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Magnetization Dynamics in Fe\textsubscript{x}Co\textsubscript{1−x} in Presence of Chemical Disorder

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Abstract: In this paper, we present a theoretical formulation of magnetization dynamics in disordered binary alloys, based on the Kubo linear response theory, interfaced with a seamless combination of three approaches: density functional-based tight-binding linear muffin-tin orbitals, generalized recursion and augmented space formalism. We applied this method to study the magnetization dynamics in chemically disordered Fe\textsubscript{x}Co\textsubscript{1−x} (\(x = 0.2, 0.5, 0.8\)) alloys. We found that the magnon energies decreased with an increase in Co concentration. Significant magnon softening was observed in Fe\textsubscript{20}Co\textsubscript{80} at the Brillouin zone boundary. Magnon–electron scattering increased with increasing Co content, which in turn modified the hybridization between the Fe and Co atoms. This reduced the exchange energy between the atoms and softened down the magnon energy. The lowest magnon lifetime was found in Fe\textsubscript{50}Co\textsubscript{50}, where disorder was at a maximum. This clearly indicated that the damping of magnon energies in Fe\textsubscript{x}Co\textsubscript{1−x} was governed by hybridization between Fe and Co, whereas the magnon lifetime was controlled by disorder configuration. Our atomistic spin dynamics simulations show reasonable agreement with our theoretical approach in magnon dispersion for different alloy compositions.

Keywords: chemical disorder; magnetization dynamics; alloy

1. Introduction

The dynamics and damping of magnetic excitations play a pivotal role in many modern day spintronic devices through the exploration of the nature of magnon dispersion and spin transport in pristine metals, semiconductors and their alloys. Band theory of ferromagnetism based on itinerant electrons has successfully predicted the magnetic properties of metallic systems, including transition metals, Heusler alloys and rare-earth magnets. However, these theoretical investigations have been a controversial subject for modern science due to the dual character of the d-electrons. Their ground state is described by the band-like itinerant electrons at \(T = 0\). However, many open questions arise regarding the more general finite temperature ab initio approach, which includes spin excitation [1,2]. The spin wave theory of magnetism includes the fluctuations that switch from band theory of ferromagnetism to the spin dynamics approach [3–7].

A first principles study of the spin correlation function and magnon lifetimes of disordered magnetic systems has become important for both fundamental and technological interests. Recent advancements in experimental techniques have enabled us to probe magnon dynamics [8–15]. In a recent experiment using the spin-polarized electron energy loss spectroscopy (SPEELS) technique, it was shown that magnon energies were reduced (referred to as magnon softening) for a single ferromagnetic monolayer of Fe on W, compared with pure bulk Fe [16]. Theoretical predictions based on an itinerant electron model are in contradiction with the above experimental finding. Theoretical limitations have hindered a correct understanding of the fundamental nature of spin-wave excitations. The...
structural relaxation between the Fe and W layers influences the hybridization between Fe and W states, which modifies the exchange interactions [17]. Disordered local moments (DLM) configuration reduces the exchange interaction strengths. This leads to significant magnon softening compared with bulk Fe. The magnetic ordering from Dzyaloshinskii–Moriya interactions (DMI), as a consequence of spin–orbit coupling, also leads to magnon softening. For the bulk system, these effects can be ignored.

Continuum and atomistic models have been developed to study spin dynamics in real materials. In micromagnetic simulations, a continuum model of magnetization is considered at a length scale much bigger than the interatomic distance [18]. Regarding atomistic models based on first principles methods, one may directly solve the time-dependent density functional equation, which requires huge computational cost and time to make realistic predictions for materials. The standard approach used to simulate the time evolution of the spin texture is to propagate the Landau–Lifshitz–Gilbert (LLG) equation [19,20]. However, the Hamiltonian required for solving the LLG equation contains several terms involving magnetic exchange and anisotropy. It should be mentioned that for a disordered magnetic alloy, the exchange interaction between atoms may strongly depend on the chemical composition [21–24]. Therefore, the magnon spectrum and lifetimes in a material depend on the configuration of the magnetic state via the local atomic environment [25]. Another important parameter in the LLG equation is the Gilbert damping parameter, which can be calculated by first principles electronic structure calculations. Two successful models in this regard are the breathing Fermi surface model (BFS) [26] and the torque correlation model (TCM) [27]. Unfortunately, neither of these models are parameter-free, nor do they really shed light on the microscopic origins of damping. Finally, it was found that damping arose from microscopic scattering processes [28–30]. Ebert et al. calculated the Gilbert damping parameter for the bcc Fe$_x$Co$_{1-x}$ alloy from a CPA approach via linear response theory [30]. However, the microscopic origins of damping is still being uncovered.

