Perspective

# Polarization transport in ferroelectrics

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The duality between electric and magnetic dipoles in electromagnetism only partly applies to condensed matter. In particular, the elementary excitations of the magnetic and ferroelectric orders, namely magnons and ferrons, respectively, have received asymmetric attention from the condensed matter community in the past. In this Perspective, we introduce and summarize the current state of the budding field of "ferronics." We argue that the introduction of dipole-carrying elementary excitations allows the modeling of many observables and potentially leads to applications in thermal, information, and communication technologies.

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## I. FERROELECTRICITY AND MAGNETISM

Ferroelectrics are materials that exhibit permanent and switchable electric polarization resulting from the ordering of electric dipoles below possibly high critical temperatures. Comparing and contrasting the behavior of ferroelectrics with that of magnets, in which magnetic dipoles order, can enhance our understanding of both material classes [1,2]. Due to their high dielectric constant, ferroelectrics are commonly used in (power) capacitors. Their polarization can be manipulated by external electric fields and temperature, leading to applications such as electrocaloric cooling via thermodynamic cycles similar to those used in magnetocalorics [3]. Pyro- and piezoelectricity can be utilized for sensor and energy harvesting applications [4]. The finite coercivity of the switchable order makes ferroelectrics useful materials for nonvolatile memories that complement (and compete with) magnetism-based devices [5].

The polarization of a polar material is affected by the details of its surface, but the distinction between a ferroelectric and an unpolarized dielectric is a well-defined bulk property that can be computed from first principles [6]. The analogies between ferroelectricity and magnetism are well understood for equilibrium thermal properties in terms of the entropy of the dipolar order. Surprisingly, the excited states of ferroelectrics or "ferrons" [9] and their nonequilibrium transport properties attract much less attention than the equivalent states in magnets, known as magnons [7,8]. In this Perspective, we summarize the state of the art of "ferronics" with a focus on potential applications.

## II. FERROELECTRIC MATERIALS

The exchange interaction drives magnetism by favoring parallel spins over antiparallel ones. The magneto-dipolar interaction is relatively weak and causes secondary effects,

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such as the formation of domains. In ferroelectrics, there is no exchange interaction but the dipolar interaction is stronger by a factor of $1/(\alpha^2\epsilon_r) \approx 10^3$, where $\alpha = 1/137$ is the fine-structure constant and $\epsilon_r$ is a high-frequency relative dielectric constant [10]. The electric dipolar energy drives ferroelectricity in some but not all cases. "Improper" ferroelectricity [12] accompanies a structural or magnetic phase transition in which electrostatics does not play a leading role. Since there is no unifying physical origin, many types of ferroelectrics exist. In the following, we discuss a few selected examples.

*Order-disorder ferroelectrics.* In materials with bistable protons or loosely locked molecular dipoles, the entropy increases with temperature driving a second-order phase transition that destroys a ferroelectric state at a critical temperature. This phenomenology is similar to that of magnetic systems and can be modeled by Ising pseudospin Hamiltonians [13,14].

*Displacive ferroelectrics.* Most ferroelectric phase transitions, including that of the iconic perovskite $BaTiO_3$, are associated with an inversion symmetry-breaking structural phase transition triggered by a soft transverse optical phonon. The Landau-Devonshire theory, based on a parameterized Landau free energy, is an appropriate description of the ferroelectric phase transitions in these materials [10].

*Hyperferroelectrics.* Hyperferroelectrics are a class of semiconducting ferroelectrics with a polarization that remains stable under an unscreened depolarization field [11].

*Shift-induced ferroelectricity* was reported recently in atomically thin bilayers of two-dimensional materials with hexagonal noncentrosymmetric unit cells such as $WTe_2$ [15] and BN [16–18]. Here a small slide or twist between the two monolayers reverses an electric dipole by mutual electric polarization and charge transfer. Landau theory [19] and first-principles calculations [20] explain the observed high critical temperatures and low switching fields.

