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Competition of lattice and spin excitations in the temperature dependence of spin-wave properties

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The interplay of magnons and phonons can induce strong temperature variations in the magnetic exchange interactions, leading to changes in the magnetothermal response. This is a central mechanism in many magnetic phenomena, and in the new field of Spin Caloritronics, which focuses on the combination of heat and spin currents. Boson model systems have previously been developed to describe the magnon-phonon coupling, but until recently studies rely on empirical parameters. In this work we propose a first principles approach to describe the dependence of the magnetic exchange integrals on phonon renormalization, leading to changes in the magnon dispersion as a function of temperature. The temperature enters into the spin dynamics (by introducing fluctuations) as well as in the magnetic exchange itself. Depending on the strength of the coupling, these two temperatures may or may not be equilibrated, yielding different regimes. We test our approach in typical and well known ferromagnetic materials: Ni, Fe, and Permalloy. We compare our results to recent experiments on the spin-wave stiffness, and discuss departures from Bloch’s law and parabolic dispersion.

INTRODUCTION

The interaction of ionic vibrational and magnetic degrees of freedom (magnon-phonon coupling or MPC) influences many physical properties in magnetic and strongly correlated materials. Beyond providing a playground to fundamental science, MPC holds promise in many applied fields such as nanoelectronics (to boost efficiency and reduce power consumption), sensors and actuators (magneto-thermal response can be combined with multiple external inputs: B field, T, strain, current...) or to reduce magneto transport response: spin currents can experience reduced resistance due to coupling with coherent acoustic vibrations, giving resonant magnetoelastic deformations. The explicit calculation, and then engineering, of MPC will be central to the development of devices operating at room temperature and above.

Since the 1950s, MPC has been studied using model Hamiltonians. Only very recently Fransson et al have published a more complete theory of MPC interaction terms and their possible symmetries. The interaction governs changes in the frequency and relaxation time of magnons as a function of temperature, and conversely the change in phonon frequencies and lifetimes with the magnetic state. Over the past decade the physics of electrical to spin transport conversion has expanded dramatically. The new field of Spin Caloritronics characterizes different transport phenomena, which depend on the coupling between thermal vibrations, charge and spin. Creative experiments have led to the discovery of new physical effects (Spin Peltier, Spin Seebeck, Spin-dependent Seebeck, etc) which combine temperature gradients, internal and external magnetic fields, heat currents, charge and spin, and in which energy conversion depends (at least in part) on intrinsic magnetic excitations, along with their coupling to phonons and electrons. Conversely, temperature can also have a strong direct effect on the magnetic excitations themselves, as demonstrated, e.g., in Ni deposited on VO2, where heat drives a softening of a VO2 phonon mode, which couples to the magnetic response in Ni.[14]

MPC is much more delicate than electron-phonon coupling, as the quasi-particles can have similar energies and momenta - neither is a universally small perturbation of the other. An accurate measurement or calculation of MPC is pivotal for the correct description of the thermodynamical properties of magnetic materials, the free energy will contain magnetic and vibrational contributions, often of the same magnitude. The MPC strength is maximal when spin-waves and elastic waves have the same frequency and wave number, i.e. when there is a crossing in the two dispersion curves.[12] At the intersection, the system shows neither a magnon nor a phonon, but rather a magneto-elastic excitation.

In the following, we present calculations of the combined effects of phononic (T_p) and magnonic (T_m) temperatures on the spin-wave (SW) dispersion, stiffness, and Curie temperatures of Fe, Ni and disordered Ni_81Fe_{19} (permalloy or simply Py) by combining first principles methods with model Hamiltonians. Inspired by Ref.[9], we present a theory that includes several effects on the magnetic exchange couplings. In addition to a first principles description of MPC, we also take into account disorder within the Virtual Crystal Approximation (VCA) to calculate Heisenberg exchange integrals. This is non-trivial and adds chemical/structural...
disorder into the interplay between the magnetic and thermal excitations.

As a first step in our analysis, we consider the effect of $T_p$ on the SW dispersion by means of Linear Spin-Wave Theory (LSWT). We then take into account the effects of thermal fluctuations in the magnetic system, within the quasi-harmonic approximation and the Atomistic Spin Dynamics (ASD) formalism\cite{21,22}, which allows for thermal noise and SW-SW interactions. In this way, we can consider separately the effects of the phonon and magnon temperatures, and study their influence on the SW energies, and implicitly gauge the MPC strength. We demonstrate the effect of the phonon temperature on the magnon dispersion, and show that, in Py, it competes with the spin fluctuations and renormalizes the acoustic mode. This introduces an unexpected non-monotonic behavior of the magnetic response as a function of $T$, and defines an optimal temperature window for SW generation in spintronics applications.

Several previous works have taken a different approach by mixing spin and molecular dynamics potentials, combined with parametrized radially dependent magnetic exchange integrals, in particular by the Dudarev group\cite{23}. This introduces an entropic effect on the magnetic exchange couplings, which then affects the total magnetic response in alloy systems. For Fe, they show very good agreement with experimental Curie temperatures. The exchange interactions are strongly simplified, assuming a $1/r^3$ functional dependence obtained from fitting two sets of calculated exchange integrals\cite{24}. These exchange integrals are truncated to second nearest neighbors, and do not take into account the detailed and oscillatory behavior of the exchange further out, which is relevant in many different magnetic materials. Here we consider the full range of exchange integrals, but consider the thermal vibrational effects in a more averaged way, as described below.

