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Authors

Chernyshev, AL

Rozhkov, AV

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Heat transport in spin chains with weak spin-phonon coupling

A. L. Chernyshev¹ and A. V. Rozhkov^{2,3}

¹*Department of Physics and Astronomy, University of California, Irvine, California 92697, USA*

²*Moscow Institute of Physics and Technology, Dolgoprudny, Moscow Region, 141700 Russia*

³*Institute for Theoretical and Applied Electrodynamics,
Russian Academy of Sciences, Moscow, 125412 Russia*

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The heat transport in a system of $S = 1/2$ large- J Heisenberg spin chains, describing Sr_2CuO_3 and SrCuO_2 cuprates, is studied theoretically at $T \ll J$ by considering interactions of the bosonized spin excitations with optical phonons and defects. Treating rigorously the multi-boson processes, we derive a microscopic spin-phonon scattering rate, which adheres to an intuitive picture of phonons acting as thermally populated defects for the fast spin bosons. The mean-free path of the latter exhibits a distinctive T -dependence reflecting a critical nature of spin chains and gives a close description of experiments. We invoke a naturalness criterion of realistically small spin-phonon interaction, by which our approach stands out from previous considerations that require large coupling constants to explain the data and thus imply a spin-Peierls transition, absent in real materials.

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The one-dimensional (1D) spin chains are among the first strongly-interacting quantum many-body systems ever studied [1, 2] and they remain a fertile ground for new ideas [3] and for developments of advanced theoretical and numerical [4, 5] methods. A number of physical realizations of spin-chain materials [6–10] has allowed for an unprecedentedly comprehensive comparisons between theory, numerical approaches, and experimental data [11–13]. Current theoretical challenges for these systems include their dynamical, non-equilibrium, and transport properties [14–20]. The transport phenomena are particularly challenging as the couplings to phonons and impurities, perturbations that are extrinsic to the often integrable spin systems, become crucial [21–26].

In this Letter, we address the problem of heat transport in 1D spin-chain systems by considering coupling of spins to optical phonons and impurities and having in mind a systematic experimental thermal conductivity study in the high-quality single-crystalline large- J spin-chain cuprates, Sr_2CuO_3 and SrCuO_2 , that has been recently conducted [27–30]. Several attempts to develop a suitable formalism to describe this phenomenon have been made in the past [24–26]. However, these approaches either relied on unrealistic choices of parameters [24, 26] or offered only qualitative insights [24, 25].

Below we attempt to bridge the gap between experiment and theory. We argue that the heat conductivity by spin excitations can be quantitatively described within the bosonization framework with the large-momentum scattering by optical phonons or impurities. For weak impurities, scattering grows stronger at lower temperature, a feature intimately related to a critical character of the $S = 1/2$ Heisenberg chains [26]. Taking into account multi-spin-boson processes, it follows naturally from our microscopic calculations that scattering by phonons bears a close similarity to that by weak im-

purities, only the phonons are thermally populated and thus control heat transport at high T . This is also in accord with a physical picture of phonons playing the role of impurities for the fast spin excitations. Within this picture, the transport relaxation time is the same as spin-boson scattering time and the corresponding mean-free path fits excellently the available experimental data. Further systematic extensions of our theory to include multi-phonon scattering that can influence thermal conductivity at higher temperature are briefly discussed.

Finally, we emphasize an important physical constraint on the strength of spin-phonon coupling of magnetoelastic nature [31, 32], which is weak in the materials of interest. While an estimate of this coupling can be made microscopically, a simple phenomenological evidence for this criterion is the absence of the spin-Peierls transition in real compounds down to very low temperatures. Our theory easily satisfies the proposed constraint, setting itself apart from the previous approaches [24, 26]. We thus provide a microscopic, internally consistent description of thermal transport and scattering in 1D spin chains, which satisfies naturalness criteria by having weak spin-phonon coupling and conforming to an analogy between phonon and impurity scatterings.

Spin-phonon coupling Hamiltonian.—The nearest-neighbor Hamiltonian of an $S = 1/2$ Heisenberg chain magnetoelastically coupled to phonons is

$$\mathcal{H} = \sum_{\langle ij \rangle} J(\mathbf{r}_i - \mathbf{r}_j) \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where $\langle ij \rangle$ denote nearest-neighbor lattice sites. A standard Jordan-Wigner transformation with the subsequent bosonization [11] and the lowest-order expansion in lattice displacements brings it to the following form

$$\mathcal{H} = \sum_k \varepsilon_k b_k^\dagger b_k + \mathcal{H}_{\text{s-ph}}, \quad (2)$$

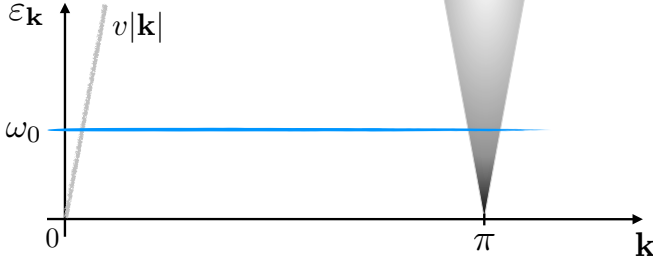


FIG. 1: (Color online) Schematics of the spectra of bosonic excitations in a large- J , $S = 1/2$ Heisenberg spin chain (dispersive branch $\varepsilon_{\mathbf{k}} = v|\mathbf{k}|$ and a continuum at $\mathbf{Q} = \pi$) and of the dispersionless optical phonon branch ω_0 (horizontal line).

where $b_k^{(\dagger)}$ are spin-boson operators of the excitation with $\varepsilon_k = v|k|$ sketched in Fig. 1, velocity is $v = \pi Ja/2$, k is the 1D momentum, and a is the lattice spacing. Hamiltonian $\mathcal{H}_{\text{s-ph}}$ describes a large-momentum, $q \approx Q = \pi/a$, spin-boson scattering by phonons

$$\mathcal{H}_{\text{s-ph}} = \frac{2\lambda}{\pi a^2} \int dx \mathbf{U}_x(x) \cos(\hat{\Phi}(x) + Qx), \quad (3)$$

where $\lambda = a\partial J/\partial x$, x is the direction along the chains, the lattice displacement field $\mathbf{U}(x)$ is associated with the optical and zone boundary phonons, and the spin-boson field $\hat{\Phi}(x) = \sqrt{\pi} \sum_k e^{ikx} (b_k^\dagger + b_{-k}) / \sqrt{L|k|}$, in which L is the linear size of the chain and we used Luttinger-liquid parameter $\mathcal{K} = 1/2$ for the Heisenberg case [33]. Small-momentum scattering is deliberately ignored, as the corresponding vertex carries small in-plane momentum of the phonon and leads to negligible scattering effects [26].

