

## Chiral Asymmetry of the Spin-Wave Spectra in Ultrathin Magnetic Films

L. Udvardi<sup>1,2</sup> and L. Szunyogh<sup>1,\*</sup>

<sup>1</sup>*Department of Theoretical Physics, Budapest University of Technology and Economics, Budafoki út 8. H1111 Budapest, Hungary*

<sup>2</sup>*BME-HAS Group of Solid State Physics, Budapest University of Technology and Economics, Budafoki út 8. H1111 Budapest, Hungary*

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We raise the possibility that the chiral degeneracy of the magnons in ultrathin films can be lifted due to the presence of Dzyaloshinskii-Moriya interactions. By using simple symmetry arguments, we discuss under which conditions such a chiral asymmetry occurs. We then perform relativistic first principles calculations for an Fe monolayer on W(110) and explicitly reveal the asymmetry of the spin-wave spectrum in the case of wave vectors parallel to the (001) direction. Furthermore, we quantitatively interpret our results in terms of a simplified spin model by using calculated Dzyaloshinskii-Moriya vectors. Our theoretical prediction should inspire experiments to explore the asymmetry of spin waves, with a particular emphasis on the possibility to measure the Dzyaloshinskii-Moriya interactions in ultrathin films.

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It is by now well established that relativistic effects play a fundamental role in the magnetism of nanostructures, in particular, for thin films and finite deposited nanoparticles. Over the past two decades, a vast number of experimental and theoretical studies has been published to explore related phenomena such as magnetic anisotropies, spin-reorientation phase transitions, and noncollinear magnetic orderings [1–8].

The antisymmetric exchange interaction between two magnetic atoms  $E_{\text{DM}} = \mathbf{D}_{ij}(\mathbf{M}_i \times \mathbf{M}_j)$ , where  $\mathbf{M}_i$  and  $\mathbf{M}_j$  denote the spin moments of the atoms labeled by  $i$  and  $j$ , was proposed 50 years ago by Dzyaloshinskii [9] and Moriya [10]. The  $\mathbf{D}_{ij}$  is called the Dzyaloshinskii-Moriya vector, being identical to zero if the sites  $i$  and  $j$  experience inversion symmetry. It was put forward just about ten years ago that an enhanced Dzyaloshinskii-Moriya interaction at surfaces or interfaces can give rise to novel phenomena in nanomagnetism such as to noncollinear interlayer coupling [11,12], to unidirectional competing magnetic anisotropies [13], or to stabilization of noncollinear (chiral) magnetic orderings [14,15].

A breakthrough in this field happened when the resolution of spin-polarized scanning tunneling microscopy enabled detection of magnetic pattern formation on the atomic scale in monolayer-thin films. Such periodic modulations have been observed for Mn monolayers deposited on W(110) and W(001) and could successfully be interpreted in terms of a combination of relativistic first principles calculations and a simple micromagnetic model as the consequence of large DM interactions [16,17]. By using the same theoretical basis, it was even possible to explain the homochirality of the domain walls in two monolayers of Fe on W(110) [18], in agreement with previous experimental observation [19].

In this Letter, we investigate a consequence of the DM interactions on the spin-wave spectra in ultrathin films, not

yet explored in the literature. We argue that the chiral degeneracy of the spin-wave (SW) spectrum can be lifted due to the DM interactions and discuss under which conditions such a chiral asymmetry occurs. Based on relativistic first principles calculations, we explicitly evidence the asymmetry of the SW spectrum of an Fe monolayer on W(110) in the case of wave vectors parallel to the (001) axis. By emphasizing the possibility of probing the DM interactions in ultrathin films, we impel experiments to explore the proposed effect.

