

When and how can structural chirality and electron spin be coupled?

1. Comparing electrical magnetochiral anisotropy and chirality-induced spin selectivity

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2. Chirality induced spin selectivity in chiral crystals

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Structurally chiral materials lack mirror symmetry but time-reversal invariant while ferromagnetic materials do not have time-reversal symmetry. Therefore, structural chirality and ferromagnetism have been considered as separate, unrelated entities, but possible connection between them has been suspected for more than a century partly because both cause the rotation of the linear polarization of incident light.

In 2012, Naaman and co-workers discovered the phenomenon called chirality-induced spin selectivity (CISS) where electric current passing through chiral molecules exhibits strong spin polarization which depends on the chirality (right-handedness or left-handedness) of the molecules [1]. Motivated by the discovery of CISS, interaction between chiral bio-molecules and ferromagnetic substrates was studied to investigate the origin of their homochirality. In fact, adsorption of chiral molecules on ferromagnetic substrates (double-stranded DNA molecules Co and Ni) was found to depend on the enantiomer (left-handed or right-handed) and on the magnetization directions of the substrates (parallel or antiparallel to the surface normal) [2]. The growth of one enantiomer of an RNA precursor over the other was also reported to depend on the magnetization direction of Fe_3O_4 substrates [3].

CISS was observed not only in assemblies of organic chiral molecules mentioned above but also in inorganic chiral crystals, such as the paramagnetic phase of $\text{Cr}_{1/3}\text{NbS}_2$, where the Cr atoms are intercalated in spiral forms [4], transition-metal disilicides NbSi_2 and TaSi_2 with chiral structures, [5], and chiral crystals of Te [6]. Various possible mechanisms have been discussed so far, such as orbital Edelstein effect [7], vibration of chiral molecules [9], electron-phonon coupling at molecule-substrate interfaces [8], and chirality-induced spin current [10], but the problem remains elusive [11]. Because the chiral molecules do not contain heavy elements, spin-orbit coupling (SOC) had been thought to play only a minor role although

it had been difficult to conceive other origins that could lead to finite coupling between the structural chirality and the spin polarization.

In the first paper, Rikken and Avarvari have examined relationship between CISS measured through magnetoresistance (CISS-MR) and a closely related phenomenon, electrical magneto-chiral anisotropy (eMChA). In eMChA as shown in Fig. 1, electrical resistance R depends on the handedness of the conducting sample and on the relative orientation of electrical current \mathbf{I} and magnetic field \mathbf{B} or magnetization \mathbf{M} . In CISS, the electron transmission coefficient T depends on the handedness of the conducting sample and on the relative orientation of charge velocity \mathbf{v} and spin polarization \mathbf{S} . The relationship between R , \mathbf{I} , and \mathbf{B} in eMChA and that between T , \mathbf{v} , and \mathbf{S} in CISS are given by the same forms up to the first order as indicated in Fig. 1. However, note that crucial data of CISS-magnetoresistance (CISS-MR) and eMChA have been taken outside the first-order range and are dominated by non-linear effects.

Difficulties in experimentally identifying the microscopic mechanisms of CISS lie in the diversity of experimental conditions, ranging from thermodynamic equilibrium, steady state, to transient state. For example, Onsager’s reciprocal relation, which is applicable to diffusive transport starting from thermal equilibrium, may not apply to CISS-MR where electron transmission rather than diffusive transport may dominate. They pointed out that recent CISS-MR and eMChA data are not consistent with each other, and tentatively attributed the inconsistencies to the extrinsic effect of interfaces between the chiral samples and the electrodes in CISS-MR samples.

In the second paper, Yang *et al.* focused on inorganic crystals with chiral structures and carried out first-principles calculations to identify the still-unknown microscopic mechanism of CISS. They have found that SOC induces both the spin and orbital polarization of the charge current and that both increase with traveling distance until they saturate. This suggests that, even if SOC is very weak as in the organic chiral molecules on substrates, strong spin polarization may be realized after the charge current has travelled a long distance. Spin polarized current thus produced leads to an eMChA when probed by magnetic electrodes [12]. Yang *et al.* have also found that the spin polarization is more sensitive to the SOC strength than the orbital polarization.

Recently, “magnetochiral tunneling effect”, an STM version of CISS, was observed using a magnetized tip to detect spin-polarized current from the chiral crystal $\text{Co}_{1/3}\text{NbS}_2$ in the paramagnetic state [13]. The recent observation of strong x-ray circular dichroism (XCD) from the chiral crystal Ni_3TeO_6 in the paramagnetic state might be understood as an x-ray-photon version of CISS [14] (distinct from x-ray magnetic chiral dichroism (XMChD), an effect observed with fixed x-ray polarization but reversed magnetization direction [15]).

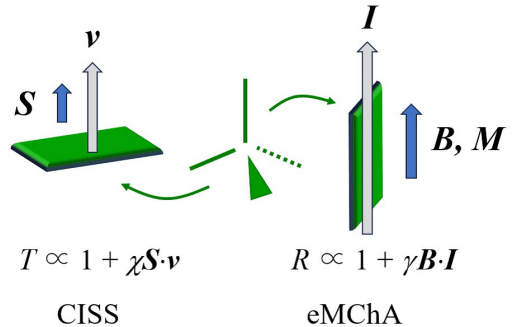


Figure 1: Schematic pictures of CISS and eMChA measurements on chiral materials. Charge transmission coefficient T and resistivity R are given to the first order in \mathbf{v} , \mathbf{S} , \mathbf{I} and \mathbf{B} (for definition, see the text). χ and γ change signs depending on the chirality of the sample.

Here, the incident x rays were supposed to break time reversal symmetry of the Ni_3TeO_6 crystal in analogy to charge current in CISS. According to “symmetry-operational similarity” proposed by Cheong [16], large spin polarization may have become symmetry-allowed under x-ray irradiation. It is interesting to note that the simultaneous breaking of time-reversal and chiral symmetries as discussed above has also been proposed to occur in the pseudogap phase of the high T_c cuprates through an intra-unit-cell loop current order [17] which may be realized by a pair-density-wave (PDW) order [18, 19]. The chiral structural distortion induced by the loop current order may lead to the chirality of phonons, too, and to the intriguing thermal Hall effect reported recently [20].

All the chiral materials exhibiting “CISS” discussed above are non-magnetic or paramagnetic, but the XCD of Ni_3TeO_6 was found to be enhanced when entering its collinear antiferromagnetic (AFM) phase [14]. Because the collinear AFM order preserves effective time-reversal symmetry (invariant under time reversal plus translation by a lattice unit vector), one has to go beyond the symmetry argument and explore microscopic mechanisms that enhance XCD in the AFM phase. More generally, effects of AFM order on CISS have not been studied yet, but may provide interesting problems that can be explored in future studies.

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