Dynamical magneto-electric response in topological materials

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Axion field induced topological magneto-electric response has attracted lots of attentions since it was first proposed by Qi et al. in 2008. Here we find a new type of anti-commutative magneto-electric response \(\beta^5(\omega)\), which can induce a dynamical magneto-electric current driven by a time-varying magnetic field. Unlike the Chern-Simons Axion term, this magneto-electric response term is gauge-independent, and manifests in the systems breaking the symmetries of the time-reversal, inversion and mirror. In particular, we predict Mn\(_2\)Bi\(_2\)Te\(_5\) as a material candidate to observe dynamical magneto-electric current.

**Introduction.** Magneto-electric response has been discovered more than a century ago, which describes that an electric field can induce the magnetization or a magnetic field can induce the polarization in certain materials. Recently, topological magneto-electric response has been discussed with an effective action \(S_0 = \left(\frac{\pi}{\theta}\right) \int d^3 x dt \mathbf{E} \cdot \mathbf{B}\)[1], similar to Axion in the Standard Model of particle physics, where \(\theta = \pi\) or \(\theta = 0\) represents the topological nontrivial or trivial term in a material[1-9]. Generally, topological insulators with the time-reversal symmetry \(\mathcal{T}\) and the inversion symmetry \(\mathcal{P}\) have \(\theta = \pi\), while Axion insulators are the materials still maintaining \(\theta = \pi\) without the \(\mathcal{T}\) and \(\mathcal{P}\) symmetries. Physically, the magneto-electric response \(\theta\) term in a crystal material has a non-Abelian expression of the momentum-space Chern-Simons form [1, 10]. With \(\theta = \pi\) in Axion insulators, some novel physical effects have been proposed, such as the half-integer anomalous Hall effect\[11, 12\]. These novel magneto-electric-response-relevant phenomena have attracted lots of attentions in recent theoretical and experimental studies[13], which includes the studies of the antiferromagnetic material Mn\(_2\)Bi\(_2\)Te\(_4\)[14-17].

In this Letter, we propose a new type of magneto-electric response term \(\beta^5(\omega)\), originated from the anti-commutative correlation of the magnetization and the polarization. This term not only induces the interface magneto-electric responses including the surface charge and the surface anomalous Hall responses, similar to the behaviors in the Axion insulators [1, 10], but also gives rise to a dynamical magneto-electric current response driven by a time-varying magnetic field \(j^\beta = 2\beta^5(\omega) \partial_t \mathbf{B}\), with \(\beta^5(\omega)\) denoting the linear response coefficient. We discuss the origin and required symmetry-breaking terms of this dynamical magneto-electric response, and propose an effective model to describe the main features of this phenomenon. We also propose some small bandgap topological materials, such as Mn\(_2\)Bi\(_2\)Te\(_5\), as possible candidates as well as a feasible experimental setup to observe the dynamical magneto-electric response.

**Electrodynamics of dynamical magneto-electric effect.** Considering a system with a longitudinal magneto-electric coupling, the total Lagrangian can be derived from the linear response theory[18] and reads:

\[
\mathcal{L}(t) = \frac{1}{2} \left[ \varepsilon_0 \mathbf{E}^2(t) - \frac{1}{\mu_0} \mathbf{B}^2(t) \right] - \rho \phi + \mathbf{j} \cdot \mathbf{A} - \alpha^\lambda(t) \mathbf{E}(t) \cdot \mathbf{B}(t) - \mathbf{E}(t) \int \xi(t, t') \mathbf{B}(t') dt' - \mathbf{B}(t) \int \xi^\lambda(t, t') \mathbf{E}(t') dt'.
\]

(1)
Here, the last three terms represent the magneto-electric response due to magneto-electric fields coupled with Bloch electrons in materials. Specifically, \( \alpha(t) \) denotes the simultaneous magneto-electric response, and \( \xi(t, t') \), \( \xi'(t, t') \) represent the retarded magneto-electric response. In the following, we assume that the electric field \( \mathbf{E}(t) = B_0 e^{-i\omega t} \) and magnetic field \( \mathbf{B}(t) = B_0 e^{-i\omega t} \) are time-harmonic variables with frequency \( \omega \), after Fourier transform from \( t - t' \) to frequent \( \omega \), the general charge response and current response can be derived from the Euler-Lagrange equations[19]:

\[
\delta \rho = -\nabla \left[ \alpha^\lambda + \xi(\omega) \right] \cdot \mathbf{B}(t) \tag{2}
\]

\[
\delta \mathbf{j} = \nabla \left[ \alpha^\lambda + \xi'(\omega) \right] \times \mathbf{E}(t) + \partial_t \left[ \alpha^\lambda + \xi(\omega) \right] \mathbf{B}(t) + 2 \beta^\xi \partial_t \mathbf{B}(t). \tag{3}
\]