*Electronic ferroelectrics.* Ionic displacements cause permanent polarization in the above material classes. The inertia of the atomic masses slows down the switching dynamics. Ferroelectricity caused by charge ordering without rearrangement of the lattice [21–23] promises a much faster response. Even though the microscopic physics is very different, the phenomenology of polarization accumulation and transport should be the same as for phonon-based ferroelectrics [24].

*Ferroelectric conductors.* According to conventional wisdom electric conduction and ferroelectricity are incompatible because of the efficient screening of bulk charges by mobile electrons. Nevertheless, ferroelectric conductors and metals do exist in special materials with incomplete screening [25]. The perpendicular polarization in $WTe_2$ [15] or bilayer graphene [23] can exist because two-dimensional conduction electrons cannot screen a permanent dipole oriented perpendicularly to the planes. A two-dimensional metal such as graphene also does not screen a homogeneous perpendicular polarization of a close-by ferroelectric [19].

*Antiferroelectrics.* The condensation of a Brillouin zone center (edge) phonon precedes the ferroelectric (antiferroelectric) order. Ferrielectricity and more complex textures [26] may emerge at phase transitions that do not need to be displacive.

*(Anti)ferroelectric topological insulators* have been predicted to allow control of band topology by applied electric fields [27,28].

*Multiferroics.* In multiferroic materials, the orders in the electric and magnetic polarizations coexist [29]. "Electromagnons" are the excitations in multiferroic materials in the presence of ac electric fields that may carry electric dipoles [30–32]. Electric and magnetic dynamics are coupled by the Moriya-Dzyaloshinskii interaction [31,32].

*Electrets* are materials with a metastable macroscopic electric polarization that cannot be switched [33], in contrast to conventional ferroelectrics with spontaneously formed bistable ground states.

*Polar materials* with broken inversion symmetry but without spontaneous electric polarization may still be piezo- and pyroelectric, i.e., develop transient polarizations under stress and heat. Such materials also may support the ferron excitations discussed in more detail below. Here we concentrate on ferroelectrics because their switchability allows control of larger polarization currents and the phase transition to the inversion-symmetric paraelectric phase quenches the polarity.

## III. FERRONICS

Ferronics is the study of the elementary excitations of the ferroelectric order or "ferrons" [19,34–38], analogous with the subfield of magnonics in magnetism [7,8]. In contrast to spin waves that look the same in most ferromagnets, the excitations of ferroelectrics reflect the richness of their ground states. Figure 1 is a schematic of the symmetry-restoring ferron excitations in selected materials.

*Ferrons.* A ferron in state $i$ is a bosonic excitation that carries electric polarization in the form of a finite dipole $\mathbf{p}_i$. By a direct derivation [36] or the Hellmann-Feynman theorem, the latter follows from the dependence of its frequency $\omega_i = \epsilon_i/\hbar$ on an applied electric field $\mathbf{E}$ (parallel to the polarization)

$$\mathbf{p}_i = -\frac{\hbar\partial\omega_i}{\partial\mathbf{E}}. \quad (1)$$

For comparison, the magnetic moment of a magnon $j$ in simple ferromagnets with frequency $\omega_j$ reads $\mathbf{m}_j = -\hbar\partial\omega_j/(\partial\mathbf{B})$, where $\mathbf{B}$ is a magnetic field along the

