the well-established thermal properties of phonons and electrons, the size effect on magnon thermal transport remains elusive. Here, we quantitatively measure the role of grain size on magnon thermal transport in the nanostructured quantum magnet La_2CuO_4. Samples with controlled grain sizes are synthesized using a bottom-up method. The measured magnon thermal conductivity decreases as the grain size is reduced. Analysis using a two-dimensional magnon transport kinetic model, combined with optical and electrical measurements, suggests that magnon-boundary scattering effectively restricts the magnon mean free path, suppressing magnon thermal conductivity. These results offer valuable insights into the prediction and control of magnon transport at the nanoscale, especially for applications in quantum information processing and energy conversion.

INTRODUCTION

Magnons are quasiparticles, which are associated with wave-like disturbances in ordered spin systems, and can carry both thermal energy and spin angular momentum.[1,2] High values of magnon-mediated thermal conductivity (κ_m) have been reported in several magnetic cuprates,[3,4] which are promising for thermal management applications. Magnon thermal transport also plays an important role in the spin Seebeck effect,[5] which describes the creation of a spin current in a magnetic material by a temperature gradient. This effect could be harnessed for waste heat recovery and efficient thermoelectric energy conversion.[6] In the field of spintronics, the spin of electrons can be harnessed to store, process, and communicate information. The incorporation of magnons into advanced spintronic devices offers prospects for lower power consumption and diminished heat generation.[7] Therefore, an improved understanding of magnon transport, in particular on the micro- and nano-scale, is pivotal for the development of innovative applications in both quantum information

processing and energy conversion.[8–12]

In contrast to the established thermal transport properties of phonons and electrons, length scales for thermal transport and scattering mechanisms of magnons are not yet well understood.[13] As electronic devices continue to shrink to nanoscale dimensions, so size effects on thermal transport become increasingly important.[14] For example, nanostructured thermoelectric materials have improved performance for waste heat recovery and refrigeration applications.[15] In general, smaller grain sizes lead to increased scattering of phonons at boundaries, which can reduce the phonon mean free path (MFP) and result in lower lattice thermal conductivity (κ_{L}).[16] Size effects on phonon heat transport have been observed in several experimental studies.[17–19] However, measurements of the influence of grain size on magnon thermal transport have been hindered by the presence of defects in samples, such as $Sr_{14}Cu_{24}O_{41}$ polycrystals[20] and microrods[21], and $Ca_9La_5Cu_{24}O_{41}$ polycrystals.[22] For these samples, the processing methods that were used resulted in increased defect concentrations, leading to reduced values of κ_{m} that were dominated by magnon-defect scattering. It is therefore critical to synthesize high quality samples, in which the effects of magnon-boundary scattering can be studied.

The cuprate-based quantum magnet La_2CuO_4 is a well-known antiferromagnetic perovskite-related compound, in which the magnetic moments of neighboring Cu^{2+} ions are aligned antiparallel to each other in the ab-plane below the Néel temperature of ~320 K.[23–27] This two-dimensional (2D) square-lattice quantum magnet shows several interesting physical properties, including superconducting behavior[28] and a thermal Hall effect.[29,30] Furthermore, thermal transport measurements on single crystals[4,31] have revealed high values of κ_{m} in the ab-plane. However, magnon thermal transport in La_2CuO_4 nanostructures is not yet well understood.

Here, we investigate the influence of grain size on magnon thermal transport in nanostructured La_2CuO_4 synthesized by a bottom-up approach. High quality La_2CuO_4 nanorods (NRs) and nanoparticles (NPs) are prepared by a wet chemical method. The nanostructures are consolidated into dense pellets with different grain sizes by using spark plasma sintering (SPS). Our thermal conductivity (κ) measurements show that a decrease in grain size results in a reduction of κ_{m}. Analysis of the experimental data using a kinetic model for 2D magnon transport confirms the role of magnon-boundary scattering in suppressing κ_{m} in La_2CuO_4. The growth mechanism of the La_2CuO_4 nanostructures is also discussed based on a template method. Our findings provide new insight into the factors that influence magnon thermal transport on the nanoscale, and lay the groundwork for the heterogeneous integration of magnetic nanostructures in energy and quantum information applications.