The dynamical relaxation of various complex systems have also been a focus of considerable interest. In particular, spin relaxation in transition metal-based alloys have received considerable attention over the years. The relaxation time for ferromagnetic transition metals such as Fe, Co and Ni is strongly spin-dependent. The spin-averaged relaxation time in these metals is much shorter than in noble metals. The analysis of the peak-positions and broadening of magnon excitation provides us information on the magnon energy and lifetime, respectively. Magnon energy decays exponentially, such as $\exp(-\gamma t/2\hbar)$, where $\gamma$ represents the intrinsic line width of the Lorentzian peak in magnon spectral density and $\hbar$ is the reduced Plank constant. The magnon lifetime $\tau = 2\hbar/\gamma$ is usually defined as the time in which the amplitude drops to $e^{-1}$ of its initial value. This is similar to the concept of a quantum mechanical broadening parameter and lifetime of an energy eigenstate [31–35].

The physical picture underlying the spin-dynamics method, which we present here, is not completely new, but their implementations in the presence of chemical and magnetic interactions within first principles accuracy are of fundamental importance. Augmented space formalism (ASF) has been successfully used to describe the effects of different disorders [36–46]. Here, we applied ASF for the simulation of spin dynamics, which enabled the characterization of magnetic excitations in the presence of chemical disorder within Kubo linear response formalism, in combination with a density functional-based first principles approach. The Hamiltonian parameters obtained from first principles theory provides a reliable tool for the analysis and even prediction of complex collective modes of magnetic materials. Our aim is to probe the magnon dynamics in chemically disordered Fe$_x$Co$_{1-x}$ alloys.

This article is organized as follows: In Section 2, we describe the Hamiltonian for spin dynamics for binary alloys in the presence of chemical disorder. We use the augmented space recursion (ASR) approach to tackle the disordered Hamiltonian, techniques for the calculation of the adiabatic magnon lifetime and an atomistic scheme for the numerical integration of the equations of motion. We present computational details to describe the
2. Methodology

2.1. Spin Transport at Low Temperatures

We analysed the spin transport in ferromagnetic alloys. We began with a ferromagnetic sea as our unperturbed state. A spin flip is an excitation or fluctuation in that state. We used the XXZ Heisenberg Hamiltonian with a random distribution of the exchange parameters described by spin waves or magnons that can be envisaged as spin patterns against the XXZ Heisenberg Hamiltonian with a random distribution of the exchange parameters.

The linear response to an external homogeneous disturbance was described in terms of two-particle Green’s functions within the Kubo linear response theory [47]. If a spin system is disturbed by an external field that causes a perturbation in the XXZ Heisenberg Hamiltonian, then the spin response function \( \Gamma(\vec{R} - \vec{R}', t - t') \) is related to the disturbance by a spin–spin correlation function \( C(\vec{R} - \vec{R}', t - t') \) [47–49], which is given by:

\[
\Gamma(\vec{R} - \vec{R}', t - t') = \frac{i}{\hbar} \Theta(t - t') \langle \Phi_0 | [\sigma(\vec{R}, t'), \sigma(\vec{R}, t)] | \Phi_0 \rangle
\]

where \( \Theta(t) \) is the Heaviside step function and \( | \Phi_0 > > \) is the ground state. The dynamical structure factor \( S(\vec{q}, \omega) \) is the Laplace transform of the spin correlation function \( C(\vec{R} - \vec{R}', t - t') \).

