1. Title of the Project

Spiral, Noncoplanar and Chiral Magnetic Ordering in the Double Exchange Model on the Triangular Lattice

2. Applicant

Maria Azhar

3. Institute

Theory of Condensed Matter
Zernike Institute for Advanced Materials
University of Groningen
Nijenborgh 4
9747 AG Groningen
The Netherlands

4. Abstract

Spiral, noncoplanar and chiral spin configurations lead to nontrivial topological effects and unconventional transport properties that have physical manifestations very important for new-generation technology and spintronic devices. Recently, helicoidal spiral, noncoplanar, and chiral magnetic structures have gained interest for spintronics applcations [1-3], particularly for application in devices that use electric current for more efficient switching of magnetization with a smaller bit size, instead of the bulkier conventional magnetic storage that uses magnetic fields to read or write data. In many multiferroic materials, a spiral ordering is induced by competing interactions, and the resulting inversion symmetry breaking can induce an electric dipole moment [4-7] through a mechanism completely different from those working in conventional ferroelectrics. Such examples give incentive to not only the search for new and technologically relevant material properties, but also for newer mechanisms that can lead to those properties and contribute to our understanding of condensed matter physics, for the engineering of better and more efficient materials.

5. Duration of the project

4 years, starting from September 2014

6. Personnel

6.1 Senior Scientists

Prof Dr Maxim Mostovoy, supervision, 10%

6.2 Junior Scientists

Maria Azhar OIO full-time

7. Cost Estimates

7.1 Personnel positions

One OIO position for 4 years

7.2 Running budget

5k€ per year for conferences, summer schools, and maintenance

7.3 Equipment

No additional budget is required for equipment.

7.4 Budget summary (in k€)

		2014	**2015**	**2016**	**2017**	**2018**	**Total**
Position	PhD student	15	50	50	50	35	200
	Postdocs	-	-	-	-	-	-
	Technicians	-	-	-	-	-	-
	Guests	-	-	-	-	-	-
Costs	Personnel	15	50	50	50	35	200
	Running budget	3	5	5	5	2	20
	Equipment	-	-	-	-	-	-
Total							220

8. Research Program

8.1 Inroduction to spiral and noncoplanar spin textures

Spiral, noncoplanar and chiral spin configurations lead to nontrivial topological effects and unconventional transport properties Recently, helicoidal spiral, noncoplanar, and chiral magnetic structures have gained interest for spintronics applcations [1-3]. Heterostructures consisting of ferromagnetic and spiral layers can be used for the manipulation of electronic spins. A relatively small magnitude of spin-polarized current flowing through such a device can allow the switching of the magnetization of the ferromagnetic layers through the spin transfer torque [8,9]. This holds potential for the electric field control of magnetic memory.

In spiral magnets, both the time and the space inversion symmetry is broken - in centrosymmetric crystals the space symmetry is broken spontaneously and in non-centrosymmetric crystals the symmetry breaking is enforced by the crystal lattice. The lack of inversion symmetry is evident in that “left-handed” and “right-handed” spirals are not equivalent (see Figure 1) and are obtained from each other by the operation of space inversion (the simultaneous reversal of all the space coordinates). In many multiferroic materials, a spiral ordering is induced by competing interactions, and the resulting inversion symmetry breaking switches on the Dzyaloshinskii-Moriya interaction in the expression for the free energy, inducing an electric dipole moment [4-7] through a mechanism completely different from those working in conventional ferroelectrics (see Figure 2). Multiferroics hold potential for technological applications with their ability for a 4-state logic (with the two polarization states and two magnetization states), as magnetoelectric sensors, and for electric field control of magnetic memory.

