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

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I. INTRODUCTION

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 paper, 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.

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Since the 1950s, MPC has been studied using model Hamiltonians [1–4]. Only very recently, Fransson et al. [5] 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 [6–10]. The new field of Spin Caloritronics characterizes different transport phenomena, which depend on the coupling between thermal vibrations, charge, and spin [11,12]. 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 VO$_2$, where heat drives a softening of a VO$_2$ phonon mode, which couples to the magnetic response in Ni [13].

MPC is much more delicate than electron-phonon coupling, as the quasiparticles 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 [1,14,15]: The free energy will contain magnetic and vibrational contributions, often of the same magnitude. The MPC strength is maximal when spin waves (SWs) and elastic waves have the same frequency and wave number, i.e., when there is a crossing in the two dispersion curves [16]. At the intersection, the system shows neither a magnon nor a phonon, but rather a magnetoelastic excitation.

In the following, we present calculations of the combined effects of phononic ($T_p$) and magnonic ($T_m$) temperatures on the SW dispersion, stiffness, and Curie temperatures of Fe, Ni, and disordered Ni$_{81}$Fe$_{19}$ (permalloy or simply Py) by combining first-principles methods with model Hamiltonians. Inspired by Ref. [3], 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 [17] (VCA).
for the electronic ground state and vibrational properties calculation, and within the coherent potential approximation (CPA) to calculate Heisenberg exchange integrals. This is nontrivial 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 quasiharmonic approximation and the atomistic spin dynamics (ASD) formalism [20,21], 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 nonmonotonic 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 exchange integrals, in particular by the Dudarev group [22]. 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 [3,23]. 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. [4,24], 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) [25–27]. Very recently [28], the temperature dependence of the SW stiffness was determined experimentally for thin films using ferromagnetic resonance (FMR) 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: First, we present our formalism for the thermal dependence of the magnetic Heisenberg model, followed by the LSWT for disordered systems, and, finally, ASD and Curie Temperature calculations. The last sections report our numerical results, discussions, and conclusions.
1. Thermal-dependent exchange integrals

We parametrize the magnetic Hamiltonian of the system, mapping electronic structure calculations on a Heisenberg Hamiltonian of the form

$$\mathcal{H} = - \sum_{(i\rho,j\eta)} J^{\rho\eta}_{ij} \mathbf{S}^\rho_i \cdot \mathbf{S}^\eta_j, \quad (1)$$

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 disordered, single-atom species to deal with disorder. \(J^{\rho\eta}_{ij}\) is the exchange coupling between spins located at different sites and \(\mathbf{S}^\rho\) are the spins associated with a given sublattice. The exchange-coupling constants are calculated as a function of the interatomic positions through the \(ab\) \textit{initio} SPRKKR code [31].

The phonon bath temperature \((T_p)\) is introduced by calculating, \textit{ab initio}, the atomic mean-square thermal displacement \(\langle u^2(T_p) \rangle\) as implemented in PHONOPY [30] and obtained from the quasiharmonic approximation in the second quantization formalism. Chemical disorder is introduced for Py (1) in the \(J^{\rho\eta}_{ij}\) values using the CPA method, which returns all possible interactions among different sites and \(2\) in the phonons and thermal displacement using the VCA.

To calculate the effect of temperature on the exchange integrals, the Heisenberg exchange parameters, \(J^{\rho\eta}_{ij}\), 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 constant, 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 the \(J^{\rho\eta}_{ij}\) values using the CPA method, which returns all possible interaction as implemented in PHONOPY [30] and obtained from the quasiharmonic approximation in the second quantization formalism. Chemical disorder is introduced for Py (1) in the \(J^{\rho\eta}_{ij}\) values using the CPA method, which returns all possible interactions among different sites and \(2\) in the phonons and thermal displacement using the VCA.

