Electron as soliton: Nonlinear theory of dielectric polarization

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We describe a class of theories of dielectric polarization and a species of solitons in these theories. The solitons, made entirely out of the polarization field, have quantized values of the electric charge and can be interpreted as electrons and holes. A soliton-antisoliton pair is an exciton. We present numerical evidence that the elementary soliton is stable.

Absorption of light by dielectrics is one of the oldest problems in quantum theory. Because a typical frequency of visible light is much larger than the maximal frequency of phonons, absorption of even one photon must result in many phonons. Frenkel’s idea was that the immense suppression, naively expected for such a process, can be overcome if there is an intermediary—an electronic excitation of the solid (an exciton). An alternative to this point of view would be to imagine that phonons produced by light absorption initially form a highly coherent, nearly classical state. Then, the effective nonlinearity is enhanced by the large amplitude of the phonon field in the final state, resulting in a sizable absorption rate.

In the present Letter, we would like to argue that these two points of view are, in fact, complementary to each other. Namely, if the phonon field is treated as a nonlinear system, there are solitons of this field that are naturally interpreted as electrons and holes, and a soliton-antisoliton pair can be interpreted as an exciton.

We wish to stress from the outset that our results are quite different from the familiar polaron picture. In the latter case, there are two fields, which describe electrons and phonons separately, and the polaron is a bound state involving both. In our case, there is no separate electron field—electrons emerge as nonlinear excitations of phonons, somewhat similarly to how a baryon emerges as a nonlinear excitation of pions in Skyrme’s model. An even closer similarity is with the solitons of the sine-Gordon (SG) model, which can be viewed as fermions of the massive Thirring model.

Neither of these similarities is complete, however. Both in the SG case and in Skyrme’s model, solitons exists directly in the continuum limit, while in our case, at least in the present version of the theory, they can only live on a lattice.

One reason to pursue the picture of electrons as solitons is that it allows one to think about various processes of interest in nanoscale dielectrics entirely in classical terms. For example, one may want to know if a highly excited excitonic state relaxes to the lowest-energy exciton via carrier multiplication (i.e., production of more excitons) or via emission of multiple phonons. By viewing electrons as solitons of the phonon field, we obtain a classical approximation, which can be used to simulate this process on a computer.

Quantum physics of these solitons, in particular, the question of their spin and statistics, also promises to be interesting. In the present Letter, however, we limit ourselves to problems of the classical theory: arguing the existence of solitons and describing their simplest properties. While a plausible existence argument can be given analytically, as further evidence we also present results of a numerical study.

Consider then the field $\theta_n(x, t)$ of an optical phonon. We will consider in parallel spatial dimensions $d = 2$ and $3$ (planar and bulk dielectrics, respectively). The number of components of $\theta_n$, i.e., the number of values that $n$ takes, is equal to $d$. We assume that $\theta_n$ is defined on the faces (for $d = 3$) or edges (for $d = 2$) of a rectangular lattice with spacings $a_n$. Up to a constant factor, $\theta_n$ is the component of dielectric polarization in the $n$-th direction.

For notational simplicity, we will often use continuum notation for lattice derivatives. Thus, for example, the electric charge density (in suitable units) will be written as a divergence,

$$\rho(x, t) = \nabla \cdot \theta(x, t) \equiv \partial_n \theta_n(x, t),$$

but this should be read to mean

$$\rho(x, t) = \sum_{n=1}^{d} \frac{1}{a_n} \left[ \theta_n(x + n, t) - \theta_n(x, t) \right],$$

where $n$ is the primitive vector in the $n$-th direction. Note that, since $\theta_n$ live on the faces (or edges), $\rho$ naturally lives in lattice cells (i.e., on the lattice dual to the original). Hence the tilde in eq. (2).