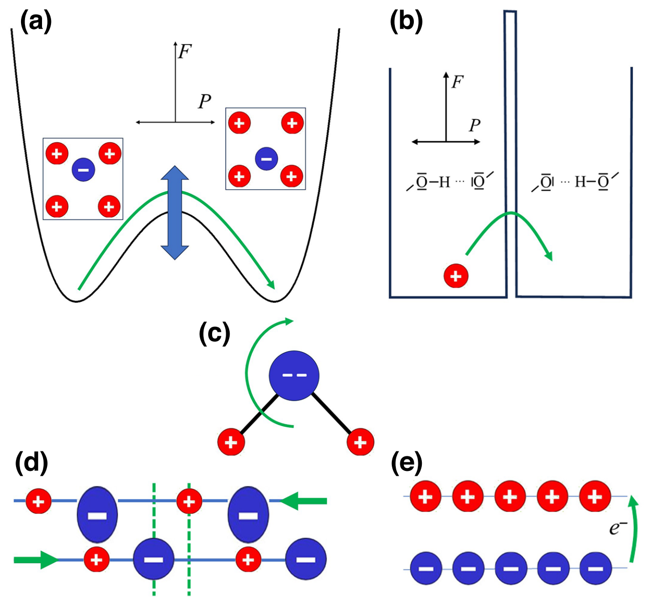


FIG. 1. The green arrows indicate symmetry-restoring ferron excitations in selected ferroelectric materials. (a) Sketch of the soft-phonon excitation in the free energy landscape of a displacive ferroelectric with a temperature-dependent barrier (blue arrow). (b) In typical order-disorder ferroelectrics, the symmetry is restored by the hopping of protons in hydrogen bonds. (c) Thermally activated rotations destroy the order of permanent molecular dipoles. (d) The rigid relative shift of the bilayers reverses the electric polarization in a double-well potential. (e) In electronic ferroelectrics, the symmetry is restored by the motion of electrons, not the ions.

magnetic order. In the absence of anisotropies value is $m_j = -2\mu_B$, where $\mu_B$ is the Bohr magneton [39]. A similar simple relation does not exist for the ferron polarization, however.

The temperature dependence of the equilibrium polarization is

$$\Delta \mathbf{p}_0 = \mathbf{p}_0(T) - \mathbf{p}_0(0) = \sum_i \mathbf{p}_i f_P(\epsilon_i, T), \tag{2}$$

in terms of the sum over the eigenstates of the system, where $f_P(\epsilon_i, T) = \exp[\epsilon_i/(k_B T) - 1]^{-1}$ is the Planck distribution and $k_B$ is Boltzmann's constant. The excitations of ferroelectrics (and ferromagnets) suppress the order; hence $p_0(T) \leq p_0(0)$.

The ferrons of most ferroelectrics are also phonons, but not all phonons are ferrons. The optical phonons in dielectrics with inversion symmetry including the paraelectric phase of ferroelectrics above the phase transition temperature scale like $\delta\omega_{\mathrm{op}} \sim -E^2$ and therefore cannot harbor ferrons. Ferron excitations exist only as anharmonic oscillators in crystals with intrinsic or induced broken inversion symmetry. Ferron modes can be transverse (Goldstone-like) or longitudinal (Higgs-like) as illustrated

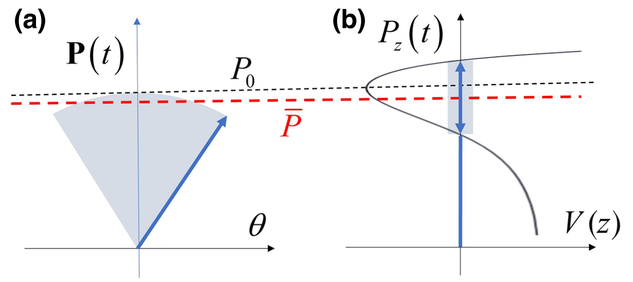


FIG. 2. Sketch of (a) a "Goldstone" transverse and (b) a "Higgs" longitudinal ferron mode. In (a) the modulus of the electric dipole $P_0$ is conserved during transverse oscillations, as in stable molecular dipoles. The projection on the equilibrium polarization $|\overline{P}| < |P_0|$. In (b) the length of the dipole fluctuates in an anharmonic potential $V(z)$ that also reduces the average value $|\overline{P}| < |P_0|$.

in Fig. 2, as well as acoustic or optical. In complex displacive ferroelectrics, we expect that the soft polar phonons dominate the polarization dynamics while other phonons form an inert background.

In the following, we focus on ferroelectrics because their switchability implies "larger" ferrons and associated functionalities for technological applications.

*Models.* In most materials, ferroelectricity is associated with the movement of charged ions. The ferrons are a subset of the phonon spectrum and accessible to lattice dynamical calculations with an harmonic corrections that endow the excitations with an electric dipole. A very simple model system is a one-dimensional chain with two atoms in the unit cell [34,35]. $\mathbf{p}_i$ depends strongly on band index and crystal momentum. It is mainly carried by optical modes, but in transport phenomena, the much larger group velocity of the sound waves can compensate for their smaller dipoles.

