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Mode Softening, Ferroelectric Transition, and Tunable Photonic Band Structures in a Point-Dipole Crystal

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We study the photonic band structure of cubic crystals of point dipoles. It is shown that in contrast to earlier claims these systems cannot have an omnidirectional photonic band gap. For sufficiently large plasma frequencies, however, they exhibit softening of photonic bands, leading to (anti)ferroelectric ordering of the dipoles and the possibility to open and tune directional band gaps by external electric fields. The model studied may be realized through lattices of quantum dots.

The study of wave propagation through a periodic structure has had a long history, and still plays an important role in condensed matter physics. A conceptually simple example with a rich variety of physical properties is the propagation of electromagnetic waves through crystals of point dipoles. The oldest use of this model is the concept of the polariton and the polariton stop gap, describing the optical modes in atomic and molecular crystals [1,2]. More recently, crystals of point dipoles have also been used to model the propagation of light through photonic crystals. These are materials in which the refractive index is modulated with a period of the order of the wavelength of light. The multiple scattering in these crystals gives rise to the opening of photonic band gaps [3,4], in analogy to the electronic band gaps in semiconductors; photonic band gaps have been reproduced using lattices of point dipoles [5]. Photonic band gaps have interesting fundamental and technological consequences, such as the possibility to modify spontaneous emission [6], and the development of perfect mirrors and novel wave guides [7]. Other intriguing phenomena that have been predicted for point-dipole crystals are a negative refractive index and subwavelength lensing [8].

Recent advances in the fabrication of semiconductor and metallic quantum dots have opened new possibilities to realize materials that are well represented as crystals of point dipoles. The Mie or plasma resonances of the individual dots act as the dipolar transitions, with a wavelength much larger than the dot size [9]. It has been realized that arrays of closely spaced quantum dots, considered as point dipoles, may be used as subwavelength guides for electromagnetic energy (plasmonics) [10,11]. It has also been suggested that quantum dots forming two-dimensional lattices may spontaneously polarize as a consequence of the interdot dipolar interactions, even for lattice constants of the order of 100 nm [12].

Combining several of the above concepts, one may envision photonic crystals whose band structures soften as a consequence of electromagnetic interactions between unit cells, leading to a dipole-ordered state at some critical interaction strength. Close to this instability, the band structure is expected to be very sensitive to external electric fields. This opens the possibility to tune photonic band gaps, a highly desired feature in photonics, for which several alternative methods have been suggested [13–18].

In this Letter, motivated by the above ideas, we perform a detailed study of the photonic band structure of cubic (sc, fcc, and bcc) point-dipole crystals. The dipole-light interaction couples the dipole excitations with a crystal wave vector $\mathbf{k}$ to photons with wave vectors $\mathbf{k} + \mathbf{G}$, where $\mathbf{G}$ is an arbitrary vector of the reciprocal lattice. This coupling, on the one hand, gives rise to the photonic bands and, on the other hand, results in long-range (retarded) interactions between the dipoles. We show that, in contrast to earlier claims [5], omnidirectional gaps do not occur in the band structure of these lattices. We also show that for sufficiently strong interactions the lowest photonic bands may soften at a particular symmetry point of the Brillouin zone, which leads to a dipole-ordered state, and we demonstrate the opening and tunability of directional band gaps when switching external electric fields close to the transition to this ordered state.

We consider a periodic array of transition dipoles, treated as harmonic oscillators with a frequency ω_0, coupled linearly to the electromagnetic field. The Lagrangian reads (cf. Ref. [1])

$$L = \int dV \left[\frac{\mathbf{E}^2 - \mathbf{H}^2}{8\pi} + A^0 \nabla \cdot \mathbf{P} + \frac{\mathbf{A}}{c} \cdot \dot{\mathbf{P}} \right]$$
$$+ \frac{2\pi n}{\omega_p^2} \sum_j (\dot{\mathbf{p}}_j^2 - \omega_0^2 \mathbf{p}_j^2). \tag{1}$$

