Orbital angular momentum-driven anomalous Hall effect

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Article

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Orbital angular momentum-driven anomalous Hall effect

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Abstract

Orbital angular momentum (OAM) plays a central role in regulating the magnetic state of electrons in non-periodic systems such as atoms and molecules. In solids, on the other hand, OAM is usually quenched by the crystal field, and thus, has a negligible effect on magnetisation. Accordingly, it is generally neglected in discussions around band topology such as Berry curvature and intrinsic anomalous Hall conductivity (AHC). Here, we present a theoretical framework demonstrating that crystalline OAM can be directionally unquenched in transition metal oxides via energetic proximity of the conducting $d$ electrons to the local magnetic moments. We show that this leads to ‘composite’ Fermi-pockets with topologically non-trivial OAM textures. This enables a giant Berry curvature with an intrinsic non-monotonic AHC, even in collinearly-ordered spin states. We use this model to explain the origin of the giant AHC observed in the forced-ferromagnetic state of EuTiO$_3$ and propose it as a prototype for OAM driven AHC.
Introduction: The anomalous Hall effect (AHE) is a variant of the Hall effect that arises in magnetic conductors even in the absence of an external magnetic field. AHE has both extrinsic contributions, i.e. skew-scattering mechanisms, and intrinsic contributions, from the geometric and topological properties of electronic states as formulated by Berry\cite{berri1}. In a solid, we can treat the momentum space of the lattice, also known as the Brillouin zone (BZ), as a parameter space which in turn defines a manifold of electronic states that has a defined connection and curvature, commonly known as Berry connection and Berry curvature (BC)\cite{berri2}, respectively. Heuristically the Berry connection can be seen as the $k$-space dual of the magnetic potential, which gives us some intuition as to its connection to the AHE, and how we can have a Hall current in the absence of an external magnetic field. BC in a system can typically be attributed to energetically proximate, or even degenerate, energy eigen-states that are guaranteed by the symmetries and degrees of freedom of the system. Typically the focus is on degrees of freedom such as spin or valley index, OAM is rarely considered as it is typically quenched or of a trivial nature in solids.

To explore the role of OAM in BC, we examine a crystal structures which possesses a collinear spin angular momentum (SAM) texture, but OAM varies across the BZ due to externally-imposed anisotropies in the underlying CF. The resulting OAM texture generates an emergent spin-orbit (SO) field that varies non-trivially over $k$-space, despite the collinearity of SAM, leading to composite FPs (energy iso-surfaces of each band) with regions of different orbital character separated by sharp boundaries. This, in turn, generates a large OAM-dependent BC, dominating the contribution to the intrinsic AHC. As an example, we apply our model to the forced ferromagnetic (FFM) phase of the rare-earth transition-metal oxide perovskite EuTiO$_3$. Despite its collinear SAM, we show this system exhibits non-collinear OAM resulting from the symmetry breaking of the underlying CF, directionally controllable by an external magnetic field. Such an external control on the directional anisotropy of the OAM enables us to directly control the OAM induced BC through the magnetic field orientation. Through this scheme, we explore the Fermiology and Berryology of the low energy bands, hosting the charge carriers, and find they are directionally warped by an RKKY interaction between Eu-4$f$ moments, mediated through Ti-3$d$ states. This is shown to lead to an intrinsic non-monotonic AHC that have been experimentally observed for EuTiO$_3$ but so far eluded rigorous explanation\cite{eu1,eu2}.

