Dynamics, Selection Rules and Dzyaloshinsky-Moriya Interactions in Strongly Frustrated Magnets

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Anisotropic spin-spin interactions of the symmetry described by Dzyaloshinsky and Moriya are generally considered weak, as they depend on the spin-orbit couplings. In frustrated spin systems with singlet ground states they can, however, have rather strong effects. We discuss recent results related to two gapped spin systems: $CuGeO_3$ and $SrCu_2(BO_3)_2$ in particular. In the first compound the Dzyaloshinsky-Moriya interactions effectively lower the symmetry of the magnetic unit cell and this leads to doubling of the low frequency mode. In the second case, the Dzyaloshinsky-Moriya interactions also split the lowest magnon mode linearly in the spin-orbit coupling. In addition, the relatively weak Dzyaloshinsky-Moriya interactions can dominate the dispersion.

Consideration of the selection rules for optical transitions show that while the Dzyaloshinsky-Moriya interactions can explain much of the dynamics, they do not explain the observed transition amplitudes. This leads to a review of recent calculations of anisotropic spin-phonon couplings. We discuss how this leads to a novel mechanism to explain the ESR intensities in the spin gap systems discussed. Selection rules for this novel mechanism involving coupling to the electric field of the resonant probe are discussed and relation to polarised neutron experiments briefly mentioned.

§1. Introduction

In strongly frustrated magnets with singlet ground states Dzyaloshinsky-Moriya interactions [1),2)] $\sum_{i,j} \vec{D}_{i,j} \cdot (\vec{S}_i \times \vec{S}_j)$, (with sum over neighbours i and j), can have marked effects on the dynamics even though they are generally considered to be a relatively weak perturbation of the isotropic exchange. By strongly frustrated we mean systems that have singlet ground states separated by a gap. This may be associated with a spin-Peierls distortion, as exemplified by the compound $CuGeO_3$ or purely geometric frustration, as in the case of the compound $SrCu_2(BO_3)_2$ which is close to a model system for the Shastry-Sutherland model in two dimensions.

Moriya [2)] estimated that the magnitude of the Dzyaloshinsky-Moriya vector between two sites is related to the isotropic exchange J by the relation $D = (\frac{\Delta g}{g})J$ if it is allowed by symmetry, where $\Delta g = g - 2$ is the measure of the strength of spin-orbit interactions, about .1 in the case of the copper oxides. J is the isotropic Heisenberg exchange. We remark that one can find exceptions to the rule that the Dzyaloshinsky-Moriya interaction should be much smaller than the isotropic exchange in model calculations, essentially by involving superexchange with copper-oxygen-copper angles close to $\frac{\pi}{2}$, in which case J is exceptionally small. These

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exceptions involve fine-tuning and have, as yet, not been shown to be relevant to real systems. Local symmetries or approximate symmetries may of course give a Dzyaloshinsky-Moriya coupling that is much smaller: if the relevant exchange J comes from superexchange paths that have an inversion symmetry $\vec{D}$ must vanish, and if this inversion symmetry is only weakly broken $\vec{D}$ must be small.

We shall argue nonetheless that $\vec{D}$ can dominate certain features of the dynamics because:
(i) it is the leading source of spin anisotropy in zero field, and
(ii) it may lower the spatial symmetry of the effective magnetic model.
In each case it may be expected to allow transitions forbidden by the original spin or space symmetries. Selection rules are necessary to determine experimentally the $\vec{D}$ vectors and see what processes are allowed and distinguish from the effects of other anisotropies, for example staggered g tensors in finite magnetic field. In some cases the Dzyaloshinsky-Moriya interaction alone does not permit transitions that have actually been observed, and this will lead us to consider a higher order of anisotropy: "dynamical Dzyaloshinsky-Moriya", in which the spin anisotropy is generated by distortions of the equilibrium lattice linear in the phonon coordinates. By a perturbative treatment of this coupling we derive an effective operator purely in terms of spin-operators and again give selection rules. These may explain optical transitions, at wave vectors $q = 0$, observed by ESR and infrared absorption, and, for finite values of q, mixing of nuclear and magnetic neutron scattering amplitudes.