Here the plasmon frequency ω_p plays the role of a coupling constant, n is the density of dipoles and the polarization density is given by $\mathbf{P}(t, \mathbf{x}) = \sum_j \mathbf{p}_j(t)\delta(\mathbf{x} - \mathbf{R}_j)$, where $\mathbf{R}_j$ are the coordinates of the lattice sites. Imposing the Coulomb gauge $\nabla \cdot \mathbf{A} = 0$ and eliminating A^0, we obtain

$$L = \frac{1}{8\pi} \int dV \mathbf{A} \cdot \left(-\frac{1}{c^2} \frac{\partial^2}{\partial t^2} + \Delta \right) \mathbf{A}$$
$$+ \frac{2\pi n}{\omega_p^2} \sum_j (\dot{\mathbf{p}}_j^2 - \omega_0^2 \mathbf{p}_j^2) + \frac{1}{c} \sum_j \dot{\mathbf{p}}_j \cdot \mathbf{A}(\mathbf{R}_j)$$
$$+ \frac{1}{2} \sum_{l \ne j} \frac{3(\mathbf{p}_l \cdot \mathbf{R}_{lj})(\mathbf{p}_j \cdot \mathbf{R}_{lj}) - \mathbf{R}_{lj}^2 \mathbf{p}_l \cdot \mathbf{p}_j}{R_{lj}^5}, \quad (2)$$

where $\mathbf{R}_{lj} = \mathbf{R}_l - \mathbf{R}_j$. The third term describes the coupling of dipoles to transverse electromagnetic field modes, while the last term is the static dipole-dipole interaction.

This Lagrangian gives rise to coupled equations of motion for dipolar excitations with wave vector $\mathbf{k}$ from the first Brillouin zone and photons with wave vectors $\mathbf{k} + \mathbf{G}$, where $\mathbf{G}$ is an arbitrary vector of the reciprocal lattice. Eliminating the photon field we obtain an effective equation of motion for dipoles

$$\sum_b M_{\omega\mathbf{k}}^{ab} p_{\omega\mathbf{k}}^b = 0, \quad (3)$$

where $a, b = x, y, z$, $\mathbf{p}_{\omega\mathbf{k}} = \sum_j \int dt e^{i\omega t - i\mathbf{k} \cdot \mathbf{R}_j} \mathbf{p}_j(t)$, and

$$M_{\omega\mathbf{k}}^{ab} = (\omega_0^2 - \omega^2)\delta^{ab}$$
$$+ \omega_p^2 \left[\sum_{\mathbf{G}} L_{\omega\mathbf{k}+\mathbf{G}}^{ab} - \int_{BZ} \frac{d^3 q}{v_{BZ}} L_{\omega\mathbf{k}+\mathbf{G}+\mathbf{q}}^{ab} \right],$$

with $L_{\omega\mathbf{k}}^{ab} \equiv (\omega^2 \delta^{ab} - c^2 k^a k^b)(\omega^2 - c^2 k^2)^{-1}$. The dispersion $\omega_{\mathbf{k}}$ of the coupled excitations is found from

$$\det M_{\omega_{\mathbf{k}}\mathbf{k}}^{ab} = 0. \quad (4)$$

Effects of dissipation due to nonradiative decay on the band structure [19], are not considered here; as a result, all solutions to Eq. (4) are real. In fact the calculation of bands in our model is similar to the calculation of the polariton dispersion [1]. The only subtlety is the regularization of the sum over $\mathbf{G}$ in Eq. (3) resulting from the self-interaction of the point dipoles. Subtracting the self-energy is accomplished by subtracting from $L_{\omega\mathbf{k}+\mathbf{G}}^{ab}$ the average of $L_{\omega\mathbf{k}+\mathbf{G}+\mathbf{q}}^{ab}$ over the first Brillouin zone [with volume $v_{BZ} = (2\pi)^3 n$]. Apart from the regularizing term, Eq. (4) is identical to those obtained in Refs. [5,20] by more complicated methods (inspection reveals that the regularization used in Ref. [20] does not remove all terms divergent at large G). In what follows we use dimensionless variables measuring $\mathbf{k}$ in units of $2\pi/a$ and ω in units of $2\pi c/a$. For a simple-cubic (sc) lattice a is the lattice period. For bcc and fcc lattices, which can be represented as, respectively, 2 and 4 interpenetrating cubic sublattices, a is the period of the sublattice.