Results

Fermiology of EuTiO$_3$: EuTiO$_3$ is a rare-earth perovskite\cite{eu3} that has the property of separate conduction (Ti) and magnetic (Eu) centres, allowing it both a high carrier mobility (in excess of 3000cm$^2$V$^{-1}$s$^{-1}$ at 2K)\cite{eu4} and metamagnetism\cite{eu5}. The system naturally has a weak anti-ferromagnetic order (AFM) at temperatures below 5.5 K\cite{eu6,eu7} which effectively can be regarded as a paramagnetic (PM) phase, as depicted in Fig. 1a. Under a low magnetic field (2.1 T), EuTiO$_3$ transitions to a FFM order\cite{eu8,eu9,eu10} as depicted in Fig. 1b. This
leads to a complete spin-polarisation of carriers for a broad range of carrier densities, \( n_c \), as experimentally verified through magnetotransport measurements\(^7\) for \( n_c \) up to \( 4 \times 10^{19} \text{cm}^{-3} \). We have schematically illustrated the resulting change in the electronic structure in Fig. 1c and d. The low energy conduction bands here are dominated by the Ti-3\(d\) – \(t_{2g}(d_{xy}, d_{xz}, d_{yz})\) sub-manifold, owing to the octahedral symmetry (\(O_h\)) of the host perovskite structure. Magnetising the system along (001) direction further splits the \(t_{2g}\) states into two branches \(t'_{2g}\) made of \(\{d_{xy}, d_{yz}\}\) and \(t''_{2g}\) made of \(d_{xy}\) as depicted in Fig. 1e and f (which will be discussed in further detail later on). Due to this magnetic phase transition, these states undergo a tetragonal warping\(^5\) despite the cubic symmetry of the CF. This, as will be discussed later, is due to the emergence of a directional OAM term \(L\), which when added to the SAM \(S\), modifies the total angular momentum \(J = S + L\), as shown in Fig. 1g and h.

It is worthwhile comparing EuTiO\(_3\) to its twin compound SrTiO\(_3\), which shares the same crystal structure and similarly has high carrier mobility, but lacks any magnetic ordering\(^{15-17}\). In SrTiO\(_3\), like EuTiO\(_3\), the Ti \(t_{2g}\) states form a conduction manifold close to the Fermi energy (\(E_F\))\(^{18,19}\). As such, the SrTiO\(_3\) \(t_{2g}\) bands depicted in Fig. 2a are analogous to those in undoped AFM EuTiO\(_3\) where there is no net spin polarization. However, in EuTiO\(_3\) the Eu\(^{2+}\) 4\(f\) electrons form a nearly flat sub-gab manifold 0.5 eV from the conduction band minimum (CBM), with an admixture of O 2\(p\) and Ti 3\(d\) states\(^7,19\) (see the Supplementary information for a full band structure and density of states). These 4\(f\) orbitals are half-filled, hence exhibiting a large spin moment of \(S = 7/2\). They are also highly localised and interact with each other with a weak AFM ordering which only becomes significant at low temperatures (\(T < 5.5\)K)\(^9-11\). By doping, we access the Ti \(t_{2g}\) states which act as a mediator for an RKKY interaction between the Eu 4\(f\) states due to their energetic proximity\(^{12,14}\) and the small admixture of Ti 3\(d\) states in the Eu 4\(f\) manifold. More precisely speaking, Eu-4\(f\) electrons are exchanged through the Ti 3\(d\) states with another Eu centre, which is considerably ‘easier’ if the spin of all three bands matches. As such, it is energetically favourable for the Eu-4\(f\) and Ti-t3\(d\) states involved in the RKKY interaction to adopt the same direction of spin. With the application of an external magnetic field above 2.1 T, the 4\(f\) states align along the field and adopt a FFM (spin-up) order through the RKKY interaction. Furthermore, the Ti 3\(d\) spin-up states are brought down in energy, spin polarizing the \(t_{2g}\) conduction states as shown schematically in Fig. 1d and as can be observed in the band structure Fig. 2a. As the magnetic order arises from the ‘separate’ Eu magnetic centres, the \(t_{2g}\) states retain high mobility, and due to the localised nature of the 4\(f\) moments, we have direct control over the spin of the \(t_{2g}\) states via manipulation of the applied magnetic field. For the case of (001) magnetization, which is what is considered in Fig. 1 and Fig. 2, the \(t_{2g}\) manifold near the CBM form FPs with three distinct shapes\(^7\) as shown in Fig. 2. The FPs intersect each other in the BZ, resulting in two distinct types of symmetry protected crossings in the \(k_x-k_y\) plane (shown in Fig. 2a). The first type are Weyl nodes, occurring...
between states of opposite spin along $\Gamma \rightarrow X$. The second type, which are part of nodal lines, are formed by the same spin states along $\Gamma \rightarrow M$ and are protected by the planar mirror symmetry of EuTiO$_3$ see Fig. 2b.