Here, the charge and current responses originate from magneto-electric effect are present, while the ordinary charge and current responses originate from the simultaneous magneto-electric response, and \( \alpha^\lambda(t, t) \) is a novel effect not studied before. Comparing with previous studies, the first term describes the surface half integer anomalous Hall effect in an Axion insulator [1, 10], and the second term describes the dynamical axion effect [11, 12]. Both of these effects should be corrected by the retarded magneto-electric response \( \xi(t) \) and \( \xi'(t) \). The third term describes the time-dependent magneto-electric effect (DME) \( \delta^\beta = 2 \beta^\xi \partial_t \mathbf{B} \) which only depends on the anti-commutative correlation of the magnetization operator \( \hat{\mathbf{M}} \) and the polarization operator \( \hat{\mathbf{P}} \) [20]. The main focus of this work is the dynamical magneto-electric effect driven by a time-varying magnetic field denoted by the third term in Eq. (3). As shown in the Fig. 1, for the topological magneto-electric response term \( \alpha(t) \) (including the case of an Axion insulator), the polarization current \( j^p \) and bound current \( j^b \) cancel each other. Meanwhile, for the retarded magneto-electric response \( \xi(t) \) and \( \xi'(t) \), a dynamical magneto-electric current \( \delta^\lambda = 2 \beta^\xi \partial_t \mathbf{B} \) exists, which is a novel effect not studied before.

**General magneto-electric response.** The general magneto-electric response can be obtained from the linear response theory [21], including both the simultaneous magneto-electric response \( \alpha(t, t) \) and the retarded magneto-electric response \( \xi(t, t') \) and \( \xi'(t, t') \). The operator forms of these coefficients read as follows:

\[
\alpha^\lambda(t, t) = -\left| \frac{\partial^2 H}{\partial \mathbf{E} \partial \mathbf{B}} \right|_{\theta t>0} \alpha^\lambda(t)
\]

\[
\xi(t, t') = -\frac{i}{\hbar} \Theta(t-t') < \left[ \hat{\mathbf{P}}(t), \hat{\mathbf{M}}(t') \right] >_{\theta 0}
\]

\[
\xi'(t, t') = -\frac{i}{\hbar} \Theta(t-t') < \left[ \hat{\mathbf{M}}(t), \hat{\mathbf{P}}(t') \right] >_{\theta 0}
\]

where \( \hat{\mathbf{P}} = -\partial H/\partial \mathbf{E} \) is the polarization operator, and \( \hat{\mathbf{M}} = -\partial H/\partial \mathbf{B} \) is the orbital magnetization operator. The time variable \( t \) in the brackets represents the measurement time, and \( t' \) represents the time of an external electric or a magnetic field. Then, the first term \( \alpha^\lambda(t, t) \) is a response function at the same time, and the Maxwell relations in \( \alpha^\lambda(t, t) \) make \( \mathbf{B} \) and \( \mathbf{E} \) commutative in relevant Lagrangian \( \mathcal{L}^\lambda = -\alpha^\lambda(t, t) \mathbf{E}(t) \cdot \mathbf{B}(t) \). Thus, \( \alpha^\lambda(t) \) represents the commutative magneto-electric response. The second term and the third term describe the magneto-electric response with different times, \( \mathbf{P}(t) = \int \xi(t, t') \mathbf{B}(t') dt' \), \( \mathbf{M}(t) = \int \xi'(t, t') \mathbf{E}(t') dt' \), and the related Lagrangian can be written as \( \mathcal{L}^\xi(t) = -\int [\mathbf{E}(t) \xi(t, t') \mathbf{B}(t') + \mathbf{B}(t) \xi'(t, t') \mathbf{E}(t')] dt' \), where \( \mathbf{B} \) and \( \mathbf{E} \) are not commutative necessarily, for different times.

Comparing to notations of the optical conductivity in the linear response theory [21], \( \lambda(t, t) \) term is the interband Drude term, which also includes the Chern-Simons Axion term \( \alpha^\lambda = (e^2/2\pi \hbar) \theta \) [22]. As for the retarded magneto-electric responses \( \xi(t, t') \) and \( \xi'(t, t') \), the explicit formula can be obtained similar to the current-current correlation in the Kubo formula. To be specific, after Fourier transform from \( t - t' \) to frequent \( \omega \), these coefficients read:

\[
\xi_{ij}(\omega) = e \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_{n \neq m} \frac{(f_{nk} - f_{mk}) A_{imn} M_{jm}^i}{\varepsilon_{nk} - \varepsilon_{mk} + i\omega + \gamma}
\]

