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Magnetoelastic fingerprints of metamagnetism in FeRh

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Abstract

Recently, there has been renewed interest in the magnetism of FeRh that extends well beyond the extensively studied, yet not fully understood, antiferromagnetic–ferromagnetic transition. Despite its popularity, fundamental magnetic properties—including magnetocrystalline anisotropy and magnetoelasticity—remain insufficiently characterized from both theoretical and experimental studies. In this work, we first used Density Functional Theory to analyze these properties in single crystal FeRh for both antiferromagnetic and ferromagnetic phases at zero temperature, revealing significant qualitative and quantitative differences between them. We then attempted to develop a magnetic interatomic potential for a classical spin-lattice model, coupling spin dynamics and molecular dynamics, that reproduces the magnetoelastic phenomenology obtained from first principles calculations. Our results constitute a step towards a better understanding of the complex magnetoelasticity of FeRh and open the door

to more accurate large-scale atomistic modeling of similar materials undergoing antiferromagnetic-ferromagnetic first-order phase transitions.

Keywords: Density Functional Theory, magnetoelasticity, spin-lattice model

1 Introduction

More than a century after the advent of quantum mechanics, the intricate interactions between electrons and phonons in solids continue to produce a remarkable diversity of phenomena that drive modern condensed-matter research. Contemporary efforts to probe phase transitions in functional materials have uncovered a broad spectrum of emergent effects with substantial technological and commercial relevance, ranging from transport control systems to thermal energy conversion and storage applications. Of particular significance are phase transitions in magnetic materials, where the strong coupling among charge, lattice, and spin degrees of freedom can lead to first-order phase transitions (FOPTs). These magnetostructural phase transitions arise from the pronounced interplay between the crystal lattice and the magnetic moments of electrons [1], resulting in concurrent magnetic and structural transformations that can be induced by variations in temperature, applied magnetic field, or mechanical strain. Such coupled transitions frequently generate amplified functional responses of technological importance, including colossal magnetoresistance [2], the giant magnetocaloric effect [3], and giant magnetostriction [4].

Among the relatively small class of magnetostructural materials, the chemically ordered intermetallic compound FeRh, around the equi-atomic $\text{Fe}_{50}\text{Rh}_{50}$ composition, stands out due to its structural simplicity, high symmetry, and a FOPT temperature close to or above room temperature, which together make it an ideal platform for both fundamental studies and practical engineering. Interest in FeRh magnetism has recently been revitalized, extending far beyond the actively investigated antiferromagnetic (AF) to ferromagnetic (FM) transition occurring near 320–370 K [5]. This renewed attention stems from the diverse pathways available to manipulate the transition—through magnetic fields, mechanical strain, or ultrafast optical excitation [6–9]—as well as from the complex underlying physics involving hysteresis, phase coexistence, kinetic effects, and disorder [10–12]. Distinct from many other magnetocaloric or magnetostrictive systems, FeRh exhibits a readily accessible first-order AF→FM metamagnetic transition accompanied by a measurable lattice expansion of approximately 1% and a pronounced magnetoelastic coupling. Despite the interest and numerous studies on this material, there is still a lack of both theoretical and experimental data on some relevant magnetic properties such as magnetocrystalline anisotropy (MCA) and magnetoelasticity (ME) [13–15].

Many models have been proposed to try to explain the fundamental behaviour of the FOPT in FeRh. One of the first attempts was the exchange inversion model suggested by Kittel [16], which failed to account for the large entropy change in the AF-FM phase transition and the elastic properties [17]. A more advanced model was proposed by Gruner *et al.* [17], based on the Blume-Capel Hamiltonian [18, 19], explaining the

phase transition through the instability of the rhodium magnetic moment using Monte Carlo simulations. Gruner’s model was successfully used by Maat *et al.* to describe the field dependence of the shift in transition temperature [20]. An important step towards understanding the magnetic interactions of FeRh was achieved by Mryasov [21] by means of first-principles calculations, which were used later by Barker *et al.* to implement a spin model for FeRh including effective higher-order, non-Heisenberg exchange interactions capable to describe the phase transition using atomistic spin dynamics (ASD)[22]. Recently, Barker approach has been successfully used to study the thickness dependence of phase transition and magnetic properties of FeRh thin films [13, 23]. We should also highlight the Landau-Heisenberg Hamiltonian model for FeRh developed by Derlet [24], where longitudinal and transverse moment fluctuations are considered for both the Fe and Rh atoms.

In these previous works, anisotropic ME was not studied, while MCA was only included in the model by McGrath *et al.* [13]. Therefore, in the present work we first study MCA and ME of single crystal FeRh for both AF and FM phases using Density Functional Theory (DFT), then we develop a coarse-grained spin-lattice model, coupling spin dynamics and molecular dynamics (SD-MD) [25], that reproduces the DFT results.

2 First principles calculations

In this section, we describe electronic structure calculations of FeRh that have been performed using VASP software [26–28]. VASP is a plane-wave (PW) basis set implementation of DFT within the Projector Augmented Wave (PAW) approximation [29]. Calculations were done using the generalized gradient approximation of Perdew, Burke, and Ernzerhof (PBE) [30] to the exchange-correlation part of the energy functional. We used PAW PBE potentials version 5.4 with 14 and 15 valence electrons for Fe and Rh, respectively. The size of the PW basis set was determined by an energy cut-off of 470 eV. The reciprocal space was sampled with a uniform mesh of k-points [31] with a separation between the k-points of $0.1 \text{ \AA}^{-1}$. For the total energy summation, we used the Methfessel-Paxton method [32], with a smearing width of 0.1 eV and total energy convergence per formula unit was set to 10^{-7} eV.

2.1 Isotropic magnetoelasticity

The spontaneous volume magnetostriction ω_s at zero-temperature of a magnetic material with ordered magnetic configurations (AF or FM) is defined as:

$$\omega_s^{AF(FM)} = \frac{V_{AF(FM)} - V_{PM}}{V_{PM}}, \quad (1)$$

where $V_{AF(FM)}$ is the equilibrium volume of the AF or FM phases, and V_{PM} is the equilibrium volume of the paramagnetic phase at $T = 0$ K.

To calculate the total energies of the AF and FM phases we used cubic unit cells, see Fig.1. For the FM configuration the unit cell has the Pm-3m symmetry (space group No. 221), with one Fe atom at the Wyckoff site 1a, and one Rh atom at site 1b.

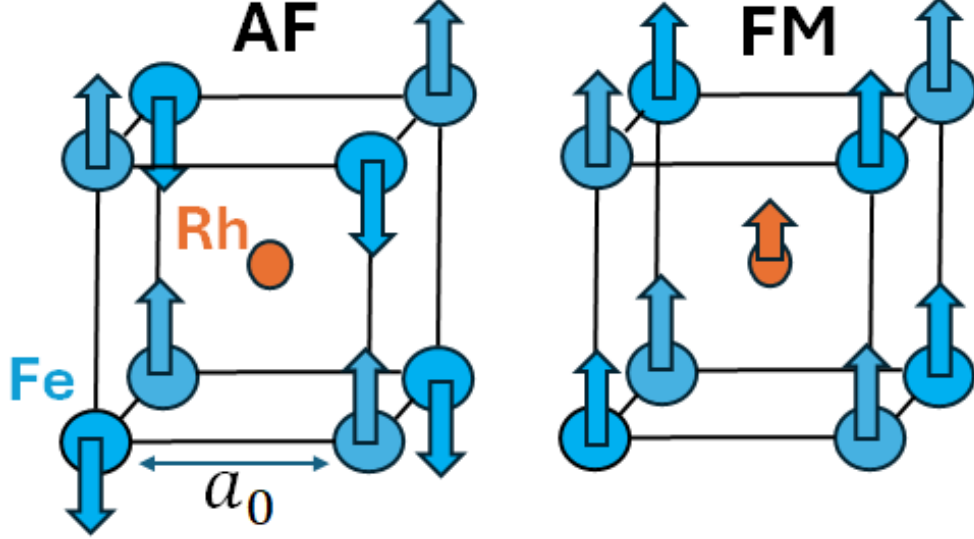


Fig. 1 Unit cell of FeRh with AF (left) and FM (right) magnetic order.