We remark that in the frustrated cases we are discussing, effects such as the splittings may appear *linearly* in the strength of the Dzyaloshinsky-Moriya coupling: this is in contrast to the case of ordered antiferromagnets where, for example the contribution to the energy of a weakly ferromagnetic state is quadratic in the spin-orbit strength. In that case the exchange anisotropy, which is also quadratic,[2] may compensate at least in special cases.[3),4] Here the exchange anisotropy is of higher order and can safely be neglected.

This paper will review material presented in greater detail elsewhere, either for the static Dzyaloshinsky-Moriya[5] and the dynamic.[6),7]

§2. Dynamics: Examples of the influence of Dzyaloshinsky-Moriya

In this section we will discuss two cases where the direction of the Dzyaloshinsky-Moriya vectors can be predicted from the knowledge of the structure and have marked effects on the dynamics, producing an effective doubling or tripling of the low frequency mode, as observed in inelastic scattering of neutrons or in absorption of light.

2.1. $CuGeO_3$

We first consider the case of $CuGeO_3$, that has been much studied as the first inorganic example of a spin-Peierls system. In fact analysis of the magnetic susceptibility has shown that this system is, in addition to being a spin-Peierls system,

magnetically frustrated. It may be described by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_{1D} + \mathcal{H}_t, \tag{2·1}$$

$$\mathcal{H}_{1D} = \sum_{i,j} J_c(1+\delta(-1)^{i+j})\vec{S}_{i,j}\cdot\vec{S}_{i+1,j} + J_{2c}\vec{S}_{i,j}\cdot\vec{S}_{i+2,j}, \tag{2·2}$$

$$\mathcal{H}_t = \sum_{i,j} J_b\vec{S}_{i,j}\cdot\vec{S}_{i,j+1}, \tag{2·3}$$

c and b refer to crystalline axes of strongest and next-to-strongest magnetic exchange. The argument i is in the chain direction c, and j in the transverse direction b. The low energy magnetic excitations are well described by an alternating exchange $J_c(1\pm\delta)$ and second-nearest-neighbour coupling J_{2c} and an interchain coupling J_b. The numerical values of the couplings can be estimated, including the effects of interchain coupling $J_b/J_c = 0.15$, as $J_{2c}/J_c = 0.2$, dimerization $\delta \approx .065$, $J_c = 12.2$ meV.[8] The observation of a second mode[9),10)] (called an "optical mode" by the experimentalists), with weak intensity, was initially attributed to a slight difference in the dimerization of alternate chains[8] but in fact is more convincingly attributed to the Dzyaloshinsky-Moriya interactions. The alternation of dimerization from chain to chain (the factor $(-1)^{i+j}$) to give a chequer-board structure, is responsible for the fact that the mode is out of phase with the stronger mode. From the observed structure[6] the Dzyaloshinsky-Moriya vectors should be in the $\vec{c}$ direction, act between spins in the perpendicular (b) direction and alternate:

$$\mathcal{H}_{\perp}^{\mathrm{DM}} = \sum_{i,j}(-1)^j D_b\vec{c}\cdot(\vec{S}_{i,j}\times\vec{S}_{i,j+1}). \tag{2·4}$$

By making a rotation of the spin axes about the plane perpendicular to the Dzyaloshinsky-Moriya vectors, following an argument of Affleck and Oshikawa,[11),7)] the magnetic response can be deduced from that without the Dzyaloshinsky-Moriya interactions.

