# Static and Dynamical Dzyaloshinsky-Moriya interactions in gapped spin systems

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Anisotropic spin-spin interactions of Dzyaloshinsky and Moriya symmetry are generally considered weak, as they depend on the spin-orbit couplings. In spin systems with gapped ground states they can, however, have rather strong effects. We will discuss recent results related to the results of neutron scattering and ESR for $SrCu_2(BO_3)_2$. Inclusion of the Dzyaloshinsky-Moriya interactions can explain much of the dynamics of the systems discussed, in particular the splitting and dispersion of the triplet modes. Some effects remain to be explained, however. Symmetries of the crystal lead to the prediction of zero intensity for transitions, for example between the ground state and the triplets observed in ESR.

We present recent calculations of the effects of anisotropic terms generated dynamically, i.e. linearly in the phonon coordinates. We discuss how this leads to a novel mechanism to explain the ESR intensities. For polarized neutron scattering experiments, we can calculate the mixing of nuclear and magnetic scattering amplitudes by such a term and how such mixing should be observed.

## §1. Introduction

In quantum spin $\frac{1}{2}$ magnets with the leading anisotropies in powers of the spin-orbit coupling are Dzyaloshinsky-Moriya interactions [1),2)] $\sum_{i,j} \vec{D}_{i,j}.(\vec{S}_i \times \vec{S}_j)$, (with sum over neighbours $i$ and $j$), which appear in first order and anisotropy in the exchange, which appears in second order. In the copper oxides such terms are an order of magnitude weaker than the isotropic exchange $D = (\frac{\Delta g}{g})J$ with $\Delta g = g - 2$ yet we will see that they can have strong effects on the dynamical response. In a gapped system with singlet ground states, unlike an ordered antiferromagnet, the relatively weak Dzyaloshinsky-Moriya terms will not close the gap to allow, for example, weak ferromagnetism with a spontaneously broken symmetry. The interactions can have important consequences in addition to an explanation of transitions otherwise forbidden by overall spin rotation symmetry. They may give splittings *linear* in the spin-orbit strength which cannot be cancelled by the higher order terms in the exchange [3),4)]. They may also enlarge the effective magnetic unit cell, giving extra branches to excitations. Furthermore the dispersion of excitations due to the stronger isotropic exchanges may be strongly reduced by frustration, allowing the splittings of the Dzyaloshinsky-Moriya terms to dominate. We will illustrate these points by referring to recents experiments and theory in the geometrically frustrated $SrCu_2(BO_3)_2$.

While the Dzyaloshinsky-Moriya interactions lower the spin symmetry allowing

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certain transitions forbidden from a completely isotropic singlet state, it turns out that there are transitions observed that should be forbidden even in their presence. This leads us to consider a higher order of anisotropy: "dynamical Dzyaloshinsky-Moriya ": spin-phonon terms in the Hamiltonian which modulate not only the exchange strength but in addition modify the anisotropic terms of Dzyaloshinsky-Moriya form. This is especially interesting when the terms generated are forbidden in the equilibrium structure. We can treat such generalised spin-phonon terms perturbatively and for each operator corresponding to a physical process generate an effective operator in terms of spin-operators. 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.

This paper will review material presented in greater detail either for the static Dzyaloshinsky-Moriya [5),6)] and the dynamic [7),8)].

## §2. Dynamics: Examples of the influence of Dzyaloshinsky-Moriya in $SrCu_2(BO_3)_2$

$SrCu_2(BO_3)_2$ is exciting in that it can be considered as planes of spins $\frac{1}{2}$ interacting via the Hamiltonian of the Shastry-Sutherland model in two dimensions. The interaction between planes is via couplings that are both weak and frustrated. The Shastry-Sutherland 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 [9)]. Furthermore this eigenvector is the ground state even for the relatively large value of the relative coupling $J'/J = 0.62$. While the ground state does not change the dynamics are strongly renormalized by the weaker coupling, and in fact the coupling is close to the estimated value $J'/J \approx 0.68$ where there is a quantum phase transition, possibly to a plaquette state [10)]. This ratio is estimated either from the susceptibility [11)] or the ratio of the energies of singlet states, seen in Raman scattering, to triplet energies, seen by magnetic neutron scattering [5)].