We note that boson-boson scattering cannot dissipate the heat current [21, 22] and thus is neglected.

Self-energy and relaxation rate.—Assuming the spin-phonon coupling to be small, a conjecture discussed below in detail [32], one can consider only the second-order spin-boson self-energy in Fig. 2(a) given by

$$\Sigma_k(\tau) = -\frac{2\lambda^2}{\pi a^4 |k|} \int dx e^{ikx} D(\tau, x) \langle e^{-i\hat{\Phi}(0,0)} e^{i\hat{\Phi}(\tau,x)} \rangle, \quad (4)$$

where $D(\tau, x) = \langle \mathbf{U}_x(0,0) \mathbf{U}_x(\tau, x) \rangle$ is the phonon propagator and the second-quantization of lattice displacement field is standard [35]. We exploit the large values of J compared to a typical phonon energy scale (in cuprates $J/\Theta_D \sim 10$), which allows us to neglect dispersion of the phonon branches near the π -point in Fig. 1. Then, the lattice-displacement correlator is fully local in space [33] and separates into a sum over phonon branches ℓ that have non-zero projections of their polarizations, $\xi_{\mathbf{q}\ell}^x$, on the chain axis x . Considering for simplicity only one longitudinal phonon with the energy ω_0 , see Fig. 1, and reserving the right to add more phonon branches later, we obtain $D(\tau, x) = a\delta(x)F_\tau(\omega_0)/2m\omega_0$ with

$$F_\tau(\omega_0) = n_0 e^{\omega_0 \tau} + (n_0 + 1) e^{-\omega_0 \tau}, \quad (5)$$

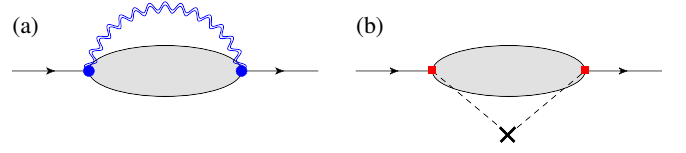


FIG. 2: (Color online) Multi-boson diagrams contributing to the scattering rate of spin bosons on (a) phonons and (b) weak impurities. Shaded ellipses represent a set of diagrams involving arbitrary number of spin bosons in the intermediate state. Solid and wavy lines are the Green's functions of spin bosons and phonons, respectively.

where $n_0 = 1/(e^{\omega_0/T} - 1)$ is the phonon distribution function, m is the mass of the unit cell, and $\hbar = k_B = 1$.

For the bosonic field correlator in the spin-phonon self-energy (4) in Fig. 2(a), we note an immediate similarity to the second-order T -matrix for the weak impurity scattering in Fig. 2(b), which also generates a large-momentum transfer [26]. The correlator can be evaluated at $x \rightarrow 0$ and $T \ll J$ [26, 33] and leads to

$$\langle e^{-i\hat{\Phi}(0,0)} e^{i\hat{\Phi}(\tau,0)} \rangle \approx \frac{\pi T}{J |\sin(\pi T \tau)|}. \quad (6)$$

Then the self-energy at Matsubara frequency ω_n is

$$\Sigma_k(\omega_n) = -g_{\text{sp}}^2 \cdot \frac{2TJ}{a|k|} \int_0^\beta d\tau \frac{e^{i\omega_n \tau} - 1}{|\sin(\pi T \tau)|} F_\tau(\omega_0), \quad (7)$$

where we introduced a naturally appearing *dimensionless* spin-phonon coupling constant $g_{\text{sp}} = \lambda/(aJ\sqrt{2m\omega_0})$ [31, 32]. For the spin-boson scattering rate we need the imaginary part of the self-energy that is analytically continued to real frequencies. The transformations allowing to perform integration in (7) exactly are delegated to [33]. Here we simply list the answer

$$\text{Im}\Sigma_k(\omega) = -g_{\text{sp}}^2 \frac{2J}{a|k|} (2n_0 + 1) (1 - f_+ - f_-), \quad (8)$$

where $f_\pm = 1/(e^{\omega \pm \omega_0} + 1)$. The fermionic distributions can be seen as a result of a re-fermionization of bosons via a multiple-boson scattering. The result (8) can be expanded in ω/T , yielding

$$\text{Im}\Sigma_k(\omega) \approx -g_{\text{sp}}^2 \frac{2J\omega}{a|k|T} \cdot \frac{1}{\sinh(\omega_0/T)}, \quad (9)$$

which holds exceptionally well for all $\omega \lesssim T$ of interest. Generally, the single-particle scattering rate (9) should differ from the transport relaxation rate, but for the impurity-like scattering the two become the same.

Mean-free path.—Then, the on-shell approximation, $\omega = \varepsilon_k$, in (9) yields the inverse spin-boson mean-free path, $1/\ell = 1/v\tau$, due to spin-phonon scattering

$$\left(\frac{\ell_{\text{sp}}}{a}\right)^{-1} = g_{\text{sp}}^2 \frac{2J}{T} \cdot \frac{1}{\sinh(\omega_0/T)}. \quad (10)$$

This result is k -independent and thus can be compared directly to the transport mean-free path extracted from thermal conductivity data [27, 29]. We note that the $1/T$ prefactor in (10) is strongly reminiscent of the result for the scattering on weak impurities [26, 36, 37]: $(\ell_{\text{imp}}/a)^{-1} = n_{\text{imp}} (\delta J/J)^2 (J/T)$, where n_{imp} is the concentration of such impurities and δJ is the strength of impurity potential. Clearly, this scattering gets stronger at lowering T , down to the Kane-Fisher scale, $T_{\text{KF}} \propto \delta J^2/J$, below which weak impurity becomes a strong scatterer, similar to a chain break [38]. This behavior is a consequence of a critical character of spin chains [26, 39]. Since phonons should be seen as weak impurities by the fast spin excitations, it is natural that the spin-phonon scattering yields the same $1/T$ prefactor in (10).