We start our study with simple considerations based on classical spin waves. If the atomic magnetic moment in the ground state of a ferromagnetic monolayer is  $\mathbf{M}_0 = M\mathbf{e}_0$ , with  $\mathbf{e}_0$  being a unit vector, then a spin wave of wave vector  $\mathbf{q}$  and chirality index (rotational sense)  $c = \pm 1$  is defined by the magnetic orientations  $\mathbf{e}_i(\mathbf{q}, c) = \mathbf{n}_1 \cos(\mathbf{q}\mathbf{R}_i) \times \sin\theta + c\mathbf{n}_2 \sin(\mathbf{q}\mathbf{R}_i) \sin\theta + \mathbf{e}_0 \cos\theta$ , where  $\mathbf{n}_1 \perp \mathbf{e}_0$  and  $\mathbf{n}_2 = \mathbf{n}_1 \times \mathbf{e}_0$  are unit vectors,  $\mathbf{R}_i$  is the position vector of site  $i$ , and  $\theta$  is the relative angle between the moments and  $\mathbf{e}_0$ . Inspecting the energy of the SW in terms of an extended Heisenberg model containing tensorial exchange interactions [20], it turns out that only the antisymmetric exchange interactions give rise to a chirality-dependent contribution:

$$E_{\text{DM}}(\mathbf{q}, c) = c \sin^2\theta \sum_{i \neq j} (\mathbf{D}_{ij} \cdot \mathbf{e}_0) \sin[\mathbf{q}(\mathbf{R}_i - \mathbf{R}_j)]. \quad (1)$$

The above expression also implies that only the components of the DM vectors parallel to  $\mathbf{e}_0$  influence the SW energy and that a reversed chirality can be converted into a propagation of the SW in the opposite direction:  $E_{\text{DM}}(\mathbf{q}, -c) = E_{\text{DM}}(-\mathbf{q}, c) = -E_{\text{DM}}(\mathbf{q}, c)$ .

The orientations of the DM vectors in a ferromagnetic monolayer have been analyzed for different 2D lattices in Refs. [14,21]. In particular, if the lattice has a twofold rotational symmetry, such as in the case of the (001) and

(011) surfaces of cubic lattices, all of the DM vectors lie in-plane. Clearly, from Eq. (1), a chiral asymmetry of the SW occurs then only for an in-plane ground-state magnetization. Furthermore, if the ground-state magnetization is in a mirror plane of the monolayer, no chiral asymmetry applies for wave vectors along  $\mathbf{e}_0$ .

In order to demonstrate the chiral asymmetry of the SWs, we have chosen a ferromagnetic Fe monolayer deposited on W(110), since (i) it exhibits an in-plane ground-state magnetization [22,23] and (ii) as for the Fe double layer [18] or for a Mn monolayer on W(110) and W(001) [16,17,24], large DM interactions are expected.

Our notation used for the principle axes of a bcc(110) plane are shown in Fig. 1 depicting the structure of the real lattice and the surface Brillouin zone with the high symmetry points. The experimental bcc W lattice constant  $a = 3.165 \text{ \AA}$  was chosen for the in-plane lattice constant (along the  $Y$  axis) throughout the system. All of the interlayer distances were fixed to the ideal bcc(110) value  $d = \sqrt{2}a/2 = 2.238 \text{ \AA}$ , but the interlayer distance between the Fe and the topmost W layer was relaxed by  $-12.9\%$  ( $d_{\text{Fe-W}} = 1.949 \text{ \AA}$ ) according both to experiment [25] and to theory [26]. The calculations were performed in terms of the fully relativistic screened Korringa-Kohn-Rostoker (SKKR) method [27] by using the local density approximation and the atomic sphere approximation. It should be noted that the SKKR method makes use of the semi-infinite geometry of the substrate; therefore, the calculations are not affected by ambiguities related to a supercell or film geometry.

We calculated magnetic anisotropy energies  $E(001) - E(1\bar{1}0) = 2.11 \text{ meV}$  and  $E(110) - E(1\bar{1}0) = 0.41 \text{ meV}$ . This implies that, in agreement with other theoretical works [28,29] and with the experiment [22], the ground-state magnetization of FeW(110) is in-plane with an easy axis along the  $(1\bar{1}0)$  direction, and the hard axis is along the  $(001)$  direction. It should be noted that the magnetostatic dipole-dipole interaction also favors the  $(1\bar{1}0)$  direction by about  $0.01 \text{ meV}$  with respect to the  $(001)$  axis and by  $0.11 \text{ meV}$  with respect to the  $(110)$  direction [30].