For the AF phase we used a $2 \times 2 \times 2$ supercell of the FM cubic cell with the Fm-3m symmetry (space group No. 225), with four Fe atoms (spin up) at the Wyckoff position 4a, four Fe atoms (spin down) at Wyckoff position 4b, and 8 Rh atoms at site 8c.

The volume of the PM phase was estimated using supercells with a quasi-random distribution of collinear spins within the Special Quasi-random Structure (SQS) approach [33]. We generated two sets of the SQS supercells containing 32 atoms and one $4 \times 4 \times 4$ (128 atoms) SQS using the implementation of the Monte-Carlo (MC) simulated annealing method implemented in the *mcsqs* code [34]. For the first set quasi-random distributions of 16 Fe and 16 Rh atoms with spins aligned along the quantization axis, and 16 Fe and 16 Rh atoms with spins aligned oppositely to the direction of the quantization axis were generated. While the initial spin distribution of the Fe atoms is preserved during calculations, the final distribution of the Rh atoms is different from the initially generated one, and is determined, most likely, by the spin arrangement of nearby Fe atoms. For the second set of the SQS configurations, a quasi-random distribution of only Fe atoms was generated. The equilibrium volume of each SQS supercell was obtained by performing a set of volume calculations and fitting the energy versus volume data $E = E(V)$ to the Rose-Vinet equation of state (EOS) [35]. In Fig.2, we show the results of total energy DFT calculations $E_0 = E_0(V)$ for the FM, AF, and PM (modeled as an SQS $4 \times 4 \times 4$ supercell). The average equilibrium volume of the PM phase modeled by 32 atoms SQS supercells is $\langle V_{SQS} \rangle = 26.971 \text{ \AA}^3$ in agreement with the equilibrium volume of the 128 atoms SQS. From these estimated equilibrium volumes, we obtain the following values for the spontaneous volume magnetostriction: $\omega_s^{AF} = -7.6 \times 10^{-3}$ and $\omega_s^{FM} = 9.1 \times 10^{-3}$. These results are in good agreement with those extrapolated from the experimental data obtained by neutron diffraction. Fig.3 shows the temperature dependence of the experimental ω_s and the

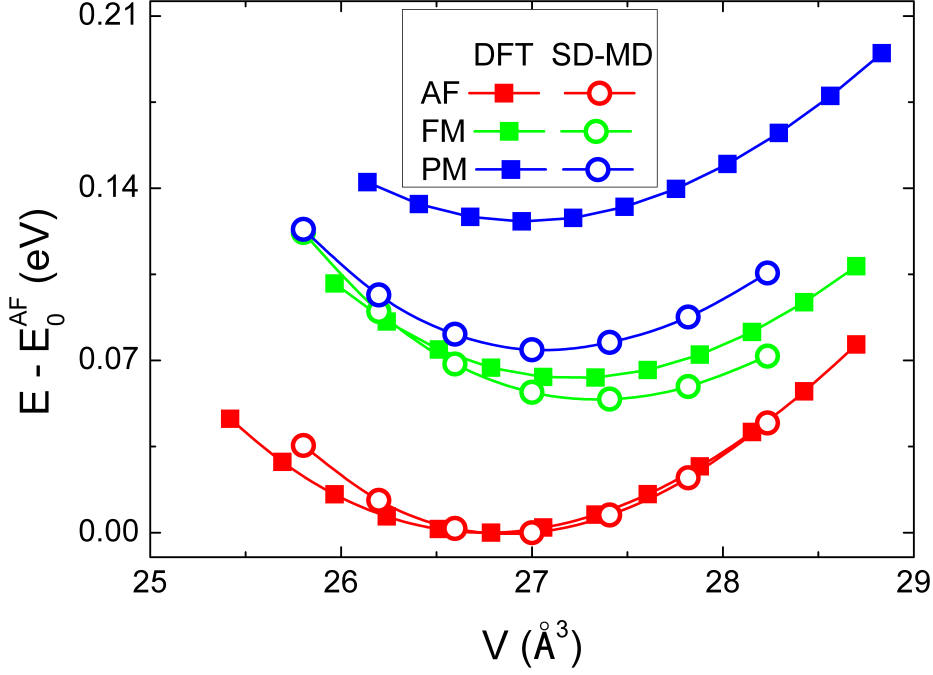


Fig. 2 Calculated total energy $E_0 = E_0(V)$ (per formula unit) of simulated AF (red color), FM (green color), and PM (blue color) phases using DFT (squares) and SD-MD (circles).

corresponding extrapolation to 0 K for both the FM and AF phases. According to the mean-field estimation, the extrapolated values at 0 K are $\omega_s^{\text{AF}} = -8 \times 10^{-3}$ and $\omega_s^{\text{FM}} = 5.3 \times 10^{-3}$ which are quite close to the calculated values of ω_s quoted above obtained from the equilibrium volume using DFT. Similar values were numerically found by Valiev *et al.* ($\omega_s^{\text{AF}} = -20 \times 10^{-3}$ and $\omega_s^{\text{FM}} = 10 \times 10^{-3}$) [15].

2.2 Anisotropic magnetoelasticity

To obtain magnetoelastic constants b_1 and b_2 from ab initio calculations we use the MAELAS code with mode 2 based on the strain-energy method [36] in combination with VASP for electronic structure calculations including spin-orbit coupling (SOC) [27, 37]. The main idea of the method proposed in MAELAS is that the difference in total energy for two specific magnetization directions of a deformed unit cell represents the linear dependence on strain from which the i -th anisotropic magnetoelastic constant b_i can be obtained by linear fitting [36]. For a cubic crystal, such magnetization

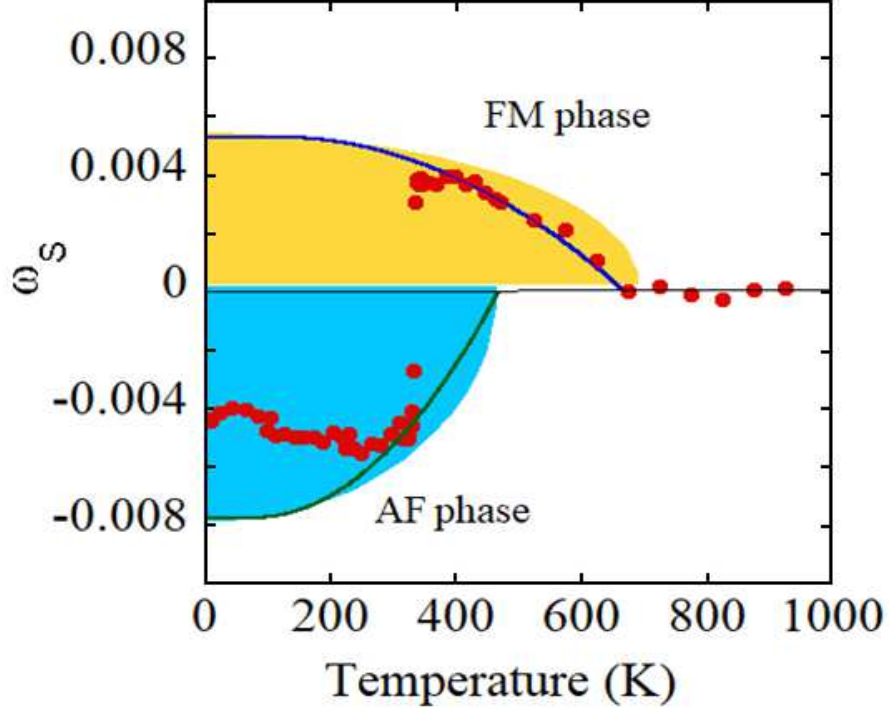


Fig. 3 Temperature dependence of the spontaneous volume magnetostriction ω_s for both the FM and AF phases. Red points correspond to the experimental data obtained by neutron diffraction. Solid lines show the extrapolation from the observed dependence of ω_s on the square of the magnetization (M^2). The magnetic moment μ was taken according to the mean-field approximation, assuming an iron magnetic moment of $3 \mu_B$ at 0 K in both magnetic phases.