While the other thermal factor in (10), $1/\sinh(\omega_0/T)$, does not coincide with the phonon population n_0 , both have the same high- and low- T asymptotes. For $T \ll \omega_0$, the mean-free path (10) exhibits activated behavior, $\ell_{\text{sp}} \sim e^{\omega_0/T}$, similar to findings of other works [24, 25].

In addition to the considered scattering mechanisms, the low- T spin thermal conductivity in real materials is limited by strong defects that act like chain breaks [27, 28, 30]. The corresponding mean-free path is an average length of a defect-free chain segment, $1/\ell_{\text{b}} = n_{\text{b}}$, where n_{b} is the concentration of these defects.

Comparison with experiments.—Figure 3 shows the T -dependence of the mean-free path of 1D spin excitations in Sr_2CuO_3 and SrCuO_2 [27, 29]. The data are extracted from the thermal conductivity measurements via a kinetic relation, $\ell(T) = \kappa(T)/\nu C_V(T)$, using theoretical values [40] for the specific heat of spin chains $C_V(T) [\propto T \text{ at } T \ll J]$. Because of high purity, the mean-free path exceeds $10^3 a$ at low T , with the difference between two compounds due to residual concentrations of defects. The two sets of data become quantitatively very close at higher T , implying that a similar scattering is dominating propagation of heat in both materials [29].

Figure 3 shows our successful fits of the data by combining spin-phonon (10) and strong-impurity scatterings, $\ell^{-1} = \ell_{\text{sp}}^{-1} + \ell_{\text{b}}^{-1}$, via Matthiessen's rule [27]. We note that the low- T part of the data, $T \lesssim 40$ K, has a large uncertainty due to subtraction of the phonon part of thermal conductivity, see [27, 29], and can be fit with an equal success by a combination of weak and strong impurities, $\ell^{-1} \approx \ell_{\text{imp}}^{-1} + \ell_{\text{b}}^{-1}$. Since it is a secondary issue for our study, the simplest account of this regime by strong impurities suffices. To fit the spin-boson mean-free path above 40 K, we assume that the spin-bosons are scattered by two phonon modes with $\omega_{0,1} = 300$ K and $\omega_{0,2} = 740$ K. Of the two, the first roughly corresponds to the longitudinal zone-boundary phonon and the second to the high-energy stretching mode [41, 42], both likely having the strongest coupling to spin chains. In (10) we used the value of $J = 2600$ K [43] and the spin-phonon coupling constants are $g_{1,\text{sp}} = 0.020(1)$ and $g_{2,\text{sp}} = 0.10(1)$ with the

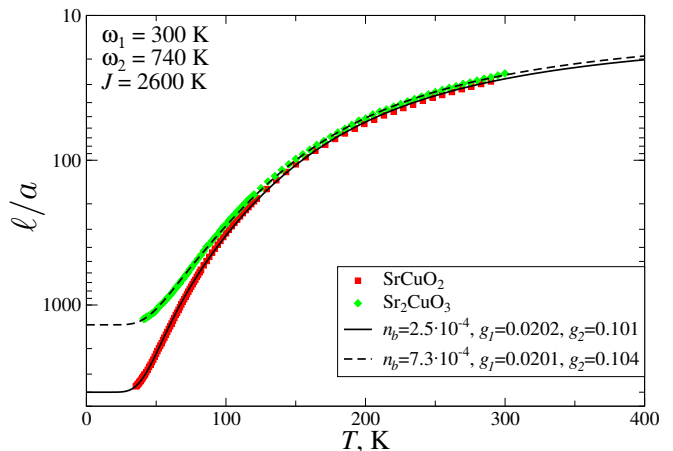


FIG. 3: (Color online) Mean-free path of spin excitations in Sr_2CuO_3 and SrCuO_2 [27–29] (symbols). Lines are theory fits, see text. Concentrations of strong impurities, n_{b} , phonon energies, $\omega_{0,i}$, and spin-phonon coupling constants, $g_{i,\text{sp}}$, are as indicated in the graph.

difference between parameters for Sr_2CuO_3 and SrCuO_2 occurring in the third significant figure.

Bounds on spin-phonon coupling.—We now discuss physical bounds on the spin-phonon coupling constant $g_{\text{sp}} = \lambda/(Ja\sqrt{2m\omega_0})$. As discussed in [31, 32], the constant is a product of two parameters, one characterizing the change of J by atomic displacement, $\gamma = \lambda/J = a(\partial J/\partial x)/J$, and the other is an amplitude of zero-point atomic motion relative to a lattice constant [35], $\alpha = \hbar/\sqrt{2ma^2\omega_0}$, where \hbar is made explicit and m is the reduced mass associated with the phonon mode ω_0 . Parameter α is small, while γ can be large [31, 42] because the superexchange is sensitive to the interatomic distance. For to cuprates, one can estimate $\alpha \approx 0.01$. The superexchange parameter has a larger uncertainty, with indirect studies giving a range of $\gamma = 3–14$ [42, 44] and a consideration of a wider class of materials suggesting an upper limit of $\gamma \leq 20$ [31]. Thus, the microscopic upper bound on the spin-phonon coupling constant in 1D cuprates can be put at $g_{\text{sp}}^{\text{max}} \approx 0.2$, justifying the weak-coupling treatment of the spin-boson scattering on phonons in (4).