Noncoplanar magnetic orders with non-zero scalar chirality give rise to unusual transport and magnetoelectric properties and are attracting interest in the fields of multiferroics and topological insulators [10-12]. Mott insulators with a nonzero scalar chirality $\mathcal{X}_{12,3} = \mathbf{S}_1 \times \mathbf{S}_2 \cdot \mathbf{S}_3$ show the existence of nonzero orbital currents and the spins are coupled to both an electric field and a magnetic field [11]. There is experimental evidence of the Topological Hall Effect arising from effective magnetic fields induced by chiral spin orders [13-16]. When an electric current passes through a noncoplanar spin texture, the electronic spins align at each lattice site with the localized magnetic moments and this change in direction of the electronic spin induces a Berry phase $i\Omega/2$ in the wavefunction of the electron where $\Omega = \boldsymbol{S_1} \cdot \boldsymbol{S_2} \times \boldsymbol{S_3}$ is the solid angle enclosed by the noncoplanar spin configuration. Effectively the electrons behave as in the presence of an applied magnetic flux, and a topological contribution to the Hall resistivity appears. This coupling between charge and spin can be exploited, for example, for electric field control of magnetic memory, for the motion of domain walls in “racetrack memory” [17] or for the comparatively faster and more efficient information flow of skyrmions on a nanoribbon [2].

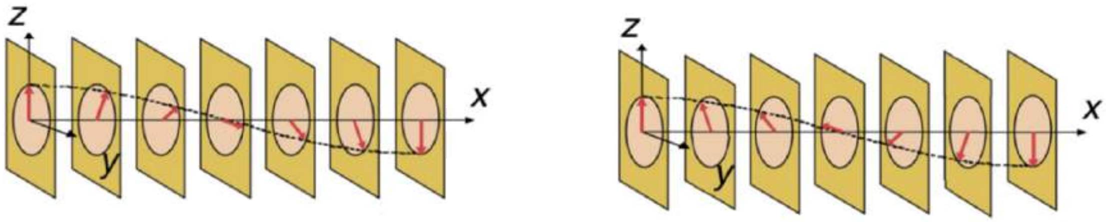

Figure 1 *A representation of magnetic moments in “left-handed” and “right-handed” spiral magnetic ordering*

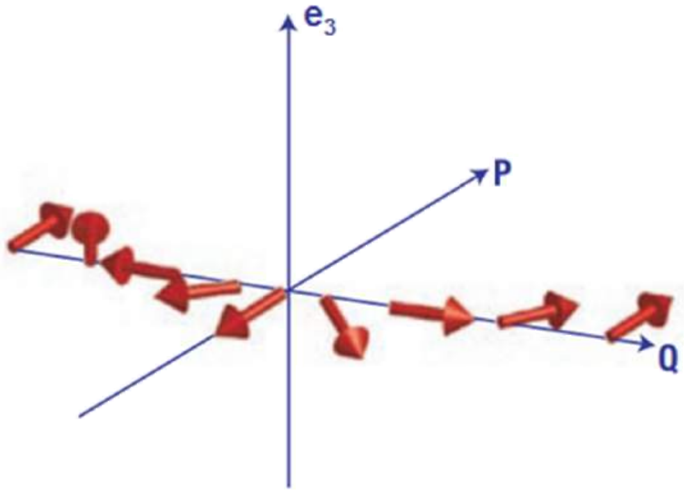

Figure 2 *The electric dipole moment* **P** *induced by a spiral magnetic ordering in a cubic lattice,* $\parallel \boldsymbol{e_3} \times \mathbf{Q}$ *, where* **Q** *is the wavevector of the spiral and* $\boldsymbol{e_3}$ *is the spin rotation axis.*

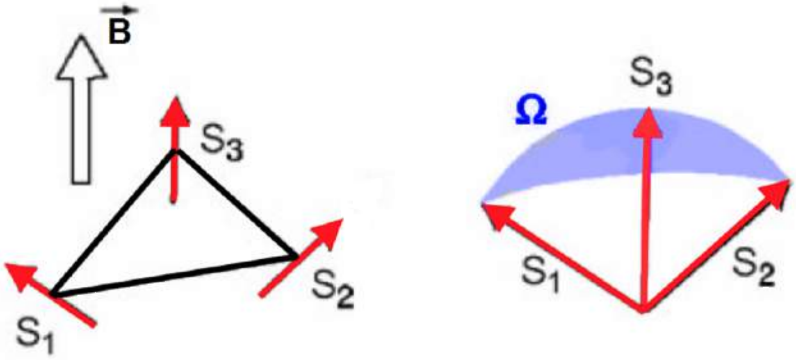

Figure 3 *The Topological Hall Effect: Electronic spins moving on a background of noncoplanar spins on a lattice feel an effective magnetic field*