Our work goes in the same direction as two other first principles approaches, namely, Refs.\cite{21} and\cite{22}, trying to fill the gap between the fully ordered ferromagnetic and the disordered paramagnetic states. In this paper we show explicitly how temperature reduces the magnetic exchange amongst the nearest neighbors and increases the coupling between remote ones. The effective MPC that we find in Permalloy is consistent with the “phonon drag” theory of the Spin Seebeck Effect (SSE)\cite{25,26}. Very recently\cite{27}, the temperature dependence of the SW stiffness was determined experimentally for thin films using ferromagnetic resonance measurements. We show below that the dependence at very low $k$ (SW stiffness) only shows the magnon scattering, while the full dispersion should be sensitive to phonons as well. In general, thermal corrections improve the calculated Curie temperature with respect to experiment, but with very different magnitudes for different materials.

The paper is organized as follows: firstly we present our formalism for the thermal dependence of the magnetic Heisenberg model, followed by the Linear Spin Wave Theory for disordered systems, and finally, Atomistic Spin Dynamics and Curie Temperature calculations. The last sections report our numerical results, discussions and conclusions.

![Diagram](image)

**FIG. 1:** A schematic showing the methodology used in this article. The green squares represent the use of a code or method, the circles represent processing of data from a code/method and the arrows show the transfer of data with the text next to the arrow representing what data is being transferred. All the symbols are explained in the Methods section.

### METHODS

In this section we detail how we calculate the phonons, magnons, and their effective coupling. The spin dynamics are treated with two formalisms, linear spin wave theory, and atomistic spin dynamics, which we have extended, the former for disorder (Appendix A), and both for finite lattice temperature.

Fig. 1 shows the a schematic of the different approaches we have followed, which are explained in detail below. All structures are initially relaxed with **ABINIT**\cite{28} to give the ground state lattice parameters. This structure is fed into **PHONOPY**\cite{29} whereby the average atomic displacements are calculated as a function of temperature. The
same lattice parameters are also taken and an atom in the
unit cell is perturbed by \( \Delta x \) to give a new structure
which is subsequently used by SPRKRR. The SPRKRR
code then calculates the Heisenberg exchange parameters
as a function of the perturbation. The size of the displace-
ment as a function of temperature is combined with
the displacement dependent \( J_{ij} \) are then combined to
give \( J_{ij}(T_p) \). These exchange constants are then used in
both linear spin-wave theory and atomistic spin dynamics
(R-ASD for perturbed \( J_{ij} \) and ASD where the perturba-
tion is zero). From (R)-ASD the temperature dependent
magnetization and spin-waves are output and the Curie
temperature and exchange stiffness calculated, respec-
tively. The LSWT does not take into account changes in
the length of the magnetization and therefore only looks
at the effect of the phonon renormalization on the spin-
waves (and then stiffness).

1. Thermal dependent exchange integrals

We parametrize the magnetic Hamiltonian of the sys-
tem, mapping electronic structure calculations on a
Heisenberg Hamiltonian of the form:

\[
\mathcal{H} = - \sum_{\langle i, j \rangle} J_{ij}^{\rho\eta} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}
\]

where \( i \) and \( j \) represent atomic sites and the \( \rho, \eta \) index refer to the atomic species. In the Hamiltonian eq. (1)
each species is treated with an explicit sublattice. In
this framework, both CPA and VCA consider a disor-
dered, single-atom, species to deal with disorder. \( J_{ij}^{\rho\eta} \)
is the exchange coupling between spins located at different
sites, and \( \mathbf{S}_i \) are the spins associated with a given sub-
lattice. The exchange coupling constants are calculated
as a function of the interatomic positions through the
ab-initio SPRKRR code.[2]

The phonon bath temperature \( (T_p) \) is introduced by
calculating, ab-initio, the atomic mean square thermal
displacement \( \sqrt{\langle u^2(T) \rangle} \) as implemented in PHONOPY[3] and
obtained from the quasi-harmonic approximation in
the second quantization formalism. Chemical disorder is
introduced for Py 1) in the \( J_{ij}^{\rho\eta} \) values using the CPA
method, which returns all possible interactions among
different sites, and 2) in the phonons and thermal dis-
placement using the VCA.

To calculate the effect of temperature on the exchange
integrals, the Heisenberg exchange parameters, \( J_{ij}^{\rho\eta} \)
are calculated for a set of distorted unit cells where the atom
at the origin of the cell is displaced from its equilibrium
position in steps from 1\% to 5\% of the cubic lattice con-
stant, in the \( x \) direction. As the unit cells are all cubic,
we only consider one displacement (along the \( x \) axis).
Checks for forward and backward displacement, and with
smaller steps (0.5\%) were carried out to validate num-
erical aspects. For each displacement \( \Delta x \) we calculate the
exchange integrals \( J_{ij}^{\rho\eta}(\Delta x) \).

The results are then fit to a second order polynomial
with respect to the displacement:

\[
J_{ij}^{\rho\eta}(\Delta x) \sim J_{ij}^{\rho\eta}(0) + \frac{1}{2} \frac{\partial^2 J_{ij}^{\rho\eta}}{\partial u^2} \Delta x^2 \tag{2}
\]

By choosing \( \Delta x = \sqrt{\langle u^2(T) \rangle} \) we obtain a temperature
dependent \( J_{ij}^{\rho\eta}(T_p) \equiv J_{ij}^{\rho\eta}(\sqrt{\langle u^2(T_p) \rangle}) \).