**Directional quenching of orbital angular momentum:** Figure 2c-k shows the orbital character, SAM, and OAM projected onto the FPs at a typical $n_e = 4 \times 10^{19}$ cm$^{-3}$ (corresponding to $E_F = 66$meV). Here, we see distinct regions of different orbital character; a simple collinear spin texture; and regions with (un)quenched OAM. We are primarily interested in the ‘light’, more dispersive, 2nd and 3rd pockets as these will dominate transport properties. Given the collinearity of the pockets’ SAM, the behaviour of the SO term $\Delta_{SO} = \mathbf{L} \cdot \mathbf{S}$ must be entirely dependent on the OAM texture, which is in turn related directly to the orbital character. Specifically, in regions dominated by $d_{xy}$, OAM is strongly suppressed. This varying SO term leads to a characteristic compression and elongation of the 2nd and 3rd pockets along the $z$-direction, respectively. This contrasting behaviour is due to the relative orientation of SAM and OAM, which are anti-parallel and parallel for the 2nd and 3rd pockets, respectively. Accordingly, the 2nd pocket gains a lower $\Delta_{SO}$, thereby becoming more dispersive than the 3rd pocket in the $k_x$-$k_y$ plane, manifesting as the above-mentioned directional distortions.

These distortions are quantified by the plot given in 3a, where the cross-sectional area of the pockets varies with the angle of the plane of the cross-section. As these cross-sectional areas are directly related to the Shubnikov de-Haas oscillations in EuTiO$_3$, this FP warping is observable in any directional anisotropy of such oscillations. To build up a clearer picture of the SO term we consider a path along the warped pocket surfaces from $k \parallel (1,1,0)$ to $(0,0,1)$, as shown in Fig. 3c and e. For the warped pockets there is a sign change in the SO term moving from regions of different orbital character due to the twisting of OAM, and the sign of the SO term is opposite between each pocket as expected.

The non-collinearity of OAM in EuTiO$_3$ is a natural consequence of symmetry constraints in perovskite structures. As already mentioned in Fig. 1c and d, the CF in a non-magnetic cubic perovskite reduces the continuous $SO(3)$ symmetry of a free ion to $O_h$, splitting the 3$d$-orbitals into two manifolds $e_g\{d_{x^2-y^2},d_{z^2}\}$ and $t_{2g}\{d_{xy},d_{xz},d_{yz}\}$. The latter, the lower energy branch, can be written out in spherical harmonics as,

\[
\begin{align*}
    d_{xy} & = \frac{i}{\sqrt{2}} (Y_2^{-2} - Y_2^2) \\
    d_{xz} & = \frac{1}{\sqrt{2}} (Y_2^{-1} - Y_2^1) \\
    d_{yz} & = \frac{i}{\sqrt{2}} (Y_2^{-1} + Y_2^1). 
\end{align*}
\]

None of these combinations alone can lead to a net OAM unless some orbital mixing is
allowed\[23\]. A (001) magnetisation can realise this. It reduces the $O_h$ symmetry to $C_{4h}$, thereby retaining only the 4-fold rotational symmetry in the $xy$ plane. Expanding $V$ as