8.2 Spiral and Noncoplanar Magnetic Order resulting from the Double Exchange Model

In many materials an incommensurate, noncollinear spiral ordering can result from competing exchange interactions, e.g. nearest-neighbour ferromagnetic and next-nearest neighbor antiferromagnetic interactions or from oscillating RKKY interactions. Also Dzyaloshinskii-Moriya (DM) interaction, four-spin exchange interations or the long range magnetic dipolar interaction can lead to spiral spin ordering or more complex noncoplanar orders, such as the skyrmion crystal that be viewed as a superposition of three spirals. A less commonly considered mechanism for noncoplanar ordering is the Double Exchange Model, which was first introduced by Zener [18] to explain the occurrence of ferromagnetism in the presence of charge carriers, in colossal magnetoresistance manganites. However the Double Exchange Model has also been shown to lead to an instability of the ferromagnetic state towards spiral ordering [19], possibly even in the limit of an infinite Hund's Rule coupling. The double exchange model describes a system of localized spins interacting with the spins of itinerant (conduction) electrons [18,20,21],

$$H_{DE} = -\sum_{ij\alpha} t_{ij}\, \psi_{i\alpha}^{\dagger} \psi_{j\alpha} - J \sum_{i\alpha\beta} \psi_{i\alpha}^{\dagger}\, \boldsymbol{\sigma}_{\alpha\beta}\, \psi_{i\beta} \cdot \boldsymbol{S}_i \,,$$

where the first term is the kinetic energy of the conduction electrons according to the tight-binding model. The electrons are described by the annihilation (creation) operator $\psi_{i\alpha}$ ($\psi_{i\alpha}^{\dagger}$). $\psi_{i\alpha}$ is the annihilation operator with spin index $\alpha = \uparrow, \downarrow$ for the itinerant electron located at the site i. t_{ij} is the hopping amplitude for an electron to hop from site j to site i. In the second term, J is the magnitude of the Hund's rule ferromagnetic coupling between the conduction electrons and the local spins, which tends to keep the electron spins parallel to the localised spins $\boldsymbol{S}_i$. $\boldsymbol{\sigma} = (\sigma^x , \sigma^y , \sigma^z)$ is a vector composed of the Pauli matrices. $\frac{1}{2}\psi_{i\alpha}^{\dagger}\, \boldsymbol{\sigma}_{\alpha\beta}\, \psi_{i\beta}$ is the spin operator for the conduction electrons on the site i and we can absorb the factor of ½ into J. This model was introduced by Zener [18] to explain the strong correlations between the movement of charge and the spin polarization of the magnetic lattice ions, in the perovskite manganites, e.g $R_{1-x}A_xMnO_3$ or $R_{2-2x}A_{1+2x}Mn_2O_7$ where R and A are the rare earth and alkali earth metals, respectively. It has since been widely used to describe systems with colossal magnetoresistance such as $La_{1-x}A_xMnO_3$ (A=Ca, Sr). Unlike the parent compound $LaMnO_3$, the partially doped lanthanum manganites are spontaneously magnetized and show a dramatic change of other properties as the doping is changed [22]. Clearly the itinerant electrons had some role to play that had to be incorporated in the Hamiltonian in the form of a spin-charge coupling – an argument that seems quite valid if we recall that electrons are particles possessing both spin and charge.

