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Disorder-induced polaritons

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Abstract

We consider phonon polariton states which may arise in disordered dielectrics due to a random light-phonon coupling. We show that these states can be described within the model of disorder-induced crossing resonance (DICR). The dispersion laws and relaxation rates of these states have been explored. Polaritons are shown to be good candidates for the experimental observation of the effects of DICR.

Keywords: Polariton; Disordered materials; Dispersion; Coupled waves; Random interaction; Level repulsion

1. Introduction

Polaritons in disordered materials have recently attracted much attention [1–4]. Different types of polaritons, such as plasmon [1], exciton [2] and phonon [3,4] polaritons, were considered. In spite of the differences between these investigations, they share one common feature. The dielectric function used in the Maxwell equations was chosen in a form that was derived for bulk ordered materials. A random parameter appeared either in the Maxwell equations or in the boundary conditions. Such an approach does not take into account the influence of disorder on the second participating system: phonons, plasmons, or excitons.

Disorder, however, affects both interacting subsystems. Moreover, fluctuations of the coupling parameter itself could be very important, particularly in the vicinity of the crossing point. At this point original dispersion curves of interacting waves cross each other and even small perturbations of the interaction may cause a significant effect. As far as we

know, these questions have not been addressed. In this paper we consider effects caused by fluctuations of the coupling parameter. In order to emphasize their role we consider a phonon polariton model, in which the mean value of the coupling parameter is equal to zero. This model describes the interaction between light and optical phonons, which would be inactive in an ideal structure. A random distortion of the structure could lead to the appearance of a local random coupling parameter, even though its mean remains zero.

We show that this situation may be described within the model of disorder-induced crossing resonance (DICR) introduced in Ref. [5]. The phenomenon of DICR reveals new challenges in the old field of wave propagation in random materials. Finding the best candidates for an experimental observation of DICR related effects is currently one of the main goals in this area. A magnetoelastic resonance in amorphous zero-mean magnetostrictive alloys was considered in Refs. [6,7]. Magnetoelastic interaction, however, is rather weak. Therefore, one needs to

construct a special material to observe DICR. The interaction between light and phonons is much stronger. The LO–TO splitting, which is a direct measure of the interaction, is of the same order of magnitude as the frequency itself in ionic crystals. Even if we assume that the local dipole moment, which arises in a random material due to structural distortion, is two orders of magnitude smaller, it will still be strong enough to result in observable effects.

In this paper we show how the Maxwell equations along with the general equations of lattice dynamics can be turned into equations describing DICR. Then, we apply the results obtained in Refs. [5,8] to the particular case of polaritons and discuss the possibility to observe the effects of interest.

2. Polaritons in disordered dielectrics. General equations

Let us consider an ensemble of ions arranged in a way allowing for optical vibrations. These kinds of vibrations may exist even in a random structure, where Voronoi's polyhedrons play the role of primitive cells in an ideal lattice [9]. The general equations of motion for such an ensemble interacting with an electromagnetic field read

$$m_\alpha \ddot{U}_\alpha^i(\mathbf{r}) + \sum_{\beta, \mathbf{r}'} D_{\alpha\beta}^{ij}(\mathbf{r}, \mathbf{r}') U_\beta^j(\mathbf{r}') = e_\alpha E^i(\mathbf{r}). \quad (1)$$

Here the indexes α, β numerate atoms within a cell, the vector \mathbf{r} indicates the position of a cell, and the upper indexes i, j refer to the Cartesian components of the vector quantities, such as the displacement vector \mathbf{U} , the dynamical matrix $\hat{D}(\mathbf{r}, \mathbf{r}')$ and the electric field \mathbf{E} . The parameters m_α and e_α are the mass and charge of the α th atom in a primitive cell.

Let us introduce the matrix $c_{\alpha\sigma}^i(\mathbf{r})$, which diagonalizes the dynamical matrix with respect to the cell indexes α, β , and let W_σ be the corresponding eigenvector. Then, the displacement vector may be written as

$$U_\alpha^i = \sum_\sigma c_{\alpha\sigma}^i(\mathbf{r}) \frac{W_\sigma(\mathbf{r})}{\sqrt{m_\alpha}}. \quad (2)$$

Eq. (1) can be reformulated in terms of the variables W_σ as

$$\ddot{W}_\sigma^i(\mathbf{r}) + \sum_{\mathbf{r}'} \hat{D}_\sigma^{ij}(\mathbf{r}, \mathbf{r}') W_\sigma^j(\mathbf{r}') = d_\sigma^i E^i(\mathbf{r}), \quad (3)$$

where \hat{D}_σ^{ij} is the dynamics matrix diagonalized with respect to the cell indexes. The vector d_σ^i , which appears in Eq. (3), plays the role of the coupling parameter between the electromagnetic waves and the lattice vibrations. This vector is defined according to the equation

$$d_\sigma^i(\mathbf{r}) = \sum_\alpha c_{\alpha\sigma}^i(\mathbf{r}) \frac{e_\alpha}{\sqrt{m_\alpha}}. \quad (4)$$

Being multiplied by a normal coordinate W_σ the vector $d_\sigma(\mathbf{r})$ gives a dipole moment of the corresponding mode.