A less restrictive, but a purely phenomenological criterion limiting the strength of the spin-phonon coupling is the absence of the spin-Peierls transition in 1D cuprates down to about 5 K ($\approx 0.002J$), where the 3D Néel ordering can be argued to preempt other transitions. Using $T_{\text{sp}} \approx Je^{-1/g_{\text{sp}}^2}$, this can be translated to the upper limit on the spin-phonon coupling $g_{\text{sp}}^{\text{max}} \approx 0.35$.

We now offer a critique of the previous considerations of thermal transport in 1D spin chains. In particular, in experimental works [27, 29, 43], the spin-phonon mean-free path is repeatedly fit by the form $\ell_{\text{sp}}^{-1} = ATe^{-\omega^*/T}$ with $\omega^* \approx 200$ K, inspired by the phonon-mediated Umklapp scenario [24]. First, most of the data in Figure 3 should be outside the quantitative accuracy range of this

expression, which is limited to $T \lesssim \omega^*/3 \approx 70$ K as the exponent is only a low- T limit of the phonon distribution function. More importantly, translating the values of A used in [27, 29, 43] to the dimensionless coupling constant via $A = g_{\text{sp}}^2/Ja$ gives $g_{\text{sp}} \approx 1$, which is exceedingly large for the perturbative treatment to hold and lies way outside the allowed range. This strong coupling also implies spin-Peierls transition at $T_{\text{sp}} \sim J$, while no such transition is observed. Likewise, our previous study, which considered small-momentum scattering on acoustic phonon branches [26], required anomalously strong spin-phonon interaction. Thus, there is a serious “naturalness” problem with previous theoretical considerations.

In the present work, the dimensionless spin-phonon coupling constants are well within the range of the microscopic expectations for cuprates, $g_{\text{sp}} = 0.02 - 0.1$, implying extremely low spin-Peierls transition temperatures. While the offered analysis of the physical bounds is not a proof of the validity of our theory, it is certainly a strong argument against the validity of the previous approaches, which require unphysically large spin-phonon coupling.

Multi-phonon scattering.—We note that for $T \gtrsim \omega_0$ the spin-boson mean-free path in (10) saturates at $(\ell_{\text{sp},2}/a)^{-1} \approx g_{\text{sp},2}^2 2J/\omega_0$. While this is not unphysical, one can still expect that the other, T -dependent terms may become important for $T \gtrsim \omega_0$. Corrections of order T/J are neglected in our derivation, see [33], since T/J is small in the relevant temperature range. Another possible source of the T -dependence is the multi-phonon scattering. Superficially, the two-phonon scattering processes have to be negligible because of the smallness of the spin-phonon coupling discussed above. However, there are factors that can compensate this smallness. First, the two-phonon scattering is less restrictive as the transverse phonons can also contribute. Second, in the non-Bravais lattices, the two-phonon processes are also amplified by the number of atoms in a unit cell, N_a . That is, for the single-phonon processes, the number of longitudinal phonons that couple to spins via (3) is N_a , of which we have chosen only two for our fits in Fig. 3. On the other hand, when a spin-boson scattering is due to emission or absorption of two phonons, the number of possible processes can be as large as $\mathcal{O}(N_a^2)$. A naïve and certainly overly optimistic estimate of their number assuming independent polarization and branch index for each phonon involved in the scattering yields $(3N_a)^2$. In cuprates [41], the total number of phonon modes is large, so that this combinatorial factor can be substantial.

A somewhat tedious, but straightforward algebra [33] yields the following result for the two-phonon scattering

$$\left(\frac{\ell_{\text{sp},2}}{a}\right)^{-1} = g_{\text{sp},2}^2 \frac{J}{T} \cdot \frac{\cosh(\omega_0/T)}{\sinh^2(\omega_0/T)}, \quad (11)$$

where $g_{\text{sp},2}^2 \propto C_2 g_{\text{sp}}^4$. When compared to (10), the result in (11) contains an extra factor $g_{\text{sp}}^2 \sim 0.01$, and a

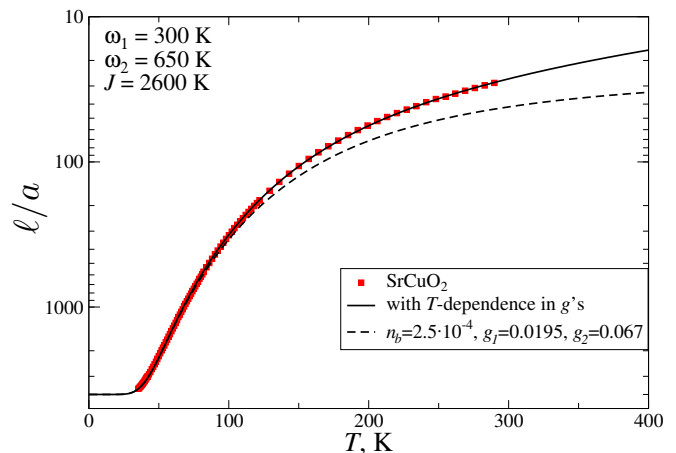


FIG. 4: (Color online) Same as in Fig. 3. Solid line includes T -dependence in the spin-phonon coupling, see text.

large combinatorial factor C_2 . Clearly, at $T \ll \omega_0$, the two-phonon mean-free path follows the same behavior as (10), thus simply renormalizing single-phonon scattering. However, at $T \gtrsim \omega_0$, it carries an extra power of T/ω_0 : $(\ell_{\text{sp},2}/a)^{-1} \approx g_{\text{sp},2}^2 JT/\omega_0^2$, thus amounting to an expansion in $T/\omega_{0,i}$, which can be argued to follow naturally from the multi-phonon scattering processes.