$$V = \sum_{m,n=-l}^{l} V_{m,n}|Y_{l}^m\rangle\langle Y_{l}^m|,$$

one can show $V$ remains invariant under such a rotation, if $V_{mn}$ satisfies $|m-n| = 4\alpha$, where \(\alpha\) is an integer (see Supplementary Information for a full mathematical discussion). This only applies to $d_{xy}$, explaining why it lacks any OAM. In contrast, $d_{xz}$ and $d_{yz}$ can mix and accordingly form an OAM texture. This situation can change if one changes the direction of magnetisation. For example, a (111) magnetisation, even though breaking the $O_h$ symmetry, retains the 3-fold rotational symmetry around this axis. As a result, all $t_{2g}$ states can equally contribute to OAM, making it less prone to directional quenching. Fig. 3.d and f confirm the (111) magnetisation exhibits less pronounced OAM quenching and less variation in the SO term, leading to less distortion of the FPs (Fig. 3.b). Hence, we have directly controlled the Fermiology and OAM texture by manipulating an external magnetic field (more details for the (111) case are given in the Supplementary information).

**OAM-driven Anomalous Hall Effect**: Each FP exhibits sharp boundaries between the regions of different orbital characters for the (001) magnetization case. Hence these separate two distinct but energetically proximate eigenstates leading to a large BC along these boundaries, contributing to an intrinsic AHC\[1,24\]. Crucially, this BC arises due to a complex OAM texture because (as discussed previously) these ‘composite’ FPs are generated by a varying SO field, despite a collinear SAM. To support this claim, we make an analytic link between the BC contribution of a state $|n\rangle(k)$, with the variation of its $L_z$ eigenvalue $\lambda_n(k)$:

$$|\Omega_n(k)|^2 \propto \sum_{i,j,p,q} \left( \partial_{k_j} \left( \frac{\partial_{k_p} \lambda_n}{\lambda_n} \right) \right)^2 - \partial_{k_p} \left( \frac{\partial_{k_p} \lambda_n}{\lambda_n} \right) \partial_{k_q} \left( \frac{\partial_{k_q} \lambda_n}{\lambda_n} \right).$$

The full derivation of this term can be found in the Supplementary Information. This term is plotted explicitly for the 2nd pocket in the left hand side of Fig. 4.a. Comparing it with the total $\Omega_n(k)$ directly calculated from the Bloch wave functions (see the insets in Fig. 4.a), we find a good agreement. This accordingly confirms the link between OAM and BC. Furthermore, from the $k \cdot p$ theory\[25\] we have that $\mathbf{L} = \nabla_r V(r) \times \mathbf{k}$, and as such we can state (at least very broadly) that there is a direct link between the CF $V(r)$ and BC.

The OAM texture and its topological nature is further elucidated in the right-hand side of Fig. 4.a. For the 2nd pocket, we can see that the $k_z = 0$ plane separates regions with a negative/positive OAM divergence above/below it, respectively. Hence, the OAM ‘flows’ from the bottom half, which acts as a source, to the upper half as a sink. The OAM itself is also depicted along the top and bottom of the pocket, to reinforce the pole/anti-pole like
behaviour of OAM in these regions.

Increasing $E_F$, we expect a systematic evolution in the magnitude and sign of $\Omega_n(k)$. Starting from the CBM, where only the 1st and 2nd pockets (which are of the same OAM character) are present, the occupied states in the BZ carry a giant positive $\Omega_n(k)$, which in turn leads to a positive AHC growing monotonously with increasing $E_F$, compare Fig. 4a, b, c and g. Once $E_F$ crosses the 3rd pocket, a negative $\Omega_n(k)$ contribution emerges due to opposite OAM character of the 2nd pocket, see Fig. 4d. The competition between these two reaches its critical point at the protected crossing between 2nd and 3rd pocket along the $\Gamma \rightarrow M$ direction, bringing AHC to its peak, as highlighted by the region I shaded in yellow in Fig. 4g. Further increasing $E_F$ leads to the domination of the negative $\Omega_n(k)$ contribution, manifested by a sign change in AHC for $n_c \sim 2 \times 10^{19}$ cm$^{-3}$, cyan region II in Fig. 4g. This trend continues until $E_F$ crosses the Weyl node along the $\Gamma \rightarrow X$ direction at $n_c \sim 2 \times 10^{20}$ cm$^{-3}$. At this point, AHC, which is now negative, shows another turning point and begins to increase, as depicted in the purple region III in Fig. 4g. Such an $n_c$-dependent sign change in AHC is consistent with the previous experimental observations, except for an overall sign difference, which is likely due to the fact we have not assigned a negative Hall conductivity for the electron carriers in our calculations. Nevertheless, our calculations demonstrate that the observed intrinsic AHC can be well described by such complex, topological OAM textures.