The Double Exchange intrinsically embodies an interplay between charge and spin. It explains the main properties of doped manganites and other materials [22-24] and is often studied in the limit of infinite Hund's Rule coupling, where usually a competing antiferromagnetic exchange leads to noncoplanar and noncollinear spin ordering. In the limit of infinite Hund's Rule coupling, the Double Exchange Model becomes simpler for numerical calculations [20] because the spin degrees of freedom for the conduction electrons are projected out. However it is known [19] that the Double Exchange Model by itself can also produce incommensurate, noncoplanar magnetic ordering, in general for an arbitrary value of J. To the second order in Q, the wavevector of the spiral, the difference between the energies of the spiral and ferromagnetic (FM) states in terms of the first order and second order corrections, $T^{(1)}$ and $T^{(2)}$ to the kinetic energy operator in the Hamiltonian, *T*, is [19]

$$\delta E = -\sum_{v} \frac{\left|\langle v|T^{(1)}|0\rangle\right|^{2}}{E_{v} - E_{0}} + \langle 0|T^{(2)}|0\rangle ,$$

where $T^{(1)}$ contains the spin-flip operator and hence describes the lowering of energy due to the mixing with excited states – a correction which is always negative in perturbation theory. The second term represents band narrowing, because the bands are widest for the ferromagnetic state. Therefore the energy difference δE between the spiral state and the ferromagnetic state can be negative under certain conditions (when the density of spin-flip excitations is high enough) and this can lead to a transition towards a spiral or generally noncoplanar state that is lower in energy.

8.3 The Double Exchange Model on the triangular lattice

Previous studies of the phase diagram for the double exchange model on the triangular lattice have been focused on commensurate magnetic orders. Recently, a non-coplanar 4-sublattice “tetrahedral” chiral phase was found by Martin and Batiste to be stabilized by nesting at 3/4 filling in the double exchange model on the triangular lattice [25]. This structure had already been proposed to describe some triangular lattice systems [26,27] and could potentially be accessible in other physical systems such as liquid surfaces or nanostructured semiconductor heterostructures. The ground state phase diagram for the double exchange model was obtained numerically by Akagi and Motome [12] and this chiral phase was found to exist at the ¼ and ¾ fillings. The competition between the ferromagnetic exchange and the antiferromagnetic superexchange on the triangular lattice and its temperature dependence at the ½ filling was studied by Kumar and van den Brink [28] and at ¼ filling in Ref. [29]. None of these authors considered the possibility of incommensurate spiral magnetic orders in the Double Exchange Model on the triangular lattice. It should be possible to obtain analytically and numerically the range of parameters for which the spiral state is the ground state for the triangular lattice. The spin spiral state has been found analytically to be stable towards phase separation for an isotropic model for small fillings $\lesssim 0.3$ [30] and also for a square and cubic lattice at small values of the Hund’s Rule coupling [31]. Therefore the spiral state could also be found to be stable for the case of the double exchange model on the triangular lattice.

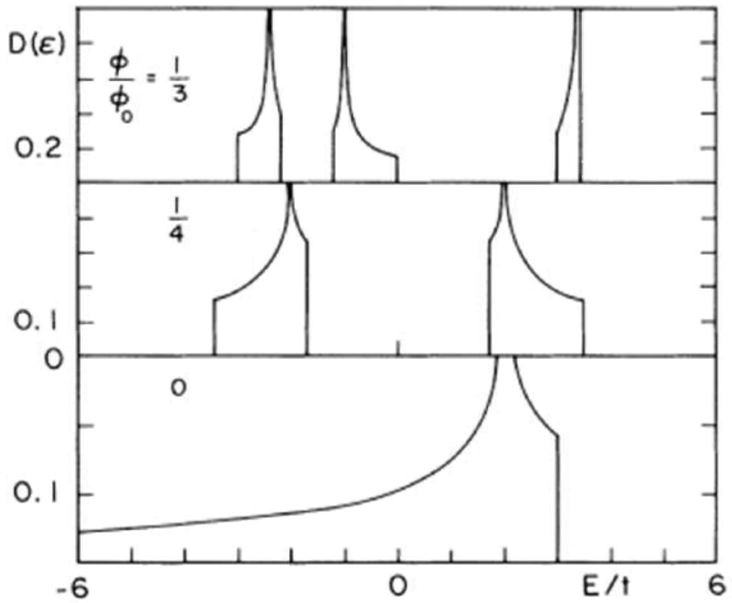

Figure 4 *Density of states in the triangular lattice with magnetic flux per triangle p/q where p and q are integers (adapted from Ref. [32]) showing that gaps are opened at special filling fractions*