2.2. Recursive Approach to Dynamical Spin Response Functions

Our starting point was a dynamical variable described by the Hermitian operator \( D(\vec{q}, t) \), following the time evolution such that \( | \Phi(t) > = D(t) | \Phi_0 > \). Our guiding equation was the Kohn–Sham equation:

\[
i \frac{\partial | \Phi(t) >}{\partial t} = H | \Phi(t) >
\]

We followed the recursion procedure described by Gagliano and Balserio [50] and Viswanathan and Muller [48,49,51,52]. We first chose a denumerable basis of representation \( \{ | \phi_n > \} \), and expanded the “wave function” on this basis:

\[
| \Phi(t) > = \sum_{n=1}^{\infty} D_n(\vec{q}, t) | \phi_n >
\]

We began with \( | \phi_1 > = a^\dagger(\vec{q}) | \Phi_0 > \), where \( a^\dagger(\vec{q}) = \frac{1}{\sqrt{\mathcal{N}}} \sum_{\vec{R}} e^{-i\vec{q}\cdot\vec{R}} a^\dagger(\vec{R}) \). In the next step, \( | \phi_2 > = H | \phi_1 > - \alpha_1 | \phi_1 > \) and orthogonality leads to \( < \phi_1 | \phi_2 > = 0 \) \( \Rightarrow \alpha_1 = \frac{< \phi_1 | H | \phi_1 >}{< \phi_1 | \phi_1 >} \).

Finally, for \( n > 2 \)

\[
| \phi_{n+1} > = H | \phi_n > - \alpha_n | \phi_n > - \beta_n^2 | n - 1 >
\]
The dynamical structure factor is then

$$i \frac{\partial D_n(\vec{q}, t)}{\partial t} = D_{n-1}(\vec{q}, t) - \alpha_n D_n(\vec{q}, t) - \beta_n^2 D_{n+1}(\vec{q}, t)$$

Taking the Laplace transformation:

$$(z - \alpha_n)D_n(\vec{q}, z) - i\delta_n0 = D_{n-1}(\vec{q}, z) + \beta_n^2 D_{n+1}(\vec{q}, z)$$

where

$$D_0(\vec{q}, z) = \frac{i}{z - \alpha_1 - \frac{\beta_1^2}{z - \alpha_2 - \frac{\beta_2^2}{z - \alpha_3 \ldots}}}$$

The dynamical structure factor is then

$$S(\vec{q}, \omega) = \lim_{\delta \to 0} \text{Re} [D_0(\vec{q}, \omega + i\delta)]$$

where $z = \omega + i\delta$.

2.3. Dynamical Spin Response Functions in the Presence of Disorder: The Augmented Space Approach

Then, we introduced disorder in the exchange parameters $J(\vec{R} - \vec{R}')$ by a set of random variables $\{n_\vec{R}\}$.

The randomness can be introduced in the exchange parameter through a local, binary random variable $n_\vec{R}$ ($\vec{R}$ can be occupied by an A or B type of atom for binary alloys), taking the values 0 and 1 with probabilities $x$ and $y = 1 - x$, respectively:

$$J(\vec{R}) = J^{AA}(\vec{R})n_\vec{R}n_{\vec{R} + \vec{x}} + J^{BB}(\vec{R})(1 - n_\vec{R})(1 - n_{\vec{R} + \vec{x}}) + J^{AB}(\vec{R})\left(n_\vec{R}(1 - n_{\vec{R} + \vec{x}}) + (1 - n_\vec{R})n_{\vec{R} + \vec{x}}\right)$$

where $\vec{R}' = \vec{R} + \vec{x}$.

The augmented space method [53–57] replaces these random variables $\{n_\vec{R}\}$ by operators $\{N_\vec{R}\}$. The eigenvalues are the random numbers taken by the variables $\{n_\vec{R}\}$ and the spectral functions are the probability densities of the operator $\{N_\vec{R}\}$ [58]. The augmented space is an outer product of real space $\Phi_\vec{q}$ (lattice space) and configuration space $\{\varnothing\}$ (random space). Then, the configuration (disorder) averaged spin response function was given by:

$$\langle \Gamma(\vec{q}, z) \rangle = \langle \Phi_\vec{q} \otimes \{\varnothing\} | (z\hat{I} - \hat{H})^{-1} | \Phi_\vec{q} \otimes \{\varnothing\} \rangle$$

where $\hat{H}$ is the disordered Hamiltonian.