In Figs. 1 and 2 we show band structures for the sc and fcc lattice, respectively. The solid lines were obtained by solving Eq. (4) [21], while the circles are solutions of the dispersion equation $\omega = k$ for free photons (poles of $L_{\omega\mathbf{k}}^{ab}$), corresponding to standing waves of the electromagnetic field $\mathbf{E}(\mathbf{x})$ with nodes on the lattice sites. The latter are superpositions of two plane waves with the wave vectors
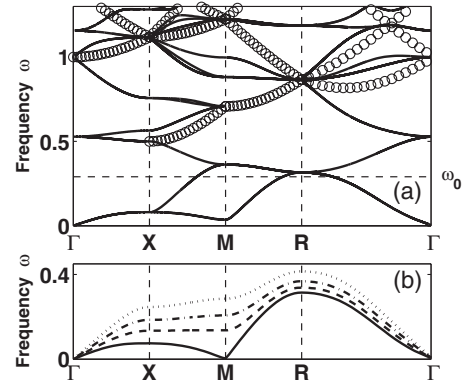

FIG. 1. (a) Band structure for the sc lattice for $g = 2.31$ ($\omega_p = 0.4$ and $\omega_0 = 0.263$), close to the softening at the M point. The solid lines are found by solving Eq. (4), and the open circles correspond to the free photon modes $\omega = k$ that have nodes on the lattice sites. (b) The evolution of the lowest band when g approaches $g_c \approx 2.35$. From top to bottom $g = 1, 1.47, 1.90$, and 2.35 ($\omega_p = 0.4$ for all bands).

$\mathbf{k}_1$ and $\mathbf{k}_2$ and polarization $\mathbf{e}$, $\mathbf{E}(\mathbf{x}) = \mathbf{e}(e^{i\mathbf{k}_1 \cdot \mathbf{x}} - e^{i\mathbf{k}_2 \cdot \mathbf{x}})$, with $\mathbf{e} \cdot \mathbf{k}_1 = \mathbf{e} \cdot \mathbf{k}_2 = 0$, $k_1 = k_2$, and $\mathbf{k}_2 = \mathbf{k}_1 + \mathbf{G}$, so that at the lattice sites $\mathbf{E}(\mathbf{R}_j) = 0$, implying that the light is not coupled to the dipoles. For all three lattices one can find lines of $\mathbf{k}_1$ and $\mathbf{k}_2$ in the momentum space passing through the Brillouin zone boundaries that are related by a symmetry transformation of the reciprocal lattice (see Table I), which give rise to bands of decoupled states with energy continuously varying between some minimal value ω_c and ∞. Because of this peculiar property of lattices of point dipoles, their band structures cannot have an omnidirectional gap for $\omega > \omega_c$, where $\omega_c = \frac{1}{2}, \frac{\sqrt{3}}{2}, \frac{1}{\sqrt{2}}$ for, respectively, the sc, fcc, and bcc lattice.

Actually, in contrast to what was reported in Ref. [5], we did not find an omnidirectional gap for $\omega < \omega_c$ for any parameter [22]. For the bands in Figs. 1 and 2 there is a path in $\mathbf{k}$ space passing through high-symmetry points, along which the frequency $\omega_{\mathbf{k}}$ continuously increases from 0 to ∞. Such a path is present for all values of the model parameters, since the degeneracy of the modes at high-symmetry points (the Γ, X, M, and R points for the sc
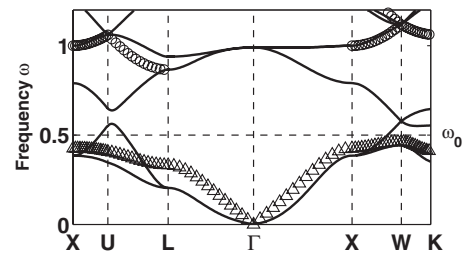

FIG. 2. Band structure for the fcc lattice for $g = 2.9$ ($\omega_p = 0.85$ and $\omega_0 = 0.5$), close to the ferroelectric transition, where the velocity vanishes at the Γ point. We also show the lowest band for a slightly weaker coupling, $g = 2.56$ (triangles).