\[
\xi'_{ij}(\omega) = e \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_{n \neq m} \frac{(f_{nk} - f_{mk}) M_{imn} A_{jm}^i}{\varepsilon_{nk} - \varepsilon_{mk} + i\omega + \gamma}
\]

where \( A_{imn} = i v_{nm}^i / (\varepsilon_{nk} - \varepsilon_{mk}) \) and \( M_{imn} = \sum_{l \neq m} (v_{nl} \times A_{lm})^i \) are the inter-band elements of the Berry connection and the orbital magnetization in the basis of the eigen-functions with the eigen-energies \( \varepsilon_{nk} \) [23]. In the following, we mainly consider the longitudinal retarded magneto-electric coefficients \( \xi_{ii}(\omega) \) and \( \xi'_{ii}(\omega) \), and for convenience one can separate the commutative part \( \alpha_{ii}^\xi(\omega) \) and the anti-commutative part \( \beta_{ii}^\xi(\omega) \) as follows:

\[
\alpha_{ii}^\xi(\omega) = [\xi_{ii}(\omega) + \xi'_{ii}(\omega)] / 2
\]

\[
= e \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_{n \neq m} \frac{(-1) (f_{nk} - f_{mk}) v_{nm}^i A_{nm}^i}{(\varepsilon_{nk} - \varepsilon_{mk})^2 - (i\omega)^2}
\]

\[
\beta_{ii}^\xi(\omega) = [\xi_{ii}(\omega) - \xi'_{ii}(\omega)] / 2
\]

\[
= e \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_{n \neq m} \frac{-i\omega (f_{nk} - f_{mk}) A_{nm}^i M_{imn}^j}{(\varepsilon_{nk} - \varepsilon_{mk})^2 - (i\omega)^2}
\]

where one can see that \( \alpha_{ii}^\xi(\omega) \) is real while \( \beta_{ii}^\xi(\omega) \) is purely imaginary. The anti-commutative magneto-electric coefficient \( \beta_{ii}^\xi(\omega) \) can give rise to new kind of magneto-electric response, which manifests in systems without the time-reversal symmetry, the inversion symmetry and the mirror symmetry.
A simple effective model. In Eq.6, we can see that $\xi(\omega)$ is an inter-band gauge-independent term (similar to $\alpha^\xi_1(\omega)$ and $\beta^\xi_1(\omega)$), which is non-zero if the time-reversal symmetry $T$ and the inversion symmetry $P$ of the system are broken, unlike the Chern-Simons Axion term. Besides, other possible symmetries such as the mirror symmetry $M$ and the mirror Axion term. When the mirror symmetry is broken, resulting in asymmetric band structure (solid lines in Fig.2 (a) & (b)). The symmetry breaking terms are associated with the factors of $\Gamma^{1,2,3}$ in the Hamiltonian $H$. In $H_0$, $d_{1,2,3} = (A_2\sin k_x, A_2\sin k_y, A_2\sin k_z)$ are odd in $k_x$, $k_y$ and $k_z$, respectively. However, $m_{1,2,3}$ in $\delta H$ are $k$-independent constants. Thus, $m_{1,2,3}$ in $\delta H$ breaks the symmetries of $P$, $T$ and mirror $M_{x/y/z}$. Meanwhile, non-zero retarded magneto-electric response $\alpha^\xi_1(\omega)$ and $\beta^\xi_1(\omega)$ appear, as shown in Fig.2 (c) & (d). Importantly, $\beta^\xi_{2x}$ and $\beta^\xi_{2y}$ have the peak values around the band edges, while decrease to zero when Fermi energy locates in the band gap. In retrospect, the values of $\alpha^\xi_{2x}$ and $\alpha^\xi_{2y}$ shows peak values when Fermi energy locates in the band gap, and decrease when Fermi surface increases. The prominent values of $\beta^\xi_{2x}$ and $\beta^\xi_{2y}$ around the band edge can be explained from the remarkable asymmetric band structure around the band edge as shown in Fig.2 (a) & (b). With increasing the Fermi energy, $H_0$ becomes dominant and $\alpha^\xi_1(\omega)$ and $\beta^\xi_1(\omega)$ decrease as shown in Fig.2 (c) & (d).