In order-disorder ferroelectrics, the ferron dipole is caused by the hopping of protons or the rotation of molecular dipoles. These are two-level systems that can be modeled by Ising pseudospins in which the tunneling between the two spin states and dipolar coupling between the dipoles govern the ferron dispersion [13,14].

In displacive and shift-induced ferroelectrics the lattice vibrations in a double-well potential reduce the modulus of the average dipole. Landau theory [10] governs the ferron dispersion and mode amplitudes of displacive ferroelectrics in the long-wavelength limit [19,36].

In electronic ferroelectrics, ferrons emerge in the Falikov-Kimball model as excitations of an excitonic insulator phase [24].

Equation (1) is a simple recipe to compute the ferron dipole by model or first-principles calculations. Many observables can be computed from the ferron energy dispersion $\epsilon_{\nu\mathbf{k}}$ for a ferron band $\nu$ and wave vector $\mathbf{k}$ and its polarization $\mathbf{p}_{\nu\mathbf{k}} = -\partial\epsilon_{\nu\mathbf{k}}/\partial\mathbf{E}$.

*Nonequilibrium.* The equilibrium polarization $\mathbf{p}_0$ density of a ferroelectric depends on temperature and the applied electric field. When the equilibrium of an isotropic medium is perturbed by gradients of temperature ($\partial_x T$) and/or effective electric field ($\partial_x E_{\text{eff}}$) parallel to the polarization in the $x$ direction, the energy ($j_q$) and polarization ($j_p$) current densities flow to restore the equilibrium. When the perturbations are sufficiently small and slow in time and space, local linear response relations (Ohm's law) apply:

$$\begin{pmatrix} -j_p \\ j_q \end{pmatrix} = \sigma_p \begin{pmatrix} 1 & S_p \\ \Pi_p & \kappa/\sigma_p \end{pmatrix} \begin{pmatrix} \partial_x E_{\text{eff}} \\ -\partial_x T \end{pmatrix}, \quad (3)$$

where $\sigma_p$ ($\kappa$) is the polarization (thermal) conductivity with units m/$\Omega$ (W K$^{-1}$ m$^{-1}$), while $S_p$ $\left(\Pi_p = S_p T\right)$ is the ferroelectric Seebeck (Peltier) coefficient with units V K$^{-1}$ m$^{-1}$ (V/m).

*Coherent ferrons.* Similar to magnons, we can study ferrons in two different limits. Propagating magnons that are coherently excited by microwaves at GHz frequencies propagate over centimeters in high-quality magnets such as yttrium iron garnet. The dominant ferrons in displacive ferroelectrics resonate at THz frequencies [40]. Moreover, resonance line broadenings are governed by viscous dissipation that scales as approximately $\omega$, whence the lifetimes of optical phonons and their group velocities may turn out to be relatively small. On the other hand, while their dipole and thus the coupling to ac and dc electric fields are weak, acoustic ferrons are attractive for experiments on propagating ferrons in crystals with low acoustic attenuation and high group (sound) velocities.

The interaction with ac electric fields leads to substantial and anisotropic anticrossings between photon and ferron branches, i.e., ferron polaritons [14]. At ferroelectric surfaces, the dipolar interaction guides the ferrons excited by localized sources into focused beams [41].

*Transport.* In order to improve upon the simple diffusion theory sketched above, the transport coefficients in Eq. (1) can be computed from the ferron spectrum and dipoles.

In constrictions such as point contacts, mean free paths may exceed the length of the current-limiting geometrical features. In that "ballistic" limit Landauer-Büttiker scattering theory is appropriate [35].