TABLE I. Free photon modes in the coupled system. Here **k** is the wave vector, $\mathbf{G} = \mathbf{k}_2 - \mathbf{k}_1$ is a vector from the reciprocal lattice, **e** is the polarization, and α is a positive real number.

	$\mathbf{k}_1$	$\mathbf{k}_2$	$\mathbf{G}$	$\mathbf{e}$	ω_c
sc	$\begin{pmatrix} \frac{1}{2} \\ \alpha \\ 0 \end{pmatrix}$	$\begin{pmatrix} -\frac{1}{2} \\ \alpha \\ 0 \end{pmatrix}$	$\begin{pmatrix} -1 \\ 0 \\ 0 \end{pmatrix}$	$\begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$	$\left(\frac{1}{2} + \alpha\right)$
fcc	$\begin{pmatrix} -\frac{1}{2} \\ \frac{1}{2} + \alpha \\ -\frac{1}{2} + a \end{pmatrix}$	$\begin{pmatrix} \frac{1}{2} \\ -\frac{1}{2} + \alpha \\ \frac{1}{2} + a \end{pmatrix}$	$\begin{pmatrix} 1 \\ -1 \\ 1 \end{pmatrix}$	$\frac{1}{\sqrt{6}}\begin{pmatrix} 2 \\ 1 \\ -1 \end{pmatrix}$	$\sqrt{(\frac{3}{4} + \alpha^2)}$
bcc	$\begin{pmatrix} -\frac{1}{2} + \alpha \\ \frac{1}{2} + \alpha \\ 0 \end{pmatrix}$	$\begin{pmatrix} \frac{1}{2} + \alpha \\ -\frac{1}{2} + \alpha \\ 0 \end{pmatrix}$	$\begin{pmatrix} 1 \\ -1 \\ 0 \end{pmatrix}$	$\begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$	$\sqrt{(\frac{1}{2} + \alpha^2)}$

lattice) cannot be made smaller, so that deformations of the bands cannot open an omnidirectional gap (similar for the bcc and fcc lattice). It is important to note that this mode degeneracy only partly results from the lattice symmetry and partly comes from the dynamics of the point-dipole model, so that this argument for the absence of a gap is not universal. For instance, for the sc lattice of point dipoles M^{ab} is a diagonal matrix at the M point [$\mathbf{k} = (1/2, 1/2, 0)$] with $M^{11} = M^{22}$. In general, however, nonzero matrix elements M^{12} and $M^{13} = M^{23}$ would also be compatible with the symmetry of this point.

When the dimensionless coupling $g = \omega_p^2/\omega_0^2$ of the dipoles to light is larger than some critical value g_c, the system becomes unstable towards a transition to an ordered state, in which the dipole operators have nonzero average values at lattice sites. The type of ordering can be inferred from the wave vector $\mathbf{Q}$, at which the excitation frequency vanishes (*softening*). If $\mathbf{Q} \neq 0$, this indicates a transition to an antiferroelectric state, in which the polarization density is modulated with the wave vector $\mathbf{Q}$. The instability towards a ferroelectric state results in a vanishing velocity of the photonlike excitations with a linear dispersion at $\mathbf{Q} = 0$. These instabilities are similar to those of the phonon spectrum (softening of phonons) in displacive (anti)-ferroelectrics at the critical temperature [23], and the instability in the Dicke model of an electromagnetic field mode coupled to an ensemble of two-level systems [24].