There exists also the possibility to find other stable commensurate states such as the “tetrahedral state” which is stabilised as the result of a generalization of the Peierls instability to orbital magnetic instabilities, and which opens gaps at ¼ and ¾ filling (see Figure 3). The effective uniform magnetic flux experienced by itinerant electrons on the triangular lattice due to the noncoplanar tetrahedral spin configuration has the value $\Phi_0/4$ per triangle which was known to open gaps in the spectrum [32]. Similarly other noncoplanar spin configurations that would correspond to integer fractions of the unit of magnetic flux, per triangle, could also lead to stable chiral orderings. The tetrahedral state was found to exhibit a nonzero scalar chirality up to a finite temperature, causing a topological contribution to the Hall Effect with consequences for the electric current control of the magnetization.

For the simplest approaches, the Coulombic repulsion between the itinerant electrons is often ignored, however, models that incorporate strong electron-electron correlations usually show rich and complex phase diagrams, leading to coupling of spin, charge and lattice degrees of freedom in such materials and the possibilities of technologically useful behavior such as collosal magnetoresistance and multiferroism. Numerically such models can be dealt with by using perturbative methods and the methods of many-body physics, or non-perturbative methods such as the Dynamical Mean Field theory, which has been applied to the double exchange model by Furakawa [33]. Since the last two decades, systems with strongly correlated electrons have gained attention for both experimentalists and theoreticians due to the interplay between orbital, spin, lattice and charge degrees of freedom. Most of the quantum magnets, heavy fermion metals, the high T_C superconductors and the collosal magnetoresistance manganites belong to this class of materials. The study of strongly correlated electron systems is however still in its infancy, despite its dramatic successes in superseding the standard Fermi liquid description for many solids. A characteristic of such systems is their rich and complicated phase diagram with implications for technological applications.

8.4 Technological applications

The double exchange model intrinsically includes the interplay between charge and spin, and was first introduced to describe the strong coupling between electrical transport and magnetization in the collosal magnetoresistance manganites [18]. The rich phase diagram of this model can contain many competing phases, implying that magnetism in double exchange systems can be controlled with a relatively low-magnitude electrical current through the spin transfer torque [3,8,9]. Conversely, conductivity at the borderline between commensurate insulating states and metallic spiral states can be controlled with an applied magnetic field as in the colossal magnetoresistance systems. Moreover, non-collinear and non-coplanar magnetic spin structures appearing in this model can result in unusual transport phenomena, such as the Topological Hall effect which can be exploited for the electric field control of magnetization or vice versa. Heterostructures consisting of ferromagnetic and spiral layers can be used for the manipulation of electronic spins. A relatively small magnitude of spin-polarized current flowing through such a device can allow the switching of the magnetization of the ferromagnetic layers through the spin transfer torque [8,9]. Recently, helicoidal spiral, noncoplanar, and chiral magnetic structures have gained interest for spintronics applcations [1-3], particularly for application in devices that use electric current for more efficient switching of magnetization with a smaller bit size, instead of the bulkier conventional magnetic storage that uses magnetic fields to read or write data. A spiral magnetic ordering breaks inversion symmetry and induces an electric dipole moment [4-7] in many multiferroic materials. A two dimensional triangular lattice described by the double exchange model can physically be realised when a magnetic field confines electrons in a plane, in semiconductor heterojunctions, and in thin films. The results from the proposed research would be relevant for the studies of these cases.