RESULTS AND DISCUSSION

Crystallographic information and structural characterization

La_2CuO_4 has a 2D magnetic structure, which consists of Cu^{2+} ions in CuO_2 planes that extend along the crystallographic a and b directions.[4] Figure 1a shows that La_2CuO_4 forms a spin-1/2 Heisenberg antiferromagnet on a square lattice, which can lead to strong antiferromagnetic intraplanar exchange coupling (J) with $J/k_B \approx 1550$ K,[32] where k_B is the Boltzmann constant. In comparison, the interplanar exchange $J_\perp$ ($J_\perp/J \approx 10^{-5}$) is negligible.[33]

La_2CuO_4 nanostructures were prepared by using hydrothermal treatment followed by calcination. Figures S1a and b in the Supporting Information (SI) show X-ray diffraction (XRD) patterns

recorded before and after calcination, respectively. After the hydrothermal process, the samples comprise $La(OH)_3$ and CuO. After calcination, a pure La_2CuO_4 phase is formed. Figures 1b and g show scanning electron microscopy (SEM) images of the final products, which were prepared under different OH^- concentrations (ρ_{OH^-}) during the hydrothermal process and include La_2CuO_4 NPs and NRs. The grain size of the NPs is nearly uniform, averaging ~350 nm. The NRs have diameters of between 50 and 250 nm, and lengths ranging from 1 to 11 μm. Energy-dispersive X-ray spectroscopy (EDX) analysis confirms a uniform distribution of La, Cu and O in the La_2CuO_4 nanostructures (Figure S2).

In order to verify the crystallographic quality of the La_2CuO_4 nanostructures, transmission electron microscopy (TEM) characterization was performed on individual NPs and NRs. Figures 1c-f and h-k confirm that the NPs and NRs are single crystals. The growth direction of the NRs is observed to be $[111]$. High-resolution TEM (HRTEM) images (Figures 1e and j) and corresponding filtered inverse fast Fourier transforms (FFTs) of the marked square regions (Figures 1f and k) show that there are almost no dislocations in the observed regions of the nanostructures.

Growth mechanism of nanostructures

In order to better control the morphology of the La_2CuO_4 nanostructures, it is important to study their growth mechanisms. Figures 2a and b show that, after the hydrothermal treatment, the NP and NR samples contain nanoparticles and a mixture of nanowires and nanoparticles, respectively. It should be noted that the nanowires form at a higher OH^- concentration. XRD results (Figure. S1a) indicate that the product comprises $La(OH)_3$ and CuO prior to calcination. The following chemical reactions take place during the hydrothermal process:

$$Cu(NO_3)_2 + KOH \rightarrow Cu(OH)_2 + KNO_3 \quad (1)$$

$$Cu(OH)_2 \rightarrow CuO + H_2O \quad (2)$$

$$La(NO_3)_3 + KOH \rightarrow La(OH)_3 + KNO_3, \quad (3)$$

where $Cu(OH)_2$ is metastable, and transforms easily into more stable CuO in the solid state at room temperature in aqueous basic solutions.[34,35] We performed EDX analysis on the NR sample before calcination. Figures 2c, d, e and f show that La is only present in the nanowires, while Cu in present exclusively in the particles. These observations suggest that the nanoparticles are CuO. In contrast, the nanowires, which can be regards as templates for the formation of La_2CuO_4 NRs, are $La(OH)_3$, as illustrated in Figure 2g.