We applied a recent relativistic extension [20] of the torque method [31] to evaluate tensorial exchange inter-

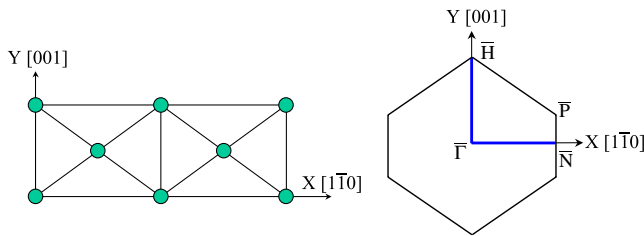


FIG. 1 (color online). Sketch of the lattice positions (left) and of the surface Brillouin zone (right) of a bcc(110) plane. The high symmetry points of the surface Brillouin zone are also labeled.

actions for FeW(110) from first principles. This method opened the way to atomistic spin-model simulations of nanostructures accounting for relevant relativistic interactions, such as the on-site magnetic anisotropy, the anisotropic symmetric exchange interaction, and the antisymmetric exchange interaction [21,24,32,33].

By using the convention  $H = -\sum_{i \neq j} J_{ij} \mathbf{e}_i \mathbf{e}_j$ , our calculated isotropic exchange interactions for the first few neighbors are  $J_{01} = 10.84 \text{ meV}$ ,  $J_{02} = -3.34 \text{ meV}$ ,  $J_{03} = 3.64 \text{ meV}$ , and  $J_{04} = 4.60 \text{ meV}$ . Note that, in particular, the nearest-neighbor interaction  $J_{01}$  is about 4 times less in magnitude than the corresponding parameter in Ref. [29]. To lend confidence to our values for  $J_{ij}$ , we performed Monte Carlo simulations and obtained a Curie temperature of about  $270 \text{ K}$ , in very good agreement with experiment ( $225 \text{ K}$ ) [22]. Note that random phase approximation (RPA) calculations in Ref. [29] provided a  $T_C$  above  $1000 \text{ K}$  which is, most likely, the consequence of the overestimated nearest-neighbor exchange interaction.

By using a canonical quantization of the linearized Landau-Lifshitz equations, we also developed a method to calculate the adiabatic SW spectra of bulk and layered systems on a relativistic first principles basis [20]. Although, within this approach, the interaction of the spin waves with the Stoner continuum is neglected, the main features of the SW spectra due to relativistic effects are expected to be well described. Notably, in the case of a monolayer, two SW solutions are obtained with the energies  $E^+(\mathbf{q})$  and  $E^-(\mathbf{q})$  that correspond to the chirality indices  $+1$  and  $-1$ , respectively.

In Fig. 2, the calculated adiabatic SW spectrum is shown along the  $X$  axis. It was demonstrated in Ref. [29] that the adiabatic SW energies and the SW dispersion obtained from RPA agree well for wave numbers as large as about  $1 \text{ \AA}^{-1}$ . We, therefore, display the adiabatic SW spectrum for only  $|q| < 1.2 \text{ \AA}^{-1}$ . Anticipated from the symmetry analysis above, since in this case the ground-state magnetization and the wave vectors lie in a mirror plane of the

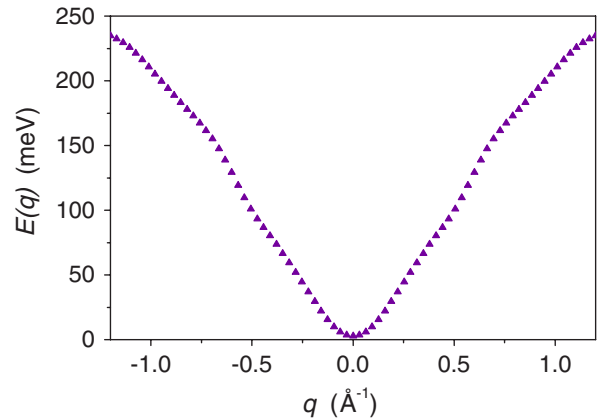


FIG. 2 (color online). Calculated adiabatic spin-wave spectrum of FeW(110) along the  $X$  axis as given in Fig. 1.