$$\begin{aligned}
S_D^{aa}(\vec{q},\omega) &= \cos^2\left(\frac{\theta}{2}\right)S^{aa}(\vec{q},\omega) + \sin^2\left(\frac{\theta}{2}\right)S^{bb}(\vec{q}-\vec{\pi},\omega),\\
S_D^{bb}(\vec{q},\omega) &= \cos^2\left(\frac{\theta}{2}\right)S^{bb}(\vec{q},\omega) + \sin^2\left(\frac{\theta}{2}\right)S^{aa}(\vec{q}-\vec{\pi},\omega),\\
S_D^{cc}(\vec{q},\omega) &= S^{cc}(\vec{q},\omega),
\end{aligned} \tag{2·5}$$

where θ is given by $\tan\theta = D_b/J_b$. $S^{\alpha\alpha}(\vec{q},\omega)$ are the dynamical structure factors for an isotropic model. $\vec{\pi} = (0,\pi)$ with respect to axes (q_c, q_b) and we neglect dispersion in the a direction as it is very weak. There should also be a weak exchange anisotropy producing an unobservably small splitting of the "acoustic mode" but we shall neglect this. Thus the "optic mode" is in fact the same mode seen at a different momentum transfer and should be visible with relative intensity: $(\frac{D_b}{J_b})^2$. From the observed intensity,[9] this gives an estimate of the magnitude of the Dzyaloshinsky-Moriya vector as $D \approx 0.4$ meV. We remark that a test of this mechanism should be the behaviour in finite field: as only the two polarizations transverse to the direction of

the Dzyaloshinsky-Moriya vector are involved in the doubling, in external magnetic field parallel to $\vec{D}$ the "optic mode" should split into two branches.

2.2. $SrCu_2(BO_3)_2$

The second case is that of Strontium Copper Borate. This compound is very interesting in that restricting first to isotropic interactions, it can be considered as planes of spins $\frac{1}{2}$ interacting via the Hamiltonian of the Shastry-Sutherland model in two dimensions. This model has the peculiarity that the product of singlet states on the closest dimers with the stronger exchange J is still an *exact* eigenvector when the frustrated second nearest neighbour interactions J' are included. [12] Furthermore this eigenvector is the ground state even for the relatively large value of the relative coupling $J'/J = 0.62$. This ratio is estimated either from the susceptibility [13] or the ratio of the energies of singlet states, seen in Raman scattering to triplet energies, seen by magnetic neutron scattering. [14] The interaction between planes is via couplings that are both weak and frustrated. When we take into account anisotropies the ground state will be perturbed. Nevertheless we have a rare example of a system with exponentially decaying magnetic correlations and a ground state that can be described as a local product of dimers with small corrections. Here we shall in fact consider a slightly idealized view of the compound, ignoring a small buckling of the planes. In this case the Dzyaloshinsky-Moriya couplings are strictly perpendicular to the planes, act between the next-nearest neighbour copper ions and, as shown in Ref. 14), give fine structure to the lowest lying magnon: i.e. a small splitting into three modes, as had been observed in the optical experiments of Nojiri et al. [15] and the neutron inelastic scattering. Taking into account renormalisation of the gap by the frustrated interactions, the splitting can be used to derive a precise numerical value of the Dzyaloshinsky-Moriya vectors $\vec{D}^c = 0.18$ meV.

The first effect of the Dzyaloshinsky-Moriya interactions is then to split the original triplet states. The more striking effect is that this splitting can dominate the dispersion. Propagation of the magnons in the Shastry-Sutherland lattice is weak: frustration of the interdimer couplings leads to a bandwidth that begins in sixth order in J'/J. The Dzyaloshinsky-Moriya interaction, in contrast, is *not* frustrated and the splitting is linear in $|\vec{D}|$. Thus the splitting due to Dzyaloshinsky-Moriya interaction is estimated to be larger than that of the dispersion due to interdimer coupling, even though that coupling is much larger. [14]