Now, the configuration-averaged dynamical operator in the disordered system can be expressed as a continued fraction of recursion coefficients $\{\tilde{\alpha}_n, \tilde{\beta}_n\}$, as:

$$\langle \langle D_0(\vec{q}, z) \rangle \rangle = \frac{i}{z - \tilde{\alpha}_1 - \frac{\tilde{\beta}_1^2}{z - \tilde{\alpha}_2 - \frac{\tilde{\beta}_2^2}{z - \tilde{\alpha}_3 \ldots}}}$$
Then, the configuration-averaged dynamical structure factor was obtained by:

\[ \ll S(\vec{q}, \omega) \gg = \lim_{\delta \to 0} \text{Re} \ll D_0(\vec{q}, \omega + i\delta) \gg \]

The self-energy \( \Sigma(\vec{q}, z) \), via the recursion method suggested by Viswanath and Müller [48,49,51,52], was given by:

\[
\Sigma(\vec{q}, z) = \frac{\beta_2^2}{z - \tilde{\alpha}_2} - \frac{\beta_3^2}{z - \tilde{\alpha}_3} - \cdots \frac{\beta_n^2}{z - \tilde{\alpha}_n} = \frac{1}{\tau(q)} T(\vec{q}, z)
\]

The terminator \( T(\vec{q}, z) \), which reflects all the singularities in the response function, was constructed from the calculated \( \{ \tilde{\alpha}_n, \tilde{\beta}_n \} \) for \( n = 1, \ldots, N \) [52,59–63]. Therefore, we used the square-root terminator [60,61] with the calculated recursion coefficients to take into account the convergence of the continued fraction. \( \text{Im}[\Sigma(\vec{q}, \omega)] \) counts the disorder-induced broadening, which provides the disorder-induced lifetime \( \tau \) of the magnon state by \( \text{Im}[\Sigma(\vec{q}, \omega)] = 1/\tau(q) \).

We studied the configuration-averaged dynamical response function of chemically disordered binary alloys, using ASR within Kubo linear response theory [53–57]. The ASR technique went beyond the usual single-site mean-field coherent potential approximation (CPA)-like approach to study the environmental effects for chemically disordered alloys. In particular, the methodology we implemented here was an admixture of the tight binding linear muffin-tin orbitals (TB-LMTO) technique, ASF and generalized recursion (GR) [48,49,51,64]. This provided an accurate computational framework for analysing dynamical response properties.

We calculated the configuration-averaged dynamical structure factors \( \ll S(\vec{q}, \omega) \gg \) of Fe\(_x\)Co\(_{1-x}\) for three different Co contents \((x = 0.2, 0.5, 0.8)\). This relates the response of the systems in the presence of an external perturbation (in our case, chemical disorder) from the self-energy. We also calculated the magnon lifetime \( \tau(q) \) of Fe\(_x\)Co\(_{1-x}\). The Brillouin zone boundary in the [001] direction of FeCo was nearly 2 Ångstrom\(^{-1}\). We neglected the effects of magnon–magnon and magnon–stoner interactions. We focused on the magnon–electron interactions to study the effects of chemical disorder on Fe\(_x\)Co\(_{1-x}\).

### 2.4. Atomistic Spin Dynamics

To compare our results, we also calculated the magnon dispersion spectra of Fe\(_x\)Co\(_{1-x}\) \((x = 0.2, 0.5, 0.8)\) from the Uppsala Atomistic Spin Dynamics (UppASD) code [6,7].

The necessary exchange parameters for UppASD were calculated in the framework of \textit{ab-initio} density functional theory (DFT). These were performed by means of the Korringa–Kohn–Rostocker Green’s function formalism, as implemented in the SPRKKR package [65]. The shape of the potential was treated by the atomic sphere approximation (ASA), whereas relativistic effects were considered by taking the fully relativistic Dirac equation. The effect of the exchange correlation part of the energy was treated by considering the generalized gradient approximation (GGA), as devised by Perdew, Burke and Ernzerhof [66]. Substitutional disorder between sub-lattices could also be studied by making the use of the CPA. Interatomic exchange interactions were calculated via the LKAG formalism [67].