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

$$J^{\rho\eta}_{ij}(\Delta x) \simeq J^{\rho\eta}_{ij}(0) + \frac{1}{2} \frac{\partial^2 J^{\rho\eta}_{ij}}{\partial u^2} \Delta x^2. \quad (2)$$

By choosing \(\Delta x = \sqrt{\langle u^2(T) \rangle}\), we obtain a temperature-dependent \(J^{\rho\eta}_{ij}(T_p) \rightarrow J^{\rho\eta}_{ij}(\sqrt{\langle u^2(T_p) \rangle})\).

A different CPA formula incorporating thermal displacements and \(\langle u^2(T) \rangle\) has recently been implemented in SPRKKR using a Debye approximation in Refs. [32,33] but does not yet allow for the calculation of the \(J^{\rho\eta}_{ij}\).

The cubic symmetry is broken by the finite displacement and subsequently restored on the exchange integrals by symmetrizing the full \(J_{ij}\) matrix with the original cubic symmetry operations along \(x, y,\) and \(z\) (we assume uncorrelated thermal displacements). The renormalization is general, in that the \(J_{ij}\) variation propagates also to integrals relative to nondisplaced 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 SWs for binary disordered systems into Appendix.

3. Atomic spin dynamics

The dynamics of each spin is governed by the phenomenological Landau-Lifshitz-Gilbert equation (LLG) [34]:

$$\frac{d\mathbf{S}_i}{dt} = -\gamma_i \mathbf{S}_i \times \left[ \mathbf{H}_i + \lambda_i \mathbf{S}_i \times \mathbf{H}_i \right], \quad (3)$$

where \(\lambda_i\) is the coupling to the \textit{magnon} thermal bath which governs return to FM equilibrium. The amplitude of the magnetic moment is given by \(\mu_i\). The effective fields, \(\mathbf{H}_i\), at the site \(i\) are determined using a Heisenberg Hamiltonian including exchange [as given in Eq. (1)] extended with anisotropy and Zeeman terms:

$$\mathcal{H}_{\text{ASD}} = -\sum_{(i\rho,j\eta)} J^{\rho\eta}_{ij} \mathbf{S}^\rho_i \cdot \mathbf{S}^\eta_j - \sum_i K_i (\mathbf{S}^\rho_i \cdot \hat{n})^2 \quad (4)$$

where \(K_i\) is the uniaxial anisotropy constant, assumed to be rather small in agreement with experimental and theoretical works [35]. \(\hat{n}\) 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 magnetic 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. (3). The effective field is given by the derivative of the Hamiltonian with respect to the spin,

$$\mathbf{H}_i = -\frac{\partial \mathcal{H}_{\text{ASD}}}{\partial \mathbf{S}_i} + \xi_i, \quad (5)$$

and includes stochastic thermal fluctuations, \(\xi_i\). These are included by incorporating a Langevin thermostat set to the desired magnonic temperature, \(T_m\). In the present paper, the noise process is assumed to be white because of the timescale 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

$$\langle \xi_i^\rho(t) \rangle = 0,$n\langle \xi_i^\rho(t) \xi_j^\eta(t') \rangle = \frac{2\lambda_i k_B T \mu_i}{\gamma_i} \delta_{ij} \delta_{\rho\eta} \delta(t - t'). \quad (6)$$

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 \(J^{\rho\eta}_{ij}(T_p)\) 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 Eq. (3) is solved iteratively in the high damping limit and time averages of the components of the spin ensemble, $\mathbf{m}_p = \frac{1}{N} \sum_{p \in \rho} \mathbf{S}_p(t)$, are taken as in Ref. [36]. 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 [37]:

$$S(k, \omega) = \frac{1}{N \sqrt{2\pi}} \sum_{\mathbf{r}, \mathbf{r}'} e^{i \mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \int_{-\infty}^{+\infty} e^{-i \omega t} C(\mathbf{r} - \mathbf{r}', t) dt,$$

where $C(\mathbf{r} - \mathbf{r}', t) = \langle S^+(\mathbf{r}, 0) S^-(\mathbf{r}', t) \rangle$ is the spin-spin correlation function of the transverse spin values ($S_x$ and $S_y$). The stochastic thermal term allows the spin system to sample all modes and the resulting spectra are analyzed to determine the frequencies. The resulting magnon dispersion curves and magnetic response are shown in Figs. 5(b) and 5(d), and commented below.