A material candidate. Based on the aforementioned model study, in order to search for materials with the dynamical magneto-electric response $\beta^\xi(\omega)$, the system needs to break the $P$, $T$ and the mirror symmetry and possesses asymmetric band structure. Here we choose the ternary chalcogenide material Mn$_2$Bi$_2$Te$_5$ as an example. The space group of nonmagnetic Mn$_2$Bi$_2$Te$_5$ is $P\overline{3}m1$ (No. 164) with symmetric operators $P$, $T$, $M$, $C_{3z}$ and $C_{2x}$ (the crystal structure of Mn$_2$Bi$_2$Te$_5$ is shown in Fig.3 (a) [12]). When the antiferromagnetic order in 001 direction is considered, a non-zero $\beta^\xi(\omega)$ exists in this system. In Fig.3 (b), we plot the band structures of Mn$_2$Bi$_2$Te$_5$ with different spin orbital coupling strength $H^{SO}$. When $H^{SO} = 0$, the band is symmetric. As the SOC strength increases, symmetry breaking of $M_z$ in orbital space occurs due to the SOC effect, resulting in an asymmetric band structure. Meanwhile, with increasing of the SOC strength, the values of $\alpha^\xi_1(\omega)$ and $\beta^\xi_1(\omega)$ become larger. As we can see in Fig.3 (b), the $k$-path of $\Gamma - K$ and $\Gamma -
FIG. 3. Result of Mn$_2$Bi$_2$Te$_5$: (a) Crystal structure and (b) the band-structures with different SOC strength $l^{SO}$ of Mn$_2$Bi$_2$Te$_5$. (c) - (d) magneto-electric response $\alpha_{xx}^\xi(\omega)$ and $\beta_{xx}^\xi(\omega)$ ( in unit of $e^2/\hbar$ ) with different energy $\hbar\omega$ ($l^{SO} = 1$ ). (e) - (f) $\alpha_{xx(yy)}^\xi(\omega)$ and $\beta_{xx(yy)}^\xi(\omega)$ ( in unit of $e^2/\hbar$ ) with SOC strength $l^{SO}$ ($\hbar\omega = 0.5$ eV ). Here $\alpha(\beta)^\xi$ is equal to $\beta(\alpha)^\xi$ due to $C_3z$ symmetry. (g) An experiment setup for observation of DME.

$K'$ show obvious differences from -0.5 eV to -0.4 eV, resulting in rapidly changing of $\alpha_{ii}^\xi(\omega)$ and $\beta_{ii}^\xi(\omega)$ in Fig.3 (c) & (d). When the chosen Fermi Energy is increased to around zero, since the band asymmetry becomes less obvious, the change of $\alpha_{ii}^\xi(\omega)$ and $\beta_{ii}^\xi(\omega)$ become slow, and show plateau in the gap. Besides, the values of $\alpha_{ii}^\xi(\omega)$ and $\beta_{ii}^\xi(\omega)$ in the gap depend on the system parameters such as $l^{SO}$, as shown in Fig.3 (e) & (f). Moreover, with increasing of SOC’s strength $l^{SO}$, the energy bands become more asymmetric, and values of $\alpha_{ii}^\xi(\omega)$’s and $\beta_{ii}^\xi(\omega)$’s are getting larger, as shown in Fig.3 (e) & (f). According to Ref. [12], the band structures of Mn$_2$Bi$_2$Te$_5$ are gapless around $l^{SO} = 0.9$, and band inversion shows up at larger $l^{SO}$, which indicates that $\alpha_{ii}^\xi(\omega)$ and $\beta_{ii}^\xi(\omega)$ can be enhanced in topological materials with band inversion. Also, as one can see in Eq. 8 & 9, $\hbar\omega$ is comparable with the band gap, so topological materials with a small band gap are better for observing the dynamical magneto-electric response.

An experiment design. Finally, we propose an experimental setup to detect the DME current $\delta j^\beta = 2\beta \partial_t B$. In Fig.3 (g), by adding an alternating magnetic field $B_z$ in the x-direction, one can measure an alternating current in the same direction. The magnetic field $B_z$ can be obtained from two light fields with different transmission directions, such the electric fields of which will cancel each other out (The thickness of the system should be smaller than the wavelength of the light fields). Ac-
According to Ref. [12], the 001-antiferromagnetic state is the ground state of Mn$_2$Bi$_2$Te$_5$, so we can observe a non-zero alternating current $J_0$ induced by alternating magnetic fields $B_z$. Furthermore, if we add a large external static magnetic fields $B_z$ in z-direction and shift Mn$_2$Bi$_2$Te$_5$ to FM state, the inversion symmetry $\mathcal{P}$ will become preserved and the anti-commutative magneto-electric response $\beta^\parallel_2(\omega)$ will vanish. Consequently, there will be no alternating current $J_0$ at a large external static magnetic field $B_z$, as shown in Fig.3 (g). For example, if we have an experimental sample of cross-section size $1 \mu$m$^2$ in z-direction with $\beta^\parallel_2 \sim 0.001 (e^2/h)$, and use 1T alternating magnetic field with $1 \text{eV}/h$ frequency, we can detect an alternating current response of $\sim 100 \mu$A.