directions and the corresponding linear dependencies are

$$\begin{aligned} \frac{1}{V_0}(E_{[100]}(\varepsilon_{xx}) - E_{[110]}(\varepsilon_{xx})) &= \frac{1}{2}b_1\varepsilon_{xx} - \frac{1}{4}K_1, \\ \frac{1}{V_0}(E_{[110]}(\varepsilon_{xy}) - E_{[\bar{1}10]}(\varepsilon_{xy})) &= 2b_2\varepsilon_{xy}, \end{aligned} \quad (2)$$

being V_0 the equilibrium volume of the magnetic supercell, K_1 the magnetocrystalline anisotropy constant, ε_{ij} the strain tensor, and $E_{[hkl]}$ total energy with magnetization direction along crystallographic direction $[hkl]$, where AF and FM order are used for each case. In order to do so, we increase the number of k-points to $36 \times 36 \times 36$ and $13 \times 13 \times 13$ for the FM and AF phases, respectively. The results are presented in Fig. 4, where, from the linear fitting of the energy versus strain data, we obtain $b_1^{AF} = 5.94$ MPa and $b_2^{AF} = -11.39$ MPa for the AF phase, while for the FM phase, we found $b_1^{FM} = -5.17$ MPa and $b_2^{FM} = -25.17$ MPa. Interestingly, the sign of b_1 is changed by magnetic order.

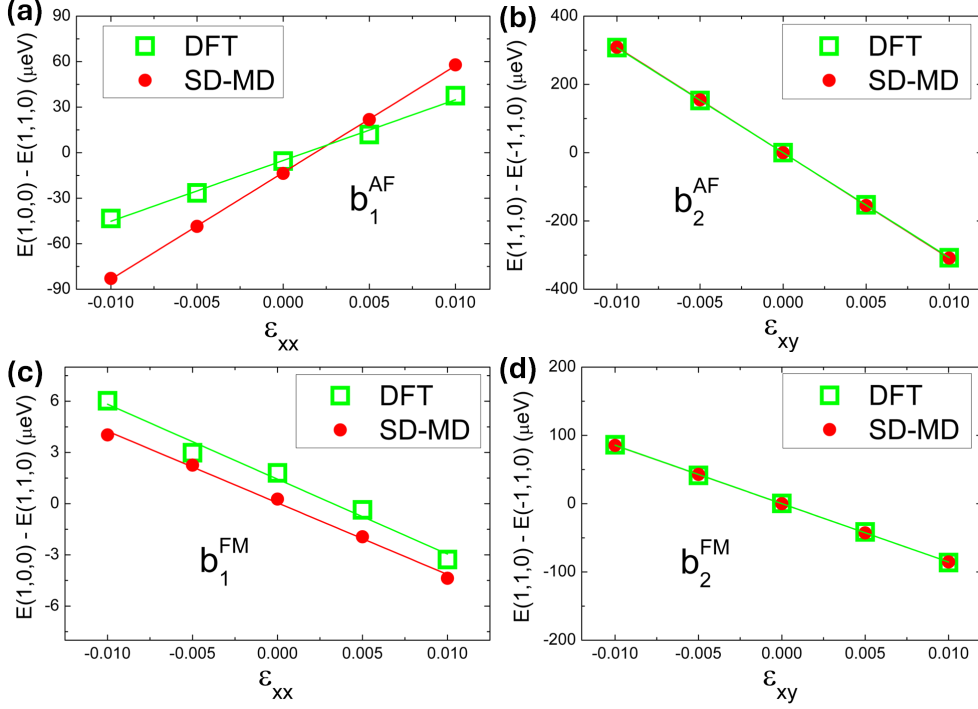


Fig. 4 Calculation of the anisotropic ME constants b_1 and b_2 for FeRh with AF (a,b) and FM (c,d) order using DFT and SD-MD. Solid line represents a linear fit to the data.

2.3 Magnetocrystalline anisotropy

Now, we use the same VASP settings as for magnetoelastic constants to find the pressure dependence of the magnetocrystalline anisotropy constant $K_1(P)$. For that reason, we evaluate the total energy E with AF or FM order along crystallographic directions $[100]$ and $[110]$ at different volumes V , and compute K_1 using

$$K_1(V) = \frac{4[E_{[110]}(V) - E_{[100]}(V)]}{V}, \quad (3)$$

where for each volume we also compute the corresponding pressure P . The results are shown in Fig. 5. As shown in Refs. [38, 39], in the low-pressure regime K_1 approximately depends linearly on the pressure

$$\frac{K_1(P)}{K_1(0)} = e^{\zeta P} \approx 1 + \zeta P, \quad (4)$$

where the notation $\frac{1}{K_1} \frac{\partial K_1}{\partial P} = \zeta$ is used and the condition $\zeta P \ll 1$, corresponding to the pressure range considered, is imposed. Thus, fitting the data to Eq. 4 allows us to obtain the $K_1(0)$ and $\frac{1}{K_1} \frac{\partial K_1}{\partial P}$ needed as input parameters for the subsequent spin-lattice model. This procedure gives the values $K_1^{AF} = 36.14 \text{ kJ m}^{-3}$

and $\frac{1}{K_1^{AF}} \frac{\partial K_1^{AF}}{\partial P} = 0.205 \text{ GPa}^{-1}$, while for the FM phase we obtain $K_1^{FM} = -5.15 \text{ kJ m}^{-3}$ and $\frac{1}{K_1^{FM}} \frac{\partial K_1^{FM}}{\partial P} = 0.464 \text{ GPa}^{-1}$. Here, we observe that the easy axis of the AF phase would correspond to [100], but for the FM phase it would be [111]. Ferromagnetic resonance measurements at $T = 425 \text{ K}$ were done by Mancini *et al.*[14] finding $K_1^{FM} = 6.4 \text{ kJ m}^{-3}$.

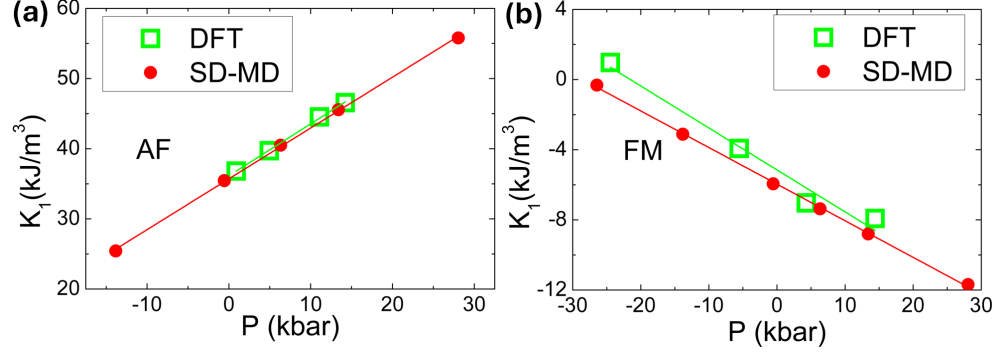


Fig. 5 Calculated pressure dependence of K_1 for FeRh with AF (a) and FM (b) order using DFT and SD-MD. Solid line represents a linear fit to the data.