For all three lattices (sc, fcc, and bcc) we find an instability at the Γ point ($\mathbf{Q} = 0$) and $g_c = 3$, indicating that for $g > 3$ the ground state is ferroelectric. The vanishing of the velocity of the transverse excitations at the Γ point is shown in Fig. 2. This instability was discussed previously in the context of a model of classical dipoles of fixed length on the sc lattice (the Lorentz model) [25]. In the long-wavelength limit the dielectric function at zero frequency reads $\epsilon(0) = (\omega_0^2 + \frac{2}{3}\omega_p^2)/(\omega_0^2 - \frac{1}{3}\omega_p^2)$. At $g = 3$, $\epsilon(0)$ diverges corresponding to a transition into a ferroelectric state. Our model with dynamic dipoles has the same instabilities as the Lorentz model, because at low frequencies the retarded interaction between dipoles, re-

sulting from their coupling to transverse photons, is small compared to the instantaneous dipole-dipole interaction.

For the sc lattice we also find another instability at the M point, which occurs at a smaller value of the coupling, $g_c \approx 2.35$. The polarization vector of the soft mode, $\mathbf{e} = (0, 0, 1)$, is transverse to the propagation vector. This softening corresponds to a C-type antiferroelectric ordering (analogous to the C-type antiferromagnetic ordering), in which dipoles form ferroelectric chains in the z direction. In each chain dipoles are either parallel or antiparallel to the z axis and neighboring chains have opposite polarization. The band structure in Fig. 1(a) has been obtained for g close to g_c. The dependence of the lower band on g is shown in Fig. 1(b). This instability is in agreement with the result of Luttinger and Tisza [26], who studied the lowest energy configurations of the Lorentz model for sc, bcc, and fcc lattices, restricting to configurations with no more than 8 dipoles in a unit cell. For the sc lattice they found that the C-type antiferroelectric ordering has the lowest energy density, $\varepsilon \approx -2.675$ (sc), measured in units of $n^2 p^2$, where p is the classical dipole moment and n is the number of dipoles per unit volume. For the bcc and fcc lattices they found lowest energy densities for antiferroelectric configurations, $\varepsilon \approx -1.986$ (bcc) and -1.808 (fcc).

We do not find any softening at nonzero wave vectors for fcc and bcc lattices that occurs before the ferroelectric instability at $g = 3$. We used our formalism to study the minimal energy states of the Lorentz model and found that for all three lattice types the incommensurate helical state

$$\mathbf{p}_j = p\left[\mathbf{e}_1 \cos\left(\frac{2\pi}{a}\mathbf{Q} \cdot \mathbf{R}_j\right) + \mathbf{e}_2 \sin\left(\frac{2\pi}{a}\mathbf{Q} \cdot \mathbf{R}_j\right) \right],$$

for $\mathbf{e}_1 \cdot \mathbf{Q} = \mathbf{e}_2 \cdot \mathbf{Q} = 0$ and $Q \to 0$ (but $Q \neq 0$) has an energy density $\varepsilon \approx -2.094$, which for the bcc and fcc lattices is lower than the energies found in Ref. [26]. We note that for infinite periodic lattices of dipoles the energy of the helical state is not a continuous function of the wave vector at commensurate values of the wave vector $\mathbf{Q}$. In particular, the energy density of the uniform ferroelectric state ($\mathbf{Q} = 0$) equals 0 for all three types of the crystal lattice and is much higher than the energy of the incommensurate state with $Q \to 0$. The energy loss for the uniform ferroelectric state with $\mathbf{Q} = 0$ results from the frustration of the long-range dipole-dipole interactions. The softening at the Γ point at $g = 3$ for the bcc and fcc lattices (see Fig. 2) thus corresponds to a transition into a state, in which the dipole ordering is *locally* ferroelectric. However, at distances much larger than 1 lattice constant the dipole polarization must vary to prevent the energy loss due to frustrated long-range interactions. We also note that the energy density of the helical state with $Q \to 0$ tends to the energy density of long thin crystals, $\varepsilon = -2\pi/3 \approx -2.094$, for which the dipole-dipole interactions are not frustrated.