The UppASD code [68] is based on the framework of LLG formalism. The temporal evolution of an atomic moment in LLG formalism is given by:

\[
\frac{d\vec{m}_i(t)}{dt} = -\frac{\gamma}{1 + \alpha} \vec{m}_i(t) \times \left[ \mathcal{H}_{\text{eff}}^i + \frac{\alpha}{m_s} \left( \vec{m}_i(t) \times \mathcal{H}_{\text{eff}}^i \right) \right]
\]

where \( \vec{m}_i(t) \) is the atomic moment on the \( i \)th site at time \( t \). \( \gamma \) is the gyromagnetic ratio and \( \alpha \) is the Gilbert damping factor, which we have assumed to be \( 3 \times 10^{-4} \). \( m_s \) is the saturation moment for the \( i \)th atom. The temperature effect in ASD was measured using a stochastic
magnetic field, included in the term of effective field $H_{\text{eff}}$. The effective field $H_{\text{eff}}^i$ on the $i$'th atom is calculated from the effective magnetic Hamiltonian given by

$$H_{\text{Mag}} = H_{\text{ex}}$$

through

$$H_{\text{eff}}^i = -\frac{\partial H_{\text{Mag}}}{\partial m_i(t)}$$

$H_{\text{ex}}$ describes the magnetic exchange interactions between the spins. $H_{\text{Mag}}$ is given by

$$H_{\text{Mag}} = \sum_{ij} J_{ij} S_i \cdot S_j$$

In order to calculate magnon dispersion relation, one should calculate spin–spin correlation function. The spin–spin correlation in an effective field is obtained by solving the LLG equation and can be written as:

$$C(R - R', t) = \langle m_R(t)m_{R'}(0) \rangle - \langle m_R(t) \rangle \langle m_{R'}(0) \rangle$$

where the ensemble average is represented in angular brackets and $k$ is the Cartesian component. The Fourier transform of the spin–spin correlation function, known as the dynamical structure factor, is written as:

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

$S(\vec{q}, \omega)$ is measured in neutron-scattering experiments.

3. Results and Discussion

Before studying the magnetization dynamics in Fe$_x$Co$_{1-x}$, we investigated the charge distribution of a body-centered, disordered Fe$_{50}$Co$_{50}$ alloy. We compared the disordered alloy with a corresponding ordered, B2-structured alloy. In Table 1, we show the chemical effects of disorder and the redistribution of charge upon alloying. In these alloys, there was a small decrease of $sp$-like charges and a corresponding small increase in the $d$-like channel in both Fe and Co. These effects were small, with the introduction of disorder. The magnetocrystalline anisotropy is the energy difference between the magnetic ground state, when the magnetic quantization (spin) axis is along the easy axis (which is along the c-axis for FeCo) and along the easy plane (which is the $ab$-plane for FeCo), i.e., $K^e = E^e - E^\parallel$. Bulk FeCo has $\zeta \sim 1$ without any tetragonal distortion. Therefore, we ignored the effects of single-ion anisotropy in FeCo alloys.

Table 1. Charge redistribution upon alloying: distribution for the ordered B2 and disordered body-centered cubic (BCC) structures for the 50-50 FeCo alloy. The charge is given in units of electronic charge $e$.

