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Quantum atmospherics for materials diagnosis

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Symmetry-breaking states of matter can transmit symmetry breaking to nearby atoms or molecular complexes, perturbing their spectra. We calculate one such effect, involving the "axion electrodynamics" relevant to topological insulators, quantitatively, and identify a signature for T violating superconductivity. We provide an operator framework whereby effects of this kind can be analyzed systematically.

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Introduction. Over the past few decades physicists have come to appreciate the importance of increasingly subtle forms of symmetry breaking in materials, often connected with topology and entanglement [1–3]. Many new states of matter characterized by such "hidden" symmetry breaking have been proposed theoretically, but concrete, unambiguous experimental manifestations have been relatively sparse. Many of the proposed states violate some combination of the discrete symmetries P, T [4]. This opens up the possibility of unusual polarizabilities, generalizing the familiar dielectric and para- or diamagnetic response parameters ϵ, μ. Those polarizabilities can support novel electromagnetic effects, which reflect the discrete symmetry breaking directly [5,6]. The effects involve virtual two-photon exchange in loops, and are intrinsically quantum mechanical. These effects lead to long-range (generalized) Casimir-type forces, also involving spin [7], but our estimates make it plausible that they are more easily accessed through spectroscopy. Two particularly interesting cases, on which we will focus especially, are boundary Chern-Simons models [8] and chiral superconductors [9]. Both these phenomena have attracted much theoretical attention, and experimental signatures of the postulated symmetry breaking should be helpful in validating candidates. We will also discuss the possibility of searching for fundamental electric dipole moments and provide a systematic operator framework for analyzing other cases of symmetry breaking.

Atmosphere from axion electrodynamics. Consider a material whose interaction with the electromagnetic field contains an action term

$$\int d^3x\,dt\;\chi_M(x)\Delta\mathcal{L}_{\rm axion} = \int d^3x\,dt\;\chi_M(x)\,\kappa\,\vec{E}\cdot\vec{B}, \qquad (1)$$

where $\chi_M(x)$ is the characteristic function of the material. This sort of interaction, an induced Chern-Simons term, was contemplated in [10], and it is realized in topological insulators [4,6,11,12], with $\kappa = j\alpha$, where j is an odd integer. (Note that while this is the most direct extrapolation of the bulk effective theory of topological insulators, there could, in principle, be additional, nonuniversal contributions to the surface action. Note also that the overall global P, T symmetry of topological insulators cannot be applied locally at boundaries.) Since $\vec{E}\cdot\vec{B}$ is a total derivative, it does not affect the bulk equations of motion. But when the spatial region occupied by the material is bounded, surface terms arise [13]. Specifically, if the plane $z = 0$ forms an upper boundary, we will have a surface action

$$\int d^3x\,dt\;\chi_M(x)\kappa\,\vec{E}\cdot\vec{B}$$
$$\to \frac{\kappa}{2}\int dx\,dy\,dt\,\epsilon^{3\alpha\beta\gamma}A_\alpha(x,y,0,t)\partial_\beta A_\gamma(x,y,0,t). \qquad (2)$$

This gives us a two-photon vertex which violates the discrete symmetries P, T locally, while preserving PT. Quantum fluctuations involving this vertex will produce a sort of P, T violating atmosphere above the material (see Fig. 1). The atmosphere induces new kinds of "Casimir" forces on bodies near the material [14–18]. It also induces new kinds of effective interactions within atoms or molecular centers, which affect their spectra. Such interactions are especially interesting, because in favorable cases the spectra can be measured quite accurately, thus plausibly rendering small symmetry-violating effects accessible.

