Nonlinear Quantum Electrodynamics in Dirac materials

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Classical electromagnetism is linear. However, fields can polarize the vacuum Dirac sea, causing quantum nonlinear electromagnetic phenomena, e.g., scattering and splitting of photons, that occur only in very strong fields found in neutron stars or heavy ion colliders. We show that strong nonlinearity arises in Dirac materials at much lower fields \( \sim 1 \text{T} \), allowing us to explore the nonperturbative, extremely high field limit of quantum electrodynamics in solids. We explain recent experiments in a unified framework and predict a new class of nonlinear magneto-electric effects, including a magnetic enhancement of dielectric constant of insulators and a strong electric modulation of magnetization. We propose experiments and discuss the applications in novel materials.

The nonlinear effects contribute to the experimentally observed high-field magnetization in the recent work on the Weyl semimetal TaAs \([29]\) and the Dirac semimetal Bi \([30]\), but the importance of this observation and its origin in the Heisenberg-Euler effect has not been recognized. In the present work we demonstrate this connection and show that the data \([29, 30]\) agree with our predictions. More importantly, we predict a new class of magneto-electric effects. The most significant is the magnetic field modulation of the Dirac material:

\[
\delta L_{HE} \rightarrow \frac{\Delta}{24\pi^2\lambda_D^3} \left[ (|\mathbf{b}| \cdot |\mathbf{e}|)^2 |\mathbf{b}|^{-1} + |\mathbf{b}|^2 \ln |\mathbf{b}| \right],
\]

in a strong \( B \), and weak \( E \)-field (See further below and also Sec. S6 of the supplement). Here, the dimensionless vectors \( \mathbf{e} \) and \( \mathbf{b} \) depend on the fine structure constant \( \alpha_D = e^2/\hbar v \) of the Dirac material:

\[
\mathbf{e}(\alpha_D) = \frac{\mathbf{UE}}{E_s(\alpha_D)}, \quad \mathbf{b}(\alpha_D) = \frac{\mathbf{U}^{-1} \mathbf{B}}{B_s(\alpha_D)}.
\]
and the critical ‘Schwinger’ electric $E_\star(\alpha_D)$ and magnetic $B_\star(\alpha_D)$ fields in the material are defined by

$$E_\star^2(\alpha_D) = \frac{v^2}{e^2} B_\star^2(\alpha_D) = \frac{\Delta}{\alpha_D \Lambda_D} = \left(\frac{\Delta^2}{e^2 \alpha_D}\right)^2.$$  (4)

Eqs. (2), (3) and (4) account for the anisotropy of real materials [32], for which the velocity tensor is a $3 \times 3$ symmetric matrix [10] $\mathbf{V} = \mathbf{v} \mathbf{u}$, with $\det(\mathbf{U}) = 1$. The term $\mathbf{UE}$ and $\mathbf{U}^{-1}\mathbf{B}$ are linear transformations of $\mathbf{E}$ and $\mathbf{B}$ respectively (See Supplement Sec. ?? [33]).

We have defined the symbols in Eqs.(3),(4) according to convention in QED. The “Dirac wavelength” $\lambda_D = \frac{hv}{\Delta}$ and the “Dirac magneton” $\mu_D = \frac{ehv}{2\Delta^2}$ replace the Compton wavelength and the Bohr magneton respectively. When the fields reach the ‘Schwinger scale’, Zeeman splitting and the potential difference at $\Lambda_D$ are equal to the half of the Dirac band-gap:

$$2\mu_D B_\star = \lambda_D e E_\star = \Delta,$$  (5)

and the nonlinearity becomes relevant. In Table I we list material parameters considered in this work. For more details see Sec. S2 and Table S2 of the Supplement [33].

The quantum contribution to the Lagrangian can be viewed as the sum of the infinite chain of 1-loop diagrams in Fig.1 that represent the polarization of the Dirac sea of electrons by external electric and magnetic fields. In this work we consider only non-magnetic crystals with inversion symmetry [40] [41] and assume the static/quasistatic approximation, $\omega, kv \ll \Delta$, where $\omega$ and $k$ are the frequency and the wave number of the external fields. Therefore our diagrams, Fig.1, have only even numbers of external E-lines and B-lines. Besides diagrams in Fig.1, there are also multi-loop diagrams suppressed by a factor of $\alpha_D/e \sim 0.03$ per each additional loop, where $e$ is the large dielectric constant mainly due to the lattice and intra-ionic polarization. For the discussion of the suppression of the multi-loop diagrams in the context of phenomena considered here, see Sec. S3 in the supplement and also Refs. [42, 43].

In Fig. 1, the first diagram quadratic in external fields is ultraviolet divergent and is equal to [24, 39]

$$\delta L_1 = \frac{\Delta}{12\pi^2 \Lambda_D^2} \ln \left(\frac{\Lambda}{\Delta}\right) \left(|e|^2 - |b|^2\right).$$  (6)

Here the subscript ‘1’ indicates contribution from the first diagram in Fig.1 and $\Lambda \sim v \Delta^{5/2} \sim 1$ eV is the ultraviolet cut-off energy, where $a$ being the lattice spacing. In QED this diagram describes the electric permittivity and magnetic permeability of vacuum and thus it is included into the definitions of the electric charge and electromagnetic fields. As a result, $\delta L_1$ does not appear explicitly in QED. However, for Dirac materials $\delta L_1$ is an explicit contribution that has to be added to the classical Lagrangian Eq. (1). Indeed, this is the contribution of the Dirac sea (valence band) to the dielectric constant and magnetic susceptibility.

Equating $(E^2 - B^2)/(8\pi) + \delta L_1$ to the classical Lagrangian (1), we find the linear dielectric constant $\epsilon_D$ and the linear magnetic susceptibility $\chi_D (\mu = 1+4\pi\chi)$:

$$\epsilon_D = 1 + \frac{2\alpha_D}{3\pi} \ln \left(\frac{\Lambda}{\Delta}\right) \mathbf{U}^2, \quad \epsilon_D \sim 3, \quad (7)$$

$$\chi_D = -\frac{\alpha_D}{6\pi^2 e^2} \ln \left(\frac{\Lambda}{\Delta}\right) \mathbf{U}^{-2}, \quad \chi_D \sim -10^{-6}. \quad (8)$$

where estimates are given for the diagonalized tensors.

Eqs. (7) and (8) define the Dirac contributions to the total dielectric and magnetic susceptibilities. The contribution (7) is relatively small compared to the total relative permittivity $\epsilon$ in Eq. (1), typically $\epsilon \sim 100$, which is primarily due to theionic (lattice) and intra-ionic contributions (See Supplement Table S3). The magnetic response (8) constitutes a significant part of the diamagnetic susceptibility, which also has contributions from lower bands and core electrons. For Bismuth, the Dirac valence band contribution (8) has been previously considered in Ref. [44].

We describe now the nonlinear effects. The diagrams in Fig.1 beyond the first one ($n \geq 2$) are convergent at arbitrarily large $|e|, |b|$ [45] and are re-summed exactly [46] to yield the 1-loop, nonperturbative Heisenberg-Euler action:

$$\delta L_{HE} = \sum_{n=2}^{\infty} \delta L_n \equiv -\frac{\Delta}{8\pi^2 \Lambda_D^2} \int_0^\infty d\eta e^{-\eta}$$

$$\times \left[ A_+ \cot(\eta A_+) A_+ \cot(\eta A_+) - \frac{1}{\eta^2} + \frac{1}{3} (A_+^2 + A_+^2) \right],$$  (9)

which accounts for crystal anisotropy, cf., Eq. (3), as well as the strong field behavior. The imaginary part of Eq.(9), obtained via its analytic continuation, captures the electric breakdown, which can be avoided in weak electric fields $|e| < 1$ ($E < E_\star$). Then, Eq. (9) can be expanded in powers of $e$. However, the magnetic field can be much larger than $B_\star$, leading to the asymptotic expression Eq. (2) (See Supplement Sec. S6). At weak magnetic fields, $|e|, |b| \ll 1$, Eq. (9) reduces to the 2nd diagram in Fig.1,

$$\delta L_2 = \frac{\Delta}{360\pi^2 \Lambda_D} \left[ (|e|^2 - |b|^2)^2 + 7(e \cdot b)^2 \right].$$  (10)
Here we study the universal nonlinear susceptibility and eliminate all uncertainties such as the choice of ultraviolet cut-off $\Lambda$, subleading terms and contributions from other bands or core electrons.