3 Classical spin-lattice model

In this section, we aim to develop a classical spin-lattice model, that couples spin dynamics and molecular dynamics (SD-MD), capable of reproducing the properties obtained with DFT in Section 2 at zero temperature. To this end, we consider the following Hamiltonian

$$\mathcal{H}_{sl}(\mathbf{r}, \mathbf{p}, \mathbf{s}) = \sum_{i=1}^N \frac{|\mathbf{p}_i|^2}{2m_i} + \sum_{i,j=1}^N \mathcal{V}(r_{ij}) + \mathcal{V}_{mag}(\mathbf{r}, \mathbf{s}), \quad (5)$$

where $\mathbf{r}_i$, $\mathbf{p}_i$, $\mathbf{s}_i$, and m_i stand for the position, momentum, normalized magnetic moment, and mass of each atom i in the system, respectively, $\mathcal{V}(r_{ij}) = \mathcal{V}(|\mathbf{r}_i - \mathbf{r}_j|)$ is the non-magnetic part of the interatomic potential energy, and N is the total number of atoms in the system with total volume V .

The magnetic part of the interatomic potential $\mathcal{V}_{mag}$ corresponds to a magnetic Hamiltonian $\mathcal{H}_{mag}$ that includes the exchange interaction, the Néel interaction $\mathcal{H}_{N\acute{e}el}$,

the Zeeman energy, and the Blume-Capel term[17], respectively:

$$\begin{aligned}\mathcal{V}_{mag}(\mathbf{r}, \mathbf{s}) = \mathcal{H}_{mag}(\mathbf{r}, \mathbf{s}) = & -\frac{1}{2} \sum_{i,j=1, i \neq j}^N J(r_{ij}) \mathbf{s}_i \mathbf{s}_j \\ & + \mathcal{H}_{N\acute{e}el}(\mathbf{r}, \mathbf{s}) - \mu_0 \sum_{i=1}^N \mu_i \mathbf{H} \mathbf{s}_i \\ & - \sum_{i=1}^N \frac{D_i}{\mu_B^2} \mu_i^2,\end{aligned}\tag{6}$$

where μ_i is the atomic magnetic moment, μ_0 is the vacuum permeability, $\mathbf{H}$ is the external magnetic field, $J(r_{ij})$ is the exchange parameter, and D_i is the Blume-Capel constant that allows to get the correct ground state [17] which is equivalent to the first constant in the Landau term of the Landau-Heisenberg Hamiltonian [24].

The Néel interaction in Eq. (6) allows to include the magnetic anisotropic effects in the spin-lattice model through a two-ion Hamiltonian[40]

$$\begin{aligned}\mathcal{H}_{N\acute{e}el} = & -\frac{1}{2} \sum_{i,j=1}^N \{g(r_{ij}) + l_1(r_{ij}) \left[(\mathbf{e}_{ij} \mathbf{s}_i)(\mathbf{e}_{ij} \mathbf{s}_j) - \frac{\mathbf{s}_i \mathbf{s}_j}{3} \right] \\ & + q_1(r_{ij}) \left[(\mathbf{e}_{ij} \mathbf{s}_i)^2 - \frac{\mathbf{s}_i \mathbf{s}_j}{3} \right] \left[(\mathbf{e}_{ij} \mathbf{s}_j)^2 - \frac{\mathbf{s}_i \mathbf{s}_j}{3} \right] \\ & + q_2(r_{ij}) \left[(\mathbf{e}_{ij} \mathbf{s}_i)(\mathbf{e}_{ij} \mathbf{s}_j)^3 + (\mathbf{e}_{ij} \mathbf{s}_j)(\mathbf{e}_{ij} \mathbf{s}_i)^3 \right]\},\end{aligned}\tag{7}$$

where $\mathbf{e}_{ij} = \mathbf{r}_{ij}/r_{ij}$, and

$$\begin{aligned}l_1(r_{ij}) &= l(r_{ij}) + \frac{12}{35} q(r_{ij}), \\ q_1(r_{ij}) &= \frac{9}{5} q(r_{ij}), \\ q_2(r_{ij}) &= -\frac{2}{5} q(r_{ij}).\end{aligned}\tag{8}$$

Here $l(r_{ij})$ and $q(r_{ij})$ are dipole and quadrupole terms which are intended to describe effects induced by spin-orbit coupling like the anisotropic magnetostriction and MCA, respectively [39]. The term $g(r_{ij})$ can be used to shift the ground state energy of the exchange interaction. In the present model we do not use it, so we set $g(r_{ij}) = 0$.

In the case of a collinear state ($\mathbf{s}_i \cdot \mathbf{s}_j = 1$), Eq. 7 is simplified to

$$\begin{aligned}\mathcal{H}_{N\acute{e}el}^{\uparrow\uparrow}(\mathbf{r}, \mathbf{s}) = & -\frac{1}{2} \sum_{i,j=1, i \neq j}^N \{g(r_{ij}) + l(r_{ij}) \left[(\mathbf{e}_{ij} \mathbf{s}_i)^2 - \frac{1}{3} \right] \\ & + q(r_{ij}) \left[(\mathbf{e}_{ij} \mathbf{s}_i)^4 - \frac{6}{7} (\mathbf{e}_{ij} \mathbf{s}_i)^2 + \frac{3}{35} \right]\},\end{aligned}\tag{9}$$

for FM ordered spins [39], while assuming antiparallel pair spins ($\mathbf{s}_i \cdot \mathbf{s}_j = -1$) we obtain

$$\mathcal{H}_{N\acute{e}el}^{\uparrow\downarrow}(\mathbf{r}, \mathbf{s}) = -\frac{1}{2} \sum_{i,j=1, i \neq j}^N \{g(r_{ij}) - l(r_{ij}) \left[(\mathbf{e}_{ij} \mathbf{s}_i)^2 - \frac{1}{3} \right] + q(r_{ij}) \left[\frac{13}{5} (\mathbf{e}_{ij} \mathbf{s}_i)^4 + \frac{6}{7} (\mathbf{e}_{ij} \mathbf{s}_i)^2 + \frac{11}{35} \right] \}, \quad (10)$$

for AF order [41].

The spatial dependences of $J(r_{ij})$, $l(r_{ij})$, and $q(r_{ij})$ are described using the Bethe-Slater curve $\Lambda(r_{ij})$, as implemented in the SPIN package of LAMMPS [25, 42]

$$\Lambda(r_{ij}) = 4\alpha_{\Lambda} \left(\frac{r_{ij}}{\delta_{\Lambda}} \right)^2 \left[1 - \gamma_{\Lambda} \left(\frac{r_{ij}}{\delta_{\Lambda}} \right)^2 \right] e^{-\left(\frac{r_{ij}}{\delta_{\Lambda}} \right)^2} \cdot \Theta(R_{c,\Lambda} - r_{ij}), \quad (11)$$

where $\Theta(R_{c,\Lambda} - r_{ij})$ is the Heaviside step function and the $R_{c,\Lambda}$ ($\Lambda = J, l, q$) are the cut-off radii. The parameters α_{Λ} , γ_{Λ} , δ_{Λ} ($\Lambda = J, l, q$) should be defined in such a way that $J(r_{ij})$ reproduces the correct characteristic temperature of transition from an ordered to a disordered magnetic state (Curie or Néel temperature) and the spontaneous volume magnetostriction ω_s , while $l(r_{ij})$ reproduces b_1 and b_2 , and $q(r_{ij})$ reproduces magnetocrystalline anisotropy constant K_1 with its correct dependence on pressure $K_1(P)$.