Without going into non-generic microscopic considerations, one can suggest a simple ansatz to account for the $T/\omega_{0,i}$ -expansion with the T -dependence of the spin-phonon coupling in the form $g_{\text{sp},i}(T) = g_{\text{sp},i}(1 + r_i n_{0,i})$, where $n_{0,i} = 1/(e^{\omega_{0,i}/T} - 1)$ as before. This form meets both the low- T and the high- T behavior of the two-phonon mean-free path discussed above. A fit of the SrCuO₂ data using this ansatz with $r_i = 1$ is provided in Fig. 4. The bare spin-phonon coupling constants $g_{i,\text{sp}}$ are even smaller than in Fig. 3 especially for the higher-energy mode. The result with the bare $g_{i,\text{sp}}$'s is provided for comparison. Although this figure is an illustration showing that our theory allows for systematic extensions by including multi-phonon processes, it also demonstrates a potential role of the latter in the $T \gtrsim \omega_0$ regime and thus contributes to the general description of the heat transport in spin-chain materials.

Conclusions.—We have provided a consistent microscopic theory for thermal transport and scattering in 1D spin chains, which stands out from previous attempts at such a theory by having weak spin-phonon coupling and conforming to the analogy of the phonon scattering to that on impurities. We have successfully fit the available experimental data and discussed possible extensions of our theory for higher- T .

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Heat transport in spin chains with weak spin-phonon coupling: Supplemental Material

A. L. Chernyshev¹ and A. V. Rozhkov^{2,3}

¹*Department of Physics and Astronomy, University of California, Irvine, California 92697, USA*

²*Moscow Institute of Physics and Technology, Dolgoprudny, Moscow Region, 141700 Russia*

³*Institute for Theoretical and Applied Electrodynamics,
Russian Academy of Sciences, Moscow, 125412 Russia*

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SPIN-BOSON SCATTERING ON PHONONS

Bosonization and coupling to phonons

Bosonization of the Jordan-Wigner fermion operators for the $S = 1/2$ chain proceeds via introduction of the bosonic fields Φ and Θ for the right- and left-moving fermion operators [1] according to

$$\psi_R = \frac{1}{\sqrt{2\pi a}} e^{i\sqrt{\pi}(\Phi+\Theta)}, \quad (S1)$$

$$\psi_L = \frac{1}{\sqrt{2\pi a}} e^{i\sqrt{\pi}(-\Phi+\Theta)}. \quad (S2)$$

The bilinear combination of the fermionic operators $\psi_R^\dagger \psi_L$ can be expressed as follows

$$\psi_R^\dagger \psi_L = \frac{1}{2\pi a} e^{-i\sqrt{4\pi}\Phi} = \frac{1}{2\pi a} e^{-i\sqrt{2\pi}\tilde{\Phi}}, \quad (S3)$$

where the rescaled bosonic field $\tilde{\Phi} = (1/\sqrt{\mathcal{K}})\Phi = \sqrt{2}\Phi$. The Tomonaga-Luttinger parameter $\mathcal{K} = 1/2$ corresponds to the Heisenberg limit of the model.

Coupling to phonon field that leads to the large-momentum scattering can be written as

$$\mathcal{H}_{s\text{-ph}} = \frac{\lambda}{\pi a^2} \int dx \mathbf{U}_x(x) e^{\sqrt{2\pi}\tilde{\Phi}(x) - i\pi x/a} + \text{h.c.}, \quad (S4)$$

where $\lambda = a\partial J/\partial x$ and $\mathbf{U}_x(x)$ is the x -component of the lattice displacement field \mathbf{U} for the optical and zone boundary phonons.

Spin-boson self-energy

Since we are interested in the scattering involving momenta close to the 1D zone boundary momentum $Q = \pi/a$, one can assume that the corresponding phonon has zero velocity. Then we can write the spin-boson self-energy as

$$\Sigma_k(\tau) = -\frac{2\lambda^2}{\pi a^4 |k|} \int dx e^{ikx} \langle \mathbf{U}_x(0,0) \mathbf{U}_x(\tau,x) \rangle \quad (S5)$$

$$\times \left\langle e^{-i\sqrt{2\pi}\tilde{\Phi}(0,0)} e^{i\sqrt{2\pi}\tilde{\Phi}(\tau,x)} \right\rangle.$$

Phonon correlation function

Using standard quantization of the displacement field, phonon correlation function in (S5) is

$$\langle \mathbf{U}_x(0,0) \mathbf{U}_x(\tau, \mathbf{R}) \rangle = \frac{1}{N} \sum_{\mathbf{P}\ell} \frac{e^{-i\mathbf{P}\mathbf{R}}}{2m\omega_{0\ell}} (\xi_{\mathbf{P}\ell}^x)^2 \quad (S6)$$

$$\times \left(\left\langle a_{\mathbf{P}\ell}^\dagger(0) a_{\mathbf{P}\ell}(\tau) \right\rangle + \left\langle a_{-\mathbf{P}\ell}(0) a_{-\mathbf{P}\ell}^\dagger(\tau) \right\rangle \right).$$

Assuming, for simplicity, that the dynamical force matrix at a given momenta can be diagonalized so that one of the polarizations (longitudinal) is along the chain and two remaining ones are perpendicular, then all of the relevant polarizations yield $(\xi_{\mathbf{P}\ell}^x)^2 = 1$ and the summation in (S6) will be over N_a independent longitudinal phonon modes with individual frequencies $\omega_{0,\ell}$, where N_a is the number of atoms per unit cell.

Consider coupling to an individual phonon mode with the energy ω_0 . For this case we derive

$$\langle \mathbf{U}_x(0,0) \mathbf{U}_x(\tau, \mathbf{R}) \rangle = \frac{V_0 \delta(\mathbf{R})}{2m\omega_0} \quad (S7)$$

$$\times [n_0 e^{\omega_0 \tau} + (n_0 + 1) e^{-\omega_0 \tau}],$$

$$\text{with } n_0 = \frac{1}{e^{\omega_0/T} - 1}, \quad (S8)$$

where V_0 is the volume of the unit cell, n_0 is the phonon occupation number, and we assumed that $\tau > 0$. Since we need to evaluate the phonon propagator on a single chain, one can write $V_0 \delta(\mathbf{R}) = a \delta(x)$, which leads to

$$\langle \mathbf{U}_x(0,0) \mathbf{U}_x(\tau, x) \rangle \quad (S9)$$

$$= \frac{a \delta(x)}{2m\omega_0} [n_0 e^{\omega_0 \tau} + (n_0 + 1) e^{-\omega_0 \tau}].$$

Obviously, for the case of N_a phonon modes, one needs to reinstate the summation over them.