The transition to a ferroelectric state largely affects the low-frequency part of the band structure of arrays of point dipoles. However, the divergency of the dielectric constant
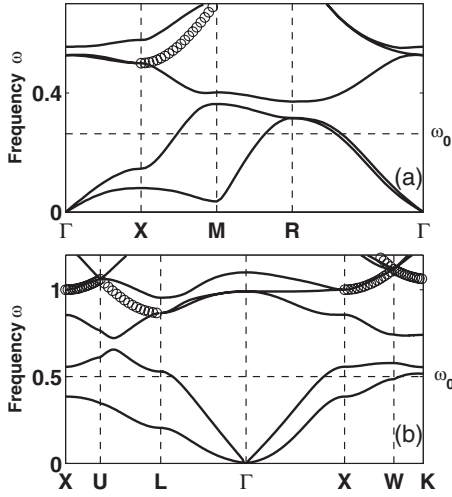
FIG. 3. Band structure (a) for a sc lattice of dipoles for $\omega_p = 0.4$ and $\omega_0 = 0.263$, with $\lambda_1 = 0.1$ and $\lambda_2 = 0.05$ in an electric field in the $(1, -1, 0)$ direction with a strength $E_{ext} = 0.006$, (b) for an fcc lattice of dipoles for $\omega_p = 0.5$ and $\omega_0 = 0.85$, with $\lambda_1 = 0.0$ and $\lambda_2 = 0.08$ in an electric field $E_{ext} = 0.03$ in the $(-1, 1, 1)$ direction.

at the ferroelectric transition can be used to manipulate the bands at high frequencies, e.g., by applying an external electric field E_{ext}. For dipoles with nonlinear dynamics, an average electric polarization $\langle \mathbf{p} \rangle$, induced by an electric field, changes the symmetry of the dipolar interactions, which affects the photonic bands. For instance, an array with a cubic symmetry allows for two quartic terms, $\frac{\lambda_1}{4} \times (p_x^4 + p_y^4 + p_z^4)$ and $\frac{\lambda_2}{2}(p_x^2 p_y^2 + p_y^2 p_z^2 + p_z^2 p_x^2)$, in the Lagrangian for the dipoles. Such higher-order terms are, actually, necessary for stabilization of ordered dipole states.

An external electric field applied along one of the cubic axes will change the symmetry from cubic to tetragonal. Such symmetry changes most strongly affect degenerate photonic bands and can open directional gaps at high-symmetry points. For example, an electric field in the $(1, -1, 0)$ direction applied to the sc lattice of dipoles lifts the double degeneracy of the photon frequency ω at $\mathbf{k} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$: $\omega_\pm^2 \approx \omega^2 - (\frac{1}{2}\lambda_2 + \frac{3}{2}\lambda_1 \pm \lambda_2)\langle p \rangle^2$, and opens the gap $\Delta = \omega_+ - \omega_- \approx |\lambda_2|\langle p \rangle^2/\omega \propto E_{ext}^2/(g_c - g)$. In Fig. 3(a) we show the band structure for the sc crystal for parameters used to plot Fig. 1(a), in the presence of an electric field along the $(1, -1, 0)$ direction, obtained by solving the linearized dipole-photon equations for $\lambda_1 = 0.1$, $\lambda_2 = 0.05$, and $E_{ext} = 0.006$. One can see a gap opening at the M and R points. Similarly, an electric field along the direction $(-1, 1, 1)$ direction applied to the fcc lattice opens a gap at the W point [see Fig. 3(b)]. The field effects are strongly enhanced close to the ferroelectric instability, where the dielectric susceptibility is large.

In conclusion, we studied a simple model for a photonic crystal, namely, a cubic crystal of point dipoles, and calculated the photonic band structure. We have shown that in such crystals, omnidirectional photonic band gaps do not occur. We have also shown that the long-range interactions between the dipoles may give rise to a dipole-ordered state, which manifests itself through the softening of the lowest photonic band at one of the symmetry points of the Brillouin zone. The resulting state is ferroelectric (fcc, bcc) or antiferroelectric (sc). Near the instability point, the band structure is very sensitive to external electric fields, a property that may be used to open and tune a directional band gap. The realization of actual systems in which the condition for softening and tunability applies, hinges on the preparation of crystals of small building blocks with strong dipole transitions; quantum dots with a sufficiently high plasmon frequency seem to be the best candidates [8,12].

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