| Alloy Component | Fe | Fe$_{0.5}$Co$_{0.5}$ | Co |
|-----------------|----|---------------------|----|
| Atomic radius   | $R_0 = 2.64$ Å | $R_0 = 2.60$ Å     |    |
| Charge          | $sp$ | $d$ | Tot | $sp$ | $d$ | Tot |
| Atomic state    | 2.0  | 6.0 | 8.0 | 2.0  | 7.0 | 9.0  |
| B2 ordered      | 1.44 | 6.52 | 7.96 | 1.46 | 7.58 | 9.04 |
| BCC disordered  | 1.43 | 6.55 | 7.99 | 1.43 | 7.63 | 9.06 |
We were interested in calculating the ensemble-averaged dynamical structure factor \( \langle S(\vec{q}, \omega) \rangle \) to understand the magnetization dynamics in disordered Fe\(_x\)Co\(_{1-x}\) alloys. Figure 1a–c shows the momentum-resolved dynamical structure function \( \langle S(\vec{q}, \omega) \rangle \), convoluted with a Lorenzian function with three disorder concentrations (\( x = 0.2, 0.5, 0.8 \)) along the symmetric \( \Gamma - H \) direction. Irrespective of the disorder concentration, for low \( \vec{q} \) values, the spectral intensity, estimated from the peak value of the Lorenztian, was high. The spectral intensity dropped to minimum values for intermediate \( \vec{q} \), after which it again rose with increasing \( \vec{q} \). At the same time, the peak width monotonically increased with increasing energy, as well as wave vectors (\( \vec{q} \)). The broadening of the peak in \( \langle S(\vec{q}, \omega) \rangle \) for larger \( \vec{q} \) values came from a magnon–electron scattering mechanism due to chemical impurity. The itinerant collective excitations of spin waves (magnon) lost their energy as a consequence of chemical disorder. A large damping of magnons in the tetragonally distorted bulk FeCo compound was also previously predicted [69,70].

![Figure 1](image1.png)

**Figure 1.** The dynamical structure factors \( \langle S(\vec{q}, \omega) \rangle \) for a selection of wave vectors \( \vec{q} \), varying from 0.1 to 0.9 along the symmetric \( \Gamma - H \) direction in the Brillouin zone of bcc Fe\(_x\)Co\(_{1-x}\) alloys with (a) \( x = 0.2 \), (b) \( x = 0.5 \) and (c) \( x = 0.8 \), respectively. In order to identify the peak position, the dynamical structure function \( \langle S(\vec{q}, \omega) \rangle \) was convoluted with a Lorentzian function, normalized to unity.

Here, we elaborate on the technique of peak finding from \( \langle S(\vec{q}, \omega) \rangle \). After configuration averaging, in order to identify the position of the intensity peaks of the dynamical structure factor \( \langle S(\vec{q}, \omega) \rangle \), we convoluted the respective intensity profiles of \( \langle S(\vec{q}, \omega) \rangle \) for each \( \vec{q} \) vector with a Lorentzian function, normalized to unity. In order to justify this technique, we applied it to calculate magnon dispersion of the bulk bcc Fe (see Figure 2a). This was to show that the implemented technique was able to qualitatively predict our experimental findings.

Then, we employed a different technique for the chemically disordered Fe\(_x\)Co\(_{1-x}\) alloys. We calculated the magnon dispersion of disordered Fe\(_x\)Co\(_{1-x}\) alloys along the \( \Gamma - H \) direction, and studied the effects of chemical disorder on it. We found quadratic dependence upon \( \vec{q} \) for the spin wave spectrum of magnon, as shown in Figure 2b. Bulk Fe had a steeper magnon dispersion compared with the disordered structure. This was expected due to the absence of magnon–electron scattering in the disorder-free material. The interesting point to note here is that the steepness of magnon energy gradually decreased with increasing Co concentration. Fe\(_{80}\)Co\(_{20}\) exhibited higher magnon velocity for weaker scattering effects, and vice versa.
Figure 2. (a) Magnon dispersion spectrum for bulk bcc Fe along the \( \Gamma - Y \) direction from ASR. For comparison, the experimental spin-wave spectrum of bulk bcc Fe by Lynn (Ref. [71]) and data from another experimental study corresponding to a spin-wave stiffness constant of \( D = 280 \text{ Å}^2 \) by Mook et al. (Ref. [72]) are displayed. The blue triangles represent theoretical data by Halilov et al. (Ref. [73]). All comparisons were consistent with the data from ASR [72]. (b) Magnon dispersion spectrum for bulk bcc Fe\(_{20}\)Co\(_{80}\) alloys along the high symmetric \( \Gamma - H \) direction in the Brillouin zone for three different Co concentrations from ASR. This shows magnon softening in the Fe\(_{20}\)Co\(_{80}\) alloy, compared with the others. (c) Magnon dispersion spectra for disordered Fe\(_{20}\)Co\(_{80}\) from ASR and ASD.