The form of the Lagrangian density that governs dynamics of $\theta_n$ should ultimately be based on the properties of the material. For our present purposes, it will be sufficient to use the simplest form, which we write as the first few terms of a derivative expansion:

$$L = -V(\theta) - \frac{1}{2C} (\nabla \cdot \theta)^2 - \frac{\mathcal{L}}{2} (\partial_t \theta)^2 + \frac{1}{2} \mathcal{M}_{mn} (\partial_t \theta_m \theta_n)^2,$$

where $C > 0$, $\mathcal{L}$, and $\mathcal{M}_{mn}$ are constants. In what follows, we limit ourselves to searching for static solutions of the theory, and for these the terms containing time derivatives do not matter. They would be crucial, however, if we were to study the phonon spectrum.

The potential $V$ contains no derivatives of $\theta_n$ and describes the dielectric response of the medium to uniform
static fields. The only feature of it essential here is a certain periodicity, so we use the simple cosine form (which in one dimension would be the potential of the SG model):

\[ V(\theta) = \mu^2 \sum_n \frac{1}{\sigma_n^2} \left[ 1 - \cos(\sigma_n \theta_n) \right]. \tag{4} \]

In (4), \( \mu^2 \) is a parameter, and \( \sigma_n \) is the area of the faces (for \( d = 2 \), length of the edges) that are orthogonal to the \( n \)-th direction. At small \( \theta_n \), \( V \approx \frac{1}{2} \mu^2 \sigma_n^2 \), which allows one to relate \( \mu^2 \) to the bulk dielectric constant \( \epsilon \): \( \mu^2 = \frac{\epsilon^2}{\pi(\epsilon - 1)} \), where \( \epsilon = 14.4 \) eV Å is the electron charge squared.

The second and third terms on the right-hand side of (3) describe the capacitive and inductive effects due to short-scale charge separation. In particular,

\[ l_p = \frac{1}{\mu \sqrt{C}} \tag{5} \]

is a characteristic length for spatial variations of the polarization charge. For a material with a high degree of spatial symmetry, where we expect the expression (3) to apply, \( C \), together with \( L \) and \( M_{mn} \), can in principle be determined by fitting the phonon spectrum.

Note that if \( \mu \) is small (i.e., \( \epsilon \) is large), as in many semiconductors, \( l_p \) can be much larger than the atomic scale. In this case, in nanocrystals of sufficiently small size, effects of spatial dispersion may be quite significant.

On the experimental side, deviations of measured excitonic spectra from predictions of the “envelope” theory (in which electrons are considered point-like) have been reported for PbSe crystals with diameters smaller than 7 nm (3).

The short-scale effects encoded in the values of \( C \), \( L \), and \( M_{mn} \) should be distinguished from effects due to macroscopic electromagnetic (e.-m.) fields that can exist in a dielectric. To take those fields into account, we would need to add to (3) the term

\[ L' = \frac{|e|}{2\pi} \left[ A_0 \nabla \cdot \theta + \frac{1}{c} \mathbf{A} \cdot \partial_t \theta \right], \tag{6} \]

where \( A_0 \) and \( \mathbf{A} \) are the scalar and vector potentials, \( e \) is the electron charge, and \( c \) is the speed of light in vacuum. In \( d = 3 \), this term also contributes to the phonon frequency. For the moment, we are not interested in macroscopic e.-m. fields of solitons, so in what follows we omit the Lagrangian (6). Effects of impurities are also readily incorporated into the theory, by adding a term \( v(x) \nabla \cdot \theta(x, t) \), where \( v(x) \) is the impurity potential. For now, these will be neglected as well.

An important property of eq. (3) is the absence of “transverse capacitances”, i.e., terms of the form \( (\partial_n \theta_n)^2 \) with \( m \neq n \). As we will see, such terms lead to a linear (confining) potential between solitons. Since our goal is to describe a material in which electrons and holes can be separated away to large distances, these terms are in fact forbidden. On the other hand, we could add terms of the form \( (\partial_n \sin(\sigma_n \theta_n))^2 \). At small \( \theta_n \), these are indistinguishable from the forbidden terms, but at larger \( \theta_n \) they display the periodicity property that causes the tension of the “string” connecting solitons to vanish. If these terms are relatively small, then adding them to (3) will only deform our solution a little, and for the present work we have left them out. In applications to specific materials, they can be readily included.