Both TaAs and Bi have nonzero chemical potential and hence have conduction electrons. Therefore at weak magnetic fields both compounds show magnetic oscillations. The conduction electrons freeze and the oscillations disappear at $B > 5 T$ in Bi [30] and $B > 10 - 13 T$ in TaAs [29]. In these ranges of $B$, we can compare the data with our predictions. In Fig. 2a the points show magnetization quasi-linear in the applied $B$-field is investigated. Here we study the universal nonlinear susceptibility and eliminate all uncertainties such as the choice of ultraviolet cut-off $\Lambda$, subleading terms and contributions from other bands or core electrons.

We now consider novel magneto-electric effects. The

$F(|b|) = \ln |b|, \ |b| \gg 1.$ (11)

The dimensionless function $F(|b|)$ in the full range of magnetic fields obtained by numerical integration of Eq. (9) is shown in Supplement Fig. S2. Strong and weak field limits of $F$ follow from the actions given by Eqs. (2) and (10) respectively.

The total magnetic susceptibility of the Dirac valence band is the sum of the linear susceptibility, Eq. (8) and the nonlinear contribution, $\chi = \chi_D + \delta \chi$. When $\ |b| \gg 1$ we have

$\chi = -\frac{\alpha_D}{12\pi^2 c^2} \frac{v^2}{c} \ln \left( \frac{c\Lambda^2}{\epsilon} |b| \right).$ (12)

Here $\chi$ depends on $B$ but not on $\Delta$, and is well-defined in the limit $\Delta = 0$, as in the Weyl semimetal TaAs [29].

According to Eqs. (11),(12) the magnetic susceptibility is nonlinear, i.e. it depends on magnetic field. Remarkably, this Dirac linearity has been recently observed, but its connection to nonlinear electrodynamics was not identified. Here we show its origin in the Heisenberg-Euler effect. The magnetization of Weyl semimetal TaAs has been measured up to $B = 30 T$, Ref. [29], and magnetization of Dirac semimetal Bi has been measured up to $B = 60 T$, Ref. [30]. In Zhang et al. [29] the valence band contribution to magnetization at $E = 0$ was considered [47], and in the high magnetic field limit, the magnetization quasi-linear in the applied $B$-field is investigated. Here we study the universal nonlinear susceptibility and eliminate all uncertainties such as the choice of ultraviolet cut-off $\Lambda$, subleading terms and contributions from other bands or core electrons.

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nonlinear dielectric constant is
\[
\delta \epsilon_D = 4\pi \frac{\partial^2 \delta L_{\text{HE}}}{\partial E \partial E} = \mathcal{U}^2 \frac{\alpha_D}{3\pi} G_i(|b|); \tag{13}
\]
\[
|b| \ll 1 : \quad G_i(|b|) = \frac{1}{3} |b|^2, \quad G_\perp(|b|) = -\frac{2}{15} |b|^2
\]
\[
|b| \gg 1 : \quad G_i(|b|) = |b|, \quad G_\perp(|b|) = -\ln(|b|).
\]

Here the index \( i = ||, \perp \) relates the relative orientation of \( \mathbf{e}, \mathbf{b} \) [48]. Dimensionless functions \( G_i(|b|) \) in the whole range of \( \mathbf{b} \) obtained by numerical integration of (9) are plotted in Supplement Fig. S2. Its strong and weak field limits define actions given by Eqs. (2) and (10) respectively. The dependence of the dielectric constant on the applied magnetic field is a novel magneto-electric effect. For \( \mathbf{b} \parallel \mathbf{e} \) the contribution \( \delta \epsilon_D \) is positive and can be very large, while for \( \mathbf{b} \perp \mathbf{e} \) the contribution \( \delta \epsilon_D \) is negative. The expressions for arbitrary angle between \( \mathbf{b}, \mathbf{e} \) and the relation to the angle between applied fields \( \mathbf{B} \) and \( \mathbf{E} \), which is generally different due to properties of the anisotropy transformation are given in Sec. S7. Furthermore, according to (1), (10) there is a nonlinear contribution quadratic in the electric field,
\[
\delta \epsilon_D(E) = \mu^2 \frac{2\alpha_D}{15 \pi} |\mathbf{e}|^2, \quad |\mathbf{b}| = 0, \tag{14}
\]
which is suppressed by \( |\mathbf{e}|^2/|\mathbf{b}| \) when \( |\mathbf{b}| \gg 1 \). Notably, at \( |\mathbf{e}|, |\mathbf{b}| \ll 1 \), contributions (13) and (14) add up.

The magnetic field induced variation of the dielectric constant in Eq. (13) scales as \( \delta \epsilon_D \propto 1/|\mathbf{B}| \propto \Delta^{-2} \). Thus, the effect is most significant in a small band-gap Dirac insulators. In Fig. 2b we plot our predictions for Bi_{0.9}Sb_{0.1}. For \( \mathbf{e} || \mathbf{b} \) the effect is enormous, \( \delta \epsilon_D \approx 10 \) Tesla. For \( \mathbf{E} \perp \mathbf{B} \) the effect is smaller and has the negative sign. In the same Fig. 2b we also plot predictions for \( \delta \epsilon_D \) in Pb_{0.5}Sn_{0.5}Te. This compound has larger gap and therefore the effect is smaller, but still observable.

One more novel magneto-electric effect is the dependence of magnetization, on the applied electric field. The electric field dependent magnetization \( M^{(e)} = \frac{\partial \mathbf{M}}{\partial B} \), in units of “Dirac magnetons” per “Dirac volume”, reads
\[
4\pi M^{(e)} = \mathcal{U}^{-1} \frac{\mu D}{|b|} \frac{\mu_0}{3\pi \lambda_D^2} |\mathbf{e}|^2 D_i(|b|); \tag{15}
\]
\[
|b| \ll 1 : \quad D_i(|b|) = \frac{2}{3} |b|, \quad D_\perp(|b|) = -\frac{4}{15} |b| |b| \ll 1 : \quad D_i(|b|) = 1, \quad D_\perp(|b|) = -\frac{1}{|b|^2}.
\]

The direction of the magnetization (15) in a Dirac crystal is defined by the vector \( \mathbf{b} \) and depends on crystal anisotropy as described by Eq. (3). Dimensionless functions \( D_i(|b|) \) in the whole range of \( \mathbf{b} \) obtained by numerical integration of (9) are plotted in Fig S2 in Supplementary material. For \( \mathbf{b} \parallel \mathbf{e} \) the magnetization is large and paramagnetic, while for \( \mathbf{b} \perp \mathbf{e} \) the magnetization is diamagnetic [48]. Magnetization (15) is quadratic in the applied electric field and as a function of magnetic field, saturates when \( |\mathbf{b}| \gg 1 \).

To enhance the magnetization in Eq. (15) one needs the electric field as strong as possible. However, the field is limited by the dielectric strength, \( E_d \) of the material, beyond which dielectric breakdown occurs. The breakdown probability (rate of Zener tunneling by electric field per unit volume) is obtained from Eq. (9) [25] and found to be \( P \propto |\mathbf{e}|^2 e^{-\pi/|\mathbf{e}|} \) (See Sec. S4). The most important here is the exponential dependence, which universally applies to both the Dirac spectrum and quadratic dispersion. Thus, one expects that \( E_d \) is proportional to \( E_\ast \). Taking two band insulators, diamond \( (2\Delta \approx 5.5 eV, E_d \approx 10^5 V/cm) \), and silicon \( (2\Delta \approx 1.14 eV, E_d \approx 3 \times 10^5 V/cm) \), as reference materials, we observe that the dielectric strength scales as \( E_d \propto \Delta^2 \). Therefore \( E_d \) is a fixed fraction of \( E_\ast \). Significant \( E \)-dependent magnetic effects Eq. (15) can then be observed for \( |\mathbf{e}| \approx 0.1-0.3 \) [49]. Furthermore, as usual in solids, setups with huge built-in electric fields in the insulating regime can be explored [50].