The concentration of OH^- ions during the hydrothermal process is a crucial factor governing the formation of nanostructures. First, a higher OH^- concentration results in an increased PH value and a higher chemical potential in the supersaturated solution, which, in turn, magnifies the difference between the free energies of the liquid and solid phases, providing a more powerful thermodynamic driving force for the system to achieve equilibrium.[36] The nucleation process is then accelerated, leading to the formation of a greater number of $La(OH)_3$ crystal nuclei. Furthermore, OH^- ions can bind selectively to specific crystal facets of the $La(OH)_3$ nuclei during the hydrothermal process, acting as crystal facet inhibitors.[37] This selective attachment of OH^- ions can modify the growth rates of different facets, resulting in the formation of nanowires.[38,39]

We also observed that the heating rate during calcination has a significant influence on the length and surface smoothness of La_2CuO_4 NRs. Figure S3 shows that the use of a higher heating rate results in the formation of longer NRs with smoother surfaces. This behavior is particularly evident in Figure 1g for a heating rate of 3 K/min. Intriguingly, at a lower heating rate nano-bamboos with

dislocations are observed (Figure S4), in agreement with a previous study.[40] Their formation is attributed to the fact that a lower heating rate cannot provide an optimal surface energy, which causes the nanostructures to fracture.

Optical, Seebeck coefficient and thermal property measurements

In order to study the thermal transport of La_2CuO_4, the nanostructures were pressed into dense pellets using SPS. For comparison, La_2CuO_4 powders were also prepared with a larger grain size using solid-state reaction (SSR). The densities (ρ) of the NP, NR and SSR pellets were measured as 6.23, 5.81 and 5.19 g/cm^3, respectively, corresponding to relative densities of 90%, 84%, and 75%. Figure 3 shows that the average grain sizes (G) in the NP, NR and SSR samples after SPS are approximately 560 nm, 780 nm and 2.9 μm, respectively. Additionally, Figure S5 demonstrates that all three constituent elements (La, Cu, and O) are uniformly distributed across all three samples, with no secondary phases detected.

Raman scattering measurements were carried out to examine the defects in the three samples after SPS, as shown in Figure 4a. A broad peak, which can be attributed to two-magnon scattering,[41] becomes discernible at ~3200 cm^{-1}. This type of scattering arises from two magnons near the center of the Brillouin zone, which have equal but opposite wave vectors (k). Previous studies show that the presence of defects, such as oxygen vacancies, significantly influences the possibility of two-magnon interactions,[41] leading to variations in the intensity of two-magnon peaks.[21] Our Raman spectroscopy data, which show similar two-magnon peak positions and intensities for the three samples, suggest that they have similar defect concentrations. In addition, as discussed in Note S6, our Raman data exhibit a high degree of consistency with the positions of phonon peaks below 1500 cm^{-1} as compared to previous studies.[42,43] Considering that the presence of lattice defects can affect the specific heat (C_p) at lower temperatures, we proceeded to measure C_p for each sample.[44] As shown in Figure 4b, the C_p values for all three samples, especially the low-temperature regime, are closely aligned. This observation suggests that the quality of the samples is comparable. Furthermore, it has been found that the addition of oxygen to La_2CuO_4 can lead to hole doping, which can serve as the primary source of mobile defects.[45] Accordingly, we conduct measurements of the Seebeck coefficient (S), which is inversely proportional to the concentration of holes.[46] The schematic of the measurement setup for S and κ is illustrated in Figure 4c. Figure 4d shows that the S values of the three samples are positive, with magnitudes that are comparable to each other, especially close to room temperature. This result further suggests that the concentration of mobile defects induced by additional oxygen in each sample is similar. By combining Raman spectroscopy, specific heat, and Seebeck coefficient measurements, it is evident that the concentration of defects is at a similar level across all three samples.

Figure 4e shows the value of κ measured for the three La_2CuO_4 samples parallel (in-plane, IP) and perpendicular (out-of-plane, OP) to the SPS uniaxial pressure direction. Possible sources of error include radiation from the sample and heat leaks from the shoe assemblies, among others. Further details about the uncertainty analysis can be found in Note S7. Intriguingly, the κ values for the NR sample exhibit significant anisotropy when compared to the SSR and NP samples, which, in contrast, display nearly isotropic behavior. Over the temperature range 50 to 350 K, the values of κ for the NR-IP measurement are approximately 1.5 times greater than for the NR-OP measurement. This anisotropy in κ can be attributed to the NRs having a greater tendency to align along the in-plane direction during SPS, as shown schematically in the inset of Figure 4e. The presence of a

pronounced texture in the NR sample is also confirmed by XRD analysis, as shown in Figure S7, where the strong diffraction intensity of the (111) peak is measured on the plane parallel to the press direction.