The magnon softening is clearly visible in Fe\(_{20}\)Co\(_{80}\) at the zone boundary. This is due to the fact that magnon–electron scattering modifies the exchange energy between Fe and Co moments. The chemical disorder between the Fe and Co atoms influenced the hybridization between Fe and Co states, which reduced the exchange interactions. This is shown in Table 2. The strength of exchange interaction for Fe\(_{x}\)Co\(_{1-x}\) gradually decreased with increasing Co content. This clearly indicates the softening of Fe\(_{x}\)Co\(_{1-x}\) with increasing Co content. The chemical disorder between Fe and Co also increased damping, which enhanced magnon softening. Ebert et al. calculated the Gilbert damping parameter from first principles theory for bcc Fe\(_{x}\)Co\(_{1-x}\) alloys [30]. Damping was at a minimum for 20\% Co content and gradually increased with increasing Co content. This led to strong magnon softening in the Fe\(_{20}\)Co\(_{80}\) alloy. Our results are also consistent with an experiment by Oogane et al. on bcc Fe\(_{x}\)Co\(_{1-x}\) alloys [74].

Table 2. The calculated nearest-neighbour exchange parameters in the framework of ab-initio density functional theory using the Korringa–Kohn–Rostocker Green’s function formalism, implemented in the SPRKKR package [65].

| Alloy     | Fe-Fe (meV) | Fe-Co (meV) | Co-Co (meV) |
|-----------|-------------|-------------|-------------|
| Fe\(_{80}\)Co\(_{20}\) | 2.065       | 2.302       | 1.740       |
| Fe\(_{50}\)Co\(_{50}\) | 2.083       | 2.117       | 1.418       |
| Fe\(_{20}\)Co\(_{80}\) | 1.919       | 1.880       | 1.208       |

For completeness, we also computed the magnon dispersion spectrum along the \( \Gamma - H \) direction in the Brillouin zone. We compared the magnon spectra of disordered Fe\(_{20}\)Co\(_{80}\) between ASR and ASD, taking the first nearest-neighbour exchange interaction into consideration (as shown in Figure 2c). The ASD simulations were performed with a system size of 120 \( \times \) 120 \( \times \) 120 at a temperature of 0.1 K. ASD followed similar characteristic behavior with increasing Co concentrations. It was observed that the softening became stronger near the zone boundary (higher \( \vec{q} \) values). The steepness of dispersion reduced as magnon–electron scattering increased. It was also observed in the profile of \( \ll S(\vec{q}, \omega) \gg \) in Figure 1a. Both the numerical techniques exhibited the same effect.

To justify our results further, we performed ASD simulations for the nearest neighbour (Figure 3a–c) and 12 neighbor exchange interactions (Figure 3d–f). The qualitative features in the magnon spectra for the first and 12 neighbor shells were similar. The softening of the magnon spectra for the Fe\(_{20}\)Co\(_{80}\) alloy was similar to that obtained from the ASR calculations as shown in Figure 2c. Additionally, we observed some branching in dispersion along \( H - N \) for higher Co concentrations. A stronger tendency for branching was visible...
at the N point of the Brillouin zone. However, we focused only on the magnon softening in $\Gamma - H$ direction. This strong magnon softening was in contradiction with a previous study based on an itinerant electron model at $T = 0$ K [75,76]. Our findings present the possibility that a disordered Fe$_x$Co$_{1-x}$ may not be a simple itinerant ferromagnet. The effects of spin correlations are important for this system. This behaviour is commonly interpreted as a transition from conductivity-like behaviour. This reflects the dominance of intra- and interband transitions. This is related to the broadening of electron energy bands caused by an increase in scattering events with concentrations of Co. An increase in the Co content in Fe$_x$Co$_{1-x}$ led to more impurity-scattering. This was responsible for band broadening, which increased the damping of magnon. This effect completely suppressed the conductivity-like behaviour in the low-temperature regime, and increased scattering due to chemical disorder.