§3. Selection rules for Dzyaloshinsky-Moriya interaction

In the optical experiments of Nojiri et al., [10),15)] the resonance is from the ground state to the excited magnetic states. The observation of absorption requires some anisotropies: as the ground state without anisotropies is a spin singlet the operator corresponding to coupling with the probe magnetic field $\vec{h} \cdot \sum_i S_i$ applied to the ground state vanishes. As the Dzyaloshinsky-Moriya interaction mixes in non-singlet components the matrix elements to excited states may be non-zero. We can first estimate which ones are non-zero, and the dependence of the absorption strengths on external field, if we consider strictly local symmetries. [16),5)] This means we con-

sidering two spins in external uniform applied field $\vec{H}$ and with different possible polarizations of the resonating probe field $\vec{h}$:

$$\mathcal{H}(\vec{S}_1, \vec{S}_2) = J\vec{S}_1 \cdot \vec{S}_2 + \vec{D} \cdot (\vec{S}_1 \times \vec{S}_2) - \vec{H} \cdot (\vec{S}_1 + \vec{S}_2) - \vec{h}(t) \cdot (\vec{S}_1 + \vec{S}_2), \tag{3·1}$$

where $\vec{D}$ is the Dzyaloshinsky-Moriya vector and $\vec{H}$ is the external magnetic field. If $\vec{H}$ is parallel to $\vec{D}$ then the component of total spin in their common direction (z let us say) is a constant of the motion. Therefore only polarisations of $\vec{h}$ perpendicular to $\vec{H}$ or $\vec{D}$ will give absorption to states with $\Delta S^z = \pm 1$. The strength of absorption will be independent of the field as the eigenvectors do not change with H. For $\vec{H}$ perpendicular to $\vec{D}$, the total spin along the axis of $\vec{D}$ is no longer a constant of the motion: therefore there will be field dependence of the absorption of the three different components of the resonating field. These general properties are clearly shared by the lattice model but the exact dependencies for $\vec{H}$ perpendicular to $\vec{D}$ must be calculated. Explicit results are given in Ref. 5). Such selection rules are used to verify the direction of the Dzyaloshinsky-Moriya vector, especially in the case when symmetry alone cannot uniquely determine its direction. In addition there are further constraints we can call "lattice selection rules" which depend on the overall pattern of Dzyaloshinsky-Moriya vectors. Applied to the two structures we are considering, these have interesting consequences: in the case of $CuGeO_3$, from the argument we have mentioned of a rotation of axes of the spin variables, only the "optic mode" should be visible at $q = 0$. This was in agreement with older results,[17),18)] but the recent results of Nojiri et al.[10)] showed that both modes were visible. Similarly in the case of the $SrCu_2(BO_3)_2$, a lattice symmetry (reflection in a diagonal followed by rotation by π) leads to a zero amplitude for excitation of the triplet states, even in the presence of the Dzyaloshinsky-Moriya couplings. Of course, there are additional anisotropies due to slight buckling of the planes and anisotropies of the g tensors, but nevertheless the amplitude of the absorption in the two cases is somewhat surprising and this leads us to consider an alternative explanation in terms of a dynamical Dzyaloshinsky-Moriya interaction.

§4. Dynamical Dzyaloshinsky-Moriya interaction

We shall now consider a general anisotropic spin-phonon couplings corresponding to modulation of the exchange by linear coupling to lattice distortions. The term in the Hamiltonian coupling the phonon and spin operators is:

$$\mathcal{H}' = \sum_{ijd\alpha\beta} g_d^\alpha u_{id}^\alpha \vec{S}_i \cdot \vec{S}_j + d_d^{\alpha\beta} u_{id}^\alpha (\vec{S}_i \times \vec{S}_{j+1})^\beta, \tag{4·1}$$

where u_{id}^α is the α component of the displacement operator of atom d in unit cell i, g_d^α and $d_d^{\alpha\beta}$ are, respectively, the isotropic spin-phonon coupling and the dynamical Dzyaloshinski-Moriya interaction.

A typical case in Copper oxide is that there are frequently bridges of Cu_2O_2 with inversion symmetry in the equilibrium state. In the presence of a phonon, the

atomic positions may move so as to instantaneously remove the inversion symmetry, generating a Dzyaloshinsky-Moriya anisotropy.