In order to correct for the porosity effect, the solid thermal conductivity (κ_s) can be calculated using the Maxwell-Eucken relation.[47] This relation has been widely used for calculating κ_s for various materials and is found to be valid for materials with porosity less than 65%.[48] Given that the porosity (Φ) of the SSR, NR, and NP samples is 25%, 16%, and 10%, respectively — all well below the 65% threshold — the porosity effect can be appropriately corrected using the Maxwell–Eucken relation. Accordingly, the κ_s of three samples can be calculated using the following relationship[48]

$$\kappa = \kappa_s \frac{\kappa_p + 2\kappa_s + 2\Phi(\kappa_p - \kappa_s)}{\kappa_p + 2\kappa_s - \Phi(\kappa_p - \kappa_s)}, \quad (4)$$

where κ_p is the pore thermal conductivity. Since our measurements were conducted under ultra-high vacuum, κ_p should be equal to 0. More details about correction for porosity effect can be found in Note S9. The calculated values of κ_s for the three samples are shown in Figure 4f, together with the single-crystal data along the ab plane reported by Hess *et al.*[4] and Sun *et al.*[31] It should be noted that a discrepancy exists in κ values reported for La_2CuO_4 single crystals. This discrepancy may be attributed to the different qualities of the single crystals. In addition, for $T <$ 250 K, the κ_s values of the SSR sample are higher than the single crystal data reported by Hess *et al.*[4] Notably, κ_m is negligible below ~50 K due to the energy gap for magnon dispersion.[4,49,50] We therefore fitted the data at lower temperature to the Debye–Callaway model for three-dimensional (3D) phonon transport and extrapolated the fitting results at higher temperature,[51] as shown in Figure 4f. The details of the fitting procedure and the reliability of the Debye-Callaway model for κ_L are given in Table S1, Notes S10 and S11.

Magnon thermal transport analysis

The κ_m values of the samples can be determined from[4]

$$\kappa_m = \kappa_s - \kappa_L . \quad (5)$$

Figure 5a shows that the texture effect persists in the NR sample even after correcting for the porosity effect and eliminating the phonon contribution. Over the temperature range 50 to 350 K, $\kappa_{m,NR-IP}$ exceeds $\kappa_{m,NR-OP}$, with the difference widening with increasing temperature. Moreover, the anisotropy of κ_m increases as the temperature rises. At 350 K, the value of $\kappa_{m,NR-IP}$ ≈5.3 $Wm^{-1}K^{-1}$ is nearly twice that of $\kappa_{m,NR-OP}$ ≈2.9 $Wm^{-1}K^{-1}$.

Since La_2CuO_4 exhibits a 2D magnetic structure and its spin-plane (ab-plane) has extremely high symmetry, the value of κ along the c-axis (κ_c) is determined only by phonons, while the value of κ in the ab-plane (κ_{ab}) is uniform.[54,55] The intrinsic magnon thermal conductivity (κ_m^i) can be expressed in the form[4]

$$\kappa_m^i = \frac{3}{2}\left(\kappa^{poly} - \kappa_L^{poly}\right) = \frac{3}{2}\kappa_m^{poly} = \frac{3}{2}\frac{\left(2\kappa_{m,IP} + \kappa_{m,OP}\right)}{3}, \quad (6)$$

where κ_m^{poly} is the magnon thermal conductivity of polycrystals.[4] Detailed discussion of Equations 5 and 6 can be found in Note S12. Figure 5b shows the obtained κ_m^i data for the three polycrystalline samples and two single crystals, displaying an increase with temperature. At 300 K, the κ_m^i values for the SSR, NR and NP samples are approximately 9.6, 6.8 and 5.5 $Wm^{-1}K^{-1}$, respectively. The order of the magnitudes of the values of κ_m^i is consistent with the grain size order. In addition, the values of κ_m^i of the three samples are all lower than the single-crystal values

of 10.5 $Wm^{-1}K^{-1}$ reported by Hess *et al.*[4] and 11.1 $Wm^{-1}K^{-1}$ reported by Sun *et al.*[31]. This phenomenon, which is observable across the entire temperature range of 50 to 350 K, provides strong evidence for a grain size effect on magnon thermal transport. The only exception occurs when $T <$ 250 K, when the value of κ_{m}^{i} reported by Hess *et al.*[4] is lower than the value of κ_{m}^{i} for the SSR sample. This discrepancy may be attributed to a different stoichiometric ratio in the samples, for example resulting from the concentration of oxygen interstitial defects.[56] The presence of interstitial oxygen atoms may reduce the value of κ_{m} in La_2CuO_4.