6. Numerical details

Thermal displacements $\sqrt{\langle \omega^2(T) \rangle}$ as a function of $T$ have been calculated from PHONOPY [30] in $2 \times 2 \times 2$ supercells. The electronic ground-state properties and forces have been calculated through density-functional theory using ABINIT [29]. The local density approximation and the generalized gradient approximation return very similar results for the electronic ground state as well as for phonon frequencies. The latter was employed with a set of FHI pseudopotentials [38], with a plane-wave energy cutoff set to 40 Ha, a gaussian electronic smearing of 1 meV, and a 123 Monkhorst-Pack grid to sample the Brillouin zone [39]. The ground-state energy was converged in all cases below $10^{-14}$ Ha. In the case of Py, the VCA was used to simulate disorder with the exact ratio (81% of nickel and 19% of iron) between species imposed. Harmonic phonon frequencies have also been calculated within density-functional perturbation theory as implemented in ABINIT [29] and agree well for these very simple one-atom unit cells. The phonon dispersion curve for VCA Py is shown in Fig. 2 in comparison with experimental results for disordered Ni$_3$Fe from Ref. [40].

The magnetic exchange integrals have been obtained using the spin-polarized relativistic Korringa-Kohn-Rostoker method as implemented in the Munich SPRKKR code [31,41], version 6.3. A generalized gradient approximation exchange-correlation functional is employed by Perdew-Burke-Érnzerhof [42]. 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 cutoff $l_{\text{max}} = 3$, and 60 points on the complex energy path. All calculations were converged to 0.1 mRy of total energy. Temperature-dependent exchange integrals

![FIG. 2. Py phonons energies calculated within DFPT in the harmonic approximation from VCA compared to experimental results from Ref. [40].](image)
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 $1/r^2$). It is important to notice that the first-neighbor interaction is generally the one most affected by the coupling with phonons (conforming with intuition), and that the $J$ decrease in absolute value.

**FIG. 3.** Temperature-dependent Heisenberg exchange integrals (times reduced radius cubed) vs reduced radius and for different values of the phonon temperature. (a) Fe (top) and Ni (bottom) (b) Py. Three interactions are present: Fe-Fe (top), Ni-Ni (middle), and Ni-Fe (bottom).

**FIG. 4.** Temperature dependence of the exchange interactions for the first-four neighbor shells shown explicitly for the Fe-Fe, Ni-Ni, and Ni-Fe interactions in Permalloy.

### 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(a). In Py, there are two magnon modes [37], one optical and one acoustic, with a form determined by the mixture of the Ni-Ni, Fe-Fe, and Ni-Fe exchange [cf. Eqs. (A15) and (A14)]. We set $S_0 = \langle S_i \rangle = 1$ and $v_0 = 0.81$ and $v_1 = 0.19$ in the $\tilde{J}_{ij}^{\text{opt}}$ 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 $k$.

### 3. Atomic spin stiffness

The effect of the spin temperature on the SW spectrum is introduced with the ASD and R-ASD, and shown in Fig. 5(c). The resulting dispersion curve is quite different from that of the phonons in the LSWT case [Fig. 5(a)], 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 0 K and 600 K curves in Fig. 5(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 ($\Gamma \rightarrow H$) compared to the LSWT, decreasing in frequency when $T = T_p = T_m$ increases. In Figs. 5(c) and 5(d), we present the easiest combination of temperatures of the different systems (phonon and magnon), setting $T_m = T_p$. This corresponds to the case where the phonons and magnons are completely equilibrated. Reference [25] shows that this is the case for the average global temperature in yttrium iron garnet upon pumping of heat into either bath. Other cases with $T_m \neq T_p$ are considered below (Fig. 7).