In the opposite "diffuse" regime, a semiclassical linearized Boltzmann equation leads to microscopic expressions for the parameters of the diffusion theory. When a nonequilibrium polarization or polarization accumulation (or ferron chemical potential) $\delta p = p(x,t) - p_0$ relaxes over a time $\tau_p$, the conservation relation for the polarization $\partial_x j_p = -(\partial_t - 1/\tau_p)\delta p$ leads to a diffusion equation for $\delta p$ that can be solved with appropriate boundary conditions [34,36]. $\tau_p$ can be very long provided the ferron and normal phonon band density of states do not overlap [14]. The effective field $E_{\text{eff}}$ is the sum of applied and internal fields including a diffusion term proportional to $\partial_x \delta p$. Its gradient is necessarily transient on a time scale $\tau_p$, as is the field-driven thermal current or "polarization Peltier effect". The polarization Seebeck effect is observable in terms of dc thermovoltages induced by a temperature gradient [34,35]. While both the Peltier effect and thermovoltage vanish with $\tau_p$, the polarization current induced by a temperature gradient persists and can be detected, e.g., by its stray magnetic fields [35].

## IV. EXPERIMENTS

Wooten *et al.* [42] reported experimental evidence of ferron excitations in terms of an electric field dependence of the thermal (or heat) conductivity $\kappa$ (Fig. 3) and velocity $v$ of pressure waves in a lead zirconium titanate (PZT)-based ferroelectric device. The observed field dependence is consistent with the relation $\kappa = (1/3)Cv^2\tau$, where the volumetric heat capacity at constant pressure $C$ is constant above the Debye temperature. Assuming that the scattering time $\tau$ does not depend on field, $d\ln\kappa/dE = 2d\ln v/dE$ as observed. These results indicate that the effect is intrinsic, i.e., caused by the field modulation of acoustic phonons that carry an electric dipole.

The original theories [34,36] are not appropriate to model these results since (i) PZT is a displacive ferroelectric and (ii) in the Landau model the dipoles are carried exclusively by a single band of longitudinal soft optical phonons at high frequencies and small group velocities. The piezoelectric strain, i.e., the linear contraction of the lattice by an applied electric field in ferroelectrics, modulates the sound velocities in particular via the mode- and frequency-dependent Grüneisen coefficients and thereby the thermal conductivity. The model predicts that $d\ln v/dE = -(d_{33} + 2d_{31})\gamma$, where the piezoelectric coefficient $d_{33}$ ($d_{31}$) measures the strain along (normal to) $\mathbf{E}$ and $\gamma = -d\ln\omega/d\ln V$ is the Grüneisen constant, i.e., the derivative of the phonon frequency $\omega$ with respect to the volume $V$. For the known material parameters for PZT the model explains the observations remarkably well. Below the Debye temperature $C \propto v^{-3}$ and $d\ln\kappa/dE = 2d\ln v/dE$ changes sign. Moreover, while $d_{33} + 2d_{31} > 0$ in PZT, it is negative in $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$, potentially enabling the design of complementary heat switches.

Preliminary data on nonferroelectric materials with large piezoelectricity/electrostriction and high dielectric constants, such as $SrTiO_3$, do not show any field dependence, which confirms the role of ferrons.

Analogously, the modulation of the sound velocity in ferromagnetic materials by magnetostriction [43] may affect the thermal transport under an applied magnetic field [44].

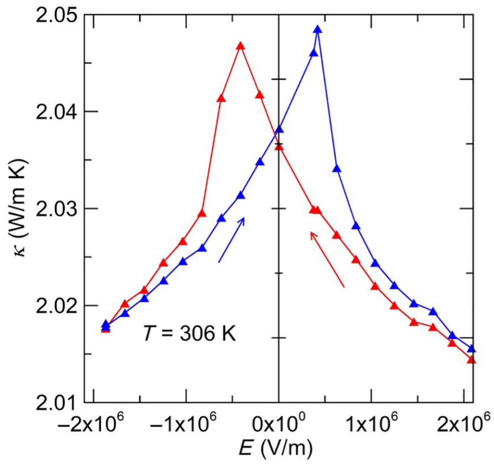


FIG. 3. Electric field dependence of the thermal conductivity of the ferroelectric PZT (adapted from Ref. [42]).

## V. POTENTIAL APPLICATIONS

*Thermal management.* In the footsteps of spin caloritronics, the science and technology of coupled spin, heat, and charge currents [45,46], ferrons offer strategies for thermal control and power generation.