We calculated the average magnon MFP (l_{m}) using the κ_{m}^{i} based on a kinetic model for 2D magnon transport,[4,45,57] which takes the following form

$$\kappa_{\mathrm{m}}^{i} = \frac{k_{\mathrm{B}}^{3}T^{2}l_{\mathrm{m}}}{2\pi\hbar^{2}v_{0}c}\int_{\frac{\Delta}{k_{\mathrm{B}}T}}^{\infty} x^{2}\sqrt{x^{2}-x_{0}^{2}}\,\frac{e^{x}}{(e^{x}-1)^{2}}\,\mathrm{d}x \ , \tag{7}$$

where c =13.2 Å is the lattice constant of La_2CuO_4 perpendicular to the spin planes, v_0 is the spin wave velocity of ~1.287×10^5 m s^{-1},[32] $\hbar$ is the reduced Planck constant, Δ is the spin gap of the magnon branch, $x = \frac{\hbar\omega}{k_{\mathrm{B}}T}$ and $x_0 = \frac{\Delta}{k_{\mathrm{B}}T}$. Based on a previous inelastic neutron scattering study, it is known that there are two magnon branches with Δ_1/k_{B} =26 K and Δ_2/k_{B} =58 K.[49] In order to ensure that the magnons are fully excited, Δ_2 was chosen for calculation. Figure 5c shows l_{m} calculated for the polycrystalline samples, compared with the single-crystal data. l_{m} for the SSR sample is much larger than that for the NR and NP samples. At 300 K, $l_{\mathrm{m,SSR}} \approx$708 Å is nearly 1.5 and 1.8 times larger than $l_{\mathrm{m,NR}} \approx$479 Å and $l_{\mathrm{m,NP}} \approx$386 Å, separately. Smaller grain sizes introduce a higher boundary scattering rate, which in turn reduces l_{m}. Moreover, values of l_{m} for single crystals reported by Hess *et al.*[4] and Sun *et al.*[31] are higher than that of our polycrystalline samples above 250 K.

In order to better understand the origin of the suppressed values of κ_{m}^{i} and l_{m} in the polycrystalline samples, we calculated κ_{m} by including a magnon boundary scattering contribution (l_{b}) to the total MFP in the form

$$l_{\mathrm{m}}^{-1} = l_{\mathrm{xtl}}^{-1} + l_{\mathrm{b}}^{-1}, \tag{8}$$

where l_{xtl} is the magnon MFP in the single crystal. Figure 5d shows the experimental values of κ_{m} plotted as a function of grain size at T =300 K together with the calculated values obtained using Equations 7 and 8. The low-temperature data can be found in Figure S10a. It is clear that the calculated κ_{m}^{i} decreases as the grain size decreases. The experimental results follow a similar trend. However, there are obvious deviations between experimental and calculation results. Previous research has revealed the presence of various defects, such as edge dislocations and planar defects, in $Sr_{14}Cu_{24}O_{41}$ polycrystals produced by a solid state reaction followed by SPS.[21] Such defects are also anticipated to be present in our samples.