So far we have diagonalized the dynamical matrix with respect to the cell indexes only. This means that the normal coordinates, $W_\sigma(\mathbf{r})$, as well as the matrix $c_{\alpha\sigma}^i(\mathbf{r})$ retain dependence upon a position, \mathbf{r} , and reflect only the local structure of a particular cell, which can fluctuate from cell to cell.

The local symmetry of a cell actually determines the coupling parameter defined by Eq. (4) through the matrix $c_{\alpha\sigma}^i(\mathbf{r})$. If the symmetry allows for a central symmetry mode, this parameter is equal to zero for this mode. If the local symmetry is randomly distorted, a coupling parameter, which is different in its value and its direction for different cells, arises. By this means we can consider components of the vector \mathbf{d} as random functions. We assume that these functions have the following properties,

$$\begin{aligned} \langle d_i \rangle &= 0, \\ \langle d_i(\mathbf{r}) d_j(\mathbf{r}') \rangle &= \langle d^2 \rangle \delta_{ij} K(\mathbf{r} - \mathbf{r}'). \end{aligned} \quad (5)$$

The first of these equations reflects the assumption that we are dealing with a mode which would be inactive in an ideal structure. The second one suppose that the local distortions of the structure are isotropic on average and may be described by one correlation function $K(\mathbf{r} - \mathbf{r}')$. This function can be characterized by a correlation radius, r_c , which determines the distance scale at which correlations between two points exist. The value $(\langle d^2 \rangle)^{1/2}$ occurring in Eq. (5) is the r.m.s. of the fluctuations and may be considered as an effective coupling parameter.

Eq. (3) must be complemented by the Maxwell equation for an electromagnetic field. It may be written as

$$\frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} + \nabla(\nabla \cdot \mathbf{E}) - \Delta \mathbf{E} = -\frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2}, \quad (6)$$

where \mathbf{P} is the density of the polarization. It can be presented in the form

$$\mathbf{P}^i(\mathbf{r}) = \frac{1}{a^3} \sum_{\sigma} d_{\sigma}^i(\mathbf{r}) W_{\sigma} + \frac{\epsilon_{\infty} - 1}{4\pi} \mathbf{E}^i, \quad (7)$$

where a is the mean size of a cell. The first term in this equation describes the ionic contribution to the polarization expressed in terms of the normal coordinates, W_{σ} . The second term takes into account the electronic part of the polarization, where ϵ_{∞} is the high-frequency dielectric permeability. Using Eq. (7) and the Maxwell equation (6) we arrive at a system of equations describing coupled states of an electromagnetic field and lattice vibrations

$$\ddot{W}_{\sigma}^i(\mathbf{r}) + \sum_{r'} \hat{D}_{\sigma}^{ij}(\mathbf{r}, \mathbf{r}') W_{\sigma}^j(\mathbf{r}') = d_{\sigma}^i(\mathbf{r}) E^i(\mathbf{r}), \quad (8)$$

$$\frac{\partial^2 E^i}{\partial t^2} + \tilde{c}^2 \nabla(\nabla E^i) - \tilde{c}^2 \Delta E^i = -\frac{4\pi}{\epsilon_{\infty} a^3} \sum_{\sigma} d_{\sigma}^i \ddot{W}_{\sigma}, \quad (9)$$

where $\tilde{c} = c/\epsilon_{\infty}$. These equations implicitly describe the interaction between electromagnetic waves and phonons. Unlike previous studies we take into account the possible randomness of both the coupling parameter and the phonon dynamical matrix, but we neglect the fluctuations of the electronic part of the dielectric constant.

3. Dispersion laws, attenuation rates, and susceptibility

In this section we briefly discuss the characteristics that describe the properties of the average amplitude of the waves. Doing so we only allow for randomness of the coupling parameter and neglect the fluctuations of the parameters, which describe the inner properties of the phonon and photon subsystems.