### 4. Spin-wave stiffness

To compare to experiments, we consider the SW stiffness $D$ ($\omega_\infty \approx Dk^2$ 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 $M(T)$ presuming Bloch’s law holds, or through the temperature dependence of FMR frequencies for standing waves in thin films, presuming the films have bulklike magnon dispersions. The latter two were used in Ref. [28] and yield very different values ($\sim 250$ vs 450 meV Å$^2$) 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 $ka = 1.2$, as represented by the vertical dashed line in Figs. 5(a) and 5(c). The resulting $D(T)$ is shown in Fig. 5(b) for LSWT (phonon temperature only), and Fig. 5(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. 4). We conclude that, as the temperature
FIG. 5. Left: SW dispersion relation in Permalloy for a range of effective phonon temperatures calculated (a) within LSWT (no magnetic disorder) and (c) within R-ASD with both magnon and phonon contributions. Right: temperature dependence of the SW stiffness within LSWT (b) and using ASD (d) with renormalized exchange integrals (green circles) and with $J_{ij}^{\text{nn}}$ values for $T_p = 0$ K (blue triangles). Dashed lines with open symbols in (d) are stiffness values extracted using a fit to Bloch’s law (which underestimate, as found in Ref. [28]), the red crosses with error bars in (d) represent the fit of $D(T)$ in Ref. [28]. Phonon thermal effects are invisible in the experimental SW stiffness, which probes only the very long wavelength limit.

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. 5(d)] the stiffness monotonically decreases, while in the R-ASD, the introduction of finite $T_p$ induces a nonmonotonic 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 [28]. 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. [28]), and is therefore sensitive only to a single nonzero $k = 2\pi/d$, 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 resymmetrization, 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 nonparabolic [as shown in Fig. 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 nonmonotonic 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 dispersion, to test our proposal of a strong thermal effect on the SW dispersion far from the acoustic $k \sim \Gamma$. Figure 5(d) also shows fits of $D$ from $M(T)$ as dashed lines. In this case, both ASD and R-ASD produce monotonically decreasing $D(T)$ and a strong underestimate as found experimentally in Ref. [28]. This difference reflects the limited validity of Bloch’s law, both in theory and experiment: $M$ is averaged over $k$ and gives a less accurate value of $D$. Even at low $T$, fitting $M(T)$ is much less reliable than FMR, at least for Py. Compared to $D$, $M(T)$ always decreases with $T$, and $M$ averages fluctuations over many length and timescales.

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. 6. The ASD Curie temperature is derived as in Ref. [36]. We consider here the two cases: ASD [using \( J_{ij}^{\text{nn}}(0 \text{ K}) \) values—empty symbols in Fig. 6] and the R-ASD (using the temperature-dependent exchange constants, \( J_{ij}^{\text{nn}}(T_p) \), calculated previously with \( T_p = T_p \)). This magnetization contains the combined effect of the phononic and magnetic temperatures for the R-ASD case (closed symbols in Fig. 6).

The Curie temperatures from the different models used and from theoretical and experimental literature are given in Table I. We also report the result in the Mean Field Approximation (MFA) as in Ref. [44], 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 \( T_c = 656 \text{ K} \) using \( J_{ij}(0 \text{ K}) \), consistent with other works using CPA [45], 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 \( J_{ij}^{\text{nn}} \) with lattice temperature is crucial. A mean field calculation with \( J_{ij}^{\text{nn}}(0 \text{ K}) \) and \( J_{ij}^{\text{nn}}(T_p) \) 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 enables more interactions with phonons and (2) our method is biased towards zone edge phonons, which seem to have a stronger coupling in Py, whereas in Fe and Ni the long wavelength phonons seem to be dominant. In the future, we will look at mode-resolved MPC and larger KKR unit cells, to verify these distinctions.