3.1 Non-magnetic part of the interatomic potential

To describe the non-magnetic part of the interatomic potential, i.e., term $\mathcal{V}(r_{ij})$ in Eq.5, we use the same Lennard-Jones potential as Gruner *et al.* [17]

$$\mathcal{V}(r_{ij}) = 4\epsilon \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 \right], \quad (12)$$

where ϵ and σ are parameters that take the values $\epsilon_{nn} = 25.23$ mRy and $\sigma_{nn} = 2.32$ Å for the nearest neighbors (NN) pair interaction, while for the next nearest neighbors (NNN) they are $\epsilon_{nnn} = 25.23$ mRy and $\sigma_{nnn} = 2.67$ Å. This part of the interatomic potential allows to describe mechanical properties such as elastic constants (C_{ij}) and bulk modulus (B). Our analysis of this potential gives $C_{11} = 424$ GPa, $C_{12} = 140$ GPa, $C_{44} = 137$ GPa, and $B = 226$ GPa. These values differ somewhat from the DFT results $C_{11} = 262$ GPa, $C_{12} = 164$ GPa, $C_{44} = 109$ GPa, and $B = 197$ GPa, retrieved from the Materials Project database [43, 44], with a significant deviation observed for C_{11} .

3.2 Magnetic part of the interatomic potential

Due to the complexity of the magnetic order transition between AF to FM in FeRh, whereby Rh atoms become magnetic, it is convenient to start first by developing the

magnetic part of the interatomic potential for the AF phase, and later extend the model to include the corresponding features of FM order.

3.2.1 Antiferromagnetic phase

In the AF phase of FeRh, the magnetic moment of Rh is zero ($\mu_{Rh} = 0$), so we only need to consider Fe-Fe pair interactions at this stage. The Fe-Rh interactions will be included in the model in Section 3.2.2 for the analysis of the FM case. For the sake of simplicity, we will consider only Fe-Fe magnetic pair interactions only up to NN for both exchange and Néel interactions, which corresponds to a simple cubic (sc) symmetry, see Fig. 1.

From the analysis of the Mean Field Approximation (MFA) and Néel model [41, 45] for an sc structure in an AF state it is found

$$J^{Fe-Fe}(r_0) = -\frac{k_B T_N}{2},$$

$$r_0 \frac{\partial J^{Fe-Fe}}{\partial r} \Big|_{r=r_0} = -\frac{\omega_s^{AF}(C_{11} + 2C_{12})V_0}{3n_{Fe-Fe}}, \quad (13)$$

where k_B is the Boltzmann constant, C_{11} and C_{12} are the elastic constants, T_N is the Néel temperature (transition temperature between AF and PM), r_0 is the equilibrium distance to the NN Fe-Fe which is equal to the equilibrium lattice parameter a_0 , and n_{Fe-Fe} is the number of Fe magnetic interacting atoms in the equilibrium volume V_0 that have been included in the model. For example, in the unit cell of FeRh with volume $V_0 = a_0^3$ we have $n_{Fe-Fe} = 1$.

Similarly, the Néel dipole term between NN Fe-Fe atoms $l^{Fe-Fe}(r_{ij})$ describes anisotropic magnetoelastic constants b_1^{AF} and b_2^{AF} at the AF order. Applying the Néel energy expression given by Eq. 10 for the sc case with AF order we find

$$l^{Fe-Fe}(r_0) = \frac{V_0 b_2^{AF}}{2n_{Fe-Fe}}, \quad r_0 \frac{\partial l^{Fe-Fe}}{\partial r} \Big|_{r=r_0} = \frac{V_0 b_1^{AF}}{n_{Fe-Fe}}. \quad (14)$$

Next, the Néel quadrupole term $q^{Fe-Fe}(r_{ij})$ simulates $K_1^{AF}(P)$ at AF order. Using again Eq. 10 for the sc case with AF order we obtain

$$q^{Fe-Fe}(r_0) = \frac{5V_0 K_1^{AF}}{26n_{Fe-Fe}},$$

$$r_0 \frac{\partial q^{Fe-Fe}}{\partial r} \Big|_{r=r_0} = \frac{15V_0 K_1^{AF}}{26n_{Fe-Fe}} \left[1 - \frac{B}{K_1^{AF}} \frac{\partial K_1^{AF}}{\partial P} \right]. \quad (15)$$

Lastly, to obtain the parameters α_Λ , γ_Λ , δ_Λ ($\Lambda = J, l, q$) of the Bethe-Slater curves in Eq. 11 for the Fe-Fe pair interactions we apply the procedure described by Nieves

et al.[39]

$$\begin{aligned}\delta_\Lambda &= r_0, \\ \alpha_\Lambda &= \frac{e}{8} \left[2\Lambda(r_0) - r_0 \frac{\partial \Lambda}{\partial r} \Big|_{r=r_0} \right], \\ \gamma_\Lambda &= \frac{r_0 \frac{\partial \Lambda}{\partial r} \Big|_{r=r_0}}{r_0 \frac{\partial \Lambda}{\partial r} \Big|_{r=r_0} - 2\Lambda(r_0)},\end{aligned}\tag{16}$$

where e is Euler's number, and again $\Lambda = J, l, q$. The cut-off radii $R_{c,\Lambda}$ should be between NN and NNN of Fe atoms, so that only NN of Fe are included in such interactions ($R_{c,\Lambda}^{Fe-Fe} = 3.5$ Å). The parameters $J(r_0), l(r_0), q(r_0)$ as well as derivatives $\frac{\partial J}{\partial r} \Big|_{r=r_0}, \frac{\partial l}{\partial r} \Big|_{r=r_0}, \frac{\partial q}{\partial r} \Big|_{r=r_0}$ are given by Eqs.13,14 and 15, respectively. The calculated values of the parameters in the Bethe-Slater curves for the Fe-Fe interactions are shown in Table 1, where we used as input data the values of ω_s^{AF} , $K_1^{AF}(P)$ and b_i^{AF} from the DFT results found in section 2, the mechanical properties B and C_{ij} from the Lennard-Jones potential computed in Section 3.1, and $T_N = 405$ K [15].

Table 1 Parameters of the Bethe-Slater curves using the SD-MD model for FeRh.

SD-MD parameters	Fe-Fe	Fe-Rh
$R_{c,J}$ (Å)	3.5	2.9
α_J (meV/atom)	-114.118	11.069
γ_J	0.896	-0.684
δ_J (Å)	3.003	2.6
$R_{c,l}$ (Å)	3.5	2.9
α_l (μeV/atom)	-995.365	-1205.733
γ_l	0.343	0.986
δ_l (Å)	3.003	2.6
$R_{c,q}$ (Å)	3.5	2.9
α_q (μeV/atom)	55.272	53.968
γ_q	0.986	0.989
δ_q (Å)	3.003	2.600

3.2.2 Ferromagnetic phase

In this section, we aim to extend the model in order to describe FM properties by using the same Fe-Fe interactions obtained in Section 3.2.1, and adding the Fe-Rh pair interactions. To do so, we need first to model the magnetic moment of Rh as a function of the magnetic state. Based on the observation that $\mu_{Rh} \simeq \chi_{Rh} h_{ex,Fe}$, where $h_{ex,Fe}$ is the exchange field from the neighboring Fe sites and χ_{Rh} is an effective susceptibility [46], we assume the i -th moment of Rh is proportional to the average

moments of its 8 NN atoms of Fe

$$\mu_{Rh}^i = \mu_{Rh}^{FM} \langle s_{Fe}^i \rangle = \frac{\mu_{Rh}^{FM}}{8} \sum_{j=1}^8 \frac{\mu_{Fe,j}^i}{\mu_{Fe,j}^i} \quad (17)$$

where $\mu_{Rh}^{FM} = 1\mu_B$ is the magnetic moment of Rh at the zero-temperature FM state, and μ_B is the Bohr magneton, see Fig.6. Hence, the two limiting cases for the AF and FM states, i.e., $\mu_{Rh} = 0$ and $\mu_{Rh} = 1\mu_B$ respectively, are recovered.