A different CPA formula incorporating thermal displace-
ments and \( \langle u^2(T) \rangle \) has recently been implemented
in SPRKRR using a Debye approximation in Refs.[4,5]
but does not yet allow for the calculation of the \( J_{ij}^{\rho\eta} \).

The cubic symmetry is broken by the finite displace-
ment and subsequently restored on the exchange integrals
by re-symmetrizing the full \( J_{ij} \) matrix with the original
cubic symmetry operations along \( x, y, \) and \( z \) (we assume
uncorrelated thermal displacements). The renormaliza-
tion is general, in that the \( J_{ij} \) variation propagates also
to integrals relative to non displaced atoms - this is also a
novelty compared to most previous calculations (except
supercell approaches) which are two-body, and usually
only vary \( J \) with the interatomic distance.

2. Linear Spin Wave Theory

We develop the theory of Linear Spin Waves for binary
disordered systems into Appendix [1]

3. Atomistic Spin Dynamics

The dynamics of each spin is governed by the
phenomenological Landau-Lifshitz-Gilbert equation
(LLG)[5,6]

\[
d\mathbf{S}_i/dt = -\gamma_i/(1 + \lambda_i^2)\mu_i \mathbf{S}_i \times [H_i + \lambda_i \mathbf{S}_i \times \mathbf{H}_i], \tag{3}
\]

where \( \lambda_i \) is the coupling to the magnon thermal bath
which governs return to FM equilibrium. The amplitude
of the magnetic moment is given by \( \mu_i \). The effective
fields, \( H_i \), at the site \( i \) are determined using a Heisenberg
Hamiltonian including exchange (as given in Eq. (1)) ex-
tended with anisotropy and Zeeman terms:

\[
H_{\text{ASD}} = - \sum_{\langle i, j \rangle} J_{ij}^{\rho\eta} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i K_i (\mathbf{S}_i \cdot \hat{n})^2 - \sum_i \mu_i \mathbf{S}_i \cdot \mathbf{B}, \tag{4}
\]

where \( K_i \) is the uniaxial anisotropy constant, assumed
to be rather small in agreement with experimental and
theoretical work[5,6]. \( \hat{n} \) is the direction of the easy axis
taken here to be in the \( z \) direction. The final term in
Eq. (4) is the Zeeman interaction with the applied mag-
netic field, \( \mathbf{B} \). Based on a real space formalism, the
magnetic moments, \( \mu_i \), are assumed to be localized on
a given atomic site, \( i \), with their time-dependence given
by the phenomenological LLG Eq. (6). The effective field is given by the derivative of the Hamiltonian with respect to the spin:

$$H_t = -\frac{\partial H_{\text{ASD}}}{\partial S_i} + \zeta_t,$$

(5)

and includes stochastic thermal fluctuations, $\zeta_t$. These are included by incorporating a Langevin thermostat set to the desired magnonic temperature, $T_m$. In the present work the noise process is assumed to be white because of the time-scale of equilibrium properties, where the heat bath (phonon or electron system) acts much faster than the spin system. The correlators of the process are defined through the fluctuation dissipation theorem as:

$$\langle \zeta^\alpha (t) \rangle = 0$$

$$\langle \zeta^\alpha (t) \zeta^\beta (t') \rangle = 2\lambda_j k_B T \mu_\alpha \delta_{ij} \delta_{\alpha \beta} \delta(t - t').$$

(6)

The $\alpha, \beta$ represent cartesian (spin) components and $i, j$ represent spin indices.

In ASD, unlike LSWT, disorder is taken into account by having a large supercell with 131,072 single species atoms placed randomly on a lattice (fcc for Py), such that the desired composition is reached: Ni and Fe atoms do not occupy the same sites. For pure Fe and Ni, the values of the moments were $\mu_{Fe} = 2.50 \mu_B$ and $\mu_{Ni} = 0.655 \mu_B$, respectively. For Py, the magnetic moments were obtained from ground state SPRKKR calculations and were $\mu_{Fe} = 2.637 \mu_B$ and $\mu_{Ni} = 0.628 \mu_B$. The phonon bath temperature is again included through the $\mathcal{J}_{ij}^{\mu}(T_p)$ parameters. This gives a renormalized version of the ASD, to which we give the acronym R-ASD.

4. Curie Temperature

To calculate the temperature dependent magnetization (and subsequently the Curie temperature) the coupled LLG equation (3) is solved iteratively in the high damping limit and time-averages of the components of the spin ensemble, $m_\rho = \frac{1}{N_s} \sum_{i \rho} S_i(t)$, are taken as in Ref[32]. The system is initially equilibrated and a further period of time is then simulated to determine the average magnetization, which is monitored until convergence in both the mean and the variance is obtained.

5. Spin-waves

The temperature-dependent magnon frequencies are determined by calculating the dynamic structure factor[33]
A generalized gradient approximation exchange-correlation functional is employed by Perdew-Burke-Érnzerhof (PBE). A Spin-polarized Scalar-Relativistic (SP-SREL) Hamiltonian was used with full potential on a Brillouin-zone grid of $22 \times 22 \times 22$ k-points, orbital momentum cut off $l_{\text{max}} = 3$ and 60 points on the complex energy path. All calculations were converged to 0.1 mRy of total energy. Temperature dependent exchange integrals $J_{ij}^{\text{en}}(T)$ are then obtained for each possible interaction of atoms in the central unit cell with all neighbors within a sphere of 3.5 times the lattice constant. Only one type of interaction is possible in pure Fe and Ni, whereas three different interactions are present in Py. Note that the chemical disorder in Py is described with a VCA approximation while the magnetic disorder comes from CPA. We used the conventional unit cells for the calculations: 4 atom fcc for Ni and Py, and 2 atom bcc for Fe.