8.5 Research Goals

In the proposed PhD project, we plan to discover novel kinds of noncoplanar and chiral magnetic ordering that could be stabilized especially on the triangular lattice, that also lead to interesting topological effects and properties relevant for technological applications.

8.5.1 Incommensurate spiral ordering on the triangular lattice from the Double Exchange Model

The main model used to describe the proposed system of localized and itinerant (conduction electron) spins on the triangular lattice would be the Double Exchange Model, which has usually been studied either in its ferromagnetic limit of infinite Hund's Rule coupling, or in the presence of a competing antiferromagnetic exchange. However it was shown that in general, for an arbitrary value of the Hund's Rule coupling, even the simple Double Exchange model can also cause an instability of the ferromagnetic state towards a generally incommensurate noncoplanar or spiral spin ordering [19]. Previous studies of the Double Exchange model on the triangular lattice have focused on commensurate ordering [12,25], and spiral or incommensurate ordering has not been considered yet, therefore one of the goals of this project is to identify and study the incommensurate magnetic ordering on the triangular lattice as described by the Double Exchange model. To increase the accuracy of the description for real materials, more than one orbital per atom can be considered and the effects of thermal fluctuations can be included.

8.5.2 Robust chiral and noncoplanar phases in spin-charge coupled systems

In [32] it has been shown that the kinetic energy for conduction electrons is a minimum for one quantum of applied magnetic flux, per particle, on the triangular lattice and the square lattice. This occurs due to the opening of gaps at special filling fractions, which lowers the energies of the occupied states. Noncoplanar magnetically ordered states can produce an effect on the conduction electrons similar to that of an applied magnetic flux and produce robust states lowered in energy in a mechanism similar to the Peierls structural distortion. It has been shown that a chiral tetrahedral ordering is stabilized on the triangular near ¼ filling due to a generalized Kohn anomaly [34] and at ¾ filling due to a generalized Peierls magnetic distortion [25,32]. Such mechanisms could be universal for spin-charge coupled systems and this can be investigated through numerical calculations for magnetic structures proposed to be suitable under topological and symmetry considerations on various lattices, corresponding to one quantum of applied magnetic flux per particle.

8.5.3 Skyrmions in the presence of an applied magnetic field

One notable example of a magnetic ordering which has been observed in real materials and is also topologically protected against small perturbations, is the skyrmion. The Double Exchange model in the arbitrary J limit, without the presence of frustration or a competing antiferromagnetic exchange, has not yet been used to describe skyrmion states on the triangular lattice. In real materials, an applied magnetic field has often stabilized a skyrmion crystal state in a range of temperatures [35] so leaves open the possiblility of the discovery of a skyrmionic phase in the triangular lattice under the Double Exchange Model. This would have implications for the design of materials that utilise skyrmions for technologically relevant applications [2].

8.5.4 Electron-electron correlation effects

Finally. electron-electron correlation effects can be included in the model. Numerically this scenario can be dealt with by using perturbative methods through the methods of many-body physics, or non-perturbative methods such as Dynamical Mean Field theory. Materials with strong electron-electron correlations show rich and complex phase diagrams which can be exploited for technological applications.

8.6 Planning and timing of intermediate steps

Time (year)	**Activity**
1st/2nd year	Learning the theoretical methods and finding stable magnetic orderings of topological relevance
3rd year	Design of materials exhibiting technologically interesting properties
4th year	Writing of thesis based on publications

9. Collaborations within the Zernike Institute

Solid State Materials for Electronics
Dr Graeme Blake and Dr Beatriz Noheda can collaborate for the synthesis and characterisation of the materials (bulk and thin films) displaying the predicted novel magnetic phases.

Physics of Nanodevices
Dr Bart van Wees working on metallic spintronics and fundamental spintronic phenomena. Dr Tamalika Banerjee working on spintronics of functional materials.

Optical Condensed Matter Physics
Dr Ron Tobey can provide his expertise of optical masurements to study phase transitions in the materials.