Spin-boson correlation function

We now turn to the spin-boson part. We have previously obtained the spin-boson correlation function in [2]. Here we provide a more rigorous derivation of it. First,

$$\left\langle e^{-i\sqrt{2\pi}\tilde{\Phi}(0,0)} e^{i\sqrt{2\pi}\tilde{\Phi}(\tau,0)} \right\rangle = e^{-\pi \langle [\tilde{\Phi}(0,0) - \tilde{\Phi}(\tau,0)]^2 \rangle}. \quad (S10)$$

Thus, we are to calculate $g(\tau)$ defined by

$$g(\tau) = - \left\langle \left[\tilde{\Phi}(0,0) - \tilde{\Phi}(\tau,0) \right]^2 \right\rangle \quad (\text{S11})$$

$$= 2 \left\langle \tilde{\Phi}(0,0)\tilde{\Phi}(\tau,0) \right\rangle - 2 \left\langle \tilde{\Phi}(0,0)\tilde{\Phi}(0,0) \right\rangle.$$

Therefore, we have

$$g(\tau) = \frac{2T}{L} \sum_{\omega_n} (e^{i\omega_n\tau} - 1) \sum_{k \neq 0} \frac{v}{\omega_n^2 + v^2k^2}, \quad (\text{S12})$$

with $\omega_n = 2\pi Tn$, $n = \text{integer}$.

Discarding the vanishing $\omega_n = 0$ term in (S12), we have the following identity for any $\omega_n \neq 0$

$$g(\tau) = \frac{2T}{L} \sum_{\omega_n \neq 0} (e^{i\omega_n\tau} - 1) \sum_{k \neq 0} \frac{v}{\omega_n^2 + v^2k^2} \quad (\text{S13})$$

$$= 2T \sum_{\omega_n \neq 0} \int_{-\infty}^{+\infty} \frac{vdk}{2\pi} \frac{(e^{i\omega_n\tau} - 1)}{\omega_n^2 + v^2k^2} e^{-\pi a|k|/2}.$$

Here the exponent $e^{-\pi a|k|/2}$ is used to set an ultraviolet cutoff. Next, we rewrite $g(\tau)$ as

$$g(\tau) = \frac{T}{\pi} \int_{-\infty}^{+\infty} \frac{dy}{1+y^2} \sum_{\omega_n \neq 0} \frac{e^{i\omega_n\tau} - 1}{|\omega_n|} e^{-y|\omega_n|/J}, \quad (\text{S14})$$

where y is the dimensionless integration variable and we used $v = \pi Ja/2$. The summation over ω_n can be performed with the help of an identity

$$\sum_{\omega_n > 0} \frac{e^{\omega_n(i\tau - y/J)}}{\omega_n} = -\frac{1}{2\pi T} \ln \left(1 - e^{2\pi T(i\tau - y/J)} \right). \quad (\text{S15})$$

As a result we obtain

$$g(\tau) = \frac{1}{2\pi^2} \int \frac{dy}{1+y^2} \ln \left[\frac{(1 - e^{-2\pi Ty/J})^2}{1 + e^{-4\pi Ty/J} - 2e^{-2\pi Ty/J} \cos(2\pi T\tau)} \right] \approx \frac{1}{2\pi^2} \int \frac{dy}{1+y^2} \ln \left[\frac{sy^2}{sy^2 + 4 \sin^2(\pi T\tau)} \right], \quad (\text{S16})$$

where $s = (2\pi T/J)^2 \ll 1$. The remaining integral can be found in [3], which gives

$$g(\tau) = \frac{1}{\pi} \ln \frac{\sqrt{s}}{2|\sin(\pi T\tau)| + \sqrt{s}}. \quad (\text{S17})$$

Exponentiating $g(\tau)$, we arrive to the following expression for the spin-boson correlator in (S5)

$$\left\langle e^{-i\sqrt{2\pi}\tilde{\Phi}(0,0)} e^{i\sqrt{2\pi}\tilde{\Phi}(\tau,0)} \right\rangle \quad (\text{S18})$$

$$= \frac{2\pi T}{2J|\sin(\pi T\tau)| + 2\pi T} \approx \frac{\pi T}{J|\sin(\pi T\tau)|},$$

where in the last expression we neglected contributions of order $\mathcal{O}(T/J)$.

Evaluation of the self-energy

Using the phonon propagator (S9) and the spin-boson correlation function (S18) obtained above and transforming the spin-boson self-energy $\Sigma_k(\tau)$ in (S5) to the Matsubara frequency domain yields $\Sigma_k(\omega_n)$ given by

$$\Sigma_k(\omega_n) = - \left(\frac{\lambda^2}{2ma^2\omega_0} \right) \left(\frac{2T}{Ja|k|} \right) \quad (\text{S19})$$

$$\times \int_0^\beta d\tau \frac{e^{i\omega_n\tau} - 1}{|\sin(\pi T\tau)|} [n_0 e^{\omega_0\tau} + (n_0 + 1)e^{-\omega_0\tau}].$$

Therefore, we need to evaluate the following integral

$$I_\varepsilon(\omega_n) = \int_0^\beta d\tau \frac{e^{i\omega_n\tau} - 1}{\sin(\pi T\tau)} e^{\varepsilon\tau}, \quad (\text{S20})$$

for $\varepsilon = \pm\omega_0$. Here we used that $\tau > 0$.