**Discussion**

To elaborate on the role of the $\Gamma \rightarrow M$ protected crossings in the BC and how this relates to the OAM texture we note that these are part of a nodal line in the $k_x$-$k_y$ plane that begins along this direction, and at our plotted carrier density ($n_c = 4 \times 10^{19}$ cm$^{-3}$) intersects the FP as a pair of protected crossings, which we expect to be topological features of the BC, acting as quantized monopoles. At this $n_c$, one of these crossings occurs at an azimuthal angle $\phi = 49.3^\circ$ from the $k_x$ axis. Considering the divergence of OAM over a path (shown as a black line on Fig. 4a on the FP) that intersects this crossing (Fig. 4h) we can see that it becomes non-analytic at the degeneracy, showing these are as much topological features of the OAM texture as they are of the BC. Hence the inclusion of the nodal line, and thus moving between the regions I and II of $\Omega_z$, corresponds directly to a change in OAM texture for the FP.

In conclusion, we showed that under certain conditions, CFs in transition metal perovskites could induce directionally unqueched OAM, giving rise to complex OAM textures. Such non-trivial, topological OAM textures were shown to generate large BC, with an intrinsic AHC, even in systems with collinear spin textures. In discussing OAM in EuTiO$_3$, we explained previous observations of warped Fermiologies and a non-monotonic AHC arising from $k$-space varying OAM. Accordingly, we proposed EuTiO$_3$ as a prototypical example of OAM driven Berryology and AHE. Additionally, We demonstrated how the energetic proxi-
imity between the local moments and charge carriers affords us easy control over a system’s magnetic and topological properties. Given the ubiquity of electronic structures sharing such features, our findings open a new way for designing a whole new class of OAM-driven topological devices.

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**Author contributions:** MSB conceived the project. OD performed the calculations. OD and MSB analysed the results and wrote the manuscript.

**Conflict of Interests:** The authors declare no conflicts of interests.

**Data Availability:** Underpinning data are available upon reasonable request to the corresponding authors.

II. METHODS

Bulk electronic structure calculations for EuTiO$_3$ and SrTiO$_3$ were performed within the density functional theory using Perdew-Burke-Ernzerhof exchange-correlation functional as implemented in the WIEN2K package. The relativistic effects including spin-orbit coupling, were fully taken into account. An effective Hubbard-like potential of $U_{\text{eff}} = 6$ eV for Eu was used to model the strong on-site Coulomb interaction of the Eu-4$f$ states. For both compounds, a cubic crystal structure with a lattice parameter of 3.905 Å was used. The corresponding BZ was sampled by a $10 \times 10 \times 10$ $k$-mesh. For the calculation of SAM, OAM, BC and AHC, a six-band tight-binding Hamiltonian was constructed using Maximally localized Wannier functions with the Ti $t_{2g}$ orbitals as the projection centres. The BZ integration of AHC was carried out using a $200 \times 200 \times 200$ $k$-mesh adopting an additional $10 \times 10 \times 10$ subdivision within each volume increment wherever the BC was greater than
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Figure 1. **Diagrammatic overview of our EuTiO$_3$ system.** a Crystal structure of EuTiO$_3$, in the para-magnetic (PM phase). b Crystal structure in the FFM phase for an applied field in the (001) direction, where insets for a and b show RKKY interaction and spin polarization of Ti for FFM phase, or lack thereof for PM phase. c Schematic representation of density of states (DOS) for PM phase, where $E_F$ is below Ti-3$d$ states. d Schematic for FFM phase DOS, RKKY interaction between Eu-4$f$ and 3$d$ states is shown. e Model potentials that reflect the symmetry reduction of the system. f Band diagram for Ti-3$d$ states: an isolated Ti atom has a full continuous $SO(3)$ symmetry, and the 3$d$ states remain fully degenerate; The CF of the EuTiO$_3$ system reduces the symmetry to a discrete octahedral $O_h$ point group, splitting the 3$d$ states into $e_g$ and $t_{2g}$ sub-manifolds; A magnetic field along the (001)-direction reduces the symmetry further to a $C_{4h}$ group, which has a 4-fold rotational symmetry about the z axis and a horizontal mirror symmetry, splitting the $t_{2g}$ states further and isolating the $d_{xy}$ orbital. (g-h) Angular momentum in the system. g States with $d_{xy}$ orbital character have fully quenched OAM. h) States formed from super-positions of $d_{xz}, d_{yz}$ have unquenched OAM that is uncoupled from the spin angular momentum.
Figure 2. **Fermiology of EuTiO$_3$.** a Bands structures of the Ti $t_{2g}$ states for FFM EuTiO3 (solid lines) and SrTiO3 (dashed). For FFM EuTiO3 the bands are spin polarized and for low energy only spin-up states are accessed. Protected crossing between the bands are marked on as black dots.