In order to quantify the influence of grain and defect scattering on κ_{m}, we introduce a defect scattering term (l_{d}) in the expression for the total MFP in the form[20]

$$l_{\mathrm{m}}^{-1} = l_{\mathrm{xtl}}^{-1} + l_{\mathrm{b}}^{-1} + l_{\mathrm{d}}^{-1} \quad . \tag{9}$$

According to Callaway *et al.*,[58,59] l_{d}^{-1} can be calculated from the expression $l_{\mathrm{d}}^{-1} = N_{\mathrm{d}}Ak^{4}$ for a 3D isotropic system, where N_{d} is the number of magnetic defects and A is a constant that is related to the material's properties. The origin of this relationship lies in the fact that the scattering rate depends on the wave vector as $k^2D(k)$,[60–62] where the density of states $D(k)$ is proportional to k^2 for a 3D system. Since $D(k)$ is proportional to k in a 2D system, the magnon defect scattering rate is expected to exhibit a k^3 dependence. Therefore, the MFP for magnon defect

scattering is expected to follow a dependence of the following form for 2D magnon transport in spin-plane compounds

$$l_{\mathrm{d}}^{-1} = c_{\mathrm{d}} k^3, \tag{10}$$

where c_{d} is a constant. Figure S10b shows that the experimental data can be fitted well using Equations 7, 9 and 10. The corresponding fitting parameters l_{b} and c_{d} are given in Figure 5e. A clear observation from our data is that, for different samples, the variation in l_{b} is substantially more pronounced than that in c_{d}. Although the values of l_{b} obtained for the three samples are smaller than the respective average grain sizes, the order of the fitted values for the magnon boundary scattering MFP still follows $l_{\mathrm{b,SSR}}$ (2.6 μm) > $l_{\mathrm{b,NR}}$ (630 nm) > $l_{\mathrm{b,NP}}$ (410 nm). This result indicates that, even taking defect scattering into account, the impact of grain size on magnon thermal transport remains significant. It is clear that the size effect is the primary factor that is responsible for suppressing both l_{m} and κ_{m} in nanostructured samples, a phenomenon that had not been demonstrated in previous research,[21,22] as shown in Figure 5f.

When magnons cross through grain boundaries, they have two primary pathways.[20] The first is that magnons couple with magnons across the grain boundary directly. However, this process can be significantly hindered due to the impact of the grain boundary on the short-range antiferromagnetic exchange interaction.[63] This hindrance is mainly caused by distorted or broken bonds at the boundaries, along with fluctuations in the orientation of the exchange interaction. The second way is through magnon-phonon coupling.[64–67] This coupling can be characterized by a relaxation length, representing the distance over which energy is exchanged between magnons and phonons. Magnons can couple with phonons on the same side of the boundary, in conjunction with phonon transmission across grain boundaries. This scattering mechanism is affected by the thermal resistance across the magnon-phonon relaxation length in quantum magnets.

Our experiments and analysis provide quantitative insight into the influence of grain size on magnon thermal transport in the nanostructured quantum magnet La_2CuO_4, which is considered to be a model system for spin-1/2 2D square-lattice quantum Heisenberg antiferromagnets. Nanostructured La_2CuO_4 samples with various grain sizes are prepared using a bottom-up approach. As the average grain size decreases from 2.9 μm to 560 nm, κ_{m}^{i} decreases from 9.6 to 5.5 $Wm^{-1}K^{-1}$ at 300 K. The magnon MFPs are determined from the measured κ values and are found to be suppressed with decreasing grain size. The suppression is attributed to magnon-boundary scattering, while the degree of defect scattering is similar in all of the samples, as confirmed by Raman, C_{p} and Seebeck coefficient measurements. This observation underscores a significant size effect on magnon thermal conductivity with potential applicability to magnon transport in other magnetic materials. It should be noted that our calculation does not consider the frequency dependence of boundary scattering, as observed in phonon transport.[68] Further studies, including first-principles calculations, are needed to calculate magnon thermal transport properties in nanostructures and to establish comparisons with experimental findings. Additionally, electron microscopy analyses reveal that the morphologies of the La_2CuO_4 nanostructures are strongly affected by the OH^- concentration. The formation of different nanostructures is based on template growth during the hydrothermal treatment. Our findings provide valuable insight into the size effect on magnon thermal transport, which had not been demonstrated experimentally before. They lay the groundwork for the development of new nanoscale engineering approaches for altering thermal transport processes in magnetic systems, as well as the use of bottom-up synthesis of magnetic nanostructures in functional devices for quantum information and heat transport applications.