**Conclusion.** In summary, we find an anti-commutative magneto-electric response term $\beta^\parallel_2(\omega)$, which can give rise to DME represented by $\delta \mathcal{P} = 2\beta_2^\parallel B_0$. This novel magneto-electric response originates from the retarded magneto-electric response, and the microscopic coefficients are obtained from the linear response theory. Moreover, the characteristics of this DME are analyzed by a detailed four-band model, and we find that a non-zero $\beta^\parallel_2(\omega)$ term exists in the systems without the time-reversal, inversion and the mirror symmetries. The values of $\beta^\parallel_2(\omega)$ can be increased by raising the value of SOC, which gives rise to a prominent asymmetric band structure. Finally, we predict Mn$_2$Bi$_2$Te$_5$ as a material candidate and propose an experimental setup to observe the DME current.

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SUPPLEMENTAL MATERIALS

1. Formula derivation of Euler-Lagrange equations

We starts from the Lagrangian considering retarded magneto-electric response $\xi(t,t')$
\[
L(t) = \frac{1}{2} \left[ \varepsilon_0 \mathbf{E}^2(t) - \frac{1}{\mu_0} \mathbf{B}^2(t) \right] - \rho \phi - \mathbf{j} \cdot \mathbf{A} - \alpha^2 \mathbf{E}(t) \cdot \mathbf{B}(t) \tag{S1}
\]
\[
- \mathbf{E}(t) \int \xi(t,t') \mathbf{B}(t')dt' - \mathbf{B}(t) \int \xi'(t,t') \mathbf{E}(t')dt'
\tag{S2}
\]

and relations from Maxwell equations
\[
\mathbf{B}(t) = \nabla \times \mathbf{A}(t), \tag{S3}
\]
\[
\mathbf{E}(t) = -\nabla \phi(t) - \dot{\mathbf{A}}(t), \tag{S4}
\]

then one can obtain
\[
L(t) = \frac{1}{2} \sum_i \varepsilon_0 \left[ (\partial_i \phi)^2 + (\partial_i A_i)^2 + 2 \partial_i A_i \cdot \partial_i \phi \right] - \frac{1}{\mu_0} \sum_i \left[ (\nabla A_i)^2 - \partial_i \mathbf{A} \cdot \nabla A_i \right] + (-\rho \phi + \mathbf{j} \cdot \mathbf{A}) \tag{S5}
\]
\[- \alpha^2 \left( -\nabla \phi(t) - \dot{\mathbf{A}}(t) \right) \cdot \left( \nabla \times \mathbf{A}(t) - \nabla \times \mathbf{A}(t) \right) \int \xi(t-t') \mathbf{B}(t')dt' - \left( \nabla \times \mathbf{A}(t) \right) \int \xi'(t-t') \mathbf{E}(t')dt'.
\]

Then, using Euler-Lagrange equations for scalar potential,
\[
\frac{\partial L}{\partial \phi} = \partial_t \left( \frac{\partial L}{\partial \phi} \right) + \nabla \cdot \frac{\partial L}{\partial (\nabla \phi)},
\tag{S6}
\]

where
\[
\frac{\partial L}{\partial \phi} = -\rho \tag{S7}
\]
\[
\frac{\partial L}{\partial \phi} = 0 \tag{S8}
\]
\[
\frac{\partial L}{\partial (\nabla \phi)} = -\varepsilon_0 \mathbf{E}(t) + \alpha^2 \left( \nabla \times \mathbf{A}(t) \right) + \int \xi(t-t') \mathbf{B}(t')dt' \tag{S9}
\]
\[
\nabla \cdot \frac{\partial L}{\partial (\nabla \phi)} = -\varepsilon_0 \nabla \cdot \mathbf{E}(t) + \left[ \nabla \cdot \alpha^2 \right] \cdot \left( \nabla \times \mathbf{A}(t) \right) + \nabla \cdot \int \xi(t-t') \mathbf{B}(t')dt' \tag{S10}
\]

one can obtain
\[
\frac{\partial L}{\partial \phi} = \partial_t \left( \frac{\partial L}{\partial \phi} \right) + \nabla \cdot \frac{\partial L}{\partial (\nabla \phi)} \tag{S11}
\]
\[- \rho = -\varepsilon_0 \nabla \cdot \mathbf{E}(t) + \left[ \nabla \cdot \alpha^2 \right] \cdot \left( \nabla \times \mathbf{A}(t) \right) + \nabla \cdot \int \xi(t-t') \mathbf{B}(t')dt'. \tag{S12}
\]