Anharmonic effects are not considered in this work, as a first step, justified as follows: (i) harmonic phonon frequencies agree with experimental results, (ii) the coefficient of thermal expansion for Py is modest at $12 \times 10^{-6}$ K$^{-1}$/Ref. 11 III/32A) and (iii) the fit of the $J$ to a quadratic function of displacement has a residual error below $10^{-6}$ for all cases relative displacement of at most $\sim 5\%$ at 1000K. These effects could be included by 1) generalizing the renormalization of the magnetic coupling due to several atomic displacements, and 2) by renormalizing the frequencies and mean squared displacements; these avenues will be considered in the future, but given the final thermalization procedure we expect the results would be qualitatively unchanged.

RESULTS

1. T-dependent Heisenberg Exchange Integrals

Temperature dependent exchange interactions, as a function of the reduced distance between atoms and temperature, are displayed in Fig. 3 for Fe and Ni (3a top and bottom) and for Py (3b). For Py, the variation of the magnetic exchange is shown in all three possible interactions in Py (Fe-Fe, Ni-Ni and Fe-Ni). The amplitude of the ferromagnetic coupling can either increase or decrease with $T_p$ and exchange integrals can even change sign. For Py, the amplitude of the exchange interaction between the first and second nearest neighbor interactions decreases with increasing $T_p$. Further out, the picture is more complex. The change in amplitude of $J$ in Fe and Ni is much smaller than Py, in particular for the Fe-Fe and Fe-Ni inter-sublattice interactions. We postulate that within the CPA (with several species) the atomic displacement has a stronger effect on the charge transfers between Fe and Ni components, and thus on the local exchange integrals.

The temperature dependence of the different neighbor interactions is also shown in Fig. 4 for the Fe-Fe, Ni-Ni and Ni-Fe interactions in Py (N.B.: without the $1/r_{ij}^3$).

FIG. 3: (Color online) Temperature dependent Heisenberg exchange integrals (times reduced radius cubed) vs. reduced radius and for different values of the phonon temperature.

2. LSWT

To demonstrate the net effect of the phonon temperature on the SW frequencies in Py, we report the SW modes within LSWT (no thermal bath for the spins) in Fig. 5 (panel a). In Py, there are two magnon modes, one optical and one acoustic, with a form determined by the mixture of the Ni-Ni, Fe-Fe and Ni-Fe exchange (cfr. Eqs. A15 and A14). We set $\langle S_0 \rangle = \langle S_1 \rangle = 1$ and $\nu_0 = 0.81$ and $\nu_1 = 0.19$ in the $J_{ij}^{\text{en}}$ definition. $T_p$ produces an increase in magnon energies at small $k$, for both the acoustic and the optical branches, while there is almost no change at larger $k$. 
presuming Bloch’s law holds; or through the temperature dependence of ferromagnetic resonance (FMR) frequencies for standing waves in thin films, presuming the films have bulk-like magnon dispersions. The latter two were used in Ref.\textsuperscript{29} and yield very different values (\(\sim 250\) vs \(450\) meV \(\AA^2\)) leading the authors to conclude 1) that Bloch’s law does not hold and 2) that magnetism is probably itinerant. We agree with the former, but not the latter, as shown below. To determine \(D\), we choose a fitting region up to \(\omega \rightarrow 0\), as represented by the vertical dashed line in Fig.\textsuperscript{5} panels a and c. The resulting \(D(T)\) is shown in panels b for LSWT (phonon temperature only), and d for ASD (magnons only - blue triangles) and R-ASD (both magnons and phonons - green circles). As the real dispersion is never purely parabolic outside \(\Gamma\), it is normal that different fits disagree.

We observe a monotonic increase of the stiffness with the temperature for LSWT. The near neighbor exchange integrals soften in all three channels, whereas from the fifth neighbors out many harden (Fig.\textsuperscript{4}). We conclude that, as the temperature increases in LSWT, the weight of the first neighbors’ \(J_{ij}\) becomes less important than at low temperature: the phonons can have a strong effect on the long range changes in \(J_{ij}\), which were not taken into account in previous work.

For ASD, when \(T_p = 0\) (blue triangles in Fig.\textsuperscript{5}d) the stiffness monotonically decreases, while in the R-ASD the introduction of finite \(T_p\) induces a non-monotonic trend: for low \(T\) the phonons increase \(D\) as for the LSWT, and at large enough \(T_m\), the magnon dispersion must flatten, and \(D\) decreases. The agreement for the “straight” ASD with the experimental \(D\) (red crosses, panel d) is excellent, showing that purely magnon fluctuations reproduce the temperature dependence, and there is no need to invoke questions about itinerant magnetism in Py discussed in Ref.\textsuperscript{30}. The stiffness, however, only contains very limited information in \(k\): in the experimental setup, an FMR method uses a standing SW with a wavelength equal to the film thickness \(d\) (100 nm in Ref.\textsuperscript{30}), and is therefore sensitive only to a single non-zero \(k = 2\pi/d\), which is very close to \(\Gamma\).