Consideration of "dynamical" Dzyaloshinsky-Moriya terms were in fact motivated first by experiments in inelastic neutron scattering in which by measurement of the polarisation of scattered neutrons one can probe mixed "nuclear", i.e., involving the positions of the nuclei that scatter from neutrons via the strong interaction and "magnetic", i.e., interactions from the magnetic fields generated by the spin and orbital moments of electrons. [19),20)] In this case the geometry of the experiment is such that only correlations between the two terms can give a non-zero results, and furthermore, as a rotation can be measured only an interaction with a "handedness" such as the Dzyaloshinsky-Moriya interaction can give a non-zero result. [21)]

4.1. *Consequences for optical experiments: $\vec{e}$ field absorption*

In this paper we will not consider the effect in neutron scattering (see Ref. 7)) but the analogous effect in optical absorption. The essential point is that as the spin-orbit interaction mixes orbital and spin degrees of freedom, the separation between coupling to the magnetic field and the electric fields of the probe is no longer complete: in fact the electric field, which one would expect to couple only to the dipole electric moments will also effectively couple to the spins and give absorption to excitations considered normally simply "magnetic". The effects can be calculated perturbatively in the spirit of Fleury and Loudon [22)] for Raman absorption to magnetically excited states, but with the difference that the spin-orbit interaction is included, and that the excited states are involving a phonon excitation rather than an electronic excitation. [23)] The linear Hamiltonian is applied to the ground state and the excited magnetic state of the unperturbed Hamiltonian to first order. The matrix element of the electric dipole operator between the perturbed states $0'$ and $\alpha\prime$ including $\mathcal{H}'$ can then be written as that of an effective operator acting between the unperturbed states 0 and α. This operator is purely written in terms of spin operators:

$$\left\langle \alpha' | \sum_{id} q_d \vec{u}_{id} \cdot \vec{e} | 0' \right\rangle = \left\langle \alpha \mid \sum_{ij} \gamma \vec{S}_i \cdot \vec{S}_j + \vec{\delta} \cdot (\vec{S}_i \times \vec{S}_j) \mid 0 \right\rangle, \tag{4·2}$$

$$\gamma = \sum_s \frac{\Omega_s}{\omega_\alpha^2 - \Omega_s^2} g_s (\vec{\mathcal{D}}_s \cdot \vec{e}), \tag{4·3}$$

$$\vec{\delta} = \sum_s \frac{\Omega_s}{\omega_\alpha^2 - \Omega_s^2} \vec{d}_s (\vec{\mathcal{D}}_s \cdot \vec{e}), \tag{4·4}$$

where $\vec{\mathcal{D}}_s = \sum_d q_d \vec{\lambda}_{ds(q=0)}$ is the amplitude of the instantaneous electric dipole of the unit cell due to the phonon mode s with energy $\Omega_s = \Omega_{(q=0,s)}$ which displaces the charges q_d. The final magnetic state has an energy ω_α. $g_s = \sum_{d,\alpha} g_d^\alpha \lambda_{ds}^\alpha$ is the amplitude of the variation of the magnetic exchange energy due the atomic distortions of the phonon s (λ_{ds}^α is the amplitude of the motion of the atom d, in the direction α due to the phonon s at $q = 0$). Here the sum ij is assumed to run over a set of equivalent neighbours: more generally there could be a set of γ and $\vec{\delta}$

for different inequivalent neighbours. The selection rules for the contribution of a particular phonon mode s to contribute are that:

- (i) $(\vec{\mathcal{D}}_s \cdot \vec{e}) \neq 0$: the virtual phonon s creates distortions that carry an instantaneous electric dipole $\mathcal{D}_s$. In other words, the phonon s must be optically active.
- (ii)
  - $g_s \neq 0$: The distortion of the unit cell due to the phonon s modulates the magnetic exchange between the spins. Only spin-conserving transitions at $\Delta S_{\rm tot} = 0$ are allowed.
  - $\vec{d}_s \neq 0$: The distortion of the unit cell due to the phonon s must break instantaneously the symmetry by inversion at the middle of the bond; so as to allow an instantaneous Dzyaloshinsky-Moriya interaction of amplitude $\vec{d}_s$. Transitions between different spin states $\Delta S_{\rm tot} = 0, \pm 1$ are allowed.