In general, we know that free charge density $\rho_0 \equiv \varepsilon_0 \nabla \cdot \mathbf{E}(t)$, thus we can get extra charge density response from magnetic field
\[
\delta \rho = \rho - \rho_0 \tag{S13}
\]
\[ = - [\nabla \cdot \alpha \lambda] \cdot \mathbf{B}(t) - \int \left[ \nabla \xi(t - t') \right] \cdot \mathbf{B}(t') dt'. \]

Moreover, considering the Euler-Lagrange equations for vector potential

\[ \frac{\partial L}{\partial \mathbf{A}_k} = \partial_t \left( \frac{\partial L}{\partial \mathbf{A}_k} \right) + \nabla \cdot \frac{\partial L}{\partial \nabla \mathbf{A}_k}, \]  

(S14)

where

\[ \frac{\partial L}{\partial \mathbf{A}_k} = j_k \]  

(S15)

\[ \frac{\partial L}{\partial \mathbf{A}_k} = \varepsilon_0 \left( \mathbf{A}_k + \partial_k \phi \right) + \partial_t \left\{ \alpha \lambda [\mathbf{B}(t)]_k \right\} + \partial_t \left\{ \int \xi(t - t') [\mathbf{B}(t')]_k dt' \right\}, \]  

(S16)

\[ \nabla \cdot \frac{\partial L}{\partial \nabla \mathbf{A}_k} = -\nabla \cdot \varepsilon_0 (\nabla \mathbf{A}_k - \partial_k \mathbf{A}) + \left\{ \nabla \times [\alpha \lambda \mathbf{E}(t)] \right\}_k + \left\{ \nabla \times \int \xi(t - t') \mathbf{E}(t') dt' \right\}_k, \]  

(S17)

one can obtain

\[ j = -\varepsilon_0 \partial_t \mathbf{E} + \frac{1}{\mu_0} \nabla \times \mathbf{B} + \partial_t \left\{ \alpha \lambda \mathbf{B}(t) \right\} + \partial_t \left\{ \int \xi(t - t') \mathbf{B}(t') dt' \right\} \]

\[ + \nabla \times [\alpha \lambda \mathbf{E}(t)] + \nabla \times \int \xi(t - t') \mathbf{E}(t') dt'. \]

(S18)

Furthermore, based on the relation \( j_0 \equiv -\varepsilon_0 \partial_t \mathbf{E} + \frac{1}{\mu_0} \nabla \times \mathbf{B} \), then one can obtain the current response from magneto-electric response

\[ \delta j = j - j_0 \]

\[ = \partial_t \left\{ \alpha \lambda \mathbf{B}(t) \right\} + \partial_t \left\{ \int \xi(t - t') \mathbf{B}(t') dt' \right\} + \nabla \times [\alpha \lambda \mathbf{E}(t)] + \nabla \times \int \xi(t - t') \mathbf{E}(t') dt' \]

\[ = \partial_t \alpha \lambda \mathbf{B}(t) + \nabla \alpha \lambda \times \mathbf{E}(t) + \partial_t \left\{ \int \xi(t - t') \mathbf{B}(t') dt' \right\} + \nabla \times \int \xi(t - t') \mathbf{E}(t') dt', \]

Assuming the electric field and magnetic field is time-harmonic with frequent \( \omega \), and Fourier transforming response function \( \xi(t - t') \) to \( \xi(\omega') \)

\[ \mathbf{B}(t') = \mathbf{B}_0 e^{-i\omega t'} \]  

(S20)

\[ \mathbf{E}(t') = \mathbf{E}_0 e^{-i\omega t'} \]  

(S21)

\[ \xi(t - t') = \frac{1}{2\pi} \int \xi(\omega') e^{-i\omega'(t-t')} d\omega', \]  

(S22)

one can obtain

\[ \delta \rho = \rho - \rho_0 \]

\[ = -\nabla \alpha \lambda \cdot \mathbf{B}(t) - \int \left[ \nabla \xi(t - t') \right] \cdot \mathbf{B}(t') dt', \]

in which

\[ \int \left[ \nabla \cdot \xi(t - t') \right] \cdot \mathbf{B}(t') dt' = \int \left[ \nabla \int \xi(\omega') e^{-i\omega'(t-t')} d\omega' \right] \cdot \mathbf{B}_0 e^{-i\omega t} dt' \]

\[ = \frac{\mathbf{B}_0}{2\pi} \nabla \int \xi(\omega') e^{-i(\omega-\omega')t'} dt' d\omega' \]

\[ \nabla \xi(\omega) \cdot \mathbf{B}_0 e^{-i\omega t}, \]

(S24)