Our lattice thermal effect is strongest for the acoustic mode at the zone edge, and our \(k\) resolution at the zone center is limited by ASD supercell size and statistics. Due to 1) a limited phonon supercell and 2) single atom displacements plus re-symmetrization, our calculations clearly overestimate the MPC at \(\Gamma\) (MPC should go to 0 at \(\Gamma\) due to phase space arguments for energy and momentum conservation). At larger \(k\), the SW dispersion becomes non-parabolic (as shown in Fig.\textsuperscript{5}c) and this is amplified by magnon and phonon perturbations. Both increase the higher order polynomial terms in \(k\), but have opposite effects on the dispersion. The combination produces a non-monotonic behavior in the magnon frequencies within the R-ASD. From this perspective, neutron scattering or a series of FMR measurements with smaller film thicknesses or higher harmonics of the standing waves would yield precious information on the SW

3. Atomistic Spin Dynamics

The effect of the spin temperature on the SW spectrum is introduced with the ASD and R-ASD, and shown in Fig.\textsuperscript{3}. The resulting dispersion curve is quite different from that of the phonons in the LSWT case (Fig.\textsuperscript{5}), showing the limits of the linear approximation, and in particular for higher temperatures. The inclusion of thermal spin fluctuations excites all eigenmodes of the system, and softens the magnons as the temperature of the thermostat increases. The acoustic branch is depressed at a frequency close to 200 meV (difference between 0K and 600K curves in panel c), near the Brillouin zone boundary, an effect which is much larger than the one found considering only phonon effects. The optical branch shows an opposite behavior in the whole Brillouin zone, an effect which is much larger than the one found considering only phonon effects. The optical branch softens in all three channels, whereas from the fifth neighbors out many harden (Fig.\textsuperscript{4}). We conclude that, as the temperature increases in LSWT, the weight of the first neighbors’ \(J_{ij}\) becomes less important than at low temperature: the phonons can have a strong effect on the long range changes in \(J_{ij}\), which were not taken into account in previous work.

To compare to experiments, we consider the SW stiffness \(D\) (\(\omega \rightarrow 0\) for small enough frequency), which is easier to access than the full dispersion. There are several ways to extract \(D\); through \(q \rightarrow 0\) fitting to the dispersion; through the temperature dependence of \(M(T)\)
5. Magnetization and Curie Temperature

In this section, we discuss the effect of the temperature dependent exchange interactions $J_{ij}^{\rho\eta}(T_p)$ on the magnetization and Curie temperature, $T_c$. We have calculated the equilibrium magnetization $M(T)$ and show the results in Fig. 5. The ASD Curie Temperature is derived as in Ref. 29. We consider here the two cases: ASD (using $J_{ij}^{\rho\eta}(0K)$ values - empty symbols in Fig. 5) and the R-ASD (using the temperature-dependent exchange constants, $J_{ij}^{\rho\eta}(T_p)$, calculated previously with $T_m = T_p$). This magnetization contains the combined effect of the phononic and magnetic temperatures for the R-ASD case (closed symbols in Fig. 5).

The Curie temperatures from the different models used and from theoretical and experimental literature are given in Table. We also report the result in the Mean Field Approximation (MFA) as in 29, considering both the unperturbed $J_{ij}$ and the temperature dependent $J_{ij}(T_p)$. Phonon corrections lead to a slight decrease in the ASD calculated $T_c$ for bcc Fe (from 1344 K to 1333 K), while it is increased for fcc Ni (from 409 K to 415 K). In both cases introducing the phonon temperature induces an improvement, but only by a few percent or less.

In the case of disordered Py our theoretical result outclasses previous methods: ASD returns $T_c$=656 K using $J_{ij}(0K)$, consistent with other works using CPA, while within R-ASD, $T_c$ increases to 844 K which considerably improves the agreement with experiments. This level of agreement may be fortuitous, but the amplitude of the correction shows that the renormalization of the exchange $J_{ij}^{\rho\eta}$ with lattice temperature is crucial. A mean field calculation with $J_{ij}^{\rho\eta}(0K)$ and $J_{ij}^{\rho\eta}(T_p)$ also shows a strong variation of $T_c$, suggesting the details of the spin fluctuations are secondary.

We believe the difference in the order of magnitude of the phonon correction is due to two interrelated factors: 1) the presence of an optical magnon mode in Py en-
Reduced Magnetization

![Graph showing temperature dependent magnetization curves of the two sublattices in Py: Fe (green circle) and Ni (blue triangles) calculated using ASD (open symbols) and temperature dependent (closed symbols) exchange constants (R-ASD). Full lines are Taylor expansion fits to extract the Curie temperatures (vertical lines). The grey area shows the range of Curie temperatures found experimentally, while the red vertical lines represent the Curie temperature from ASD (dashed) and R-ASD (full).](image)

We have assumed here that the fundamental postulate of their theory holds, i.e. a much faster time-scale of the electronic degrees of freedom with respect to the slow magnetic system, and further postulate that correlations of the fast ionic motion are negligible. Both of these may be incorrect, and further work on the topic is very important for the fundamentals of magnon phonon coupling.

The Heisenberg model itself has limits, but should function for Fe and Py, which present localized magnetism. There is no good definition of localized moments in Ni, but empirically the Heisenberg model seems to work, and we include it for comparison: our goal here is to investigate the interplay of vibrational and thermal effects, and they appear to work in similar ways in Ni.

A commonly proposed mechanism for the Spin Seebeck Effect is the propagation of out-of-equilibrium long-wavelength phonons in a crystalline substrate below the sample. This enables a phonon-drag-like pumping of acoustic magnons in the magnetic material, and a resulting spin current which has a non-local origin. This is a difficult hypothesis to verify experimentally, and contributed to motivate the present study: is the strength of the MPC sufficient to justify the observed spin current? In Ref. 23, a semi-empirical model confirmed this hypothesis showing that at short wavelengths the magnetoelastic modes are mixed and scatter quickly, whereas at long wavelengths, scattering is weaker. We find in Py that spin fluctuations affect magnons of all wavelengths, whereas MPC is particularly strong for intermediate to short wavelengths (k > 1.2/a), which provides an important suggestion for future SSE models.