The argument forbidding the “transverse capacitances” can be made more formal by identifying a relevant symmetry. For definiteness, we will speak about the \( d = 2 \) (planar) case; the argument for \( d = 3 \) is entirely similar. Consider a directed closed contour on the dual lattice (see Fig. 1). Imagine changing the values of \( \theta_n \) at all the edges (in \( d = 3 \), faces) the contour crosses by \( \pm 2\pi / \sigma_n \), the sign depending on whether the contour runs parallel or opposite to the \( n \)-th axis. This transformation will be referred to as adding a closed \( 2\pi \) string. The symmetry in question is the requirement that under this transformation the energy does not change.

An open \( 2\pi \) string, such as the one shown in the lower portion of the figure, does cost energy, i.e., it is an excitation. If the symmetry with respect to adding closed strings is in place, however, the string tension is zero, and the energy of an open string is accumulated only near the ends. Since a \( 2\pi \) string carries electric flux of \( 2\pi \), the ends of an open string are charged, with quantized charges equal to \( \pm 2\pi \), so this excitation can be interpreted as an electron-hole pair.

In the one-dimensional SG model, this way of creating excitations leads to a soliton-antisoliton pair that is nearly stable (the more so the larger is the distance between the solitons). Indeed, imagine starting from the state with \( \theta_n = 0 \) (in one dimension, \( \theta_n \) has only one component) and then changing \( \theta_n \) by \( 2\pi \) at all sites of a chosen segment. If one then “cools” down the system, to reach a low-energy state, the field will become smooth.

\FIG. 1: The original lattice (thin lines) and closed and open strings on the dual lattice.\
at the ends of the segment but will remain equal to $2\pi$ in the middle: due to the high potential barrier, it cannot unwind back to zero there. Similarly, in our case, we also expect formation of a stable soliton pair (and, as we discuss shortly, this has been confirmed in numerical simulations).

If, however, we have allowed the "transverse capacitances", the symmetry with respect to adding closed $2\pi$ strings would be broken. For an open string, energy would then have come not only from the ends but from the entire length and, for a large length, would be proportional to it. As a result, a pair of well separated soliton and antisoliton would not be an approximate solution: the linear potential would pull the soliton and antisoliton towards each other until they annihilate. (This has also been confirmed numerically.)

The symmetry with respect to adding closed $2\pi$ strings is local, in the sense that the number of independent transformations scales linearly with the volume of the system. Indeed, any closed loop can be viewed as a superposition of elementary closed loops, each encircling just a single site (in $d = 2$) or edge (in $d = 3$) of the original lattice. With the loop’s directionality taken into account, there are twice as many ways to add an elementary loop as there are sites or edges, respectively.

The local character of the symmetry suggests that the configuration space of our theory is not the space of all possible $\theta_n(x)$, but the space of equivalence classes with respect to adding an arbitrary number of closed $2\pi$ strings, i.e., states that differ only by the number of closed $2\pi$ strings should be considered as one state. The role of this equivalence relation will become clearer shortly, when we consider the limit in which our soliton turns into a vortex or a magnetic monopole. However, this relation is not essential in the method we have used for finding solitons.

The presence of a string, similar to Dirac’s string, suggests that our solution is related to the magnetic monopole (or the vortex in $d = 2$). Indeed, it is a generalization of those. The monopole is obtained in the limit $C \to 0$. Let us first consider this as a formal limit, i.e., as the requirement that $\nabla \cdot \theta = 0$. Then, $\theta$ is the curl of some other field, $\phi$. In $d = 2$, this field lives on the sites of the original lattice and has only one component, so that

