Non-collinear antiferromagnets and the anomalous Hall effect

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Abstract - The anomalous Hall effect is investigated theoretically by employing density functional calculations for the non-collinear antiferromagnetic order of the hexagonal compounds Mn₃Ge and Mn₃Sn using various planar triangular magnetic configurations as well as unexpected non-planar configurations. The former give rise to anomalous Hall conductivities (AHC) that are found to be extremely anisotropic. For the planar cases the AHC is connected with Weyl points in the energy-band structure. If this case were observable in Mn₃Ge, a large AHC of about \( \sigma_{zx} \approx 900 \text{(Ωcm)}^{-1} \) should be expected. However, in Mn₃Ge it is the non-planar configuration that is energetically favored, in which case it gives rise to an AHC of \( \sigma_{xy} \approx 100 \text{(Ωcm)}^{-1} \). The non-planar configuration allows a quantitative evaluation of the topological Hall effect that is seen to determine this value of \( \sigma_{xy} \) to a large extent. For Mn₃Sn it is the planar configurations that are predicted to be observable. In this case the AHC can be as large as \( \sigma_{yz} \approx 250 \text{(Ωcm)}^{-1} \).

Introduction. – The well-known Hall effect [1] is observed in all conducting materials, but is especially large in ferromagnets, where it is dominated by a contribution that is not due to the Lorentz force. The latter is dissipationless and is called the anomalous Hall effect (AHE). This effect was explained long ago by Karplus and Luttinger [2], who invoked spin-orbit coupling and perturbation by the applied electric field to expose an additional term to be added to the usual electron velocity. Rather recently this additional term was discovered to be related to the Berry curvature in momentum space [3–5]. It is an important correction to all transport properties [6] that rely on the velocity. In particular it describes the leading contribution to the AHE [7].

Usually the AHE in a ferromagnet is assumed to be proportional to the magnetization although this cannot be taken too literally. In fact, Chen et al. [8] very recently found theoretically that the AHE should be observable in certain non-collinear antiferromagnets with zero net magnetization, provided that some symmetries are absent. They predicted this effect for the cubic antiferromagnet Mn₃Ir which was calculated to have a rather large anomalous Hall conductivity. The non-collinear antiferromagnetism in Mn₃Ir is of the same kind as that described for Mn₃Sn some time ago [9]. In fact, the prediction by Chen et al. [8] for Mn₃Ir are found to be valid for the family of cubic, non-collinear antiferromagnets Mn₃Sn, Mn₃Pt and Mn₃Rh for which the calculations of the type used in ref. [8] have been repeated with comparable results.

Recent experimental work on materials that show a large exchange bias was conducted for the hexagonal compound Mn₃Ge [10]. This is one of a family of non-collinear antiferromagnets that are related to Heusler compounds. It supplies a wealth of different non-collinear magnetic configurations and, by means of small deviations in the stoichiometry, is close in the phase diagram to tetragonal ferrimagnets, thus making it an interesting case for applications.

The system Mn₃Z (Z = Ga, Sn, and Ge) can be viewed as Heusler compounds which occur in different structures. A hexagonal phase having the symmetry \( P6_3/mmc \) (space group number 194) has been grown quite some time ago by annealing the crystals at high temperatures [11–14]. Tetragonal phases were obtained for Mn₃Ga and Mn₃Ge by annealing at low temperatures [11,12,15]. These different phases have markedly different magnetic properties, the tetragonal ones being ferrimagnetic while the hexagonal crystals are antiferromagnetic with a very small ferromagnetic component. The latter form a kagome lattice with a triangular coupling. The triangular arrangement is sketched in fig. 1 omitting at this stage the directions of the magnetic moments.
The various magnetic and structural properties of $\text{Mn}_3\text{Z}$ ($Z = \text{Ga, Sn, Ge}$) were recently reviewed theoretically by Zhang et al. [16]. In the present study we focus our attention on the hexagonal phase of $\text{Mn}_3\text{Ge}$ and $\text{Mn}_3\text{Sn}$.