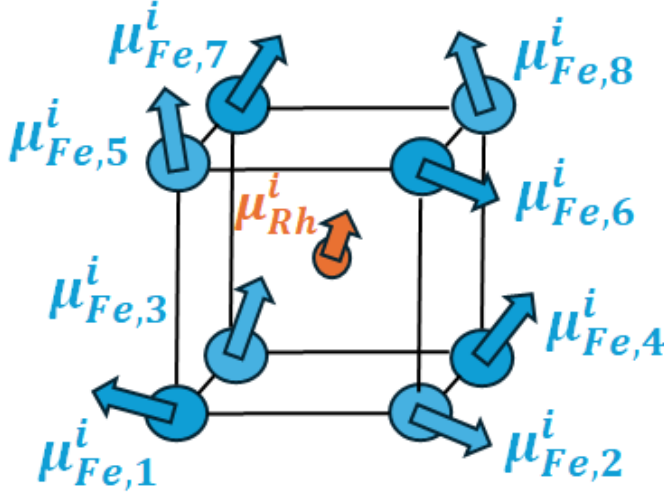


Fig. 6 Scheme showing the index convention between the i -th magnetic moment of Rh (μ_{Rh}^i) and its 8 NN atoms of Fe ($\mu_{Fe,j}^i$, $j = 1, \dots, 8$) in the unit cell of FeRh.

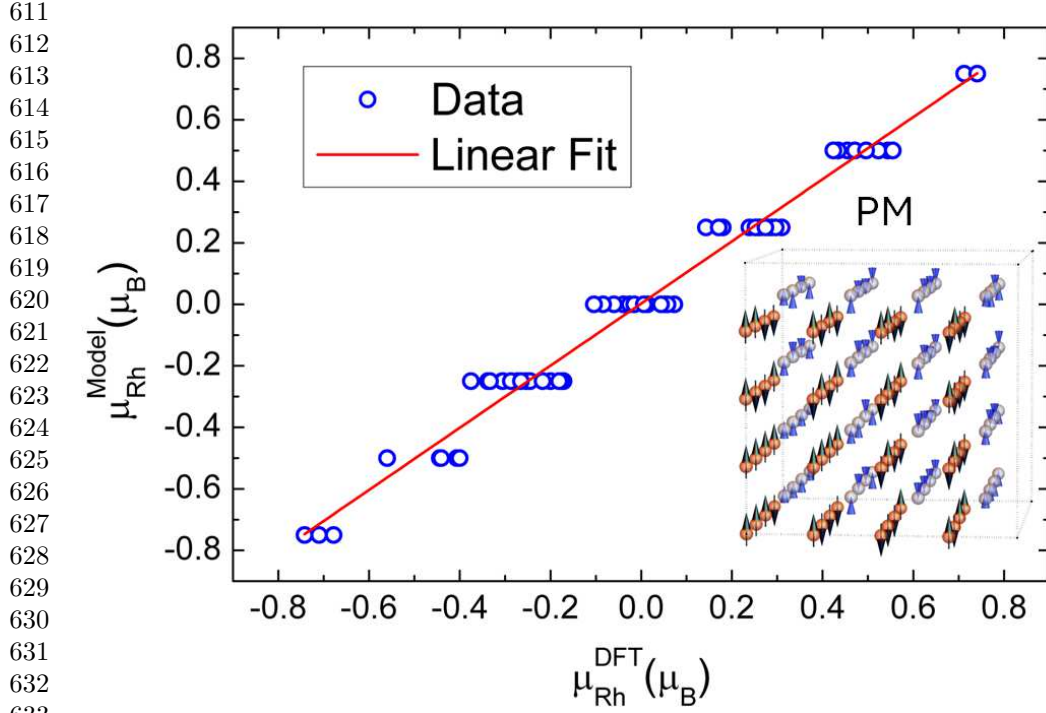
In Fig.7, we verify the accuracy of the proposed model of the magnetic moment of Rh in Eq.17 by comparing its prediction (μ_{Rh}^{Model}) with the DFT calculation μ_{Rh}^{DFT} for the PM state simulated with an SQS of 128 atoms. In this comparison, the values of $\mu_{Fe,j}^i$ in Eq.17 used to compute μ_{Rh}^{Model} were taken from the same DFT calculation to get μ_{Rh}^{DFT} . This procedure leads to "discrete" values of μ_{Rh}^{Model} since in the SQS the magnetic moments of Fe are aligned to the z-axis (Ising-like configuration, see inset of Fig.7) so that the summation over the 8 Fe atoms in Eq.17 gives multiples of $1/4$. Note that, for a PM state modeled with randomly oriented magnetic moments of Fe over the 3D directions of space, a continuous range of values for μ_{Rh}^{Model} will be obtained using Eq.17. We observe that the linear fit to $\mu_{Rh}^{Model} = a\mu_{Rh}^{DFT} + b$ in Fig.7 is quite good, finding $a = 1.01$ and $b = 0.002\mu_B$.

We can use the magnitude of the magnetic moment of Rh μ_{Rh}^i to control the strength of the Fe-Rh exchange and Néel interactions. Namely, we multiply the Bethe-Slater function Eq.11 for the Fe-Rh pair interactions by the dimensionless factor

599 μ_{Rh}^i/μ_B

$$\begin{aligned}
 600 \quad \Lambda^{Fe-Rh}(r_{ij}) &= 4\alpha_\Lambda \left(\frac{r_{ij}}{\delta_\Lambda} \right)^2 \left[1 - \gamma_\Lambda \left(\frac{r_{ij}}{\delta_\Lambda} \right)^2 \right] e^{-\left(\frac{r_{ij}}{\delta_\Lambda} \right)^2} \\
 601 \quad &\cdot \Theta(R_{c,\Lambda} - r_{ij}) \cdot \frac{\mu_{Rh}^i}{\mu_B}, \\
 602 \quad & \\
 603 \quad & \\
 604 \quad &
 \end{aligned}
 \tag{18}$$

605 where $\Lambda^{Fe-Rh} = J^{Fe-Rh}, l^{Fe-Rh}, q^{Fe-Rh}$. In the AF limit, we get $\mu_{Rh}^i = 0$ and
 606 $\Lambda^{Fe-Rh}(r_{ij}) = 0$, so that we have only Fe-Fe pair interactions, recovering the model
 607 described in Section 3.2.1 for the AF ground state. Now, we need to find the parameters
 608 $\alpha_\Lambda, \gamma_\Lambda, \delta_\Lambda$ ($\Lambda = J, l, q$) in Eq.18 that allow to reproduce the FM properties of FeRh,
 609 that is, $T_C, \omega_s^{FM}, K_1^{FM}(P), b_1^{FM}$ and b_2^{FM} .



635 **Fig. 7** Comparison between the calculation of the magnetic moment of Rh using Eq.17 and DFT
 636 for the PM state simulated with an SQS of 128 atoms. The inset shows such SQS, where orange and
 637 grey atoms correspond to Fe and Rh, respectively. Red line stands for a linear fit to the data.