### 2.1. *Splitting of the triplet mode by the Dzyaloshinsky-Moriya interaction*

It was observed in [12)], however, that the lowest triplet state is not simply renormalized in energy by $J'$ but split as well, and this was confirmed by neutron scattering which gave the dispersion. This lead us to consider the Dzyaloshinsky-Moriya interactions in $SrCu_2(BO_3)_2$ from the usual symmetry rules of Dzaloshinsky and Moriya. Apart from a small "buckling" of the planes (which we shall mention later) the presence of a centre of inversion leads to zero Dzyaloshinsky-Moriya interaction between spins of the more strongly coupled dimers, and for the more weakly interacting dimers, as the plane $(ab)$ of the spins is a mirror plane, Dzyaloshinsky-Moriya vectors are strictly perpendicular with alternate signs as shown in Figure 1, taken from reference [5)]. Inclusion of such an anisotropy will give corrections to the Shastry-Sutherland ground state but these can be estimated perturbatively. To understand the effect of the dynamics one can first ignore $J'$ and calculate the dispersion coming from the term $D$, which gives a dispersion even in linear order, corresponding

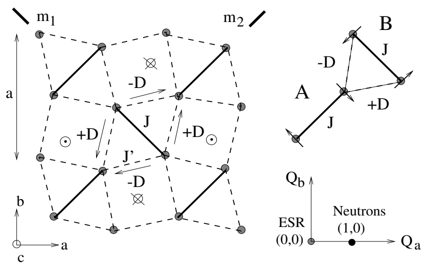


Fig. 1. Dzyaloshinsky-Moriya vectors determined from the symmetry, ignoring the weak buckling of the (ab) plane. The arrows indicate the sense of the neighbours $i$ and $j$ in the definition of the vectors which are perpendicular to the plane with directions indicated

physically to the fact the the Dzyaloshinsky-Moriya vectors are not frustrated: The dispersion of the two modes $\pm$ (each is twice degenerate with $S^z = \pm 1$) is therefore proportional to $D$ :

$$\omega_{\mathbf{q}}^{S^z=\pm1,\pm} = J \pm 2D\cos(q_a a/2)\cos(q_b a/2) \qquad (2\cdot1)$$

where $f(\mathbf{q}) = \cos(q_a a/2)\cos(q_b a/2)$. On the other hand, the Dzyaloshinski-Moriya interaction has no effect on the $S^z = 0$ component of the triplet, so that its energy remains equal to $J$ ($\omega_{\mathbf{q}}^{S^z=0} = J$). Thus for $J' = 0$ the two transverse modes will be split by $4D$. Physically the splitting into two modes results from the fact that without the Dzyaloshinsky-Moriya vectors the effective magnetic unit cell is two times smaller, as the two dimers per unit cell are equivalent, at least in the limit of small $J'$. The different signs of the Dzyaloshinsky-Moriya vector to the left and right of a given dimer lower the effective symmetry, doubling the number of modes. As $J'/J$ is not small, the numerical factor of 4 is renormalised to be a function of $J'/J$ and this we have calculated by finite size scaling on small clusters, as shown in Figures 2 and 3. Even close to the quantum phase transition the gap to the triplet while renormalised to smaller values remains large and such calculations converge quite rapidly. Using the experimental value of $J'/J$ the factor is renormalized to 2.0 [5]. From the experimental values from the optical experiments of Nojiri et al [12] and the neutron inelastic scattering [5), 13], we can deduce the absolute value of the Dzyaloshinsky-Moriya interaction $\vec{D}^c = 0.18$ meV.

The same cluster calculation can be used to estimate the dispersion (see the lower line in Figure 3) which is relatively weak as it begins at sixth order in $J'/J$. It is striking that this splitting can dominate the dispersion even though it is determined by an interaction an order of magnitue smaller than the isotropic interaction.

While this theory gives good quantitative explanation of the optical and neutron experiments with wave vector $(q_a, q_b) = (0, 0)$ the most recent high resolution experiments [13] indicate possible discrepancies near $(q_a, q_b) = (\pi, 0)$. The gap at that point is interpreted as a sizeable transverse component to the Dzyaloshinsky-Moriya vector. This had been assumed to be negligible as the buckling in the plane responsible for breaking the symmetry that would forbid it is small. We must still add the proviso that while it appears now that a transverse component should be

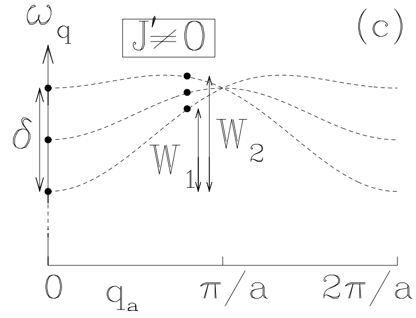


Fig. 2. For finite $J'$, exact diagonalization for a cluster of 20 spins gives the energies of two reciprocal points (the dots)