Magnetic properties of hexagonal $\text{Mn}_3\text{Ge}$ and $\text{Mn}_3\text{Sn}$ and the AHE. – The early experimental work [11–14] and later theoretical studies [16–18] illuminated the unusual and interesting magnetic structure of $\text{Mn}_3\text{Ge}$ and $\text{Mn}_3\text{Sn}$. There are various possible triangular configurations when the magnetic moments are in the basal plane sketched in fig. 1. Because the Mn-Mn bonds between neighboring layers are somewhat shorter than the in-plane bonds, the interlayer magnetic coupling is important. Various configurations are shown in the recent paper by Zhang et al. [16] and also in older works by Sticht [17] as well as by Sandratskii and one of the present authors [18]. Most of these configurations are degenerate as long as spin-orbit coupling (SOC) is ignored. This changes markedly when SOC is included in the calculations, a fact that was convincingly explained in ref. [18].

We add to the previous work a determination of the magnetic structures and band structure from the curl of the Berry connection that is given by

$$\mathcal{A}(\mathbf{k}) = \frac{i}{\hbar} \int \frac{d\mathbf{k}}{(2\pi)^3} \Omega_p(\mathbf{k}) f(\mathbf{k}),$$

where $f(\mathbf{k})$ is the Fermi distribution function, $\Omega_p(\mathbf{k})$ is the $p$-component of the Berry curvature for the wave vector $\mathbf{k}$ and the components $\ell, \mu, \nu$ are to be chosen cyclic [6]. A limit to our numerical procedure is the rather high number of $\mathbf{k}$-points, $N_k$, needed for convergence, which was found to be about $N_k = 14000$. The differentiation brings this value up to 56000 points. We set a probable error bar at approximately 25%.

Results for the anomalous Hall conductivity. – First, some general remarks on the Hall conductivity are in order. The conductivity vanishes if spin-orbit coupling is ignored in the calculations, just as stated in ref. [8]. An exception occurs for the topological Hall effect, which we discuss further down. Next, the Hall conductivity is found to be remarkably anisotropic for all configurations, for which the magnetic moments lie in the hexagonal plane. Thus, in particular, the Berry curvature vector in the $z$-direction vanishes. If the conductivity is to be measured, this will require a somewhat unusual choice of the Hall cross, i.e. the electric field must be in the $z$-direction. For $\text{Mn}_3\text{Ge}$, however, we find a prominent non-planar configuration with a sizable value of $\sigma_{xy}$. This configuration has not been anticipated before and is discussed below as topological Hall effect.

In fig. 2 we add the band structure to our discussion. The configuration shown is self-consistent and acquires no ferromagnetic component. The band structure is doubly degenerate, except for some symmetry points where it may be higher. This and the fact that the magnetic moments sketched in fig. 2 are pairwise antiparallel indicate that this is the case of a bipartite lattice where time-reversal symmetry is not broken. The AHE vanishes by symmetry. This is the normal case for antiferromagnets as was also discussed by Chen et al. [8]. Since the total energy of

Fig. 1: (Color online) Projection on the basal plane of the hexagonal crystal structure of $\text{Mn}_3\text{Z}$ ($Z = \text{Ga, Sn, Ge}$). The small circles represent $Z$ in the basal plane ($z = 0$) and in a plane at $z = 0.5c/a$. The large colored dots represent Mn in the same two planes. The triangles are drawn to guide the eye. The magnetic ordering is not shown at this stage.

Fig. 2: (Color online) Band structure of $\text{Mn}_3\text{Ge}$ for the antiferromagnetic structure whose magnetic configuration is sketched on the right. The AHE vanishes for this case. The band structure of $\text{Mn}_3\text{Sn}$ for this configuration is very similar to that of $\text{Mn}_3\text{Ge}$.
Table 1: The total energy of the listed magnetic configuration, $\Delta E$, the magnetic moments of Mn, $M_{\text{Mn}}$, in $\mu_B$, estimates of the ferromagnetic spin and orbital moments, $M_{sp}, M_{orb}$ in $\mu_B$, and the Hall conductivities, $\sigma_{yz}, \sigma_{zx}$ and $\sigma_{xy}$ in (Ωcm)$^{-1}$. THE gives the contribution from the topological Hall effect. The lattice constants were taken from experiment [12,16]: for Mn$_3$Ge $a = 0.536$ nm, $c/a = 0.80598$ and for Mn$_3$Sn $a = 0.5665$ nm, $c/a = 0.79982$.