Let us analyze the most basic case; that is, the interaction of an electron. By symmetry and dimension counting, the first-order effective P, T violating interaction with an electron, at a distance r from a planar boundary, will take the form

$$\mathcal{L}_{\rm int} \sim \frac{\alpha\kappa}{mr^2}\,\hat{n}\cdot\vec{s}\,, \qquad (3)$$

where $m, \vec{s}$ are the electron's mass and spin, and $r, \hat{n}$ are the distance and normal to the plane. Expressed using fundamental units only, as in the quoted form for topological insulators, we find the dimensional estimate

$$\mathcal{L}_{\rm int} \sim \frac{\alpha^2}{mr^2}\,\hat{n}\cdot\vec{s} \approx \left(\frac{10\ {\rm nm}}{r}\right)^2 \frac{e\hat{n}\cdot s}{m}\,10\ {\rm gauss}. \qquad (4)$$

Here we have expressed the atmospheric Zeeman-like interaction in a form which allows ready comparison with the Zeeman splitting induced by a magnetic field strength. Taken

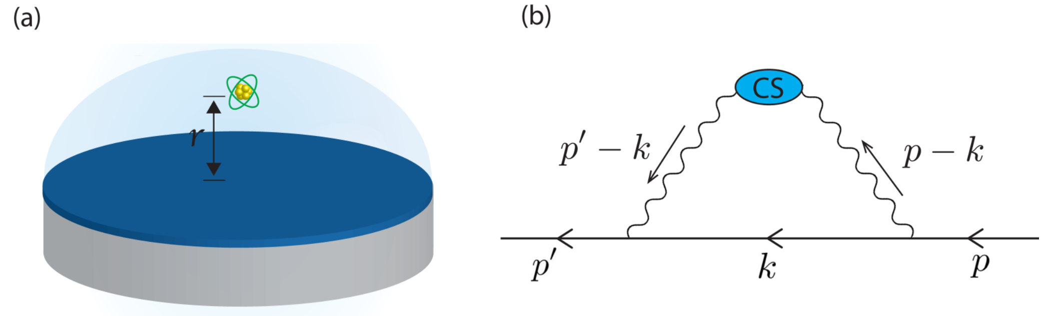

FIG. 1. (a) Illustration of quantum atmosphere induced by a Chern-Simons surface. The blue layer corresponds to the top surface described by a Chern-Simons term at $z = 0$. Due to quantum fluctuation, time-reversal symmetry-breaking effect will be transmitted to the nearby atom at the distance r from the surface. (b) Feynman diagram involving Chern-Simons vertex.

at face value, this is comfortably within the estimated sensitivity of magnetometry based on nitrogen-vacancy centers [19]—by many orders of magnitude (but see below). Note, however, that we do not generate true magnetic flux, so that superconducting quantum interference device detectors will not register (but see below).

We can check this estimate by explicit calculation, according to the Feynman diagram of Fig. 1. We find [20]

$$V(r) = \frac{\kappa e^2}{128\pi^2}\frac{1}{mr^2}\sigma_3 \rightarrow \frac{j\alpha^2}{32\pi}\frac{1}{mr^2}\sigma_3. \tag{5}$$

One might attempt to generalize this calculation to particles which possess an anomalous magnetic moment (e.g., atomic nuclei), but one encounters an ultraviolet divergence [20]. This is not a physical contradiction, because both anomalous magnetic moments and (especially) our assumed action Eq. (1) will have form factors which provide cutoffs. Also, of course, the virtual photons emitted from the material need not terminate on a single particle. For these reasons, our estimate, Eq. (4), and the result of our calculation, Eq. (5), should be regarded as encouraging, but applied with care. Dispersion relations relating spectroscopic splitting to the material's response to photons are included in the Supplemental Material [20].