We assume that only one phonon mode is coupled with an electromagnetic wave, and its coupling parameter satisfies conditions (5). Therefore, we omit the mode index for phonon variables. Under these assumptions in the first essential order of the perturbation theory one can obtain the following expressions for the averaged Green functions of transversal photons, $G_{\text{em}}(k, \omega)$, and phonons, $G_{\text{vib}}(k, \omega)$,

$$G_{\text{em}}(k, \omega) = \left(\omega^2 - \omega_k^2 - \Lambda^2 \omega^2 \int \frac{S(\mathbf{k} - \mathbf{k}_1)}{\omega^2 - \epsilon^2(\mathbf{k}_1)} d^3 k_1 \right)^{-1}, \quad (10)$$

$$G_{\text{vib}}(k, \omega) = \left(\omega^2 - \epsilon^2(\mathbf{k}) - \Lambda^2 \omega^2 \int \frac{S(\mathbf{k} - \mathbf{k}_1)}{\omega^2 - \omega(\mathbf{k}_1^2)} d^3 k_1 \right)^{-1}. \quad (11)$$

The poles of these Green functions determine the corresponding dispersion laws and damping coefficients. In the case of usual polaritons with a deterministic interaction the dispersion equations derived from Green functions (10) and (11) coincide. The situation changes when the coupling parameter fluctuates. Due to fluctuations Eq. (10) and Eq. (11) refer to different physical systems. The first of them describes a coherent electromagnetic wave coupled with scattered phonons. At the same time Eq. (11) describes states of a coherent phonon coupled with a scattered electromagnetic field.

From the experimental point of view this means that different results will be obtained depending on the method used to explore the system. If one uses electromagnetic waves to excite coupled excitations, Eq. (10) is to be used. However, if the vibrational subsystem is originally excited (by means of a neutron, for instance), then a dispersion equation following from a phonon averaged Green function will describe the situation adequately.

For electromagnetic waves Eq. (10) leads to the dispersion equation,

$$\omega^2 - \omega_k^2 - \Lambda^2 \omega^2 \int \frac{S(\mathbf{k} - \mathbf{k}_1)}{\omega^2 - \epsilon^2(\mathbf{k}_1)} d^3 k_1 = 0. \quad (12)$$

The function $S(\mathbf{k})$ is a Fourier transform of the correlation function (5), $\epsilon(\mathbf{k})$ is the initial phonon

dispersion law that we need in the long-wave limit only,

$$\varepsilon^2(\mathbf{k}) = \omega_0^2 - v^2 k^2. \quad (13)$$

The function $\omega(k) = \tilde{c}k$ describes the initial dispersion law of electromagnetic waves and

$$\Lambda^2 = 4\pi d^2 / \epsilon_x a^3.$$

We are primarily interested in the properties of electromagnetic waves. Therefore, we do not discuss the dispersion equations for phonons and for the longitudinal component of the electromagnetic field. These equations will be discussed in full elsewhere.

According to the results obtained in Ref. [5] a critical value, Λ_{cr} , of the coupling parameter, Λ , exists. Λ_{cr} separates different kinds of solutions of the dispersion equation (12). For $\Lambda < \Lambda_{cr}$ one can only find one solution corresponding to a well-defined extended mode. This solution remains continuous at the resonant point and it approaches the dispersion law of initial light far away from resonance. In the case when $\Lambda > \Lambda_{cr}$ the dispersion curve breaks down at the resonance point to form a gap,

$$\Delta = \sqrt{\Lambda^2 - \Lambda_{cr}^2}$$

In addition, a new branch of well-defined excitations appears in the vicinity of the crossing point. Both branches correspond to the same mode: a coherent electromagnetic wave coupled with scattered phonons. The dispersion curve for $\Lambda > \Lambda_{cr}$, shown in Fig. 1, describes new kinds of compounded excitations, which may be called “disorder-induced polaritons”. A more detailed general description of these excitations is given in Ref. [5].

The critical value of the coupling parameter can be obtained in the form

$$\Lambda_{cr} = \frac{v^2 k_c^2}{\tilde{c}k_r}, \quad (14)$$

where $k_c \sim r_c^{-1}$ is the correlation wave number of the inhomogeneities of the coupling parameter, k_r is the wave number corresponding to the crossing point. When deriving Eq. (14) we have used the fact that the correlation radius r_c is usually much smaller than the polariton wave lengths. In order to obtain a