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

More information and requests should be directed to and will be fulfilled by the lead contact, Dr. Xi Chen (xichen@ucr.edu).

Materials availability

This study did not produce new unique materials.

Data and code availability

The data that support this work can be found in the manuscript and the supplemental information. The raw data is available from the lead contact upon request.

Material synthesis La_2CuO_4 nanostructures were prepared by a wet chemical method, which combines a hydrothermal process with subsequent calcination. In a typical procedure, 2 g of $La(NO_3)_3 \cdot 5H_2O$ (purity $\geq$ 99.99%) and 0.54 g of $Cu(NO_3)_2 \cdot 2.5H_2O$ (purity $\geq$ 99.99%) were mixed with 30 mL of deionized water under stirring and ultrasonic irradiation. A KOH aqueous solution (12 mol/L) was then added dropwise, while stirring rapidly. It is worth noting that the preparation of La_2CuO_4 nanorods requires 20 mL of KOH aqueous solution, while the preparation of La_2CuO_4 nanoparticles requires only 10 mL of KOH aqueous solution. The co-precipitation was transferred to a 50 mL Teflon-lined stainless steel autoclave for hydrothermal treatment at 220 ˚C for 24 h. After the mixture had been cooled to room temperature, it was washed several times with deionized water and dried at 60 ˚C for 12 h. The obtained powder was calcined in air at a heating rate of 1 ˚C/min from room temperature to 400 ˚C, kept at this temperature for 1 h, heated at a rate of 3 ˚C/min to 850 ˚C and maintained at this temperature for 6 h. For comparison, we used a solid state reaction method to prepare La_2CuO_4 powders with micron-scale grains. The starting materials were powders of La_2O_3 (purity $\geq$ 99.99%, heated at 1000 ˚C for 12 h before being weighed) and CuO (purity $\geq$ 99.7%). The materials were mixed in a molar ratio corresponding to $La: Cu = 2:1$ and heated at 1050 ˚C for 48 h in air. The NP and NR powders were consolidated separately into dense pellets at 750 ˚C for 10 min under 60 MPa using SPS. The SSR powders were consolidated into a dense pellet at 850 ˚C for 10 min under 60 Mpa. The SPS process was performed in vacuum.

Material characterization The phase purity, crystal structure and texture of the samples were characterized using a PANalytical Empyrean Series 2 X-ray diffraction machine (Malvern Panalytical, Malvern, UK) with Cu Kα radiation (λ = 1.54 Å). Morphological and compositional details of the samples were investigated using a TESCAN Vega3 SBH scanning electron microscope (TESCAN, Brno, Czech Republic), which was operated at 10 kV. Energy-dispersive X-ray (EDX) spectroscopy was performed on the sample surfaces to ascertain the distribution of elements.

Structural and compositional information was obtained using a ThermoFisher Scientific Talos L120C transmission electron microscope (TEM), as well as a ThermoFisher Scientific Titan Themis 300 scanning TEM (STEM) together with EDX (ThermoFisher Scientific, Waltham, MA, USA). The STEM measurements were performed at 300 kV using an X-FEG electron source. High-

resolution TEM images were recorded using a 2048 × 2048 pixel FEI CETA-16 M CMOS digital camera, with a beam convergence semi-angle of ~0.08 mrad.

A Quantum Design Physical Property Measurement System (PPMS) was used to measure the specific heat, thermal conductivity and Seebeck coefficient of the samples between 3 and 350 K. Details of the thermal conductivity measurement and uncertainty analyses can be found in the Supplementary Information. Raman measurements were carried out using a HORIBA LabRam with a 532 nm laser at 300 K.