(S25)

thus one can obtain the charge density response

\[ \delta \rho = -\nabla \alpha \lambda \cdot \mathbf{B}(t) - \nabla \xi(\omega) \cdot \mathbf{B}(t) \]  

(S26)
\[ = -\nabla \alpha \cdot B(t) - \nabla \left( \alpha \xi + \beta \zeta \right) \cdot B(t). \]

As for current response from in Eq.S19, where

\[ \partial_t \left[ \int (t - t') B(t') dt' \right] = \partial_t \left[ \int \frac{1}{2\pi} \int \xi(\omega') e^{-i\omega'(t - t')} d\omega' B_0 e^{-i\omega t} dt' \right] \]

\[ = B_0 \partial_t \left[ \int \frac{e^{-i\omega t}}{2\pi} \int \xi(\omega') e^{-i(\omega - \omega') t} d\omega' \right] \]

\[ = \partial_t \left[ B_0 e^{-i\omega t} \xi(\omega) \right] \]

\[ = \partial_t \left[ B(t) \xi(\omega) \right], \]  

one can obtain current

\[ \delta j = \partial_t \alpha \Lambda B(t) + \nabla \alpha \times E(t) + \partial_t [B(t) \xi(\omega)] + \xi'(\omega) \nabla \times E(t) \]

\[ = \partial_t \alpha \Lambda B(t) + \nabla \alpha \times E(t) + \frac{\partial}{\partial t} (\alpha \xi + \beta \zeta) B(t) + \nabla (\alpha \xi - \beta \zeta) \times E(t) + 2\beta \zeta \partial_t B(t). \]

The current response is the central phenomenological relation for dynamical magneto-electric effect as shown in Eq. (3) of the main text. In the following, we give a microscopic derivation for the DME based on the linear response theory.

2. Linear response theory of dynamical magneto-electric effect.

For a system \( H_0 \) with perturbations, the total Hamiltonian

\[ H = H_0 + V_1(t) + V_2(t) + \cdots, \]

where \( n \) in \( V_n \) marked the \( n \)-th order perturbation. And the equation of motion for density operator reads \( i\hbar \dot{\rho}(t) = [H, \rho] \), where \( \rho_0 \) is the unperturbed density operator. According to standard perturbation procedure [cite related ref.], we replace \( \sum V_n(t) \) and \( \rho \) with \( \sum \lambda^n V_n(t) \) and \( \sum \lambda^n \rho_n(t) \) in the equation of motion, and separate terms according to the order of \( \lambda \), then we can get the equation of motion for different order of density operator:

\[ i\hbar \dot{\rho}_0 = [H_0, \rho_0] \]

\[ i\hbar \dot{\rho}_1(t) = [H_0, \rho_1(t)] + [V_1(t), \rho_0]. \]

Since the density operator here is in Schrödinger picture, it is convenient to utilize the interaction picture, \( \rho^I(t) = e^{iH_0t} \rho(t) e^{-iH_0t}. \) The equation of motion for density operator can be simplified as:

\[ i\hbar \dot{\rho}_0^I = 0 \]

\[ i\hbar \dot{\rho}_1^I(t) = [V_1^I(t), \rho_0^I(t)]. \]

After integral, the density operator can be simplified as:

\[ \rho_1^I(t) = \frac{i}{\hbar} \int_{-\infty}^{t} dt' [V_1^I(t'), \rho_0^I]. \]

For a response \( R \) excited by an excitation \( E, V \), we can identify response function [21]:

\[ \mathcal{F}^{(1)}(t, t) = \text{Tr} \left[ \rho_0(t) \frac{\partial \hat{R}(t)}{\partial E(t)} \right] \]

\[ = \langle \frac{\partial \hat{R}(t)}{\partial E(t)} \rangle_0 \]
\[ \mathcal{F}^{(1)}(t, t') = \Theta_\nu \text{Tr} \left[ \frac{\delta p_i(t) \partial V_{ij}^L(t')}{\partial E(t')} \hat{R}(t) \right] \]
\[ = -i \hbar \Theta_\nu < \left[ R(t), \frac{\partial V_{ij}^L(t')}{\partial E(t')} \right] >_0, \]

where (1) in \( \mathcal{F}^{(1)}(t, t) \) represent first order, and the second integer represent two different response functions. For magneto-electric response, \( R \) operator could be Polarization \( \mathbf{P} \), excitation \( E \) could be electric field \( \mathbf{E} \), perturbation \( V_1 = -\mathbf{M} \cdot \mathbf{B} \), or \( R \) operator could be magnetization operator \( \mathbf{M} \), excitation \( E \) could be electric field \( \mathbf{B} \), perturbation \( V_1 = -\mathbf{P} \cdot \mathbf{E} \).