Note that the selection rules involve detailed knowledge of different phonons. Directions of the vector $\vec{d}_s$ are constrained by the symmetry rules for static Dzyaloshinsky-Moriya interactions applied to structure distorted by the given phonon s from the equilibrium structure. The factors $(\vec{\mathcal{D}}_s \cdot \vec{e})$ can be measured independently from the intensity at the frequency Ω_s of the *real* phonon creation. For external magnetic field parallel to $\vec{\delta}$ the total component of spin in this common direction α say is conserved (if this is the sole form of anisotropy or, if not, if this direction is an axis of symmetry shared with the other anisotropies) and only transitions to the field-independent level $\Delta S^{\alpha} = 0$ should be observed: thus the selection rule is quite different from that for magnetic transitions with a (static) Dzyaloshinsky-Moriya interaction in the same direction. Again for a field in the transverse directions there will be transitions to the three states with magnetic field dependence that could be calculated as in Ref. 5). Note that the wave functions must include any static Dzyaloshinsky-Moriya interaction $\vec{D}$ and the matrix elements involve in general different vectors $\vec{\delta}$. From the absolute intensities one should be able to deduce the magneto-elastic constants g_s, and the components of $\vec{d}_s$. In a full comparison to experiment it is desirable to control the polarisations of the $\vec{e}$ and $\vec{h}$ fields of the probe separately. Frequently only the direction of propagation, i.e. their vector product, is controlled with respect to the crystal axes. Recent experiments by Rõõm et al.[24] of infrared absorption with polarised electromagnetic waves seem to be consistent with the selection rules enunciated: for example in $CuGeO_3$ extinction for $\vec{e} \parallel c$[25),26)] follows as the mirror planes of the equilibrium structure are maintained under an assumed distortion of the atoms along the c-axis. In $SrCu_2(BO_3)_2$ we have also found[7] good agreement with the experiments,[24] at least by using a simplified view of the structure. If, for example, $\vec{e}$ is taken in the (ab) plane and we assume that even with the virtual phonon that couples to such an electric field the (ab) plane remains a mirror plane. In this case the effective operator, by the standard symmetry arguments, will have components along the c-axis only. As argued above, there should be absorption to the $S_z = 0$ mode only, provided the external magnetic field $\vec{H} \parallel c$, and field-dependent absorption to the (static) Dzyaloshinsky-Moriya interaction split lines for $\vec{H} \perp c$.

We will not compare to the neutron case in detail [6),7)] but note that in calculating the relevant matrix element for "nuclear" scattering to a magnetic state, while the same magneto-elastic constants and vectors will enter, the vectors $\vec{\delta}$ will differ as $(\vec{\mathcal{D}}_s \cdot \vec{e})$ for example will be replaced by the phonon form factor for nuclear neutron scattering. The selection rules involve the scattering geometry, and therefore different phonons may contribute.

§5. Conclusions

We have reviewed results for the selection rules governing optical absorption, in particular in the presence of both static Dzyaloshinsky-Moriya interactions and terms generated by coupling to phonons that lower the symmetry. In the second case both nuclear and magnetic scattering amplitudes are mixed in inelastic neutron scattering, and optically, magnetic states may be excited by the electric field component of the probe. Testing of these effects can be by a full polarization experiments in both cases: in neutron scattering by polarisation of both incoming and outcoming beams, and, in the optical experiments, by controlling the polarisation of the electric and magnetic components.

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