638 For the sake of simplicity, we will consider all Fe-Rh magnetic pair interactions up
 639 to the NN, that correspond to a bcc symmetry. Using the MFA for a two-sublattice bcc
 640 structure with FM order, for which the Rh-Rh exchange interaction is not included
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(i.e. $J^{Rh-Rh} = 0$) we find [39, 45, 47]

$$J^{Fe-Rh}(r_0) = \frac{3k_B \sqrt{T_C(T_N + T_C)}}{8},$$

$$r_0 \frac{\partial J^{Fe-Rh}}{\partial r} \Big|_{r=r_0} = \frac{(\omega_s^{AF} + \omega_s^{FM})(C_{11} + 2C_{12})V_0}{4n_{Fe-Rh}}, \quad (19)$$

where n_{Fe-Rh} is the number of Fe-Rh interacting atoms in the unit cell, so that for volume $V = a_0^3$ we have $n_{Fe-Rh} = 2$. Note that in the spatial derivative of the exchange $\partial J^{Fe-Rh}/\partial r$ we included the factor $\omega_s^{AF} + \omega_s^{FM}$ since the Fe-Rh pair exchange interaction should compensate the Fe-Fe interaction that leads to an exchange magnetostriction ω_s^{AF} for the AF order and $-\omega_s^{AF}$ for the FM order.

The Néel dipole interaction between NN Fe-Rh atoms $l^{Fe-Rh}(r_{ij})$ can be used to describe the anisotropic magnetoelastic constants b_1^{FM} and b_2^{FM} at the FM order. To do so, we note that b_1^{FM} and b_2^{FM} have magnetoelastic contributions from the Fe-Rh pair interaction (b_1^{Fe-Rh} and b_2^{Fe-Rh}), as well as from the Fe-Fe with FM order ($b_{1,FM}^{Fe-Fe}$ and $b_{2,FM}^{Fe-Fe}$), so we have

$$b_1^{FM} = b_1^{Fe-Rh} + b_{1,FM}^{Fe-Fe},$$

$$b_2^{FM} = b_2^{Fe-Rh} + b_{2,FM}^{Fe-Fe}. \quad (20)$$

Here, the contribution of the Fe-Fe pair interaction with FM order corresponds to an sc symmetry [39, 45]

$$b_{1,FM}^{Fe-Fe} = -r_0 \frac{\partial l^{Fe-Fe}}{\partial r} \Big|_{r=r_0} \cdot \frac{n_{Fe-Fe}}{V_0},$$

$$b_{2,FM}^{Fe-Fe} = -\frac{2n_{Fe-Fe} l^{Fe-Fe}(r_0)}{V_0}, \quad (21)$$

where $l^{Fe-Fe}(r_0)$ and $r_0 \frac{\partial l^{Fe-Fe}}{\partial r} \Big|_{r=r_0}$ are given by Eqs.14. Therefore, inserting Eqs.21 into Eqs.20 we obtain the magnetoelastic contribution from the Fe-Rh pair interaction (b_1^{Fe-Rh} and b_2^{Fe-Rh}). Applying the Néel energy expression given by Eq. 9 for the bcc case with FM order we find [39, 45]

$$l^{Fe-Rh}(r_0) = -\frac{3V_0 b_1^{Fe-Rh}}{8n_{Fe-Rh}},$$

$$r_0 \frac{\partial l^{Fe-Rh}}{\partial r} \Big|_{r=r_0} = \frac{3V_0(b_1^{Fe-Rh} - 3b_2^{Fe-Rh})}{8n_{Fe-Rh}}. \quad (22)$$

Lastly, the Néel quadrupole interaction between the NN Fe-Rh $q^{Fe-Rh}(r_{ij})$ can be used to simulate $K_1^{FM}(P)$ for the FM order. As discussed for the dipole term, we note

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Table 2 Properties of FeRh for AF and FM order computed with SD-MD model and DFT at zero temperature. Data extrapolated with MFA in Fig.3 and from previous works are also shown.

Parameters	SD-MD(AF)	SD-MD(FM)	DFT(AF)	DFT(FM)	MFA(AF)	MFA(FM)	Prev. works(AF)	Prev. works(FM)
$\omega_s (\times 10^{-3})$	-7.6	9.7	-7.6	9.1	-8	5.3	-20[15]	10[15]
b_1 (MPa)	10.44	-4.98	5.94	-5.17				
b_2 (MPa)	-11.46	-25.35	-11.40	-25.17				
K_1 (kJ m ⁻³)	35.41	-5.93	36.14	-5.15				6.4 ¹ [14]
$\frac{1}{K_1} \frac{\partial K_1}{\partial P}$ (GPa ⁻¹)	0.204	0.344	0.205	0.464				

¹T=425K

again that the magnetocrystalline anisotropy for the FM order will have contributions from the Fe-Fe and Fe-Rh pair interactions

$$K_1^{FM} = K_1^{Fe-Rh} + K_{1,FM}^{Fe-Fe}, \quad (23)$$

where the Fe-Fe term $K_{1,FM}^{Fe-Fe}$ corresponds to the case of an sc with FM order[39]

$$K_{1,FM}^{Fe-Fe} = \frac{2n_{Fe-Fe}q^{Fe-Fe}(r_0)}{V_0},$$

$$\frac{1}{K_{1,FM}^{Fe-Fe}} \frac{\partial K_{1,FM}^{Fe-Fe}}{\partial P} = \left[1 - \frac{2V_0 r_0 \frac{\partial q^{Fe-Fe}}{\partial r} \Big|_{r=r_0}}{3n_{Fe-Fe} K_{1,FM}^{Fe-Fe}} \right] \cdot \frac{1}{B}, \quad (24)$$

where $q^{Fe-Fe}(r_0)$ and $r_0 \frac{\partial q^{Fe-Fe}}{\partial r} \Big|_{r=r_0}$ are given by Eqs.15. Hence, replacing Eqs.24 in Eqs.23 we obtain the contribution from the Fe-Rh pair interaction (K_1^{Fe-Rh}). Applying the Néel energy expression given by Eq. 9 for the bcc case with FM order we find [39]

$$q^{Fe-Rh}(r_0) = -\frac{9V_0 K_1^{Fe-Rh}}{16n_{Fe-Rh}},$$

$$r_0 \frac{\partial q^{Fe-Rh}}{\partial r} \Big|_{r=r_0} = -\frac{27V_0 K_1^{Fe-Rh}}{16n_{Fe-Rh}}$$