Our magnon dispersions implicitly contain the MPC in the temperature dependence of the magnons at different k, but also through the difference in temperature between the magnonic and phononic baths. For comparison, we have calculated of the stiffness, D, for three different cases, using the same low-k fitting of the spin-wave spectrum as above. The first case is with a fixed magnetic temperature (the temperature of the spin thermostat) of T_m=100K and a varying phonon temperature, T_p (see blue squares in Fig. 7). The second case has a fixed phonon temperature, T_p=100K and a varying temperature of the magnetic system, T_m (see red diamonds in Fig. 7). The third case corresponds to the case where the magnetic temperature, T, is temperature-dependent (green circle) and Ni (blue triangles) calculated using ASD (open symbols) and temperature dependent (closed symbols) exchange constants (R-ASD). Full lines are Taylor expansion fits to extract the Curie temperatures (vertical lines). The grey area shows the range of Curie temperatures found experimentally, while the red vertical lines represent the Curie temperature from ASD (dashed) and R-ASD (full).

### TABLE I: Curie Temperature (in K) calculated from MFA and ASD compared to previous results

<table>
<thead>
<tr>
<th>Method</th>
<th>Fe</th>
<th>Ni</th>
<th>Py</th>
</tr>
</thead>
<tbody>
<tr>
<td>Other: MFA</td>
<td>1414</td>
<td>397</td>
<td></td>
</tr>
<tr>
<td>This: MFA</td>
<td>1725</td>
<td>455</td>
<td>796</td>
</tr>
<tr>
<td>Other: ASD</td>
<td></td>
<td></td>
<td>650</td>
</tr>
<tr>
<td>This: ASD</td>
<td>1344</td>
<td>409</td>
<td>656</td>
</tr>
<tr>
<td>This: R-ASD</td>
<td>1333</td>
<td>415</td>
<td>844</td>
</tr>
<tr>
<td>Other: RPA</td>
<td>950</td>
<td>350</td>
<td></td>
</tr>
<tr>
<td>Other: R-RPA</td>
<td>1057</td>
<td>634</td>
<td></td>
</tr>
<tr>
<td>Monte Carlo</td>
<td>1065</td>
<td>615</td>
<td></td>
</tr>
<tr>
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<td>628.5</td>
<td>850</td>
</tr>
<tr>
<td></td>
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<td>871</td>
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</table>

DISCUSSION

In this section we elaborate on the limitations of our method and directions for further study.

One important aspect of the MPC is adiabaticity: the (non)conservation of energy in the process of mutual scattering. A review of the problem of adiabaticity in itinerant ferromagnets can be found in Ref. 43. We have assumed here that the fundamental postulate of their theory holds, i.e. a much faster time-scale of the electronic degrees of freedom with respect to the slow magnetic system, and further postulate that correlations of the fast ionic motion are negligible. Both of these may be incorrect, and further work on the topic is very important for the fundamentals of magnon phonon coupling.
and phononic temperature are equal, \( T_m = T_p \) (see green circles in Fig. 7, same in Fig. 5(i)). The error bars are estimated from the error in the non-linear least squares fitting procedure. The trend of the three \( D(T) \) curves is quite different, and offers therefore a way to distinguish between strong and weak MPC at low \( k \) by comparing to a “slow” adiabatic measurement. If \( D \) rises slowly or decreases with \( T_m \), or if it increases strongly with \( T_p \), then the coupling is weak and thermalization is difficult. We therefore strongly encourage a systematic experimental measurement of SW stiffnesses, in order to verify the intrinsic MPC for other materials, and validate or expand the present explanation of the SSE.

**CONCLUSION**

A new method to calculate the thermal variation of magnetic exchange couplings is introduced, and the resulting change in the SW frequencies is presented. The effects of both spin and lattice temperatures, \( T_m \) and \( T_p \), are taken into account, using the ASD method and phonon-renormalized exchange interactions, respectively. We show that phonons weaken the exchange interaction at short distances, and often harden for spins located further out, which has a non-trivial effect on the SW dispersion. \( T_p \) and \( T_m \) have competing effects on the magnon frequencies for Permalloy, which opens perspectives both for understanding and for tuning thermomagnetic behaviors. We compare the LSWT and mean field approaches, which ignore SW-SW interactions, with the ASD which includes SW fluctuations. The temperature variation of the SW stiffness reflects exclusively the zone center dispersion, and is well described with spin fluctuations only. We find that the full dispersion relation changes due to both lattice and spin fluctuations, an effect which would require neutron scattering measurements of the dispersion to confirm. The agreement of our calculations with experimental \( T_s \) suggests that the recently measured simple variation of \( D(T) \) belies the complex evolution of the full dispersion. SW stiffness values will depend strongly on the methods used to measure them, either relying on the dispersion, on Bloch’s law, or using ferromagnetic resonance measurements. We hope to stimulate further experimental investigation of the thermal evolution of SWs in Py and in other materials. This will provide a direct and simple quantification of the MPC, and is central to the understanding of spin-caloritronic effects.