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AUTHOR CONTRIBUTIONS

X.C. and S.G. designed the experiment; S.G. prepared the La_2CuO_4 nanostructures; S.G. performed XRD, SEM and TEM characterization; S.G., X.B. and R.E.D.B. analyzed the TEM data. H.L. and J.Z conducted the SPS process; S.L. performed the Raman measurement; S.G. and X.C. carried out thermal transport measurements and analysis; Y.W. performed the fitting of magnon thermal conductivity; X.C. directed the project. All of the authors contributed to the writing and editing of the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

Figure 1. Crystallographic information and structural characterization of La_2CuO_4 nanostructures

(a) Crystallographic structure of La_2CuO_4, with magnetic moments indicated by red arrows. (b, g) SEM images of NPs and NRs after calcination, prepared with ρ_{OH^-} =3 and 4.8 mol/L, respectively. The scale bars are 2 μm. (c, h) Bright-field TEM images of individual NPs and NRs after calcination. The scale bar of (c) is 200 nm, and the scale bar of (h) is 500 nm. (d, i) Selected area electron diffraction patterns. The scale bars are 5 nm^{-1}. (e ,j) HRTEM images. The scale bars are 5 nm. The insets show fast Fourier transforms (FFTs). (f, k) Corresponding filtered inverse FFTs of the square areas marked in (e) and (j). The scale bars are 1 nm.

Figure 2. Growth mechanism of La_2CuO_4 nanostructures

(a, b) SEM images of nanostructures after hydrothermal treatment. (a) Nanoparticles and (b) nanowires were formed when the OH- concentration was 3 and 4.8 mol/L, respectively. (c) HAADF images and elemental maps of NRs after hydrothermal treatment. (d, e, f) Distributions of La, Cu and O in (c). The scale bars are all 2 μm. (g) Schematic diagram of the growth mechanism for La_2CuO_4 nanorods.

Figure 3. Grain size distributions of the samples after SPS

(a-c) SEM images of the SSR, NR and NP samples after SPS. The scale bar of (a) is 10 μm, and the scale bars of (b) and (c) are 2 μm. (d) Grain size distributions of the three samples after SPS. Inset: Photograph of a La_2CuO_4 pellet prepared by SPS.

Figure 4. Optical and thermal property measurements

(a) Raman spectra recorded from the SSR, NR and NP samples after SPS, excited by a 532 nm laser. (b) Specific heat (C_p) of the SSR, NR and NP samples after SPS, compared to data reported by Syrykh et al.[52] and Sun et al.[53] Inset: low-temperature C_p in the temperature range of 3-10 K. (c) Schematic illustration of the measurement setup for thermal conductivity and Seebeck coefficient. The heat current J_q generated by a heater at one end of the sample passes through it towards a thermal bath. The temperature difference developed in the sample was determined using two pairs of thermocouples. (d) Seebeck coefficients of the SSR, NR and NP samples. (e) Thermal conductivities of the SSR, NP and NR samples measured in the perpendicular (OP) and parallel (IP) directions. The inset highlights the tendency of the nanorods to align along the IP direction after SPS. The shaded area depicts the measurement error of thermal conductivity. (f) Solid thermal conductivity of the polycrystalline samples compared to single crystal (xtl) data along the ab-plane reported by Hess et al.[4] and Sun et al.[31] The solid and dashed lines represent simulated results of the lattice thermal conductivity based on the Debye–Callaway model.

Figure 5. Size-dependent magnon thermal transport

(a) Magnon thermal conductivity of the SSR, NP and NR samples in the perpendicular (OP) and parallel (IP) directions. (b) Intrinsic magnon thermal conductivity of the SSR, NP and NR samples. For comparison, magnon thermal conductivity data of single crystal samples from the literature are also shown.[4,31] (c) Magnon MFPs of the polycrystals, shown together with the single crystal data. where the corresponding κ_m used for calculations was obtained from (b). (d) Calculated and experimental magnon thermal conductivity of La_2CuO_4 plotted as a function of grain size at T = 300 K. (e) Values of magnon boundary scattering MFP l_b (yellow area and left axis) and defect scattering parameter c_d (red area and right axis) for the SSR, NR and NP samples obtained using Equations 7 and

9 by considering both boundary and defect scattering based on the La_2CuO_4 *single crystal (xtl) data along the* ab*-plane reported by Sun et al.*[31] *(f) Size-dependent magnon thermal conductivity of* La_2CuO_4 *samples,* $Sr_{14}Cu_{24}O_{41}$ *microrods*[21] *and* $Ca_9La_5Cu_{24}O_{41}$ *polycrystals*[22] *at T = 300 K.*

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