The broadening of the magnon excitation peak ($\ll S(\vec{q}, \omega)$) provides a way to calculate the magnon lifetime $\tau$. The Fourier transform of the Lorentzian in the energy (or frequency) domain is an exponential decay of magnon lifetime, such as $\exp(-\Gamma/2\hbar)$, where $\Gamma$ represents the intrinsic linewidth of the Lorentzian peak in energy and $\hbar$ is the reduced Planck constant. The relaxing magnetic modes or patterns are labelled by $\vec{q}$, such that the average ‘size’ of the mode is $O(q^{-1})$. We obtained the magnon lifetimes from the Fourier transform of the configuration-averaged correlation function. The large broadening of $\ll S(\vec{q}, \omega)$ yielded a small relaxation time, which clearly indicated strong scattering. Figure 3g shows the lifetime $\tau$ for Fe$_x$Co$_{1-x}$ ($x = 0.8, 0.5, 0.2$) as a function of $\vec{q}$ (Angstrom$^{-1}$). We found the minimum magnon lifetime in Fe$_{50}$Co$_{50}$. This confirmed that the disorder was at a maximum in 50-50 configurations where $\ll S(\vec{q}, \omega)$ became maximally broadened for higher $\vec{q}$ values (see Figure 1b). Therefore, the magnon lifetime was not governed by hybridization between Fe and Co, but rather controlled by disorder configuration.

![Figure 3](image)

Figure 3. The figures in the top row (a–c) show the magnon dispersion of Fe$_x$Co$_{1-x}$ (where $x = 0.2$, 0.5 and 0.8) alloys with nearest-neighbour exchange parameters and the bottom row figures show the plots (d–f) with the 12 neighbour shell exchange parameters. (g) Magnon lifetimes for disordered Fe$_x$Co$_{1-x}$ alloys for $x = 0.2$, 0.5 and 0.8 from ASR.

In the current study, we propose using the ASR technique to study magnetization dynamics in chemically disordered alloys (which could also be generalised to surfaces and interfaces) and compare calculated magnon energy spectra with ASD-simulated magnon spectra. There are two different approaches within first principles theory for studying magnetization dynamics in materials: ASR and ASD. These approaches are based on two different working principles. ASD is based on the LLG theory, whereas the ASR based on the Kubo linear response theory. ASR captures the effect of hybridization between Fe and Co on the magnon softening of Fe$_{20}$Co$_{80}$ alloys (see Figure 2c), as well as magnon lifetimes in chemically disordered alloys originating from magnon band broadening (see Figure 3g).

4. Conclusions

In conclusion, we have investigated magnetization dynamics in the presence of chemical disorder using a first principles TB-LMTO-ASR method, interfaced with Kubo linear
response theory. This serves as a general and powerful computational tool for studying the effect of chemical disorder on magnetic excitations in binary random alloys. We applied our technique to study the magnetization dynamics in disordered $\mathrm{Fe}_x\mathrm{Co}_{1-x}$ alloys, which are of great fundamental and technological interest [77–80]. We showed that the magnon–electron scattering mechanism plays a crucial role in determining the exchange interaction between Fe and Co. Magnon softening increases with Co concentration as the exchange interaction decreases. Our findings were further supported by atomistic spin dynamics simulations. Moreover, we calculated magnon lifetimes from the broadening widths of dynamical structure factors. It was found that the maximally disordered alloy showed the smallest lifetime. Therefore, magnon softening depends on the hybridization between Fe and Co, whereas the magnon lifetime depends on the disorder configuration. The proposed formalism could be used to study the magnetization dynamics in any binary random alloy.

Author Contributions: Conceptualization, A.M. (The project was originally conceived by Abhijit Mookerjee (A.M.) and B.S. (Banasree Sadhukhan)); methodology, A.M. and B.S. (Banasree Sadhukhan); validation, B.S. (Banasree Sadhukhan), R.C. and B.S. (Biplab Sanyal); formal analysis, B.S. (Banasree Sadhukhan); investigation, B.S. (Banasree Sadhukhan); resources, B.S. (Banasree Sadhukhan); data curation, B.S. (Banasree Sadhukhan); writing—original draft preparation, B.S. (Banasree Sadhukhan); writing—review and editing, B.S. (Banasree Sadhukhan) and B.S. (Biplab Sanyal); visualization, B.S. (Banasree Sadhukhan); supervision, A.M. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: The data supporting for the present study are available from the corresponding author upon request.

Conflicts of Interest: The authors declare no conflict of interest.

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