Our theory is general and it can be applied to different crystal forms, magnetic cations and variations of the components and alloy fractions. Beyond magnon spectrum changes with temperature, it also allows one to assess variations as a function of impurity concentration. These results open up important perspectives for tailoring alloys, without the need for costly nanostructuring, to obtain optimal spintronics and spin-caloritronic materials in a desired temperature window. Our results indicate that phonons can also lead to deviations from Bloch’s law in measurements of the exchange stiffnesses\(^{[32]} \). Natural extensions include a fully phonon mode and wavevector dependent formalism, to explore in more detail MPC from first principles.

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Appendix A: Linear Spin Wave Theory for disordered materials

In this section we derive explicitly the LSWT for an alloy, assuming a mixture of both Ni and Fe at each atomic site, i.e. the site and species exchange constants are weighted by the composition. We begin by linearizing the exchange part of the Hamiltonian Eq. (1) assuming that the x/y oscillations are small with respect to the spin moment in the quantization direction (taken to be the z-axis). Writing the dot products in terms of the spin raising and lowering operators, \( S_i^{\pm,\rho} = S_i^x \pm iS_i^y \), which are now species dependent:

\[
S_i^{\rho} \cdot S_j^{\eta} = \frac{1}{2} \left[ S_i^{+,\rho}S_j^{-,\eta} + S_i^{-,\rho}S_j^{+,\eta} + S_i^{z,\rho}S_j^{z,\eta} \right],
\]

(A1)

where \( \rho, \eta \) are the species. To describe the spin-wave energies of a two component disordered alloy system we first write the Hamiltonian explicitly for each set of interactions:

\[
\mathcal{H} = \sum_i \sum_j \tilde{J}_{ij}^{00} \mathbf{S}_i^0 \cdot \mathbf{S}_j^0 + \sum_i \sum_j \tilde{J}_{ij}^{01} \mathbf{S}_i^0 \cdot \mathbf{S}_j^1 + \sum_i \sum_j \tilde{J}_{ij}^{11} \mathbf{S}_i^1 \cdot \mathbf{S}_j^1,
\]

(A2)

where the superscripts \((0,1)\) correspond to each species pair \((\rho, \eta)\). We assume that the magnetic moments of each species occupy the same sites (see below) and the \( \tilde{J}_{ij}^{mn} \) are corrected in the spirit of a VCA to account for composition, \( \tilde{J}_{ij}^{00} = J_{ij}^{00} \nu_{j}^{\rho} \), where \( \nu_{j}^{\rho} \) is the percentage of the species, \( \rho \). Writing the \( S_i^{\rho} \cdot S_j^{\eta} \) products in terms of the species dependent spin raising and lowering operators, the Hamiltonian becomes:

\[
\mathcal{H} = \sum_i \sum_j \tilde{J}_{ij}^{00} \left\{ \frac{1}{2} \left[ S_i^{+,\rho}S_j^{-,\eta} + S_i^{-,\rho}S_j^{+,\eta} + S_i^{z,\rho}S_j^{z,\eta} \right] + \frac{1}{2} \left[ S_i^{+,\rho}S_j^{-,\eta} + S_i^{-,\rho}S_j^{+,\eta} + S_i^{z,\rho}S_j^{z,\eta} \right] \right\} + \sum_i \sum_j \tilde{J}_{ij}^{01} \left\{ \frac{1}{2} \left[ S_i^{+,\rho}S_j^{-,1} + S_i^{-,\rho}S_j^{+,1} + S_i^{z,\rho}S_j^{z,1} \right] \right\} + \sum_i \sum_j \tilde{J}_{ij}^{11} \left\{ \frac{1}{2} \left[ S_i^{+,1}S_j^{-,1} + S_i^{-,1}S_j^{+,1} + S_i^{z,1}S_j^{z,1} \right] \right\}.
\]

(A3)

As we are neglecting the thermal effects arising from fluctuations of the magnetic moments we take the low temperature approximation for the Holstein-Primakoff transformations:

\[
S_i^{+,0} = \hbar \sqrt{2} a_i^{+,0},
\]

\[
S_i^{-,0} = \hbar \sqrt{2} a_i^{-,0}.
\]

(A4)

For ease of notation we take \( \hbar = 1 \) and absorb the spin value, \( S \), in the exchange constant (i.e. setting \( S = 1 \)). The transverse components in Eq. (A3) are then replaced by the low temperature Holstein-Primakoff transformation and we use the relation, \( S_i^{z,\rho} = S - \hat{n}_i^{\rho} \),

\[
\mathcal{H} = \sum_{i,\delta} \tilde{J}_{i\delta}^{00} \left\{ a_i^{-,0} a_{i+\delta}^{+,0} + a_i^{+,0} a_{i+\delta}^{-,0} - a_i^{+,0} a_{i+\delta}^{-,0} - a_i^{-,0} a_{i+\delta}^{+,0} \right\} + \sum_{i,\delta} \tilde{J}_{i\delta}^{01} \left\{ a_i^{-,0} a_{i+\delta}^{+,1} + a_i^{+,0} a_{i+\delta}^{-,1} - a_i^{+,0} a_{i+\delta}^{-,1} - a_i^{-,0} a_{i+\delta}^{+,1} \right\} + \sum_{i,\delta} \tilde{J}_{i\delta}^{11} \left\{ a_i^{-,1} a_{i+\delta}^{+,1} + a_i^{+,1} a_{i+\delta}^{-,1} - a_i^{+,1} a_{i+\delta}^{-,1} - a_i^{-,1} a_{i+\delta}^{+,1} \right\}.
\]

(A5)