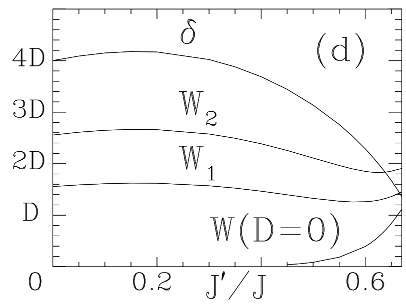


Fig. 3. Renormalization of the splitting $\delta$ and the widths $W_1$ and $W_2$ defined from Fig 2. $W$ is the bandwidth for the dispersion for zero $D$, with scale corresponding to the physical value of $D$.

included, especially in view of the behaviour in finite magnetic field [13)], a reliable quantitative estimate is not yet possible, especially as the gap is smaller, close to the estimated bandwidth from the $J'$ terms. A reliable calculation will probably need diagonalisation of larger clusters [14)].

### 2.2. *Selection rules for Dzyaloshinsky-Moriya interaction*

In the optical experiments of Nojiri et al [12)], 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 $\vec{h}.\sum_i S_i$ 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 must use the symmetries to predict which ones are non-zero [6)]. For $SrCu_2(BO_3)_2$ , a lattice symmetry (reflection in a diagonal followed by rotation by $\pi$) leads to a zero amplitude for excitation of the triplet states, even in the presence of the Dzyaloshinsky-Moriya couplings. As we have mentioned for the dispersion, there are additional anisotropies due to 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.

## §3. 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:

$$\mathcal{H}' = \sum_{ijd\alpha\beta} g_d^\alpha u_{id}^\alpha \vec{S}_i.\vec{S}_j + d_d^{\alpha\beta} u_{id}^\alpha (\vec{S}_i \times \vec{S}_{j+1})^\beta \tag{3·1}$$

where $u_{id}^\alpha$ is the $\alpha$ component of the displacement operator of atom $d$ in unit cell $i$, $g_d^\alpha$ and $d_d^{\alpha\beta}$ are, respectively, the isotropic spin-phonon coupling and the dynamical Dzyaloshinsky-Moriya interaction. For example the inversion symmetry that forbids the Dzyaloshinsky-Moriya interaction within the strongly bound dimer in $SrCu_2(BO_3)_2$ may be lifted instantaneously by a phonon mode, generating a Dzyaloshinsky-Moriya anisotropy. Consideration of "dynamical" Dzyaloshinsky-Moriya terms were in fact motivated first by experiments in inelastic neutron scattering.

### 3.1. *Polarisation analysis of inelastic neutron scattering*

In another paper in this symposium [15] L.P. Regnault had discussed how experiments with polarisation analysis in all directions of incident and scattered beams of neutrons can give new information on correlation functions. One can probe mixed "nuclear", and "magnetic" correlations [16),17]. If a rotation of the outcoming spin polarisation around some axis can be measured, only an interaction with a "handedness" such as the Dzyaloshinsky-Moriya interaction can give a non-zero result [18]. Such experiments are therefore ideal for their sensitivity to the dynamical Dzyaloshinsky-Moriya terms we have discussed here. Here we will give a simple introduction to the contribution of the terms to inelastic scattering.

Let us consider an incident beam of neutrons fully polarised, let us say in the "up" direction of an axis $z$. The spin part of the incident wave function is then

$$\psi_i = |\uparrow> \tag{3·2}$$

The neutron is scattered both by the atomic nuclei via the strong interaction and the coupling via its spin to the magnetic field generated by the magnetization of the sample. The spin part of the outgoing wave function for momentum transfer $\kappa$ is, by the Born approximation,

$$\psi_o = (\mathcal{N} + \vec{\mathcal{M}}_z^\perp)|\uparrow> + (\vec{\mathcal{M}}_x^\perp + i\vec{\mathcal{M}}_y^\perp|\downarrow> \tag{3·3}$$

$$\mathcal{N}(\kappa) = <\phi_\alpha|\sum_{id} a_d \exp i\vec{\kappa}\cdot\vec{r}_{id}|\phi_0> \tag{3·4}$$

$$\vec{\mathcal{M}}^\perp(\kappa) = <\phi_\alpha|\vec{\kappa}\times(\vec{S}_{\vec{\kappa}}\times\vec{\kappa})|\phi_0> \tag{3·5}$$