For a fixed \( \mathbf{e} = E/E_\ast \), the electric field modulated magnetization in Eq. 15 obeys \( M^{(e)} \propto B_\ast \propto \Delta^2 \), so materials with large gap are preferable, unlike in the dependence of dielectric constant on magnetic field. In Fig. 2c we plot the predicted magnetization for Pb_{0.5}Sn_{0.5}Te versus magnetic field at \( E = 10^4 \) V/cm, which corresponds to \( |\mathbf{e}| \approx 0.3 \). For the both fields, \( \mathbf{e} \) and \( \mathbf{b} \), parallel to the c-axis, the electric field driven magnetization is \( 4\pi M^{(e)} \approx 0.2 \mu T \) at \( B = 1T \). When \( \mathbf{E} \perp \mathbf{B} \), the magnetization changes sign, see Fig. 2c. In the same figure we also plot the magnetization in Bi_{0.9}Sb_{0.1} for \( \approx 0.3 \). Here the effect is smaller due to the smaller Dirac gap.

The electric field driven magnetization in Bi_{0.9}Sb_{0.1} \( (4\pi M^{(e)} \sim 10^{-8} T) \) and in Pb_{0.5}Sn_{0.5}Te \( (4\pi M^{(e)} \sim 2 \times 10^{-7} T) \) can be feasibly detected in lock-in experiments, in an applied electric field having a constant and an AC component (with frequency \( \omega \)). The induced magnetization is then characterized by contributions modulated at frequencies \( \omega \) and \( 2\omega \). Of course, the condition \( \hbar \omega \ll 2\Delta \) is assured fulfilled. Experiments on observation of \( M^{(e)} \) could also take advantage of the SQUID magnetometry, sensitive to magnetization as low as \( 10^{-15} \) T/\( \sqrt{Hz} \) [51], much lower than the predicted values.

In conclusion, Based on the Heisenberg-Euler theory of the physical vacuum we develop the theory of nonlinear electromagnetic effects in Dirac materials. We explain the results of two recent experiments on nonlinear contribution to magnetization of Dirac materials. We predict two novel magneto-electric effects and discuss possible experiments and materials for their observation.

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Supplemental Material

S1. DIRAC CONE ANISOTROPY

Here, for the sake of completeness, we provide a prescription to treat anisotropic crystals, adopted from Ref. [32]. In real crystals the velocity is a tensor that is represented by a $3 \times 3$ real symmetric matrix $\mathbf{V}$, so that the Dirac Hamiltonian reads

$$H = \beta \Delta - 1|e\phi + \sum_{i,j} \alpha_i \mathcal{V}_{ij} (p_j + |e| A_j / c), \quad i, j = 1, 2, 3.$$  \hfill (S16)

where $\alpha_i, \beta$ are $4 \times 4$ Dirac matrices and $\mathbf{1}$ is a $4 \times 4$ identity matrix. We define the Dirac velocity $\mathbf{v}$ and the anisotropy matrix $\mathbf{U}$ as

$$v^3 = |\det(\mathbf{V})|, \quad \mathbf{U} = \mathbf{V}/v.$$  \hfill (S17)

A. Dilation/contractions of space that renders the cone isotropic

The matrix $\mathbf{U}$, being a symmetric matrix with $\det(\mathbf{U}) = 1$, without loss of generality, induces a volume preserving transformation on coordinates, that is

$$\tilde{x} = \mathbf{U}^{-1}x, \quad \tilde{p} = \mathbf{U}p, \quad \tilde{A} = \mathbf{U}A, \quad d^3\tilde{x} = d^3x,$$  \hfill (S18)

with which the Dirac Hamiltonian assumes its isotropic form

$$H = v\alpha \cdot (\tilde{p} + |e| \tilde{A}/c) + \beta \Delta - 1|e|\phi.$$  \hfill (S19)

The Lagrangian, being a scalar, is invariant under this volume preserving transformation. Meanwhile, the scaled electric and magnetic fields are

$$\tilde{E} = \frac{\partial \phi}{\partial \tilde{x}} = \mathbf{U}E, \quad \tilde{B} = \mathbf{U}^{-1}B,$$  \hfill (S20)

where the equation for $\tilde{B}$ follows from the identity (repeated indices are summed)

$$\mathcal{U}_{nk} \tilde{B}_k = \mathcal{U}_{nk} \frac{\partial \tilde{A}_i}{\partial \tilde{x}_j} \epsilon_{ijk} = \frac{\partial A_s}{\partial x_m} \mathcal{U}_{im} \mathcal{U}_{js} \mathcal{U}_{nk} \epsilon_{ijk} = B_n.$$  \hfill (S21)

Finally, the physical susceptibilities $\chi, \chi^e$ in the anisotropic crystal as a function of applied fields $\mathbf{E}, \mathbf{B}$ are

$$\chi(\mathbf{E}, \mathbf{B}) = \frac{\partial^2 L}{\partial B \partial \mathbf{B}} = \mathbf{U}^{-1} \tilde{\chi}(\tilde{\mathbf{E}}, \tilde{\mathbf{B}}) \mathbf{U}^{-1}$$

$$\chi^e(\mathbf{E}, \mathbf{B}) = \frac{\partial^2 L}{\partial E \partial \mathbf{E}} = \mathbf{U} \tilde{\chi^e}(\tilde{\mathbf{E}}, \tilde{\mathbf{B}}) \mathbf{U}.$$  \hfill (S22)

The magnetization and polarization vectors and higher order susceptibility tensors transform in the usual way like above. We re-iterate a point already mentioned in the main text that even when the crystal anisotropy is not taken into account, that is $\mathbf{U} = \mathbf{1}$, the susceptibilities are intrinsically anisotropic, since they depend on the mutual alignment of electric and magnetic fields. Crystal anisotropy is an additional source of directional dependence that is relevant in the experimental context.

B. Invariance of $\mathbf{E} \cdot \mathbf{B}$ and the transformation of the angle between $\mathbf{E}$ and $\mathbf{B}$

In account of anisotropy, the normalized fields are defined as Eq. (3). We note that the scalar product of externally applied fields $\mathbf{E} \cdot \mathbf{B}$ transforms as

$$\mathbf{E} \cdot \mathbf{B} = \mathcal{U}^{-1}_{ik} \tilde{E}_k \mathcal{U}_{li} \tilde{B}_l = B_\star \mathbf{e} \cdot \mathbf{b},$$  \hfill (S23)
i.e., is proportional to the scalar product of the transformed fields. However, we note that $U$ is not a simple rotation. Hence, while scalar products are proportional, it does not preserve angles or norms taken separately. We see this when we express $\hat{e} \cdot \hat{b}$ in terms of $\hat{E} \cdot \hat{B}$ as

$$
\hat{e} \cdot \hat{b} = \frac{e \cdot b}{|e||b|} = \frac{E \cdot B}{|UE||U^{-1}B|} = \frac{|E||B|}{|UE||U^{-1}B|} \hat{E} \cdot \hat{B}
$$

(S24)

Therefore

$$
E \perp B \iff e \perp b.
$$

(S25)

We can work in a reference frame where $U$ is diagonal. However, even when $E \parallel B$ are directed along a general direction, we have

$$
E \parallel B \implies \hat{e} \cdot \hat{b} = \frac{1}{|UE||U^{-1}E|} \leq 1
$$

(S26)

The equality is satisfied when $E$ and $B$ are directed along the principle directions of the frame that diagonalizes $U$, e.g. $\hat{E} = \hat{B} = (1, 0, 0)^T$. However, for general direction, the vectors $e$ and $b$ are no longer parallel, for example, if

$$
U = \text{diag}(u, u, 1/u^2), \quad \hat{E} = \hat{B} = \frac{1}{\sqrt{3}}(1, 1, 1)^T
$$

(S27)

we obtain

$$
\hat{e} \cdot \hat{b} = \left(\frac{2}{u^2 + u^4} \left(2u^2 + \frac{1}{u^2}\right)\right)^{1/2}
$$

(S28)

For exemplary values of $u = 1, 2, 1.5, 2$, we get the angle between $\hat{e}$ and $\hat{b}$ as $28^\circ, 55^\circ, 75^\circ$ respectively.