$$\theta_n = \epsilon_{nm} \partial_m \phi$$

($\epsilon_{nm}$ is the antisymmetric unit tensor); in $d = 3$, $\phi$ lives on the edges. Changing $\phi$ by $2\pi$ at a single site creates, in terms of $\theta$, an elementary closed $2\pi$ string encircling that site. So, imposing the equivalence relation as above amounts to requiring that $\phi$ is an angular variable with period $2\pi$. Substituting eq. (7) into eq. (4), we see that the potential $V(\theta)$ becomes the energy density of the 2-dimensional XY model, which has well-known static solutions—vortices. In $d = 3$, a similar argument leads to compact electrodynamics, whose solutions are magnetic monopoles [4]. (They are “magnetic” with respect to the gauge field that replaces the scalar $\phi$ in the curl condition, not with respect to ordinary electromagnetism.)

A less formal way to view the $C \to 0$ limit is to note that in this case the screening length $l_p$, eq. (5), goes to infinity. Since the total charge of an elementary soliton is fixed (and equal to $2\pi$), the charge density $\nabla \cdot \theta$ goes to zero. Thus, if a soliton and an antisoliton are separated by a distance much smaller than $l_p$, i.e., sit well within each other’s polarization cloud, we expect them to interact as if they were monopoles (or vortices) of the respective $C = 0$ theories.

As well known, monopoles and vortices have long-range interactions, which are important in various problems of statistical physics [6]. From our present standpoint, these interactions are a feature of the $l_p \to \infty$ limit. For a finite $l_p$, the interaction range is of order $l_p$ (provided, of course, that the e.-m. interactions (6) have been switched off).

We now turn to numerical results. To search for static solutions of the theory (4), we drop the terms with time derivatives and numerically solve the relaxation equation:

$$\frac{\partial \theta}{\partial \tau} = \nabla (\nabla \cdot \theta) - \frac{1}{l_p^2} \frac{\partial V}{\partial \theta},$$

where $\tau$ is a fictitious time. To avoid influence of boundaries, we have used periodic boundary conditions.

As the initial condition, we have used an open string, such as shown in fig. 1, with various values of the electric flux. The actual solutions (endpoints of the relaxation process) are expected to have the flux quantized in integral multiples of $2\pi$—these are the values of $\sigma_n \theta_n$ at which the potential (4) has minima. It is interesting, however, to start with an arbitrary value and see how the nearest quantized value is approached. Indeed, some of the most illustrative results are obtained by starting with $\sigma_n \theta_n$ close to a maximum of the potential.

Fig. 2 shows results from a 2-dimensional $64 \times 32$ lattice for the case when the initial string has a flux of $\sigma_1 \theta_1 = 3.02\pi$; results from a 3-dimensional $64 \times 32^2$ lattice (for the same initial flux) are similar. The screening length ($l_p = 1$) in either case was about 3 times the lattice spacing (which was the same in all directions).

The initial flux is just larger than $3\pi$, a value at which the potential has a maximum. So, we expect the flux (and therefore also the charge at the string’s end) to roll to the nearest stable value, $\sigma_1 \theta_1 = 4\pi$, which is twice the minimal soliton charge. The final value of the charge at each end is indeed close to $\pm 4\pi$ (a small difference from $4\pi$ being attributable to finite-size effects), but we see that it is achieved by forming two solitons at each end, rather than a single doubly-charged one. The final state was stable on the timescale of our simulation.

We have observed formation of two solitons instead of one even when the initial string flux already was
σ_1θ_1 = 4π. We interpret this as an indication that the short-range interaction between solitons of like charge is repulsive, and a compound soliton is unstable with respect to decay into elementary ones.

To summarize, we have described a class of theories that provide a unified description of phonons and excitons in a dielectric. Phonons are small fluctuations of a polarization field, and excitons are soliton-antisoliton pairs made of that same field. We have presented numerical evidence that the elementary soliton is stable and identified the symmetry (adding closed 2π strings) that underlies both this stability and the quantization of the soliton charge.

FIG. 2: Surface plots of the charge density towards the beginning (top) and end (bottom) of the relaxation process.

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