|      | $\Delta E$ (meV) | $M_{\text{Mn}}$ | $M_{sp}$ | $M_{orb}$ | $\sigma_{yz}$ | $\sigma_{zx}$ | $\sigma_{xy}$ |
|------|-----------------|-----------------|----------|----------|---------------|---------------|---------------|
| Mn$_3$Ge |                 |                 |          |          |               |               |               |
| Figure 2 | 744             | 2.258           | 0        | 0        | 0             | 0             | 0             |
| Figure 3(b) | 1               | 2.738           | 0.007    | 0.03     | 667           | 379           | 0             |
| Figure 5(c) | 0               | 2.738           | 0.004    | 0.03     | 607           | 1             | 0             |
| Figure 7   | -27             | 2.738           | 0.002    | 0.02     | 231           | -965          | 104           |
| THE        |                 |                 |          |          |               |               |               |
| Mn$_3$Sn   |                 |                 |          |          |               |               |               |
| Figure 3(b) | 0               | 3.121           | 0.003    | 0.001    | 248           | 129           | 0             |
| Figure 5(c) | 1               | 3.120           | 0        | 0.001    | 256           | 17            | 0             |
| Figure 7   | 2                | 3.121           | 0.01     | 0.001    | 95            | 111           | 8             |
| THE        |                 |                 |          |          |               |               |               |

Fig. 3: (Color online) Two triangular magnetic configurations. Panel (a) demonstrates opposite winding (or chirality) of the magnetic moments in the two triangles. (b) Configuration of (a) after a self-consistent calculation. Note the change of the directions of the moments of atom 1 and 3, which leads to the chirality of the two triangles to become the same.

Fig. 4: (a) Band structure of Mn$_3$Ge of the opposite-chirality case given in part (a) of fig. 3. (b) Band structure of the equal-chirality case given in part (b) of fig. 3. Note especially the bands in the range from $K$ to $M$.

this non-collinear spin configuration is rather high (see table 1), we turn to more interesting cases.

We now focus our attention on another general property of a non-collinear magnetic configuration, viz. on the winding or chirality. A special choice is shown in fig. 3(a), where the opposite chirality of the two groups of the magnetic moments is easily seen. Although quite seductive the so-chosen configuration (a) is not stable, which means it “unfolds” in the self-consistent calculation to the configuration depicted in fig. 3(b). This is due to the interlayer exchange interactions that rotate two of the magnetic moments. Both parts of the configuration (b) now have the same chirality. The conductivities are finite in each case, being for the non-selfconsistent case (a) of the order of 100 (Ωcm)$^{-1}$ but much larger for (b) as seen in table 1. At first sight one tends to attribute this large change to the appearance of a very small ferromagnetic component that develops in the self-consistent calculation (see table 1). However, a different reason emerges from a special property of the band structure, which is graphed in fig. 4 for the two cases (a) and (b) of fig. 3. Drawing the attention to the band structure in the range from $K$ to $M$, we see there is nothing unusual in case (a), but in case (b) two non-degenerate bands cross at the Fermi edge. Such a crossing point has recently been named Weyl point [23] which is expected to result in interesting topological properties, especially those concerning the anomalous Hall effect. However, caution should be taken with the terminology since the work of Wan et al. [23] is on ferromagnets and the Fermi-arc surface states arising from the Weyl point. This does not apply to our case. Still, the crossing point plays a large role as we show next. At least, this point can be called “diabolic” a term used by Gradhand et al. [24].

To elucidate the role of the Weyl point we show in fig. 5(a) the spin-filtered band structure of Mn$_3$Ge and in fig. 5(b) that of Mn$_3$Sn. In both cases the self-consistent configuration depicted in fig. 5(c) was used. For Mn$_3$Ge the band crossing occurs as in fig. 4(b) between the points $K$ and $M$ (not drawn to scale). For Mn$_3$Sn, however, the bands are slightly gapped between $K$ and $M$, instead the
Fig. 5: (Color online) (a) Spin-resolved band structure of Mn$_3$Ge for the magnetic configuration shown in panel (c) along the standard symmetry lines. (b) Spin-resolved band structure of Mn$_3$Sn for the magnetic configuration shown in (c) along standard symmetry lines extended by the section Γ-K$''$. See fig. 6(b) for the definition of the label K$''$.

band crossings here occur twice between the points K$''$ and K. In both cases the bands at the Fermi energy are non-degenerate minority-spin electron bands that unfortunately, because of spin-orbit coupling, do not describe simply spin-up or spin-down electrons.