In terms of transformed vectors $e$ and $b$, Dirac materials are described by a Hamiltonian that has the usual isotropic form which simplifies calculations. In particular, for the perpendicular fields $e$ and $b$, one can apply the Lorentz transformation that eliminates the smaller of the fields, resulting in the purely electric field or the purely magnetic field case. In the case when fields $e$ and $b$ are not orthogonal, a particular choice of a Lorentz transformation leads to a Hamiltonian with a new set of electric and magnetic fields parallel to each other, for which the calculations of dielectric and magnetic susceptibilities are simplified (Section S6). Once the results are obtained in the frame where the fields are parallel, the inverse Lorentz transformation allows to express them in terms of the original non-orthogonal fields $e$ and $b$. Finally, inverse transformation to initial anisotropic system will give the results in terms of applied fields $E$ and $B$. In Sec. S7 we derive the susceptibilities for an arbitrary angle between $e$ and $b$, see Table S6 for quick reference.

### S2. COMPARISON TO THE RELATIVISTIC QUANTUM ELECTRODYNAMICS

The Dirac equation that follows from Eq. (S19), after multiplying from the left by $\beta$, is

$$
i\gamma^0(\partial_t - i|e|\phi) - v\gamma \cdot (\hat{p} + |e|\vec{A}/c)\psi - \Delta \psi = 0, \quad \gamma^0 = \beta, \quad \gamma^i = \beta\alpha^i.
$$

(S29)

If we define the effective and true 4-position and electromagnetic 4-potential respectively as

$$
effective: \quad \hat{x}^\mu = (vt, \vec{x}), \quad A^\mu = \frac{1}{v}(\phi, v\vec{A}/c)
$$

(S30)

true-relativistic: \quad \hat{x}^\mu = (ct, \vec{x}), \quad A^\mu = \frac{1}{c}(\phi, \vec{A}), \quad \text{where } \eta^\mu{}^\nu = (+, -, -, -),

(S31)

we can write down the Dirac action coupled to classical electromagnetism in the (anisotropic) material as

$$
\text{Dirac material: } S = \int dt d^3\vec{x} \left( \psi^\text{\dagger} (i\gamma^\mu(\hat{\partial}_\mu - i|e|A_\mu) - \Delta) \psi + \frac{1}{8\pi}(E^T\epsilon E - B^T \mu^{-1} B) \right).
$$

(S32)

In addition, when the band gap is inverted, as in topological insulators, there is an additional boundary term bulk-boundary term $E \cdot B$ [54] which does not effect the nonlinear response which we investigate in this letter. For this reason we assume $\Delta > 0$ with out loss of generality.
The first difference one can notice is the speed of the Dirac fermion, that is \( c \) in QED. In the material with a gap \( 2\Delta \),

\[
\text{QED: }\quad S = \int dtd^3x \left( \bar{\psi}i\gamma^\mu(\partial_\mu - i|A_\mu|) - m_e c^2|\psi| + \frac{1}{8\pi}(E^2 - B^2) \right).
\]

The first difference one can notice is the speed of the Dirac fermion, that is \( c \) in QED. In the material with a gap \( 2\Delta \),

\[
|e|^2 = \frac{\alpha_D E^2}{\Delta/\Lambda^2} \quad \text{or} \quad |b|^2 = \frac{\alpha_D B^2 v^2}{\Delta/\Lambda^2},
\]

are large, the diagrams can be exactly summed to yield Eq. (S39)
the Fermi-Dirac velocity is related to the effective mass $m^*$ (measured in terms of electron mass) and satisfies
\[
E = \sqrt{\Delta^2 + p^2v^2}, \quad E \approx \Delta + \frac{v^2p^2}{2\Delta} \Rightarrow m^*m_ev^2 = \Delta,
\]
\[
\Delta \approx 0.1 \text{ eV, } m^* = 0.01-0.5 \Rightarrow 1000 \gtrsim c/v \gtrsim 100.
\]

For this reason, the effective fine structure constant of the insulator is
\[
\alpha_D = \frac{e^2}{\hbar v} \sim \alpha \times 400 \approx 3. \tag{S35}
\]

The minimally coupled 4-potentials are different as outlined in Eq. (S30). The classical action of the electromagnetic field in the two cases are obviously different. In the material case, $\epsilon, \mu$ are symmetric matrices that define the relative permittivity and the relativity of the material, respectively. In the vacuum they satisfy $\epsilon = \mu = 1$ in the CGS units.

A more subtle difference between QED and the Dirac insulator is the definition of electric charge and electric field, when seen from a renormalization point of view. In QED the charge is normalized at zero momentum transfer, $e_0 = e_q=0$ (referred to as the running electric charge [26], which is related to the bare charge $\bar{e}$ through the ultraviolet (UV) cutoff dependent vacuum dielectric screening, $e_0 = \bar{e}/\sqrt{\epsilon\Lambda}$. However, in condensed matter, the charge is defined independently of scale and is given by $e \approx 1.6 \times 10^{-19}$ C. Nevertheless, due to RPA dielectric screening, the apparent charge at the macroscopic scale is $e/\sqrt{\epsilon}$, hence different from the bare $e$ that applies at the lattice spacing scale $q \sim \hbar\pi/a$. Definitions of the electric field are also different. In QED, the field is normalized to $\vec{E} = \sqrt{\epsilon\Lambda} \vec{E}$ so that the energy density of the field is fixed as $E^2/(8\pi)$, and the dielectric constant of vacuum is by definition equal to unity. However in condensed matter, the Lagrangian of the free field, is already written in terms of the screened field $\vec{E}$, as $eE^2/(8\pi)$. The coupling term is also written in terms of the bare charge and the screened field $e\vec{E}$, hence $e$ does not appear in the definition of the effective fine structure constant $\alpha_D = e^2/(\hbar v)$.

Importantly, the coupling term is independent of the definition, $e_0\vec{E} = e\vec{E}$, allowing us to use the 1-loop renormalized effective action of QED in the condensed matter setting. A detailed comparison of parameters in Dirac insulator to their counterparts in QED is given in Table S2.

### S3. EFFECTIVE ACTION FOR DIRAC INSULATOR

The effective action can be schematically organized in terms of multi-leg and multi-loop diagrams as in Fig. S1. The multi-loop diagrams contain at least one internal interaction line and therefore in QED, each such line contains the factor $\alpha \sim 0.01$.

We now justify the possibility to limit the calculation of the effective action to the 1-loop approach and justify neglecting multi-loop diagrams in Dirac materials, particularly insulators. In the Dirac insulator, we first note that multi-loop diagrams are UV convergent hence the polarization is dictated mostly by the low energy-momentum sector where the static dielectric constant $\epsilon$ applies [42, 43]. The Coulomb interaction is therefore screened due to $\epsilon$, which is dominated by the lattice and intra-ionic polarization contribution. In a wide range of practical situations as seen in Table S3 $\epsilon \sim 100$ and therefore the interaction lines are suppressed by the factor $\alpha_D \sim 0.03$.