10. Infrastructure

No additional financial support is needed for infrastructure.

The Millipede High Performance Cluster under the auspices of the CIT, University of Groningen may be used for its parallel processing abilities. Access is already provided to all employees of the University of Groningen.

11. Application perspective in industry, other disciplines or society

Materials showing the interplay of charge and spin have numerous technological applications, notably in the fields of spintronics and data storage. Materials that store and process data efficiently and compactly shall allow the continued fulfillment of Moore's Law and contribute to a sustainable future.

12. References

[1] Jonietz F et al 2010 *Science* **330** 1648
[2] Fert, A, Cros V, Sampaio J 2013 *Nature Nanotechnology* **8** 152
[3] Bode, M. et al. 2007 *Nature* **447**, 190–193
[4] Mostovoy M 2006 *Phys.Rev Lett.* **96** 067601
[5] Kimura T, Goto T, Shintani H, Ishizaka K, Arima T, and Tokura Y, 2003 *Nature* **426**, 55
[6] T. Kimura 2007 *Annu. Rev. Mater. Res*. **37** 387
[7] Wang K F, Liu J, and Ren Z F, 2009 *Adv. Phys*. **58** 321
[8] J. Slonczewski J 1996 *Magn. Magn. Mater.* 159 L1
[9] L. Berger 1996 *Phys. Rev. B* **54**, 9353
[10] Ohgushi K, Murakami S, Nagaosa N 2000 *Phys. Rev. B.* **62** R6065
[11] Bulaevskii L N et al 2008 *Phys Rev B* **78** 024402
[12] Akagi Y and Motome Y 2010 *J. Phys. Soc. Jpn.* **79**, 083711
[13] Taguchi Y et al 2001 *Science* **291** 2573
[14] Taguchi Y and Tokura Y 1999 *Phys. Rev. B.* **60** 10280
[15] Taguchi Y et al 2003 *Phys. Rev. Lett.* **90** 257202
[16] Machida Y et al 2007 *Phys. Rev. Lett* **98** 057203
[17] Parkin S S P, Hayashi M, Thomas L, 2008 *Science***320** 190
[18] Zener C 1951 *Phys. Rev*. **82** 403

[19] Mostovoy M 2005 *J. Phys.: Condens. Matter* **17** S753
[20] Anderson P W and Hasegawa H 1955 *Phys. Rev.* **100** 675
[21] Kubo K and Ohata N 1972 *J. Phys Soc. Japan* **33** 21
[22] Coey J M D, Viret M, von Molnar S 1999 *Adv. Phys.* **48** 167
[23] Wollan E D and Koehler W C 1955 *Phys. Rev.* **100** 545
[24] Khomskii D I and Sawatzky G 1997 *Solid State Commun.* **102** 87
[25] Ivar Martin, C. D. Batista 2008 *Phys. Rev. Lett*. **101**, 156402
[26] Kurz P, Bihlmayer G, Hirai K and Blugel S 2001 *Phys Rev Lett* **86** 1106
[27] Momoi T, Kubo K, Niki K 1997 *Phys. Rev. Lett* **79** 2081
[28] Kumar S and van den Brink J 2010 *Phys. Rev. Lett.* **105,** 216405
[29] Zhang G P , Zhang J,Zhang Q-L, Zhou J-T,Shangguan M H 2013 *Physica B* **417**, 4-8
[30] Pekker D, Mukhopadhyay I S, Trivedi N and Goldbart P M 2005 *Phys. Rev. B* **72**, 075118
[31] Yin L 2003 *Phys. Rev. B* **68**, 104433
[32] Hasegawa Y et al 1989 *Phys. Rev. Lett.***63** 8
[33] Furukawa N 2002 *Fundamental Materials Research* 1-36
[34] Akagi Y, Udagawa M and Motome Y 2012 *Phys. Rev. Lett.* **108** 096401
[35] Nagaosa N and Tokura Y 2013 *Nature Nanotechnology* **8**, 899