We can also consider the effect of applying an external electric field. Importantly, this does not in itself introduce T violation. If we apply an electric field parallel to the boundary plane, we induce a surface Hall-like current. A planar current sheet produces a spatially constant (true) magnetic field, which will be aligned (or antialigned) with the applied electric field. To maximize the induced field while avoiding cancellations between contributions from opposite sides of the material, we should use samples with effective surfaces whose linear dimensions are large compared to the distance to the test atom or complex, but small compared to the separation between surfaces. If we apply an electric field perpendicular to the boundary plane, it induces a surface magnetic charge, and thus again a magnetic field aligned or antialigned with the applied electric field, and in the same sense. The magnitudes of the magnetic fields, for moderate values of the applied electric field, can be quite substantial:

$$B \sim \kappa E \rightarrow \alpha E \approx 10^{-1}\ \text{gauss}\left(\frac{E}{10^4\,\frac{\text{V}}{\text{cm}}}\right), \tag{6}$$

where the progression from general to particular is as previously. These induced currents and fields were anticipated in [10]; here we are adding some context on their connection with symmetry and their possible experimental accessibility. They are a much more conservative application of the effective theory.

Atmosphere of superconductors. The classic signature for superconductivity is the Meissner effect, i.e., exclusion of an applied magnetic field. This signature is not ideal for discovery work, since the superconducting regions can be small and the superconductivity itself disrupted by magnetism. Spectroscopic shifts induced by Meissner response to virtual photons can offer an alternative. Such shifts were calculated in [27,28], under the assumption of T symmetry. Violation of T symmetry can induce splitting between states that are otherwise degenerate. Chiral superconductors are typical examples where time-reversal symmetry is broken due to the finite orbital angular momentum of Cooper pairs [29,30]. This leads to a state-dependent magnetic energy shift [20]

$$\begin{aligned}\delta\epsilon_n = \sum_m \int_0^\infty \frac{d\omega}{2\pi}\frac{2\epsilon_{mn}}{\epsilon_{mn}^2 - \omega^2} \\ \times \operatorname{Im}\{\langle n|D_1|m\rangle\langle m|D_2|n\rangle\mathcal{H}_{12}(z,z;\omega) \\ + \langle n|D_2|m\rangle\langle m|D_1|n\rangle\mathcal{H}_{21}(z,z;\omega)\},\end{aligned} \tag{7}$$

where $\vec{D}$ is the magnetic dipole operator, the coordinates are labeled 1, 2, z, and $\mathcal{H}$ is the frequency-dependent modification of the magnetic field correlator due to the superconductor. T violation introduces an imaginary part into $\mathcal{H}_{12}(= -\mathcal{H}_{21})$ and leads to an effective interaction which splits states of opposite angular momentum in the z direction [31]. It mimics, in other words, the effect of a Zeeman interaction with an emergent magnetic field.

Fundamental electric dipole moments. Apart from spontaneous P, T symmetry breaking in materials, we may also have intrinsic violation. That possibility is of great interest for fundamental physics [32]. A generic signature of such violation is the existence of particles having both elementary magnetic dipole moments and (small) elementary electric dipole moments. (Let us emphasize that this represents physics beyond the "standard model.") A material containing a density ρ of such particles will, in the presence of an applied electric field at temperature T, contain a density $\rho g_e \vec{E}/T$ of aligned spins, and hence an energy density $(g_m g_e/T)\rho\vec{E}\cdot\vec{B}$. Thus, we identify an alternative source of our action, Eq. (1), with $\kappa = \rho g_m g_e/T$. In this model, it is transparently clear why a normal electric field, by inducing a magnetic dipole density, yields a surface magnetic charge density. Some possible experimental arrangements to probe intrinsic symmetry-breaking effects of this kind were discussed in [33] from a very different point of view. Numerically, we have

$$B \sim \rho g_m g_e E/T \sim \left(\frac{\rho}{\frac{10^{22}}{\mathrm{cm}^3}}\right)\frac{g_e}{10^{-26}\ \mathrm{e\,cm}}\frac{E}{10^6\frac{\mathrm{V}}{\mathrm{cm}}}\frac{10^{-3}\mathrm{K}}{T}10^{-12}\ \text{gauss}, \quad (8)$$

where we have inserted the electron gyromagnetic moment, aggressive reference values of the parameters, and a reference value of the electric-dipole moment comparable to current limits. The resulting magnetic field is well within advertised sensitivities [19]. Note that in this estimate we have assumed a thermal population of the spins, for which the asymmetry is suppressed, due to the tininess of the electric moment energy splitting.