We note that the increase in the Curie temperature of Py due to the temperature-dependent exchange constants is already visible in their effect on the LSWT dispersion without magnetic disorder. In passing, Barker et al. [37] include the reduction of the magnetization directly in the LSWT, using a mean-field analysis, but their Curie temperature is strongly overestimated.

Finally, other corrections to the DFT/KKR+ASD framework are needed in general for a quantitative prediction of \( T_c \), e.g., many-body effects in the \( J_{ij}^{\text{nn}} \) [46].

IV. 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. [52]. We have assumed here that the fundamental postulate of their theory holds, i.e., a much faster timescale 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 MPC.

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 SSE is the propagation of out-of-equilibrium long-wavelength phonons in a crystalline substrate below the sample [27]. This enables a phonon-drag-like pumping of acoustic magnons in the magnetic material, and a resulting spin current which has a nonlocal origin. This is a difficult hypothesis to verify experimentally,
FIG. 7. Temperature-dependent exchange stiffness for three different nonequilibrium cases: (1) fixed magnetic (spin thermostat) temperature of $T_m = 100$ K and a varying phonon temperature, $T_p$ (blue square points), (2) fixed phonon temperature $T_p = 100$ K and a varying magnetic (red diamond points), and (3) equal magnetic and phononic temperature (green circle points). The error bars are estimated from the error in the nonlinear least-squares fitting procedure.

and contributed to motivate the present study: Is the strength of the MPC sufficient to justify the observed spin current? In Ref. [26], a semiempirical model confirmed this hypothesis showing that at short wavelengths the magnetoelastic modes are mixed and scatter quickly, whereas at long wavelengths, the 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 SW spectrum as above. The first case is with a fixed magnetic temperature (the temperature of the spin thermostat) of $T_m = 100$ K and a varying phonon temperature, $T_p$ (see blue squares in Fig. 7) given by the value on the $x$ axis. The second case has a fixed phonon temperature, $T_p = 100$ K, 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 and phononic temperature are equal, $T_m = T_p$ (see green circles in Fig. 7, same in Fig. 5(d)]. The error bars are estimated from the error in the nonlinear 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 to verify the intrinsic MPC for other materials, and validate or expand the present explanation of the SSE.

V. 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 nontrivial 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 $T_c$ 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 FMR 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 stiffness [53]. Natural extensions include a fully phonon mode and wave-vector-dependent formalism to explore in more detail MPC from first principles.

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Hamiltonian explicitly for each set of interactions: a two-component disordered alloy system we first write the Hamiltonian Eq. (1) assuming that the site and species exchange constants are weighted by the site occupancy, \( \rho, \eta \), which are the species. To describe the SW energies of terms beyond first order, and note that the pure powers of \( S \) only add an arbitrary constant to the Hamiltonian. Furthermore, we can relabel the sum over \( j \) by introducing the \( \delta \) vector, which is the translation between \( i \) and a neighbor, \( j \), so \( \sum_j \rightarrow \sum_{i,j} \). For a disordered alloy with only one type of site, we have \( \bar{J}_i = \bar{J}_{i,j} = \bar{J} \) depending on the relative displacement only. Thus we can write the Hamiltonian:

\[
\mathcal{H} = \sum_{i,k} \bar{\mathcal{J}}_{00} [a_i^{\dagger} a_{i + \delta}^{\dagger} + a_{i + \delta} + a_i + a_{i + \delta}^{\dagger} a_i^{\dagger}]
\]

We can then transform to Fourier space through \( a_i^{\dagger} a_{- k}^{\dagger} = \sum_k e^{i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} \). Substituting these into Eq. (A5) gives (for readability we write just the first term, \( \rho = \eta = 0 \))

\[
\mathcal{H}^{(0)} = \sum_{i,k} \bar{\mathcal{J}}_{00} \left[ e^{- i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} + e^{i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} + e^{- i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} + e^{i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} \right].
\]