We note that, no matter how small the interaction parameter $\alpha_D/\epsilon$, the multi-loop diagrams may come with large combinatorial factors and can render the perturbation series divergent. In both QED and condensed matter physics several examples of divergences are known. In some of these divergences, the coupling constant $\alpha$ gets enhanced by an additional parameter $R$, $\alpha \to R\alpha$. For example, in an electron gas in metals, there is Random Phase Approximation (RPA) infrared divergence, where $\alpha$ is enhanced by a large distance. This diverging series of diagrams is eventually converging, and can be re-summed, as was shown by Gell-Mann Brueckner, [61] However, in the case of Dirac insulator, the electron gas infrared divergence does not arise. Indeed, for example the expansion of contribution to energy due to interactions,
\[
E = E_0 + \langle 0|H_{ee}|0\rangle + \sum_n \frac{\langle 0|H_{ee}|n\rangle \langle n|H_{ee}|0\rangle}{E_n - E_0} + ..., \tag{S36}
\]

where $E_0$ is the interaction-independent contribution to energy, $H_{ee}$ is the interaction Hamiltonian, index zero denotes the ground state of the system and index $n$ corresponds to excited states. In an electron gas, the denominator $E_n - E_0 = \frac{k^2}{m}(q^2 + (k_1 - k_2) \cdot q)$, where $k_1$ and $k_2$ are momenta (wave vectors) of the two holes inside the Fermi sphere, $q$ is the transferred momentum, $k_2 + q$ and $k_2 - q$ are the momenta of electrons outside the Fermi sphere. This denominator is divergent in the second order perturbation theory, and higher order terms are even more divergent. For the electron gas, these perturbation series can be summed. However, for the Dirac insulator, when excitations
TABLE S3: The Dirac gap and dielectric constants of Dirac materials. When multiple numbers for $\epsilon$ are provided, anisotropy is signified (see references for more information). The linear dielectric contribution from the valence bands coming from Eq. (S40) is, by definition, already contained in these measurements.

arise only in transitions between the full valence band and empty conduction band, every denominator in series Eq. (S36) contains extra constant $2\Delta$, and divergences do not appear. Furthermore, as we consider high magnetic fields, if both spin states of the highest occupied Landau level are fully filled, excitation involves a cyclotron energy gap. Then if the cyclotron energy is much larger than the characteristic Coulomb interaction, the mixing of excited states is negligible, and the non-interacting "closed shell configuration" is essentially the ground state \cite{62}. Due to large $\epsilon$, this happens in rather small magnetic fields in our case. Such situation is relevant for our consideration of Bi, where we consider the limit $\Delta = 0$ and zero electric field. It becomes clear that electron gas RPA-like contribution need not be included in our consideration.

Another type of divergence often arising due to interactions is Lippman-Schwinger \cite{63}, Bethe-Salpeter effects \cite{64}, or, in condensed matter context, the divergence related to the exciton bound state. \cite{65–67} For exciton, when $\epsilon$ is big, binding energy is small and the size is effectively big. Small binding energy means closeness in energy to the bottom of the conduction band. For this bound state to manifest itself in susceptibilities, excitations have to be generated at frequencies of electromagnetic wave close to $2\Delta$. Thus, the related contributions can be separated from the effects at small frequencies that we mostly consider.

In susceptibilities, divergences as a result of summation of series of interaction contributions may also indicate phase transitions, such as ferromagnetism. In the presence of the gap $2\Delta$ for excitations in Dirac insulators, such phase transition is not expected. Similarly, a superconducting transition, while emerging due to binding of electrons in Cooper pairs associated with the presence of the Fermi surface (see, e.g. Abrikosov \cite{68}), is unlikely to arise in the presence of excitation gap in insulators.

We note that the one-loop Heisenberg-Euler contribution at large $B$ that we calculated, Eq. 2 of the main text, also stems from divergence of the general renormalization group type, when $\alpha$ is enhanced by the first power of log. Finally, there is potentially a divergence stemming from loop expansion that differ from the cases discussed above. It contains the fine structure constant $\alpha$ only, and is asymptotic, with combinatorial coefficients growing fast with increasing the power $n$ of the fine structure constant in diagrams with $n$ interaction lines and vertices of electromagnetic interactions. These so-called asymptotic series were considered first by Dyson \cite{69} (Also see an excellent review by Huet and co-authors \cite{70}). It is now widely accepted that for asymptotic series a summation up to an infinite order in small $\alpha$ does not make sense, and one must terminate the series after a few terms when doing practical calculations with QED, although a rigorous mathematical justification for using 1-loop QED is an open problem.

Thus, we can safely restrict ourselves to 1-loop approximation since

$$\frac{(K + 1)-\text{loops}, 2n\text{-legs}}{K\text{-loops}, 2n\text{-legs}} \sim \frac{\alpha_D}{\epsilon} \approx 0.03, \quad \text{See Eq. (S35) and Table S3.} (S37)$$

This applies to QED where $\alpha \approx 1/137$, as considered by Heisenberg and Euler, and in the condensed matter contexts, when the interaction parameter $e^2/\hbar v\epsilon$ is small at large $\epsilon$, fixed by lattice properties. We note that our predictions pass the test of comparison with available experimental results (See Fig. 2 of the main text), even in the strong field regime which can not be directly probed in QED.
The 1-loop effective action of the material follows as

\[ S_{\text{eff}} = -i \ln \det \left[ v^\gamma \mu (\tilde{\partial}_\mu - i e A_\mu) + i \Delta \right]. \]  
\( \text{(S38)} \)

This determinant is exactly calculated in the QED case [27]. By comparing the QED and Dirac insulator actions in Eqs. (S32) and (S33), we can write the 1-loop, nonperturbative, renormalized Heisenberg-Euler action in terms of the normalized fields defined in Eqs. (3) and (4) as [24, 25]

\[
\delta L_{\text{HE}} = \frac{\Delta}{8\pi^2 \lambda_D^3} \int_0^\infty d\eta \eta^2 \left[ -\frac{\eta A \cot(\eta A) \eta C \coth(\eta C) + 1}{3} \eta^2 (A^2 - C^2) \right],
\]
\[
A = -i \left[ \sqrt{(b + ie)^2} - \sqrt{(b - ie)^2} \right],
\]
\[
C = \frac{1}{2} \left[ \sqrt{(b + ie)^2} + \sqrt{(b - ie)^2} \right].
\]  
\( \text{(S39)} \)

In addition to this, there is also the cut-off dependent part of the action that is due to the UV-divergent diagram (shown in Fig. S1 in a red box) which evaluates to

\[ \delta L_1 = \frac{\Delta}{12\pi^2 \lambda_D^3} \ln \left( \frac{A}{\Delta} \right) \left| \mathbf{e} \right|^2 - \left| \mathbf{b} \right|^2. \]  
\( \text{(S40)} \)

In QED, this quantity has the same form as the classical electromagnetic Lagrangian, hence it is renormalized into the definition of electric charge. However in the material case, it gives rise to the magnetic and electric susceptibilities that we discuss below absorbed into \( \mathbf{e}, \mathbf{\mu} \) of the material. The Eq. (S39) is valid for both strong and weak fields. The contour on the complex \( \eta \) plane shall be chosen so that the poles in the integrand are avoided. The imaginary part of the action gives the breakdown probability (volume rate of particle hole creation in QED). For example, when \( \left| \mathbf{b} \right| = 0 \), we have to leading order [25]

\[ P = 2 \Im \delta L_{\text{HE}} = \frac{\Delta}{4\pi^3 \hbar \lambda_D^3} \left| \mathbf{e} \right|^2 e^{-\pi/\left| \mathbf{e} \right|}. \]  
\( \text{(S41)} \)

Meanwhile, another way to write Eq. (S38) is the standard form

\[ S_{\text{eff}} = -i \ln \det \left[ v^\gamma \mu (\tilde{\partial}_\mu - i e A_\mu) + i \Delta \right] = -i \text{Tr} \ln \left[ 1 + \left| e \right| v G \gamma^\mu A_\mu \right] - i \ln \det \left[ -i G^{-1} \right]. \]  
\( \text{(S42)} \)

Carrying out the formal diagrammatic expansion of the logarithm, we have

\[ \text{1-loop, 2n-legs} = \frac{i}{2n} \text{Tr} \left[ (\left| e \right| v G) (\gamma^\mu A_\mu) (2n) \right], \quad n = 1, 2, \ldots \]  
\( \text{(S43)} \)