Here $\phi_0$ is the ground state and $\phi_\alpha$ the state excited by inelastic scattering of the neutron. $\vec{S}_{\vec{\kappa}}$ is the Fourier transform of both spin and orbital contributions to the magnetization. The momentum transfer $\kappa$ can be chosen by the geometry of the experiment: it is frequently taken parallel to the axis of polarisation in order to eliminate the term $\vec{\mathcal{M}}_z^{\perp}$. In this configuration the spin of the scattered neutron will then be simply $\sigma_z = (|\mathcal{N}|^2 - |\vec{\mathcal{M}}|^2)/2$ In this case the "nuclear" term $\mathcal{N}$, is simply separated from the "magnetic" $\mathcal{M}$ by the amplitude of non-spin-flip to spin-flip scattering. When we resolve in energy, if there is separation of the nuclear and magnetic degrees of freedom in the Hamiltonian either $\mathcal{M}$ or $\mathcal{N}$ vanishes and the coherent scatted beam will be either fully polarised "up" or "down". If the excitations are mixed, then it becomes interesting to measure, for example, the spin in another direction. We shall consider the expectation value of the spin in a transverse direction $x$. From the Pauli matrices this is just $\sigma_x = \left[(\vec{\mathcal{M}}_x + i\vec{\mathcal{M}}_y)^*\mathcal{N} + \mathcal{N}^*(\vec{\mathcal{M}}_x + i\vec{\mathcal{M}}_y)\right]/2$. Thus by measuring in a transverse direction we have an alternative measure of the two amplitudes. It has the advantage that if one is small, the result is linear rather than quadratic and that by measuring in two perpendicular directions we can also measure relative phases of $\mathcal{M}$ or $\mathcal{N}$. Interpretation of the polarisation requires calculation of the matrix elements $\vec{\mathcal{M}}$ and $\mathcal{N}$ in a case when they are both non-vanishing. Here we shall consider the case of $\mathcal{N}$, the nuclear scattering amplitude, for an excitation considered "magnetic": ie which in the absence of spin-phonon terms would have vanishing nuclear amplitude. The matrix element of the nuclear 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 $\alpha$. This operator is purely written in terms of spin operators:

$$\langle \alpha' | \sum_{id} a_d \exp i\vec{\kappa}\cdot\vec{r}_{id} | 0' \rangle = \langle \alpha \mid \sum_{ij} \exp(i\vec{\kappa}\cdot\vec{R}_i) \left(\gamma_{\vec{\kappa}} \vec{S}_i.\vec{S}_j + \vec{\delta}_{\vec{\kappa}}.(\vec{S}_i \times \vec{S}_j)\right) \mid 0 \rangle \tag{3·6}$$

where $\vec{R}_i$ is the equilibrium position of the atom $i$ and the two terms depend on $\gamma_{\vec{\kappa}}$ and $\vec{\delta}_{\vec{\kappa}}$,

$$\gamma_{\vec{\kappa}} = i \sum_s \frac{\Omega_{\kappa s} a_s(\kappa)}{\Omega_{\kappa s}^2 - \omega_\kappa^2} g_{\vec{\kappa}s} \tag{3·7}$$

$$\vec{\delta}_{\vec{\kappa}} = i \sum_s \frac{\Omega_{\kappa s} a_s(\kappa)}{\Omega_{\kappa s}^2 - \omega_\kappa^2} \vec{d}_{\vec{\kappa}s} \tag{3·8}$$