For \( \mathcal{F}^{(1)}(t, t) \), we identify it as \( \lambda(t, t) \),
\[ \lambda(t, t) = -\left< \frac{\partial \mathbf{P}}{\partial \mathbf{B}} \right>_{E_0} < \left< \frac{\partial \mathbf{M}}{\partial E} \right>_{\mathbf{B}_0} \]
\[ = -\left< \frac{\partial^2 H}{\partial E \partial \mathbf{B}} \right>_{E_0, \mathbf{B}_0} \equiv \alpha_\lambda. \] (S38)

As for \( \mathcal{F}^{(1)}(t, t') \), we have two functions:
\[ \xi(t, t') = -i \hbar \Theta(t - t') < \left| \left[ \hat{P}_i(t), \hat{M}_j(t') \right] \right| >_0 \] (S39)
\[ \xi'(t, t') = -i \hbar \Theta(t - t') < \left| \left[ \hat{M}_i(t), \hat{P}_j(t') \right] \right| >_0 \] (S40)

then Fourier transform from \( t - t' \) to frequent \( \omega \):
\[ \xi_{ij}(\omega = \omega_1) \]
\[ = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dt dt' e^{i\omega t} \xi_{ij}(t, t') \]
\[ = -i \hbar \int \frac{dk}{(2\pi)^d} \int_{-\infty}^{+\infty} dt dt' e^{i\omega t} \Theta(t - t') \left< \left| \left[ P_{li}^j M_{mm'}^l \right] \right| >_0 \right> \]
\[ = -i \hbar \sum_{l',j,m,m'} \int \frac{dk}{(2\pi)^d} \int_{-\infty}^{+\infty} dt dt' e^{i\omega t} \Theta(t - t') \left< \left| c_{l'}^\dagger(t) c_j^\dagger(t') c_m(t') c_{m'}(t) \right| >_0 \right> \]
\[ = -i \hbar \sum_{l',j,m,m'} \int \frac{dk}{(2\pi)^d} \int_{-\infty}^{+\infty} dt dt' e^{i\omega t} \Theta(t - t') \left< \left| P_{li}^j M_{mm'}^l \right| >_0 \right> \]
\[ = -i \hbar \sum_{l',j,m,m'} \int \frac{dk}{(2\pi)^d} \int_{-\infty}^{+\infty} dt dt' e^{i\omega t} \Theta(t - t') \left< \left| M_{mn}^j P_{li}^j \right| >_0 \right> \]
\[ = i \int \frac{dk}{(2\pi)^d} \int_{-\infty}^{+\infty} dt dt' \left< \left| P_{li}^j M_{mn}^j M_{mn'}^j \right| >_0 \right> \]
\[ = i \int \frac{dk}{(2\pi)^d} \int_{-\infty}^{+\infty} dt dt' \left< \left| P_{li}^j M_{mn}^j M_{mn'}^j \right| >_0 \right> \]

where \( P_{li}^j = e r_{li}^j = e < nk | \hat{r}_i | mk > = e < nk | \hat{r}_i | mk > = e < nk | \hat{r}_i | mk > = e A_{l,m}^i = i A_{l,m}^i / (\varepsilon_{mk} - \varepsilon_{nk}) \) is inter-band Berry connection, and \( M_{mn}^j = \frac{1}{2} \sum_{l \neq m} (\mathbf{v}_{nl} \times A_{lm}) \) is inter-band elements of orbital magnetization.
\[ \xi_{ij}(\omega) = e \int \frac{dk}{(2\pi)^d} \sum_{n \neq m} \frac{(f_{nk} - f_{mk}) A_{nm}^j M_{mn}^j}{\varepsilon_{nk} - \varepsilon_{mk} + i\eta} \] (S42)
\( \xi'_{ji}(\omega) = e \int \frac{dk}{(2\pi)^d} \sum_{n\neq m} \frac{(f_{nk} - f_{mk}) M_{nm}^i M_{mn}^j}{\varepsilon_{nk} - \varepsilon_{mk} + \hbar \omega + i\eta} \)  

(S43)

Furthermore, we can get two diagonal response functions from linear combination of \( Q(\omega) \) and \( Q'(\omega) \),

\[
\alpha_{ii}^\xi = \frac{[\xi_{ii}(\omega) + \xi_{ii}'(\omega)]}{2}
\]

(S44)

\[
\beta_{ii}^\xi(\omega) = \frac{[\xi_{ii}(\omega) - \xi_{ii}'(\omega)]}{2}
\]

(S45)

These dynamical magneto-electric coefficients represent the main microscopic results of our work as shown in Eq. (8)-(9) of the main text.