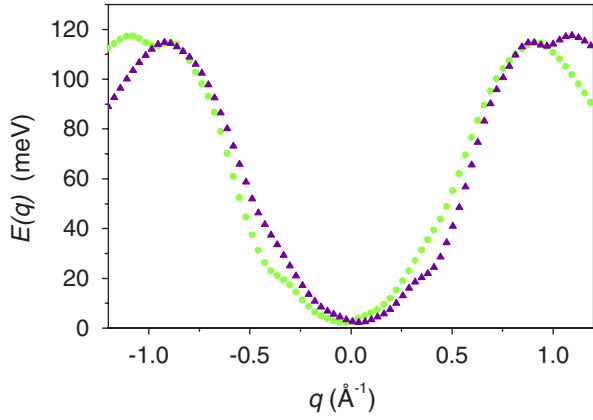


FIG. 3 (color online). Calculated adiabatic spin-wave spectra with chirality index  $+1$  (triangles) and  $-1$  (spheres) of FeW(110) along the  $Y$  axis; see Fig. 1.

system, the spectrum is degenerate, i.e.,  $E^+(q) = E^-(q)$ . Correspondingly, the SW dispersion is symmetric:  $E(q) = E(-q)$ . Note that the energy range of the dispersion in Fig. 2 is approximately half of that in Ref. [29], which we again attribute to the very different exchange interaction parameters in the two theoretical works.

Next we inspect the SW spectrum for wave vectors parallel to the (001) axis displayed in Fig. 3. Since in this case  $\mathbf{q}$  is perpendicular to the ground-state magnetization, our symmetry analysis predicts lifting of the chiral degeneracy of the spectrum, which can evidently be inferred from Fig. 3. Furthermore, the relationship  $E^+(-q) = E^-(q)$  is clearly regained. As compared with Ref. [29], again a difference by a factor of 2 in the energy range of the magnons can be noticed.

In order to demonstrate that the observed asymmetry of the SW spectrum results from the DM interactions, we performed a model calculation for  $\Delta E(q) = E^+(q) - E^-(q)$ . Our first principles calculations indicated that the DM vectors for the nearest and next-nearest neighbors, visualized in Fig. 4, are at least by an order larger in magnitude than the ones for more distant pairs. By using the method described in Ref. [20], the asymmetry of the SW energy can then be expressed as

$$\Delta E(q) = \frac{16\mu_B}{M_0} D_1^x \sin\left(\frac{1}{2}qa\right) - \frac{8\mu_B}{M_0} D_2^x \sin(qa), \quad (2)$$

where  $M_0 = 2.22\mu_B$  is the spin-magnetic moment per atom and  $D_1^x$  and  $D_2^x$  are the magnitudes of the  $x$  components (parallel to the ground-state magnetization) of the DM vectors for the nearest and second-nearest neighbors, respectively.

In Fig. 5, we plotted the asymmetry  $\Delta E(q)$  of the SW spectrum of FeW(110) along the  $Y$  axis obtained from the data in Fig. 3. As can be inferred from this figure,  $\Delta E(q)$  exhibits local extrema at about  $q = \pm 0.44 \text{ \AA}^{-1}$  with  $|\Delta E(q)| \approx 15 \text{ meV}$  and changes sign at  $q = \pm 0.83 \text{ \AA}^{-1}$ .

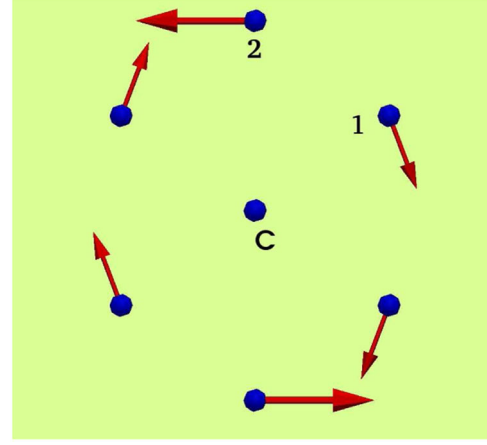


FIG. 4 (color online). Sketch of the calculated Dzyaloshinskii-Moriya vectors between an atom (C) and its nearest (1) and next-nearest (2) neighbors in an Fe monolayer on W(110).