For example, the 2-leg diagram after taking the Fourier transform \( A(q) = \int d^4 \tilde{x} A(\tilde{x}) e^{i k x} \) reads

\[ \text{1-loop, 2-legs} = \frac{i e^2 v^2}{4} \int \frac{d^4 q d^4 k}{(2\pi)^8} \text{tr} \left[ A(q) \frac{1}{v k - \Delta} A(-q) \frac{1}{v (-k + g) - \Delta} \right], \quad k^\mu = (\omega/k, \tilde{k}), \quad \tilde{k} = \gamma^\mu k_\mu. \]  
\( \text{(S44)} \)

which gives the UV divergent term Eq. (S40). The higher order diagrams can be identified by formally expanding the action Eq. (S39) in the fields. For example, if we choose \( e \parallel \mathbf{B} \), we have [27]

\[ \text{1-loop, 2n \geq 4-legs} = -\frac{\Delta}{8\pi^3 \lambda_D^3} (2n - 3)! \sum_{k=0}^{n} \frac{B_{2k} B_{2n-2k}}{(2k)! (2n-2k)!} (2\left| \mathbf{b} \right|)^{2n-2k} (2\left| \mathbf{e} \right|)^{2k}, \]  
\( \text{(S45)} \)

where \( B_{2n} \) are Bernoulli numbers. The expansion parameters are

\[ \left| \mathbf{e} \right|^2 = \alpha_D E^2 \frac{\lambda_D^3}{\Delta}, \quad \left| \mathbf{b} \right|^2 = \alpha_D B^2 \frac{\lambda_D^3}{\Delta}. \]  
\( \text{(S46)} \)

Therefore when the fields or the coupling constant are strong, the diagrammatic expansion is only schematic because the series Eq. (S45) should be summed exactly back into the original form Eq. (S39), where the non-coplanar \( \mathbf{E} \) and \( \mathbf{B} \) case is also taken into account.
\[ \lim_{n \to \infty} \left( \frac{1}{x} + \frac{1}{3} - \coth(x) \right) \rightarrow \frac{1}{x} - \frac{1}{3} + \frac{1}{3} \cdot \frac{1}{x} - \frac{1}{3} \cdot \frac{1}{x} \cdot \coth(x) \]

| \begin{array}{c|c}
 m & I_m^n(b, x) \\
 \hline
 1 & (1 + \frac{x}{2b} + \frac{1}{3} - \coth(x)) \\
 \end{array} | \begin{array}{c}
 x(x + b) \left( \frac{1}{x^2} + \frac{1}{3} - \coth(x) \right) \\
 \hline
 \end{array} | \begin{array}{c}
 \frac{1}{x^2} \cdot \frac{1}{3} - \frac{1}{3} \cdot \frac{1}{x^2} \cdot \coth(x) \\
 \end{array} |

\[ \text{TABLE S4: The functions } I_m^n(x, b) \text{ appearing in the integrands in } I_m^n(b) \text{ (Eq. (S52)) the perturbation expansion of the renormalized Lagrangian Eq. (S51)} \]

\[ \lim_{n \to \infty} \left( \ln(b) \left( \frac{1}{2} b^2 + b + \frac{1}{2} \right) + b + \frac{1}{2} + \frac{2}{b^3} - \left( \frac{\ln(b)}{b} \right) - \frac{3}{12} \right) \rightarrow \frac{1}{b^3} \left( \sum_{n=0}^{\infty} \frac{1}{n!} \right) \]

\[ \text{TABLE S5: Strong } B \text{-field expansion (up to order } 1/b^2) \text{ of the integrals } I_m^n(b) \text{ (Eq. (S52)) in the perturbation expansion of the renormalized Lagrangian Eq. (S51)} \]

**S5. REGIME OF VALIDITY OF 1-LOOP EFFECTIVE ACTION FOR A DIRAC MATERIAL**

From previous sections, firstly, we have to ensure that the 1-loop approximation holds by keeping the interaction strength under control

\[ \alpha_D \ll \epsilon \sim 100, \quad \text{(1-loop approximation holds).} \quad (S47) \]

When the magnetic field is too large, the Landau level separation energy becomes comparable to the cut-off, and the Dirac description breaks down, but at

\[ |B| \ll \frac{\Lambda^2 e}{c \hbar v^2}, \quad \text{(S48)} \]

the Dirac theory is valid. Finally when the E-field is strong, a formal asymptotic expression can be easily derived from Eq. (S39). However, in our case this becomes somewhat an academic question, because as seen from Eq. (S41) the electric breakdown probability increases significantly with strong fields, which we do not consider. At small fields we have conditions

\[ |E| \ll \frac{\Lambda^2}{c \hbar v}, \quad \text{(The Dirac theory is valid)} \quad (S49) \]

\[ |e| < 0.3, \quad \text{(avoids electric breakdown).} \quad (S50) \]

**S6. ASYMPTOTIC EXPANSION OF THE HEISENBERG-EULER ACTION**

In order to avoid electric breakdown of an insulator, we are always in the weak field Lagrangian, being quadratic in |e|^2 and \( \hat{e} \cdot \hat{b} \), can be expanded as

\[ \delta L_{HE} = \frac{\Delta}{8\pi^2 \lambda_D} \sum_{n=0}^{\infty} \sum_{m=0}^{n} |e|^{2n} (\hat{e} \cdot \hat{b})^{2m} I_m^n(b), \quad b = |b|. \quad (S51) \]

The functions \( I_m^n \) are integrals of the form

\[ I_m^n(b) = \int_0^\infty dx \frac{e^{-x/b}}{x} f_m^n(b, x), \quad (S52) \]

where the functions \( f_m^n(b, x) \) are tabulated in Table S4.

Making a change of variables \( x \to bx' \), we can recheck the low field limit of the Lagrangian in Eq. (10) of the main text

\[ \frac{8\pi^2 \lambda_D^2}{\Delta} \delta L_{HE} \rightarrow \frac{1}{45} \int_0^\infty dx \left( \frac{d}{b^3} \right) \left( b^4 + 7(\hat{e} \cdot \hat{b})^2 - |e|^2 b^2 x + \frac{2}{2} + \frac{|e|^4 x + 4 x^2}{8} \right) = \frac{1}{45} [(|e|^2 - |b|^2) + 7(\hat{e} \cdot \hat{b})]. \quad (S53) \]
FIG. S2: Dimensionless functions $F,G,D$ of Eqs. (11), (13)) and (15) versus the dimensionless magnetic field. Functions $G$ and $D$ depend on the relative orientation of electric and magnetic field. The left (right) panel corresponds to $b \parallel e$ ($b \perp e$). For arbitrary angle between $b,e$ see Table S6.

$$
\delta \chi = \frac{c^2 \alpha_D \lambda^3_D}{\Delta} U^{-2} \frac{2|b|^2/5}{\log(|b|)} \quad |b| \ll 1
$$

$$
\delta \epsilon(B) = \frac{c^2 \alpha \lambda^3_D}{\Delta} U^2 \frac{1}{5} \left( \hat{e} \cdot \hat{b} \right)^2 - \frac{1}{5} |b|^2 - \log(|b|) + \left( \hat{e} \cdot \hat{b} \right)^2 |b| \quad |b| \ll 1
$$

$$
4 \pi M^{(e)} = \frac{\mu_0 \lambda^3_D}{\Delta} \left| \epsilon \right|^2 U^{-1} \left[ \frac{1}{5} \left( \hat{e} \cdot \hat{b} \right)^2 - \frac{2}{5} \left( \hat{e} \cdot \hat{b} \right) - \frac{1}{5} \right] \quad |b| \gg 1
$$

TABLE S6: Asymptotic expressions for the nonlinear magnetic susceptibility $\chi$, magnetically modulated dielectric constant $\delta \epsilon(B)$ and the electric modulated magnetization $4 \pi M^{(e)}$ tensors for arbitrary angle between $\hat{e}$ and $\hat{b}$.