We can then transform to Fourier space through \( a_i^{\pm,\rho} = \sum_{k} e^{\mp ik \cdot r_i} a_k^{\pm,\rho} \). Substituting these into Eq. (A5) gives (for readability we write just the first term, \( \rho = \eta = 0 \)):

\[
\mathcal{H}^{00} = \sum_{i,\delta, kk'} \tilde{J}_{i\delta}^{00} \left[ e^{-i(k-k') \cdot r_i} e^{+i k' \cdot \delta} a_k^{+} a_{k'}^{-} + e^{+i(k-k') \cdot r_i} e^{-i k' \cdot \delta} a_k^{+} a_{k'}^{-} + e^{-i(k-k') \cdot r_i} e^{+i k' \cdot \delta} a_k^{-} a_{k'}^{+} + e^{+i(k-k') \cdot r_i} e^{-i k' \cdot \delta} a_k^{-} a_{k'}^{+} \right],
\]

(A6)
Sums over \( i \) cause all terms to vanish unless \( k = k^0 \) and thus the sum becomes:

\[
\hat{\mathcal{H}}^{00} = \sum_{k,\delta} \tilde{\mathcal{J}}_{\delta}^{00} \left[ e^{-ik\delta} a_k^+ \bar{a}_k^0 + e^{ik\delta} a_k^0 \bar{a}_k^+ - 2a_k^+ a_k^0 \right],
\]

We use the identity \([a^+, a^-] = 1\) and define \( \gamma_k = \sum_{\delta} e^{-ik\delta}, \) and \( \hat{J}_k = \sum_{\delta} \tilde{J}_\delta e^{-ik\delta} \) For crystals with a center of inversion symmetry (FCC, BCC), \( \gamma_k = \gamma_{-k} \), simplifying Eq. (A7) to:

\[
\hat{\mathcal{H}}^{00} = \sum_{k,\delta} \tilde{\mathcal{J}}_{\delta}^{00} [\gamma_k a_k^+ \bar{a}_k^0 + \gamma_k a_k^0 \bar{a}_k^+ - 2a_k^+ a_k^0].
\]

Again, we ignore the arbitrary constant \( \sum_{k} \gamma_k \), and take the sum over \( \delta \) inside. The Hamiltonian given by Eq. (A8) becomes:

\[
\hat{\mathcal{H}}^{00} = 2 \sum_{k} (\tilde{\mathcal{J}}_{k}^{00} \gamma_k a_k^+ \bar{a}_k^0 + \gamma_k a_k^0 \bar{a}_k^+ - 2a_k^+ a_k^0).
\]

Terms for \( \hat{\mathcal{H}}^{01}, \hat{\mathcal{H}}^{10} \) and \( \hat{\mathcal{H}}^{11} \) can be derived in a similar way. For brevity they have not been shown explicitly here but we note that we can write the Hamiltonian as a matrix product:

\[
\hat{\mathcal{H}} = \sum_{k} a_k^+ M a_k^-,
\]

where \( M \) is a \( 2 \times 2 \) matrix containing the appropriate exchange constants and:

\[
a_k^\pm = \begin{bmatrix} a_{k,0}^\pm \\ a_{k,1}^\pm \end{bmatrix}.
\]

The Hamiltonian matrix is rewritten using a Bogoliubov transformation \([10]\) which now mixes the excitations associated with Ni-Ni, Fe-Fe and Ni-Fe exchange interactions.

\[
a_k^\pm = u_k a_k^\mp + \nu_k \bar{a}_k^\mp,
\]

\[
\bar{a}_k^\pm = u_k \bar{a}_k^\mp + \nu_k a_k^\mp.
\]

With this transform the Hamiltonian can then be written as:

\[
\hat{\mathcal{H}} = \sum_{k} [\alpha_k^+ a_k^-] M_k \left[ \begin{array}{c} \alpha_k^+ \\ \beta_k^- \end{array} \right].
\]

The elements of the matrix, \( M_k \) can be found by comparing coefficients of the \( a_k^+ \rho^- \):

\[
M_k = \begin{bmatrix} \Omega_{k,0}(S_0) + \Xi_{k,00} & \Xi_{k,01} \\ \Xi_{k,10} & \Omega_{k,1}(S_1) + \Xi_{k,01} \end{bmatrix},
\]

where \( \Omega_{k,\rho}(\bar{S}_\rho) = \frac{\gamma}{\mu_\rho} \sum_R \tilde{J}_{ij}^{\rho \rho}(R)[1 - \exp(ik \cdot R)] \) is the SW frequency of the individual species (the two possible sublattices). \( \gamma \) is the gyromagnetic ratio, \( \mu_\rho \) is the spin moment amplitudes and \( \Xi_{k,\rho \eta} = \frac{\gamma}{\mu_\rho} \sum_R \tilde{J}_{ij}^{\rho \eta}(R) \langle S_\eta \rangle \). \( \langle \cdot \rangle \) represent the equilibrium value of reduced magnetisation for each species (normalised to 1 at T=0K). Here we ignore spin fluctuations and these values are fixed to 1.

We could artificially introduce a temperature dependence of the magnetisation in the LSWT but we choose not to as the ASD gives a better account of spin fluctuations as it allows for spin-wave interactions.

Upon diagonalization, the solutions to the eigenvalue equation are given by:

\[
\omega_{\pm}(k) = \frac{1}{2} \left[ \text{Tr} M_k \pm \sqrt{(\text{Tr} M_k)^2 - 4 \det[M_k]} \right],
\]

where \( \pm \) corresponds to the upper (+) and lower (−) magnon branches, \( \det \) is the determinant. The resulting magnon dispersion curves and magnetic response are shown in Fig. 5 panels a and c and commented in the main text.