Operator analysis of polarizabilities. In constructing effective theories of electromagnetism in condensed matter, there are few principles we can apply *a priori*. Nevertheless, when plausible assumptions and approximations give us tractable theories which contain few parameters, those theories can be very useful in organizing data and planning experiments. For our purposes, it is instructive to recall that textbooks of electromagnetism commonly introduce just two material-dependent parameters, ϵ and μ, to describe a wide range of observed behaviors. They can be considered as coefficients in the Maxwell action

$$\int d^3x\, dt\ \chi_M(x)\Delta\mathcal{L}_{\text{Maxwell}} = \int d^3x\, dt\ \chi_M(x)\left(\frac{\epsilon}{2}\vec{E}^2 - \frac{1}{2\mu}\vec{B}^2\right). \quad (9)$$

These are the possible terms which satisfy four sorts of conditions:

(1) They are local in space and time, containing only products of fields at the same space-time point.

(2) They are invariant under many symmetries: time and space translation, rotation, gauge.

(3) They are quadratic in fields and of lowest possible order (i.e., zero) in space and time gradients.

(4) They are invariant under P and T symmetry.

Equation (1) is an additional term we can bring in if we drop the last of those conditions. Aside from symmetry, it is also commonly ignored because it does not contribute to the bulk equations of motion, but as we have seen that reason is superficial.

The third condition is practical rather than fundamental. Indeed, terms containing higher powers of fields are the meat and potatoes of nonlinear optics [34]. But in many circumstances it is appropriate to ignore nonlinear effects. Also, it is often appropriate to consider external and effective fields which vary smoothly in space and time. With those ideas in mind, we can get a nice inventory of the possible terms which are quadratic in fields and of lowest order in space and time gradients while consistent with conditions (1)–(3) and displaying different P, T characters. We arrive at the following candidate Lagrangian densities:

(i) P *even*, T *even*: Maxwell terms, Eq. (9):

$$\mathcal{O}_E = \vec{E}^2, \quad \mathcal{O}_B = \vec{B}^2. \quad (10)$$

(ii) P *odd*, T *odd*: axion electrodynamics, Eq. (1):

$$\mathcal{O}_a = \vec{E}\cdot\vec{B}. \quad (11)$$

(iii) P *even*, T *odd*:

$$\begin{aligned}\mathcal{O}_1 &= \frac{\partial\vec{E}}{\partial t}\cdot\vec{E} = \frac{\partial}{\partial t}\frac{1}{2}\vec{E}^2,\\ \mathcal{O}_2 &= \frac{\partial\vec{B}}{\partial t}\cdot\vec{B} = \frac{\partial}{\partial t}\frac{1}{2}\vec{B}^2,\\ \mathcal{O}_3 &= [(\nabla\times\vec{E})\cdot\vec{B}],\\ \mathcal{O}_4 &= (\nabla\times\vec{B})\cdot\vec{E} = \mathcal{O}_3 - \nabla\cdot(\vec{E}\times\vec{B}).\end{aligned} \quad (12)$$

(iv) P *odd*, T *even*:

$$\begin{aligned}\mathcal{O}_5 &= [(\nabla\times\vec{E})\cdot\vec{E}],\\ \mathcal{O}_6 &= (\nabla\times\vec{B})\cdot\vec{B},\\ \mathcal{O}_7 &= \frac{\partial\vec{E}}{\partial t}\cdot\vec{B},\\ \mathcal{O}_8 &= \frac{\partial\vec{B}}{\partial t}\cdot\vec{E} = \frac{\partial}{\partial t}(\vec{B}\cdot\vec{E}) - \mathcal{O}_7.\end{aligned} \quad (13)$$