Consider Eq. (S20) for $\varepsilon = \omega_0$. Defining the complex variable $z = e^{i2\pi T\tau}$, we transform I_{ω_0} into an integral along the unit circle in the z -plane. This contour, however, is not closed, since the circle is cut at $z = 1$ by a branch-cut running along the real axis from zero to $+\infty$. Completing the unit circle to the contour \mathcal{C} , see Fig. 5, one can verify that

$$I_{\omega_0} + I_{\omega_0}^* = \frac{1}{\pi T} \int_{\mathcal{C}} dz \frac{1-z^n}{1-z} z^{-i\varphi_0 - 1/2} = 0, \quad (\text{S21})$$

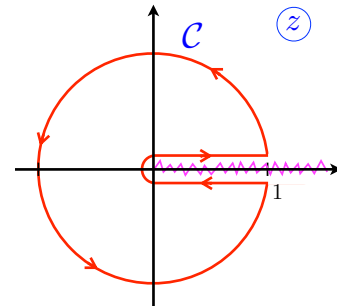


FIG. 5: (Color online) Contour \mathcal{C} for the evaluation of I_{ω_0} .

where $\varphi_0 = \omega_0/2\pi T$, and $I_{\omega_0}^r$ contains two segments of the contour \mathcal{C} along the real axis.

Thus, I_{ω_0} reduces to the integral of the real variable

$$I_{\omega_0} = -\frac{1}{\pi T} (1 + e^{\omega_0/T}) \int_0^1 dx \frac{1-x^n}{1-x} x^{-i\varphi_0-1/2}. \quad (\text{S22})$$

Method I

The integral in (S22) can be evaluated using [4], giving

$$I_{\omega_0} = -\frac{1 + e^{\omega_0/T}}{\pi T} \left[\psi \left(n - i\varphi_0 + \frac{1}{2} \right) - \psi \left(-i\varphi_0 + \frac{1}{2} \right) \right], \quad (\text{S23})$$

where $\psi(w)$ is the digamma function.

Ultimately, we need the real-frequency propagator. To obtain it, let us perform the substitution $i\omega_n \rightarrow \omega + i0$, or, equivalently, $n \rightarrow \omega/i2\pi T + 0$. In the real-frequency domain the following holds

$$I_{\omega_0} = -\frac{1 + e^{\omega_0/T}}{\pi T} \left[\psi \left(\frac{\omega}{i2\pi T} + \frac{\omega_0}{i2\pi T} + \frac{1}{2} \right) - \psi \left(\frac{\omega_0}{i2\pi T} + \frac{1}{2} \right) \right]. \quad (\text{S24})$$

Imaginary part of I_{ω_0} can be expressed in terms of elementary functions, since

$$\text{Im } \psi \left(iy + \frac{1}{2} \right) = \frac{\pi}{2} \tanh(\pi y). \quad (\text{S25})$$

The latter equality can be derived with the help of the reflection formula for the digamma function $\psi(1-x) - \psi(x) = \pi \cot(\pi x)$, in which one has to substitute $x = 1/2 - iy$. Consequently,

$$\text{Im } I_{\pm\omega_0} = \frac{1 + e^{\pm\omega_0/T}}{2T} \left[\tanh \left(\frac{\omega \pm \omega_0}{2T} \right) \mp \tanh \left(\frac{\omega_0}{2T} \right) \right]. \quad (\text{S26})$$

Method II

A substitution $x = e^{-2z}$ transforms I_{ω_0} in (S22) to

$$I_{\omega_0} = \frac{(1 + e^{\omega_0/T})}{\pi T} \int_0^\infty dz \frac{e^{-2nz} - 1}{\sinh z} e^{2i\varphi_0 z}. \quad (\text{S27})$$

Since we are, ultimately, interested in the imaginary part of $\Sigma_k(\omega)$, it is the imaginary part of I_{ω_0} that we are concerned about. This is also a point at which an analytical continuation can be made via $i\omega_n \rightarrow \omega + i0$, or,

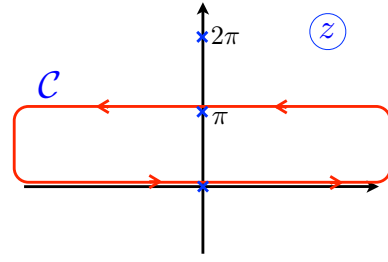


FIG. 6: (Color online) Contour \mathcal{C} for I_{ω_0} in (S28).

equivalently, $n \rightarrow -i\omega/2\pi T$. Introducing the variable $\varphi = \omega/2\pi T$ and extending the limit to $-\infty$ we get

$$\text{Im } I_{\omega_0} = \frac{(1 + e^{\omega_0/T})}{\pi T} \times \int_{-\infty}^{\infty} dz \frac{\sin(\varphi z) \cos((2\varphi_0 + \varphi)z)}{\sinh z}. \quad (\text{S28})$$

The resulting integral can be found in [5] or evaluated by using integration in a complex plane with the help of a standard trick utilizing the contour in Fig. 6. Both yield identical results, coinciding with Eq. (S26). The quantity $\text{Im } I_{-\omega_0}$ can be obtained from the latter result by simple substitution $\omega_0 \rightarrow -\omega_0$.

Scattering rate

The imaginary part of the self-energy in (S19) is proportional to a combination of $\text{Im } I_{\pm\omega_0}$ with the phonon distribution functions that correspond to the phonon absorption or emission processes

$$F(\omega, \omega_0) = n_0 \text{Im } I_{\omega_0} + (n_0 + 1) \text{Im } I_{-\omega_0}, \quad (\text{S29})$$

where n_0 is defined in Eq. (S8). The expression for $F(\omega, \omega_0)$ can be rewritten as a combination of the phonon and effectively fermionic distribution functions

$$F(\omega, \omega_0) = \frac{1}{T} (1 + 2n_0)(1 - f_+ - f_-), \quad (\text{S30})$$

$$\text{with } f_{\pm} = \frac{1}{e^{\omega \pm \omega_0} + 1}. \quad (\text{S31})$$

All of that finally yields the imaginary part of the (retarded) self-energy

$$\text{Im } \Sigma_k^{\text{ret}}(\omega) = -g_{\text{sp}}^2 \frac{2J}{a|k|} (1 + 2n_0)(1 - f_+ - f_-), \quad (\text{S32})$$

where the dimensionless spin-phonon coupling constant $g_{\text{sp}} = \lambda/(aJ\sqrt{2m\omega_0})$ is introduced.