Ferroelectric capacitors under temperature gradients generate stable surface charges, in contrast to the transient polarization offered by the pyroelectric effect. A transient polarization Peltier effect may contribute to electrocaloric cooling [34,35].

The polarization drag effect in heterostructures between ferroelectrics and metals frees device designers from the yoke of the Wiedemann-Franz law. In such structures, heat and charge currents flow in parallel but are spatially separated in different materials. Choosing a ferroelectric with a low thermal conductivity and a metal with a high electric conductivity leads to respectable thermoelectric figures of merit in van der Waals stacks [38]. In the spin Seebeck effect, the heat current in the magnet is also spatially separated from the induced transverse charge current in heavy-metal contacts, but the thermoelectric figure of merit is limited by small spin Hall angles [47].

The electric field dependence of the thermal conductivity [42] could lead to electrically controlled all-solid-state heat switches between high- and low-conducting states with important potential applications [48] in energy-saving technologies, particularly in engines with transient heating cycles such as solar-thermal power plants. Their development would enable all-solid-state power generators and refrigeration technologies, improving the efficiency of electrocaloric and magnetocaloric engines. Particularly interesting would be a set of complementary devices in a high- and low-conducting state that switches their role by an applied electric field that also powers the active electrocaloric material, which is an option offered by experiments [42].

*Logics and interconnects.* The weak dissipation of magnons makes them suitable conduits to exchange spin information and carry out logic operations [49]. Coherently excited ferrons can in principle carry out the same functions. A theory of surface ferrons predicts functionalities, such as tunable routing of focused ferron beams without the need for micro- and/or nanostructuring the surface into waveguides [41]. Since spin currents are the bread and butter of spin-torque magnetic memories [50], polarization currents might be useful in the writing and readout of ferroelectric memories [5].

*THz radiation source.* When injected into a metal, the polarization current decays on a femtosecond scale. However, when injected into an undoped semiconductor with a fundamental band gap below thermal energies, the generated electron and holes may recombine to emit photons. When the nonradiative recombination times are sufficiently long, such a structure could operate as a THz radiation source driven by waste heat.

## VI. CONCLUSIONS AND CHALLENGES

The present Perspective only scratches the surface of a full understanding of the basic physics and the applications of electric-dipole-carrying excitations in ferroelectrics and ferroelectric devices. Fundamental issues such as the formulation and observation of the polarization relaxation length must be still addressed. Some low-hanging fruits may be picked by applying the advanced insights of magnonics and mechanics to ferroelectric materials as in the examples below.

*Charge-spin-polarization coupling.* Spin-charge coupling is essential spintronics, but a polarization-charge coupling such as in the ferron drag effect [38] is much less established.

Ferroelectricity in or close to a conducting channel strongly affects the spin-orbit interaction. A switch of polarization then reverses the direction of spin Hall currents [51]. The reciprocal effect is the excitation of a ferron current by spin currents. Natural or composite multiferroic materials have been the focus of many studies in past decades, but such a coupling between polarization and spin currents has to the best of our knowledge been overlooked. A systematic study of transport phenomena in the presence of polarization and magnetic order coupled with the Moriya-Dzyaloshinskii interaction [31,32] could lead, for instance, to polarization-current detectors.

*Ferroelectric textures.* In unpoled ferroelectrics, the external electric fields are quenched by domains with opposite polarizations that are separated by domain walls. Moving domain walls facilitate ferroelectric switching by strongly suppressing critical fields. The interaction of ferrons with domain walls [20] and topological textures such as skyrmions [52] appears interesting and important. Ferroelectric domain walls may also provide waveguides for ferron excitations [53]. In analogy with magnonics, we envisage manipulation of ferroelectric textures by polarization currents, or in polar metals, even by charge currents.

*Quantum effects.* Extending the techniques of optomechanical systems [54] to ferroelectric materials would allow the quantum control of macroscopic electric polarization.

In order to progress, more experiments are sorely needed. We hope that we have been able to inspire our colleagues and stimulate collaborative efforts between experts in the fields of ferroelectrics and magnetism.

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