The strong $B$ limit is obtained if we use the fact that for a bounded function $g(x) \to 0$ when $x \to 0$ the integral converges to

$$
\int_0^{\infty} \frac{dx}{x} e^{-x^5} g(x) \to g(\infty) \ln(1/\delta), \quad \delta \to 0.
$$

Taking the derivative or integral with respect to $1/b$, we can generate the subleading or super-logarithmic terms, respectively. Performing this procedure we obtain the strong $B$ expansions of the integrals in Eqs. (S51) and (S52), as tabulated in Table S5. Reading of the leading order terms from Table S5 and substituting in Eq. (S51) we obtain Eq. (2) of the main text

$$
\frac{8\pi^2 \lambda_D^3}{\Delta} \delta L_{HE} \to \frac{1}{3} |b|^2 \log |b| + \frac{1}{3} |e|^2 |b| (\hat{e} \cdot \hat{b})^2.
$$

S7. DERIVATION OF SUSCEPTIBILITY TENSORS

Inspecting the classical Lagrangian of the electromagnetic field $L_0 = (E^2 - B^2)/(8\pi^2)$, we can derive the (linear and nonlinear) contributions to susceptibilities from Eq. (S40) and Eq. (S39). If the quantum part of the action is called $\delta L$, the differential magnetic and electric susceptibilities are

$$
\chi_{ij} = \frac{\partial^2 \delta L}{\partial B_{jai} B_i}, \quad \frac{c^2 \alpha_D \lambda^3_D}{\Delta} \frac{\partial^2 \delta L}{\partial B_i \partial B_k} U_{ik}^{-1} U_{j1}^{-1},
$$

$$
\chi_{ij} = \frac{\partial^2 \delta L}{\partial E_{jai} E_i}, \quad \frac{\alpha_D \lambda^3_D}{\Delta} \frac{\partial^2 \delta L}{\partial E_i \partial E_k} U_{ikj1}.
$$
A. Linear and low order nonlinear susceptibilities

The cutoff dependent UV contribution is

$$\delta L_1 = \frac{\Delta}{12\pi^2\chi_D} \ln \left( \frac{\Lambda}{\Delta} \right) \left( |e|^2 - |b|^2 \right).$$  \hfill (S57)

In vacuum quantum electrodynamics, this quantity has the same form as the classical electromagnetic Lagrangian, hence it is absorbed into the definition of electric charge. However in the material case, it gives rise to the isotropic valence band contribution to the magnetic and electric susceptibilities, respectively

$$(\chi_1)_{ij} = -\frac{\varepsilon}{c^2} \frac{\alpha_D}{6\pi^2} \ln \left( \frac{\Lambda}{\Delta} \right) \delta_{ij} = -\frac{\varepsilon^2}{c^2} (\tilde{\chi}_1)_{ij}.  \hfill (S58)$$

In the anisotropic case, these become

$$\chi_D = -\frac{\varepsilon}{c^2} \frac{\alpha_D}{6\pi^2} \ln \left( \frac{\Lambda}{\Delta} \right) \mathcal{U}^{-2}, \quad \varepsilon_D = 1 + 4\pi\chi_D^e = 1 + \frac{2\alpha_D}{3\pi^2} \ln \left( \frac{\Lambda}{\Delta} \right) \mathcal{U}^2.  \hfill (S59)$$

as in Eqs. (7) and (8) of the main text.

The weak field limit of Eq. (S39) contains the lowest order nonlinear terms that are quadratic in $|e|^2 - |b|^2$ and $e \cdot b$

$$\delta L_2 = \frac{\Delta}{360\pi^2\chi_D} \left[ (|e|^2 - |b|^2)^2 + 7(e \cdot b)^2 \right].  \hfill (S60)$$

from which the nonlinear differential susceptibility tensors for an isotropic system follow as

$$\delta \chi_{ij} = \frac{\varepsilon}{c^2} \frac{\alpha_D}{180\pi^2} \left[ 2|b|^2(2\hat{b}_i \hat{b}_j + \delta_{ij}) + |e|^2(7\hat{e}_i \hat{e}_j - 2\delta_{ij}) \right], \hfill (S61a)$$

$$\delta \epsilon_{ij} = \frac{\alpha_D}{45\pi} \left[ |b|^2(7\hat{b}_i \hat{b}_j - 2\delta_{ij}) + 2|e|^2(2\hat{e}_i \hat{e}_j + \delta_{ij}) \right], \quad \hat{e}_i = \frac{e_i}{|e|}, \quad \hat{b}_i = \frac{b_i}{|b|}. \hfill (S61b)$$

These quantities can be re-expressed in the anisotropic case by using Eq. (S22) as

$$\delta \chi = \frac{\varepsilon}{c^2} \frac{\alpha_D}{180\pi^2} \left( 4(\mathcal{U}^{-1}b) \otimes (\mathcal{U}^{-1}b) + 2|b|^2\mathcal{U}^{-2} + 7(\mathcal{U}^{-1}e) \otimes (\mathcal{U}^{-1}e) - 2\mathcal{U}^{-2}|e|^2 \right), \hfill (S62a)$$

$$\delta \epsilon = \frac{\alpha_D}{45\pi} \left( 4(\mathcal{U}e) \otimes (\mathcal{U}e) + 2|e|^2\mathcal{U}^2 + 7(\mathcal{U}b) \otimes (\mathcal{U}b) - 2\mathcal{U}^2|b|^2 \right). \hfill (S62b)$$

The electric modulated magnetization then is simply $M^{(e)} = \delta \chi(E)B = B, \delta \chi(E)\mathcal{U}b$ and reads

$$4\pi M^{(e)} = \frac{2\alpha_D}{45\pi \chi_D} \mathcal{U}^{-1} \left( 7e(e \cdot b) - 2b|e|^2 \right). \hfill (S63)$$

To obtain the expressions in the main text we work in coordinates where

$$\mathcal{U} = \text{diag}(U_{xx}, U_{yy}, U_{zz}). \hfill (S64)$$

For simplicity we assume $E$ and $B$ directed along principal directions, e.g. $E \parallel B \parallel \hat{x}$ for the parallel field configuration and $E \parallel \hat{y}, B \parallel \hat{y}$ for the perpendicular field configuration. When these conditions are met, we can reduce, for example the parallel weak field magnetic susceptibility as

$$(\delta \chi_{||}(E))_{xx} = U_{xx}^{-1} \frac{\varepsilon}{c^2} \frac{\alpha_D}{36\pi^2} |e|^2, \quad 4\pi (M^{(e)}))_{x} = (\mathcal{U}^{-1}b)_x \frac{2\mu_D}{9\pi \chi_D} |e|^2. \hfill (S65)$$

For a summary of susceptibilities for arbitrary mutual orientation of E&B see Table S6. As we discussed in section S1, using inverse anisotropy transformation, the results for susceptibilities can be re-written in terms of applied external fields $E$ and $B$. The angle between $e$ and $b$ as function of the angle between $E$ and $B$ is given in Eq. (S24).
B. Higher nonlinear corrections to susceptibilities

Now that we have the renormalized Lagrangian in the form Eq. (S51), with the integrals $I_n^m$ tabulates in Table S5, we can calculate the susceptibilities according to Eq. (S56), in notable cases. If the electric field is zero, we have

$$\frac{8\pi^2 \lambda_D^3}{\Delta} \delta L_{HE}(|\mathbf{e}| = 0) \rightarrow \frac{\mathbf{b}^2}{3} \ln(\mathbf{b}) + \mathbf{b} \ln(\mathbf{b}) + \ldots$$

(S66)

consistent with Eq. (2) of the main text. Therefore the contribution to magnetic susceptibility due to applied magnetic field is

$$\delta \chi(|\mathbf{e}| = 0) \rightarrow \frac{\mathbf{b}^2}{c^2} \frac{\alpha_D}{12\pi^2} \mathbf{U}^{-2} \ln|\mathbf{b}|,$$

(S67)

as in Eq. (11) of the main text.