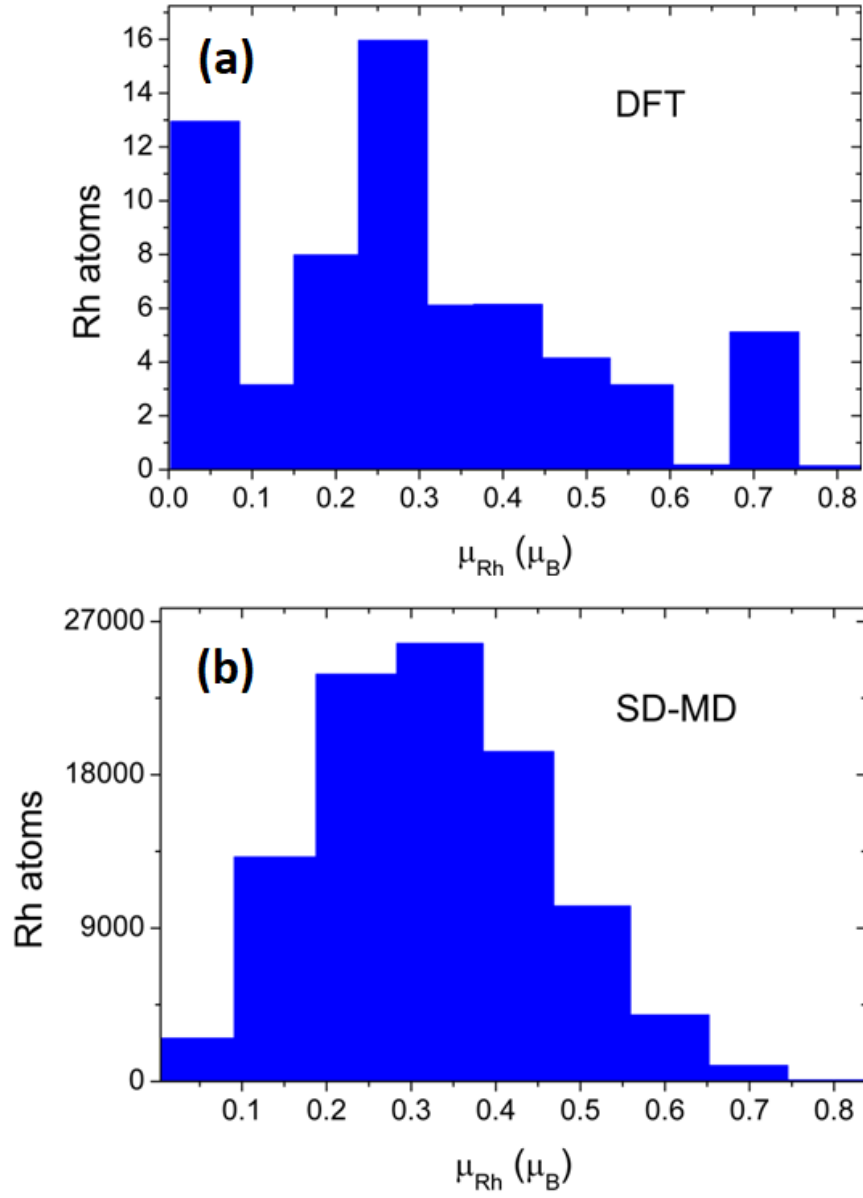
$$\cdot \left[1 - \frac{B}{K_1^{Fe-Rh}} \frac{\partial K_1^{Fe-Rh}}{\partial P} \right]. \quad (25)$$

To obtain the parameters α_Λ , γ_Λ , δ_Λ ($\Lambda = J, l, q$) of the Bethe-Slater curves in Eq.18 for the Fe-Rh pair interactions we input Eqs.19, 22 and 25 in Eqs.16, leading to the values shown in Table 1. The cut-off radii $R_{c,\Lambda}^{Fe-Rh}$ should be between NN and NNN of Fe and Rh atoms, so that only NN of Fe-Rh pair interactions are included ($R_{c,\Lambda}^{Fe-Rh} = 2.9$ Å). Here, we use as input data $\omega_s^{FM(AF)}$, $K_1^{FM}(P)$ and b_i^{FM} from the DFT results found in Section 2, mechanical properties B and C_{ij} from the Lennard-Jones potential computed in Section 3.1, $T_N = 405$ K and $T_C = 670$ K [15].

3.3 Model properties

In this section, we evaluate the properties of the spin-lattice model of FeRh and compare them with the DFT results obtained in Section 2. We start by analyzing the calculation of ω_s by computing the energy versus volume curve for the FM, AF and PM states, extracting equilibrium volume from a fit to the Vinet EOS and inserting these values in Eq.1. The obtained values are $\omega_s^{AF} = -7.6 \times 10^{-3}$ and $\omega_s^{FM} = 9.7 \times 10^{-3}$ that are in good agreement with the results found with DFT in Section 2.1, see Table 2. In Fig.2, we show the calculated energy versus volume with the spin-lattice model, where we set the Blume-Capel constant $D_{Rh} = -0.17$ eV for Rh in order to correctly obtain the AF phase as ground state, as well as similar energy gaps as in the DFT

783 calculation [17]. However, we observe that the energy difference between FM and PM
 784 states is still greater in DFT than in the model.



825 **Fig. 8** Distribution of magnetic moments of Rh at the PM state obtained using (a) DFT for an SQS
 826 with 128 atoms and (b) SD-MD for a supercell with 2×10^5 atoms.

The calculation of b_1 and b_2 using the SD-MD model is shown in Fig.4, while the analysis of $K_1(P)$ is presented in Fig.5. The values obtained are given in Table 2, where we see quite a good agreement between the model and the DFT results for all quantities, except b_1^{AF} , which is approximately twice higher in the former. Here, we checked that, assuming a larger value of b_1^{AF} a much better agreement is obtained between the DFT and the model, suggesting that this factor of 2 is related to the accuracy of the model due to the small value of b_1^{AF} . We point out that the model gives a good description of $K_1(P)$ only in the low pressure regime shown in Fig.5, where the DFT results exhibit a linear behavior.

For the PM state, all magnetic moments are randomly oriented so that the averages of their vectors are zero ($\langle \mu_{Fe} \rangle = 0$ and $\langle \mu_{Rh} \rangle = 0$), but the averages of the magnitudes of the magnetic moments are not ($\langle |\mu_{Fe}| \rangle \neq 0$ and $\langle |\mu_{Rh}| \rangle \neq 0$). In the case of Rh, the distribution of the magnitude of the magnetic moments for the PM state is determined by the local average of the moments of the 8 Fe NN to each Rh atom, which in our model will arise from Eq.17. In Fig.8, we show a comparison of the distribution of magnetic moments of Rh in the PM state between DFT for an SQS with 128 atoms (shown in the inset of Fig.7) and SD-MD for a supercell that includes 2×10^5 atoms with randomly oriented magnetic moments in 3D directions of space. We observe a similar trend in both approaches except for the onsets and tails of the distributions, where the small number of atoms included in the DFT calculation makes a fair statistical comparison difficult. Still, a good agreement is found for the average value $\langle |\mu_{Rh}^{DFT}| \rangle = 0.3\mu_B$ and $\langle |\mu_{Rh}^{SD-MD}| \rangle = 0.33\mu_B$, which combined with the results in Fig.7, suggests that Eq.17 could be a reasonable and simple way of modeling the magnetic moment of Rh in terms of the local moments of Fe.

The developed a spin-lattice model could be considered as an extension of Gruner *et al.* approach [17], where magnetic anisotropic features are included through the Néel model. The present analysis of the model was restricted to zero temperature. In previous models, the phase transition between AF to FM, as well as from FM to PM, were correctly described by Monte-Carlo simulations in Gruner *et al.* model [17], while such transitions were successfully simulated by time integration of the Landau-Lifshitz-Gilbert (LLG) equation in more advanced models like Barker *et al.* [22], where higher order exchange interactions were included. Therefore, further studies using the model presented in this work at finite temperature will be desirable.

4 Conclusions

In summary, we have performed a systematic first-principles investigation of the isotropic and anisotropic magnetoelastic properties of FeRh in its AF and FM ground states. Our results reveal pronounced qualitative differences between the two magnetic phases, demonstrating that the AF-FM transition is accompanied by a fundamental change in spin-lattice coupling. In particular, the exchange magnetostriction changes sign while retaining a comparable magnitude ($\omega_s \sim 10^{-3}$), in quantitative agreement with neutron diffraction data. In addition, both the anisotropic magnetoelastic constant b_1 and the magnetocrystalline anisotropy K_1 reverse sign between the AF and

875 FM states, highlighting a strong phase dependence of the anisotropic magnetoelastic
876 response.

877 To rationalize these findings, we developed a spin–lattice model that builds on ear-
878 lier approaches, extending them to include magnetic anisotropy within a Néel-type
879 framework. Although the present analysis is restricted to zero temperature, the model
880 establishes a direct connection between ab initio magnetoelastic parameters and atom-
881 istic simulations. Its implementation within the SPIN package of LAMMPS enables
882 large-scale simulations and provides a solid basis for future finite-temperature stud-
883 ies of the AF–FM transition, magnetoelastic effects, and their mutual interplay in
884 FeRh. More generally, the present results clarify the microscopic origin of magnetoe-
885 lasticity in FeRh and offer a transferable framework for studying coupled spin–lattice
886 phenomena in magnetostructural materials.

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