The bracketed terms are redundant, since the Faraday relation $\nabla\times\vec{E} = -\frac{\partial B}{\partial t}$ holds identically, when one expresses the fields in terms of potentials. Terms which are total time derivatives do not contribute to the equations of motion or to surface times, while terms which are total space divergences give boundary actions. Thus in the P even, T odd case we find only a boundary action, corresponding to $\mathcal{O}_4$, while in the P odd, T even case we get two terms, corresponding to $\mathcal{O}_6$ and $\mathcal{O}_7 - \mathcal{O}_8$, which affect bulk behavior. These considerations can guide the design of experiments. For example, to search for a P violating but T invariant atmosphere (and thus, to probe for states of matter with those symmetries) we might first exclude an emergent $\hat{n}\cdot\vec{s}$ interaction in a planar geometry, and then look for an emergent $\hat{n}_1\cdot(\hat{n}_2\times\vec{s})$ interaction in a more complex geometry, involving two characteristic directions. Upon applying a time-dependent electric field, we may look for an atmospheric magnetic field whose direction changes according to whether the magnitude of $\vec{E}$ is increasing or decreasing. That behavior derives from $\mathcal{O}_7$. $\mathcal{O}_5$, and $\mathcal{O}_6$, which were considered formally in [35], where they were referred to as "zilch," without proposed application.

Note that if we work directly at the level of polarizabilities, rather than actions, we can define contributions corresponding to all eight cases, and also two independent "axion" terms. Thus, for example, we might write

$$\begin{aligned}\vec{D} &= c_e\vec{E} + c_{a1}\vec{B} + c_1\frac{\partial\vec{E}}{\partial t} + c_4\nabla\times\vec{B} + c_5\nabla\times\vec{E} + c_8\frac{\partial\vec{B}}{\partial t},\\ \vec{H} &= c_b\vec{B} + c_{a2}\vec{E} + c_2\frac{\partial\vec{B}}{\partial t} + c_3\nabla\times\vec{E} + c_6\nabla\times\vec{B} + c_7\frac{\partial\vec{E}}{\partial t}.\end{aligned} \quad (14)$$

After applying the Faraday relation, we have ten independent terms, including the two conventional ones. The more restrictive Lagrangian approach seems more principled, however.

Materials that contain chiral molecules can violate P while conserving T intrinsically; indeed, many such so-called gyrotropic materials are well known [36]. The recently discovered P-violating Weyl semimetals, which display the chiral magnetic effect in transport, provide another example [37]. A possibility for more subtle, spontaneous breaking of this class, which still preserves macroscopic rotation and translation symmetry, could be a nonvanishing correlation of the type $\langle \vec{j} \cdot \vec{s} \rangle \neq 0$ between microscopic current and and spin densities which are themselves uncorrelated ($\langle \vec{j} \rangle = \langle \vec{s} \rangle = 0$). Similarly, a nonvanishing correlation of the type $\langle \vec{j} \cdot \vec{\pi} \rangle \neq 0$ between microscopic current and polarization densities which are themselves uncorrelated exhibits P even, T odd spontaneous breaking; while a nonvanishing correlation $\langle \vec{s} \cdot \vec{\pi} \rangle \neq 0$ is odd under both P and T, but even under PT, as we have mentioned before implicitly.

Summary. We have discussed how quantum fluctuations, in the presence of a material, produce a kind of atmosphere which can affect the spectra of nearby atoms. The atmosphere can be probed to diagnose properties of the material, and in particular its symmetry. We have calculated one effect of this kind, by taking the effective theory based on axion electrodynamics at face value, and found a result that is very large compared to expected experimental sensitivities. The atmosphere can be influenced in a calculable way by external fields. We displayed an operator framework in which to discuss these issues systematically, and classified the simplest nontrivial possibilities under stated, broad assumptions. Our assumptions could be relaxed, for instance to allow crystalline asymmetries, at the cost of bringing in more operators. The operator analysis suggests how to probe symmetry-breaking atmospheres experimentally, and to parametrize their properties systematically.

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