Sums over \( i \) cause all terms to vanish unless \( k = k' \) [54] and thus the sum becomes

\[
\mathcal{H}^{(0)} = \sum_{k} \bar{\mathcal{J}}_{00} \left[ e^{- i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} + e^{i k \cdot \delta} a_k^{\dagger} a_k^{\dagger} - 2 a_k^{\dagger} a_k^{\dagger} \right].
\]

(A7)

We use the identity \( [a_i^{\dagger}, a_i] = 1 \) and define \( \gamma_k = \sum_{\delta} e^{- i k \cdot \delta} \) and \( \bar{J}_k = \sum_{k'} \bar{\mathcal{J}} e^{- i k \cdot \delta} \). For crystals with a center of inversion symmetry (fcc, bcc), \( \gamma_k = \gamma_{-k} \), simplifying Eq. (A7) to

\[
\mathcal{H}^{(0)} = \sum_{k} \bar{\mathcal{J}}_{k0} [\gamma_k a_k^{\dagger} a_k^{\dagger} + \gamma_k a_k^{\dagger} a_k^{\dagger} - 2 a_k^{\dagger} a_k^{\dagger}].
\]

(A8)

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

\[
\mathcal{H}^{(0)} = 2 \sum_k (\bar{\mathcal{J}}_{k0} - \bar{\mathcal{J}}_{00}) a_k^{\dagger} a_k^{\dagger}.
\]

(A9)

Terms for \( \mathcal{H}^{(0)} \), \( \mathcal{H}^{(1)} \), and \( \mathcal{H}^{(1)} \) 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:

\[
\mathcal{H} = \sum_k a_k^{\dagger} T M a_k^{\dagger},
\]

(A10)
where $M$ is a $2 \times 2$ matrix containing the appropriate exchange constants and

$$a_k^\pm = \begin{bmatrix} a_k^\pm 0 \\ a_k^\pm 1 \end{bmatrix}. \quad \text{(A11)}$$

The Hamiltonian matrix is rewritten using a Bogoliubov transformation [55], which now mixes the excitations associated with Ni-Ni, Fe-Fe, and Ni-Fe exchange interactions:

$$a_k^0 = u_k a_k^0 + v_k a_k^1,$$

$$a_k^1 = u_k a_k^0 + v_k a_k^1. \quad \text{(A12)}$$

With this transform the Hamiltonian can then be written as

$$\mathcal{H} = \sum_k [a_k^+ \beta_k^+] M_k [a_k^0 \beta_k^0]. \quad \text{(A13)}$$

The elements of the matrix $M_k$ can be found by comparing coefficients of the $a_k^k$:

$$M_k = \begin{bmatrix} \Omega_k(0 S_0) + \Xi_k(0 S_1) \\ \Xi_k(0 S_1) \Omega_k(1 S_1) + \Xi_k(0 S_1) \end{bmatrix}, \quad \text{(A14)}$$

where $\Omega_k = \frac{1}{\mu^2} \sum_R \bar{\gamma}^{pp}(R)[1 - \exp(i k \cdot R)]$ is the SW frequency of the individual species (the two possible sublattices). $\gamma$ is the gyromagnetic ratio, $\mu$ are the spin moment amplitudes, and $\Xi_k(0 S_1) = \frac{1}{\mu^2} \sum_R \bar{J}^{pp}(R)(S_0)$. ($\cdot$) represents the equilibrium value of reduced magnetisations for each species (normalised to 1 at $T = 0$ K). Here we ignore spin fluctuations and these values are fixed to 1. We could artificially introduce a temperature dependence of the magnetization in the LSWT but we choose not to as the ASD gives a better account of spin fluctuations as it allows for SW interactions.

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

$$\omega_k(k) = \frac{1}{2} \left[ \text{Tr} M_k \pm \sqrt{\text{Tr}(M_k)^2 - 4 \det(M_k)} \right]. \quad \text{(A15)}$$

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 Figs. 5(a) and 5(c) and commented in the main text.