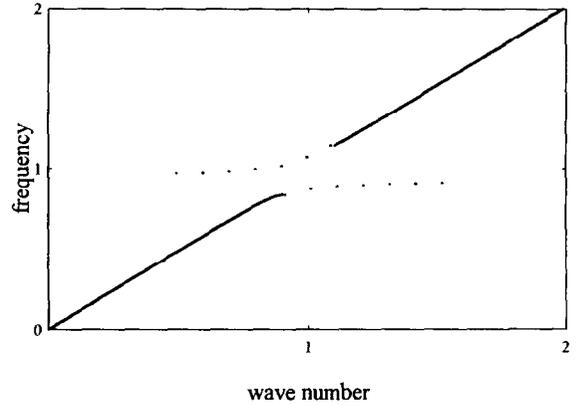


Fig. 1. Dispersion curve of a disorder-induced polariton. The solid line shows the branch that approaches the original dispersion curve of the electromagnetic wave far from the crossing point. The dotted line shows a new branch that arises due to the interaction between coherent electromagnetic waves and scattered phonons. This branch becomes ill defined in the off-resonance region. Values of the frequencies and wave numbers are normalized by ω_0 and ω_0 / \tilde{c} , respectively.

numerical estimate of the critical parameter one can notice that the parameter v in the phonon initial dispersion law is of the same order of magnitude as the velocity of acoustic phonons, $\sim 10^5$ cm/c, the parameters k_r and k_c may be taken to be equal to 10^3 cm $^{-1}$ and 10^7 cm $^{-1}$, respectively. The latter value corresponds to the correlation radius ~ 10 Å. For these values of the parameters the value of Λ_{cr} is of the order of 10^{11} c $^{-1}$. The order of magnitude of the coupling parameter in ideal ionic crystals is $\Lambda_{id} \sim 10^{13}$ c $^{-1}$. This value may be obtained from a simple estimate,

$$\Lambda_{id} \sim \left(\frac{4\pi e_{eff}^2 n}{m} \right)^{1/2}$$

(see, for example, Ref. [10]). e_{eff} in this expression is the effective charge of the corresponding mode. In the case under consideration this effective charge should vanish for an ideal configuration of a cell. However, in the presence of distortion e_{eff} can be estimated as $e_{eff} \sim e\Delta a/a$, where $\Delta a/a$ measures the degree of the distortion. In this case the parameter Λ is of the order of $\sqrt{\langle (\Delta a/a)^2 \rangle} \Lambda_{id}$. The value of $\sqrt{\langle (\Delta a/a)^2 \rangle}$ is the r.m.s. of the fluctuations of the structure. From diffraction experiments this value

is known to be $\sim 10^{-1}$ (see, for example, Ref. [9]). Therefore, the value of the coupling parameter is of one order of magnitude larger than the critical value Λ_{cr} . This estimate justifies some optimism regarding the possibility of the observation the disorder-induced polariton states.

To be more certain about this conclusion we have to compare the effectiveness of other scattering and absorption channels that may be present in the system with the considered one. The critical value of the coupling parameter, Λ_{cr} , is obtained by comparison of the strength of the interaction with the strength of the relaxation. In fact, Λ_{cr} is exactly equal to the relaxation rate due to scattering at the crossing point. The disorder-induced polaritons appear when the interaction dominates over the relaxation. The corresponding absorption and scattering rates are to be added to Λ_{cr} to give the total value of the relaxation rate and, hence, the critical value of the coupling parameters. Among other relaxation mechanisms phonon absorption and scattering are the most important ones.

The absorption rate for optical phonons in crystals is approximately two orders of magnitude smaller than the corresponding frequencies (see, e.g., Ref. [11]). This coincides with our estimate of Λ_{cr} . The scattering of phonons in disordered materials may occur due to fluctuations of the density or force parameters. The first mechanism is much stronger and the corresponding rate can be estimated as [12] $(\Delta a/a)^2(\omega_0^3/v^2k_c^2)(k/k_c)$. The value of this rate is of the order of $10^{11}c^{-1}$, which is of the same order of magnitude as the other relaxation mechanisms discussed above. So, we can conclude that other relaxation mechanisms should not considerably disturb DICR in this system. Therefore, disorder-induced polaritons can be, in principle, observed in disordered dielectrics. They are to be searched in the frequency region of phonons inactive in the corresponding ordered materials.