Note that $\vec{r}_{id}$ is the (instantaneous) position of an atom, i.e. with phonon mouvement included, in contrast to $\vec{R}_i$ (where we have suppressed the factor $d$ to indicate that we take terms including only one magnetic ion per unit cell). Thus we have integrated out the phonon motion by perturbing the excited and ground states to first order in the spin phonon coupling. We now need only calculate matrix elements of an effective spin Hamiltonian. The coefficients are defined as follows: $a_s(\vec{\kappa}) = \sum_{id} a_d \exp i\vec{\kappa}\cdot\vec{r}_{id}(\vec{\kappa}.\vec{\lambda}_{\vec{\kappa}ds})$ is the nuclear form factor of the phonon mode $s$ ($\vec{\lambda}_{\vec{\kappa}ds}$ is the displacement vector of the atom $d$, due to the phonon $s$ at $q = \vec{\kappa}$). Its magnitude is measurable independently from the intensity of *real* phonon scattering at energy $\Omega_{\kappa s} = \Omega_{(\vec{q}=\vec{\kappa},s)}$. The final magnetic state has an energy $\omega_\alpha$. $g_s = \sum_{d,\alpha} g_d^\alpha \lambda_{\vec{\kappa}ds}^\alpha$ is the amplitude of the variation of the magnetic exchange energy due the atomic distortions of the phonon $s$ ($\lambda^{\alpha}_{\vec{\kappa}ds}$ is the $\alpha$ component of $\vec{\lambda}_{\vec{\kappa}ds}$). Here the sum $ij$ is assumed to run over a set of equivalent neighbours: more generally there could be a set of $\gamma$ and $\delta$ for different inequivalent neighbours. A particular phonon mode $s$ contributes only if $a_s(\vec{\kappa}) \neq 0$: the virtual phonon $s$ creates distortions that have non-zero nuclear amplitude. That is, the phonon is not purely transverse. The two terms may then be generated provided:

- $g_{\vec{\kappa}s} \neq 0$: The distortion of the unit cell due to the phonon $s$ modulates the magnetic exchange between the spins. This term can give non-spin-flip transitions to excited singlet states at zero temperature: $\Delta S_{tot} = 0$ are allowed.
- $\vec{d}_{\vec{\kappa}s} \neq 0$: 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 $\vec{d}_{\vec{\kappa}s}$. Directions of the vector $\vec{d}_{\vec{\kappa}s}$ are constrained by the symmetry rules for static Dzyaloshinsky-Moriya interactions applied to the equilibrium structure distorted by the given phonon $s, \vec{\kappa}$. Transitions between different spin states $\Delta S_{tot} = 0, \pm 1$ are allowed.

In the second case we see that scattering to the triplet state, normally purely spin-flip will have a small component transverse from the nuclear amplitude. There is then a rotation away from the pure spin-flip direction of the spin of the scattered neutrons by an angle that is essentially $|\frac{\mathcal{N}}{\mathcal{M}}|$. This gives an estimate of the rotation of the polarisation:

$$\left(\frac{\Delta g}{g}\right)\left(\frac{\Omega_{\kappa s}E}{\Omega^2_{\kappa s}-\omega^2_{\alpha}}\right)\sqrt{\frac{\frac{d^2\sigma}{d\omega d\Omega}_{phonon\ s}}{\frac{d^2\sigma}{d\omega d\Omega}_{triplet\ \alpha}}} \tag{3·9}$$

The factor $E$ here is the modulation of the isotropic exchange and can be approximately estimated from the contribution of the phonon to change in the angle of superexchange. The factor $\frac{\Delta g}{g}$ is from the usual Moriya estimate of the anisotropic part. The ratio of inelastic cross sections $(\frac{d^2\sigma}{d\omega d\Omega})$ is, as already mentioned, measurable independently from the relative intensities of real phonon emission at the phonon frequency to the (spin-flip) magnetic scattering. The angle about which the polarisation will turn depends on the vectors $\vec{d}_{s,\vec{\kappa}}$. Estimates of the values expected for rotations [7),8)] expected in copper oxides give results that are a few degrees in the most favourable cases, and in general much smaller, essentially because of the double constraint on both the form factor of the phonon and the generation of spin anisotropy.

We have performed a similar calculation to the above for the possibility of electric field induced optical transitions [7),8)] We do not enter into details here but note that in calculating the relevant matrix element for electric dipole absorption to a magnetic state, while the same magneto-elastic constants and vectors will enter and there is an effective matrix element of the same form as at $\kappa = 0$ but the vectors $\vec{\delta}$ will differ as $a_s(\kappa)$, for example, will be replaced by an optical form factor.

## §4. Conclusions

We have reviewed effects of both static Dzyaloshinsky-Moriya interactions and terms generated by coupling to phonons that lower the symmetry in the spin gapped compound $SrCu_2(BO_3)_2$. While the observed magnetic modes are well explained by inclusion of Dzyaloshinsky-Moriya interactions, some puzzles remain. We have advanced the idea that some of these puzzles may be resolved by the dynamic terms. Such terms have the effect of mixing nuclear and magnetic scattering amplitudes in neutron scattering, and allowing excitation by the electric field component of the probe electromagnetic field to excited magnetic states. Future neutron experiments with full analysis of the spins and optical experiments using polarisation of the electric and magnetic components of the light should test these ideas.

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