To round up the physical picture we calculated the Berry curvatures in the plane that is extended enough to show the entire symmetry of the hexagonal basal plane of the Brillouin zone (BZ). The results are graphed in fig. 6(a) for Mn$_3$Ge and in fig. 6(b) for Mn$_3$Sn. The Weyl points appear distinctly at the places corresponding to the crossing points in the band structure. Closer inspection shows for Mn$_3$Sn that the spots originating from the crossings between K$''$ and K are much more pronounced than the spots seen between K and M, where the states are slightly gapped. There are more Weyl (or “diabolic”) points deep in the BZ that produce a signature similar to the one shown in fig. 6 if the vector $k_z$ characterizing the plane is appropriately chosen. The role of the Weyl points for the AHE is interesting and has been discussed controversially in the recent literature by Haldane [25], Chen et al. [26], and Vanderbilt et al. [27].

Adding to the discussion of chirality in connection with fig. 3 we observe that the chirality of the two triangles shown in fig. 5(c) is the same. This can be changed by interchanging the direction of arrows 2 and 3. After self-consisting this configuration the AHC is found to vanish. The Weyl points, however, remain visible in the Berry curvature, but the phase relations change such that the contributions to the Berry curvature cancel out in the Brillouin zone.

The topological Hall effect. – The discussion of the AHE is not yet complete. By trial and error we discovered a non-planar antiferromagnetic configuration that gives rise to a large AHC. For Mn$_3$Ge this configuration is illustrated in fig. 7, for Mn$_3$Sn it differs somewhat. For Mn$_3$Ge the total energy favors this non-collinear structure, which has not been discussed previously. Since the anisotropy is much less pronounced (see table 1) it is plausible that its AHE is easier to observe, whereas for Mn$_3$Sn it will be the planar cases that should be measurable. We could not find a Weyl point to be connected with the non-planar structures.

In connection with the non-planar magnetic configuration the contribution to the AHC arising from the topology, the so-called topological Hall effect (THE) [28] can be obtained. This is done by calculating the Hall conductivity omitting spin-orbit coupling. The results are included in table 1. For Mn$_3$Ge the value for $\sigma_{xy}$ is quite significant and is seen to be the dominant contribution to the conductivity. Its being much larger than that for...
Mn$_3$Sn is explained by the difference in the “spin chiralities”, which for non-planar configurations may be defined by $[28] \kappa = n_i \cdot (n_j \times n_k)$, where $n_i$ is the unit vector giving the direction of the moment at site $i$ and $(i, j, k)$ is chosen to be $(1, 2, 3)$ or $(3, 4, 5)$. In the case of Mn$_3$Ge the configuration shown in fig. 7 gives $\kappa \approx 0.88$, whereas for Mn$_3$Sn we obtain only $\kappa \approx 0.04$. For the planar magnetic configurations $\kappa = 0$.

Very recently exciting experimental work on the AHE in non-collinear antiferromagnetic Mn$_3$Si$_3$ [29] came to our attention. Here a topological Hall conductivity is found. Since the crystal structure of Mn$_3$Si$_3$ is considerably more complicated than that of Mn$_3$Sn or Mn$_3$Ge it appears that experimental work on the latter compounds could be quite rewarding, too.

**Summary.** – The AHE in the non-collinear antiferromagnetic compounds Mn$_3$Ge and Mn$_3$Sn is described in detail. Two different sets of magnetic configurations are relevant: triangular planar and non-planar. For Mn$_3$Sn the total energy favors a planar structure for which the AHC is predicted to be markedly anisotropic. For Mn$_3$Ge the non-planar case dominates, for which the topological Hall effect contributes significantly to the conductivity $\sigma_{xy}$.

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