We would like to point out that the only approximation that has been made in the derivation of our answer in (S32) from the Matsubara self-energy in (S5) is the omission of the $\mathcal{O}(T/J)$ terms in Eq. (S18).

At $\omega < T$ one can approximate the imaginary part of the self-energy in (S32) as

$$\text{Im } \Sigma_k^{\text{ret}}(\omega) \approx -g_{\text{sp}}^2 \frac{2J\omega}{a|k|T} \cdot \frac{1}{\sinh(\omega_0/T)}. \quad (\text{S33})$$

Finally, the scattering rate of spin-bosons on phonons is obtained by taking $\text{Im } \Sigma_k^{\text{ret}}(\omega)$ on the mass surface, $\omega = v|k|$,

$$\Gamma_k^{\text{1ph}} = -\text{Im } \Sigma_k^{\text{ret}}(v|k|) = g_{\text{sp}}^2 \frac{\pi J^2}{T} \cdot \frac{1}{\sinh(\omega_0/T)}, \quad (\text{S34})$$

where $v = \pi Ja/2$ was used as above.

This single-phonon scattering rate vanishes exponentially as a function of temperature for $T < \omega_0$ and saturates at a constant value for $T > \omega_0$.

TWO-PHONON SCATTERING

Here we extend the formalism developed above to the interaction of spin-boson with two phonons. In the limit of purely Einstein phonons this procedure is straightforward. We model the two-phonon coupling as

$$H_{2\text{ph}} = \frac{\lambda^{2\text{ph}}}{a^2} \sum_{\alpha=x,y,z} \int dx \mathbf{U}_\alpha^2 \left(\psi_L^\dagger \psi_R e^{-i\pi x/a} + \text{h.c.} \right), \quad (\text{S35})$$

$$\text{where } \lambda^{2\text{ph}} = a^2 \frac{\partial^2 J}{\partial r^2}. \quad (\text{S36})$$

Needless to say, this Hamiltonian represents a greatly simplified version of the general situation as the coupling constant $\lambda^{2\text{ph}}$ is independent of both the branch index ℓ and polarization ξ_ℓ of a phonon. The square of the phonon field, which enters this Hamiltonian, equals to

$$\begin{aligned} \mathbf{U}_\alpha^2(x) = \frac{a}{L} \sum_{q,\ell,q',\ell'} \frac{e^{i(q+q')x}}{2m\omega_0} \xi_{q\ell}^\alpha \xi_{q'\ell'}^\alpha (a_{q\ell}^\dagger + a_{-q\ell}) \\ \times (a_{q'\ell'}^\dagger + a_{-q'\ell'}). \end{aligned} \quad (\text{S37})$$

According to the Wick's theorem, the corresponding propagator can be expressed as a product of two single-phonon propagators

$$\langle \mathbf{U}_\alpha^2(0,0) \mathbf{U}_\beta^2(x,\tau) \rangle = 2 \langle \mathbf{U}_\alpha(0,0) \mathbf{U}_\alpha(x,\tau) \rangle^2 \delta_{\alpha\beta}. \quad (\text{S38})$$

The propagator itself is given by Eq. (S9). The singularity of the form $[a\delta(x)]^2$ in (S38) can be resolved through the usual prescription: $[a\delta(x)]^2 = a\delta(x)$. Consequently

$$\begin{aligned} \langle \mathbf{U}_\alpha^2(0,0) \mathbf{U}_\beta^2(x,\tau) \rangle \\ = \frac{a\delta(x)}{2m^2\omega_0^2} [n_0 e^{\omega_0\tau} + (n_0 + 1)e^{-\omega_0\tau}]^2 \delta_{\alpha\beta}. \end{aligned} \quad (\text{S39})$$

We note that for a multi-atomic unit cell the latter expression has to be multiplied by N_a^2 . In other words, the

multi-atomic unit cell enhances the contribution of the multi-phonon scattering.

Applying the same technical approach used in the single-phonon calculation above to the two-phonon case is quite straightforward. The self-energy is

$$\begin{aligned} \Sigma_k^{2\text{ph}}(\omega) = -\frac{(\lambda^{2\text{ph}})^2}{\pi a^6 |k|} \frac{3a}{2m^2\omega_0^2} \frac{\pi T}{J} \\ \times [n_0^2 I_{2\omega_0} + 2n_0(n_0 + 1)I_0 + (n_0 + 1)^2 I_{-2\omega_0}], \end{aligned} \quad (\text{S40})$$

where $I_{\pm 2\omega_0,0}$ are defined according to Eq. (S20). Then, one can obtain the scattering rate from this expression

$$\Gamma^{2\text{ph}} \propto \frac{(\lambda^{2\text{ph}})^2 v}{Jm^2 a^5 \omega_0^2} \frac{\cosh(\omega_0/2T)}{T \sinh^2(\omega_0/T)}. \quad (\text{S41})$$

Using $v = \pi aJ/2$ and introducing dimensionless spin-two-phonon coupling constant, $g_{\text{sp},2}^2 = (\lambda^{2\text{ph}}/J)^2 / (2ma^2\omega_0)^2$, we simplify the last expression to

$$\Gamma^{2\text{ph}} \propto g_{\text{sp},2}^2 \frac{J^2}{T} \cdot \frac{\cosh(\omega_0/2T)}{\sinh^2(\omega_0/T)}. \quad (\text{S42})$$

One can relate the two-phonon to the one-phonon coupling constant, $g_{\text{sp},2}^2 \propto C_2 g_{\text{sp}}^4$, where C_2 is a large combinatorial factor due to multiple phonon modes that can be involved in the scattering. At $T < \omega_0$, the two-phonon scattering rate yields the same exponential behavior as (S34). However, at $T > \omega_0$, it carries an extra power of T/ω_0

$$\Gamma^{2\text{ph}} \propto g_{\text{sp},2}^2 \frac{J^2 T}{\omega_0^2}, \quad (\text{S43})$$

thus giving a natural expansion in T/ω_0 in the multi-phonon scattering of spin-bosons.

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