The term in the Lagrangian that is second order in the electric field is

$$\frac{8\pi^2 \lambda_D^3}{\Delta} \frac{\partial \delta L_{HE}}{\partial |\mathbf{e}|^2} \bigg|_{|\mathbf{e}|=0} = - \ln(|\mathbf{b}|) \left( \frac{1}{3} + \frac{1}{2|\mathbf{b}|} \right) + (\hat{\mathbf{e}} \cdot \hat{\mathbf{b}})^2 \left( \frac{|\mathbf{b}|}{3} + \frac{1}{2|\mathbf{b}|} \left( \ln|\mathbf{b}| + 1 \right) \right) + \ldots$$

(S68)

Then the leading order magnetization contribution is obtained from $M = \partial L/\partial B$ as

$$4\pi M^{(e)} \rightarrow \frac{\mu_D}{3\pi \lambda_D^3} |\mathbf{e}|^2 \mathbf{U}^{-1} \left( -\hat{\mathbf{b}}(\hat{\mathbf{e}} \cdot \hat{\mathbf{b}})^2 + 2\hat{\mathbf{e}}(\hat{\mathbf{e}} \cdot \hat{\mathbf{b}}) - \frac{\hat{\mathbf{b}}}{|\mathbf{b}|} \ldots \right),$$

(S69)

as in Eq. (15) of the main text. The dielectric response, linear in the electric field, is due to the term in the Lagrangian that is second order in the electric field, from which we obtain the magnetic field contribution in the isotropic case as

$$\delta \varepsilon \rightarrow \frac{\alpha_D}{3\pi} \left( -\ln(|\mathbf{b}|) (\mathbf{U}\hat{\mathbf{e}}) \otimes (\mathbf{U}\hat{\mathbf{e}}) + (\mathbf{U}\hat{\mathbf{b}}) \otimes (\mathbf{U}\hat{\mathbf{b}})|\mathbf{b}| \right).$$

(S70)

The leading order electric field contribution to the dielectric tensor comes from the term that is $\sim |\mathbf{e}|^4$ in the Lagrangian

$$\delta \varepsilon_{ij} \rightarrow \frac{\alpha_D}{6\pi} \frac{|\mathbf{e}|^2}{|\mathbf{b}|^7} \left( \delta_{ij} + 2\hat{\mathbf{e}}_i \hat{\mathbf{e}}_j + |\mathbf{b}| \delta_{ij}(\hat{\mathbf{e}} \cdot \hat{\mathbf{b}})^2 + 2|\mathbf{b}|(\hat{\mathbf{e}}_i \hat{\mathbf{b}}_j + \hat{\mathbf{e}}_j \hat{\mathbf{b}}_i)(\hat{\mathbf{e}} \cdot \hat{\mathbf{b}}) + |\mathbf{b}|(\hat{\mathbf{e}}_i \hat{\mathbf{e}}_j) \right),$$

(S71)

hence small in the parameter $|\mathbf{e}|^2/|\mathbf{b}| \ll 1$.

S8. MATERIAL APPLICATIONS

A. Bismuth and Bi$_{0.9}$Sb$_{0.1}$

While our consideration is primarily for insulators, we, e.g., demonstrated that the calculated magnetic susceptibility would be valid for valence band contributions in gapless materials. However real experiments often include, particularly for insulators with small gap, a certain density of free electrons. Then analysis of experimental settings must take into account contributions from free electrons, or experimental conditions must be found when these contributions are suppressed.

Bismuth has a band gap of $2\Delta = 15.5$ meV at the L-point and a Fermi level of $\mu = 35$ meV [44] measured from the midgap point of the Dirac bands. The electronic Fermi surface is composed of 3 electron ellipsoids that lie on the binary-bisectrix (x-y) plane perpendicular to the trigonal (z) axis. There is also a hole pocket along the trigonal axis. The hole Fermi level is about $-196$ meV measured from the midgap point of the hole Dirac bands. The hole band gap is about $2\Delta = 370$ meV, [37]

The hole contribution to susceptibility in the binary-bisectrix plane is small $\chi \sim -10^{-7}$ due to the large hole band gap and the alignment of the hole ellipsoid.

The conduction bands are polarized in magnetic field, so that only the e1 ellipsoid is populated when a field is applied in the binary direction [30]. Furthermore, the e1 conduction electrons can exhibit de Haas-van Alphen
oscillations, which are suppressed above $B = 5$ T. At higher magnetic fields the nonlinear diamagnetism is identical to the insulating alloy Bi$_{0.9}$Sb$_{0.1}$, which is identical to bismuth except for the absence of the Fermi level.

Based on the band structure calculations [37], we write the diagonalized velocity tensor as

$$\mathbf{V} = \text{diag}(1.7, 1.5, 0.4)v, \quad c/v = 188,$$

where $R$ implements rotation by $120^\circ$ about the trigonal axis. The first order contribution is

$$\chi_D = -10^{-5} \begin{pmatrix} 3 & 0 & 0 \\ 0 & 3 & 0 \\ 0 & 0 & 0.5 \end{pmatrix}.$$

If we apply magnetic field in the binary $(x)$ direction we have

$$\delta \chi_{xx}(B_x = 5 \text{ T}) = 13 \times 10^{-6}.$$ 

For the full field range see Fig. 2. The other quantities (dielectric enhancement, magnetization etc.) are calculated in a similar fashion. For example the dielectric enhancement when $E \parallel B$ in $x$-direction we have

$$\delta \epsilon_{\parallel}(B_x) \rightarrow 10 B_x.$$ 

The Iwasa experiment has the reference points

$$\chi_{\text{binary}}^{\text{exp}}(50 \text{ T}) = -18.7 \times 10^{-6}, \quad \chi_{\text{bisection}}^{\text{exp}}(50 \text{ T}) = -15.4 \times 10^{-6}.$$ 

The difference in the reference levels are due to the additional contributions to linear magnetic susceptibility that are not contained in the Dirac theory. Although the logarithmic Dirac contribution in Eq. (8) constitutes a significant part of the linear diamagnetic response, non-Dirac core shell electron contributions can be equally important. Furthermore, there can be additional errors due to the choice of UV cut-off and subleading terms in the summation of the Landau levels can contribute to the total. None of the above mentioned factors affect the nonlinear response, which is dominated by the Dirac bands. Therefore, once the overall constant offset due to linear susceptibility is adjusted, we get an excellent agreement in the nonlinear behavior of the measured susceptibility in bismuth, as seen in Fig. 2a.

B. TaAs

TaAs has 12 pairs of Weyl fermions [85], 4 pairs forming hole pockets 2 meV below the Fermi level and 8 forming electron pockets 21 meV above the Fermi level. The Fermi level $E_F = 21$ meV contribution which is not accounted in our theory, becomes unimportant $B \gg \frac{4E_F^2}{\hbar v_F^2} \approx 10$ T.

We represent each pair by a single massless Dirac fermion with the velocity tensor comparable to the calculations and measurements [29, 85]

$$\mathbf{V} = \text{diag}(1.7, 1.7, 0.35)v, \quad c/v = 447$$
where the z-component is the velocity in the c-axis. Since the system is gapless we consider the situation where $E = 0$. The nonlinear susceptibility is the gapless limit is

$$\chi_D + \delta\chi \rightarrow -\frac{\alpha_D}{12\pi^2} \frac{\nu^2}{c^2} \ln\left(\frac{c\Lambda^2}{eBh
u^2}\right)\mathcal{U}^{-2} \quad (S80)$$

To compare the nonlinear behavior of our theory with the experiment by Zhang et al. [29], we take the reference level of susceptibility to be $\chi_{ref} = -\chi(30 \text{T}) = 1.56 \times 10^{-5}$.

C. Pb$_{0.8}$Sn$_{0.2}$Te

The gap as a function of the Sn content is [77]

$$2\Delta[\text{meV}] = 182 - 480x \quad (S81)$$

At $x = 0.235$ we have the inverse masses $m^{-1}_\perp = 100m^{-1}_e$ and $m^{-1}_\parallel = 11.25m^{-1}_e$ where the gap is $2\Delta = 69.1 \text{meV}$. At $x = 0.510$ we have the same inverse massed where the gap is similar $2\Delta = -62.8 \text{meV}$ (TI phase). We will base our estimates on these values. The parallel denotes the z-axis aligned in the [001] direction. The velocity tensor is then

$$\mathbf{V} = \text{diag}(1.4, 1.4, 0.5)\nu, \quad c/\nu = 580. \quad (S82)$$

There are a total of 4 Dirac fermions located at the L-points symmetric about the z-axis [001] direction of the rock salt structure. Due to the relatively large gap the most striking property is the electric field modulated magnetization. If we apply $B \parallel E$ in the x-direction with $B \sim 5 \text{T}$ and $E = E_*/3$ we have

$$4\pi(M^*_{||})_x(B_x = 5 \text{T}, E_x = 10^4 \text{V/cm}) = 0.5 \mu\text{T.} \quad (S83)$$