Apparently, these features of  $\Delta E(q)$  are fairly well reproduced by the function [Eq. (2)] when using the calculated parameters  $D_1^x = 1.42 \text{ meV}$  and  $D_2^x = 6.08 \text{ meV}$ . Note that the characteristic extrema of  $\Delta E(q)$  are determined by the DM interactions between the next-nearest neighbors, since the  $\sin(qa)$  function in the second term on the right-hand side of Eq. (2) reaches a maximum or minimum at  $|q| = \pi/2a \approx 0.49 \text{ \AA}^{-1}$ . The deviations of the asymmetry of the SW energy from this model function are related to the DM interactions between more distant pairs that add low-frequency modulations to the SW dispersion.

The magnon spectrum of FeW(110) along the  $Y$  direction has been measured very recently by using spin-polarized electron energy loss spectroscopy [34], a highly suitable technique to probe high wave vector magnetic excitations of ultrathin films. Surprisingly, the measured

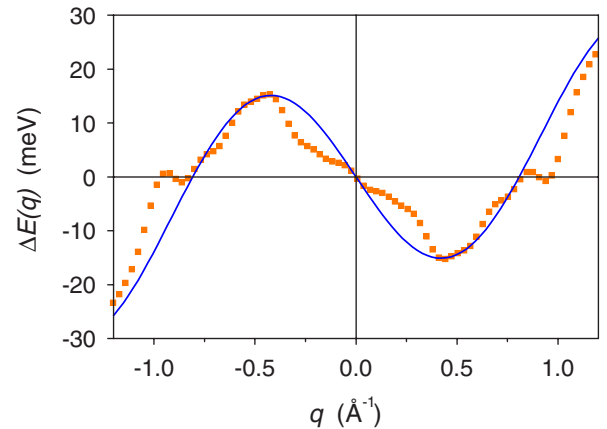


FIG. 5 (color online). Squares: Asymmetry of the spin-wave spectrum of FeW(110) along the  $Y$  axis as derived from the corresponding values in Fig. 3; solid line: the function [Eq. (2)] obtained from a second-nearest-neighbor model with the calculated DM interactions.

magnon energies are about half of the theoretical values reported here and smaller by even a factor of 4 than the calculated values in Ref. [29]. We are, however, aware of linear response calculations [35] that provided a very similar magnon dispersion along the  $Y$  axis as compared to that in Fig. 3. Thus, the low energy of the measured magnon spectrum [34] should most probably be attributed to effects not included in the first principles calculations, such as spin-charge coupling [36] or phonon-magnon interaction [37].

Considering the size of the SW asymmetry obtained from our calculations, e.g., about 20% at  $q = \pm 0.44 \text{ \AA}^{-1}$  with respect to the average energy  $[E^+(q) + E^-(q)]/2$ , we strongly suggest that it should be accessible to experiments. Indeed, preliminary measurements on FeW(110) [38] indicate the presence of an asymmetry in the magnon spectrum being quite similar in size and shape as in Fig. 5. It should also be mentioned that a related phenomenon, namely, the polarization dependence of the dynamical susceptibility, has been revealed in the paramagnetic phase of the weak antiferromagnet MnSi [39], following a theoretical prediction based on a quantum-spin chain model [40].

Further candidates for experimental observation of the proposed SW asymmetry are ferromagnetic monolayers on substrates with large spin-orbit coupling and polarizability (W, Pt, or Ir). In the case of an out-of-plane ground-state magnetization, a small magnetic field should be applied to orient the magnetization in plane, in order to fulfill the necessary condition for the chiral asymmetry of magnons. Our model calculation [see Eq. (2)] clearly implies that such experiments would serve as a unique tool to measure the Dzyaloshinskii-Moriya interactions in ultrathin ferromagnetic films, to be directly compared with the results of *ab initio* calculations. Concerning, in particular, the role of relativistic effects, such progress would clearly assist a deeper understanding of the magnetism in nanostructures.

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\*szunyogh@phy.bme.hu

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