We should note, however, that the dispersion curve, shown in Fig. 1, has been obtained under the assumption that the wave number is real, while the frequency is allowed to have an imaginary part. Realization of this assumption requires a certain way of investigation of the system. To observe this dispersion behavior one has to use an experimental method, which allows one to fix the wave number of

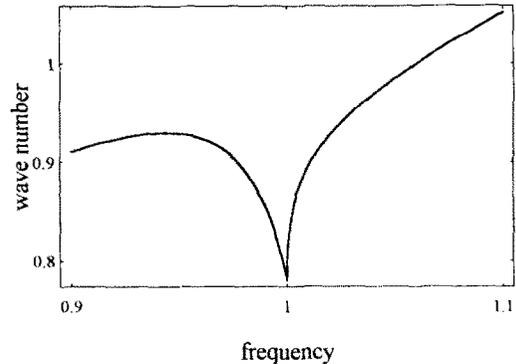


Fig. 2. Dependence of the wave number on the frequency when the frequency is a real value, while the wave number is allowed to have an imaginary part. The graph has the same units as in Fig. 1.

the excitation. This, for instance, can be Raman scattering. Other methods, where the frequency of the wave is under control and the dependence $k(\omega)$ is observed, could lead to different results. This difference is typical for systems with dispersion and attenuation. As applied to systems with DICR this was considered for the first time in Ref. [8] for magnetoelastic resonance in amorphous magnets. In the situation considered in the present paper the dependence $k(\omega)$ for the case $\Lambda > \Lambda_{cr}$ is shown in Fig. 2. Instead of the gap one can see a region of anomalous dispersion.

The coherent (average) amplitude of the disorder-induced polaritons exponentially decreases with time due to generating incoherent phonons. The relaxation time, which describes this decrease, is to be derived from the imaginary part of the dispersion curve (12). Both branches of the dispersion curve are characterized by their own relaxation times. The relaxation time corresponding to the solid line in Fig. 1 has a minimum at the resonance point. The minimum value is equal to Λ_{cr}^{-1} . On the contrary, the relaxation time of the second branch (dashed line in Fig. 1) has a maximum at the resonance. The value of this maximum is also equal to Λ_{cr}^{-1} . Therefore, these two branches at the resonance cannot be distinguished according to their relaxation properties. When moving away from the resonance the relaxation time of the first branch increases, while the relaxation time of the second one decreases. The second branch becomes eventually poorly defined far enough from the resonance.

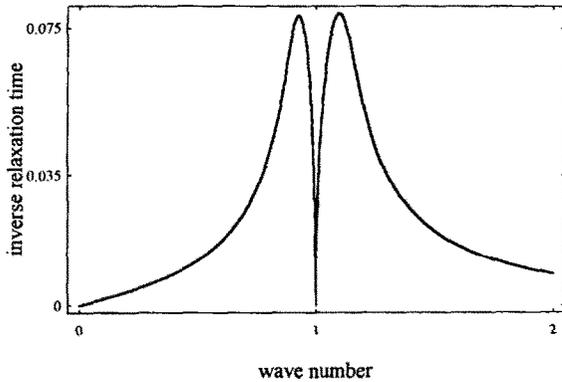


Fig. 3. Dependence of the inverse relaxation time of the coherent electromagnetic component on the wave number. The left curve corresponds to the case of negative dispersion, the right curve describes the situation when the dispersion of phonons is positive. The units are the same as in Fig. 1.

In Fig. 3 the shape of the dependence of the inverse relaxation time on the wave number is shown for the branch depicted by the solid line. The relaxation occurs only for $k < \omega_0/\tilde{c}$. It vanishes as $\sqrt{\tilde{c}k - \omega_0}$ when $k \rightarrow \omega_0/\tilde{c}$. The curve approaches the resonant point as $|k - k_r|$. If one compares the relaxation properties obtained in this paper to those derived in Refs. [5,6], it may be noted that these curves look almost like mirror images of each other. This sharp difference appears because of the sign in front of the v^2k^2 -term in the original dispersion law for optical phonons. We have a negative sign in Eq. (13), while a positive one was assumed in Ref. [5]. In the case of negative dispersion all scattering events occur at $\omega < \omega_0$, and, in contrast, for positive dispersion scattering occurs for $\omega > \omega_0$ only. This fact results in the “mirror reflection” of the relaxation curve with respect to the line corresponding to $\omega = \omega_0$, when the phonon dispersion is switched from positive to negative.

4. Conclusion

In this paper we derive general equations, which describe the interaction between light and optical phonons in disordered materials. Unlike previous studies of this problem we take into account the

randomness of the interaction parameter. To emphasize the importance of fluctuations of the coupling parameter we apply the equations derived to a special case of a zero-mean random coupling parameter. This case has a principal theoretical interest because a new kind of excitation with unusual properties arises (Ref. [5]). Besides, the developed model describes a practically possible situation of disorder-induced interaction between light and phonons, which would be inactive in an ideal crystal. Estimates support the idea that disorder-induced polaritons can appear in disordered dielectrics. In order to observe them one should explore the frequency region in which excitations inactive in ordered materials exist.

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