b Spin-up $t_{2g}$ band structure colour coded for their orbital character, $E_F$ taken to be 66 meV above the CBM, corresponding to $n_c = 4 \times 10^{19}$ cm$^{-3}$ which defines the FPs displayed. c-k FPs, going vertically moves from the 1$^{st}$ to 3$^{rd}$ pocket, horizontally goes through orbital character, spin texture, and OAM texture. Orbital character is colour-coded such that red, green and blue correspond to regions dominated by $d_{xy}$, $d_{xz}$, and $d_{yz}$, respectively. Arrows show the direction of spin and OAM over the FPs, showing that the spin texture is simple and collinear and that the regions dominated by $d_{xy}$ exhibit OAM quenching as expected.
Figure 3. **Emergent spin-orbit coupling due to directional quenching of OAM.** a and b Cross sectional area of 2nd and 3rd pockets for the (001) and (111) magnetization cases, respectively. c and d Orbital projection ($P_{dxy}$), absolute OAM ($L$) and spin-orbit coupling ($L \cdot S$) over the path $k = (0, 0, 1)$ to $k = (1, 1, 0)$ for the 2nd pocket for the (001) and (111) magnetisations, respectively. e and f Same as c and d but for 3rd. In the polar plots c-f, the radius of the dashed circles is shifted such that the origin corresponds to $r = -1$, so we can show the full range of the spin-orbit terms.
Figure 4. **Berryology of FPs in EuTiO$_3$.** a, OAM texture and BC on the 2nd pocket, calculated for $n_c = 4 \times 10^{19}$ cm$^{-3}$. Left hand side shows term from Eq. 3 effectively showing magnitude of BC. Right hand side shows divergence of OAM. Arrows show OAM on two paths along the top and bottom of the pockets respectively, showing the pole/anti-pole behaviour of OAM. Insets show numerically calculated BC for regions where BC is concentrated. b, Band structure of conduction bands, with four $E_F$ marked corresponding to carrier concentrations of 0.5, 2, 20, $50 \times 10^{19}$ cm$^{-3}$. c-f, $z$ component of BC at said $E_F$, taken in the $k_x$-$k_y$ with $k_x$ and $k_y$ varying from 0 to 0.15 $\pi/a$ where $a$ is the real space lattice constant. Black dots mark symmetry protected crossings. g, Carrier-dependence of the $xy$ component of AHC. The shaded areas correspond to the energy windows shaded with the same colours in b. h, Divergence of OAM over path on the 2nd pocket that passes through the nodal line (shown by black line in (a)), where $\theta$ is the polar angle paramatising the path.
Supplementary Files

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