PHYSICS AND CHEMISTRY
OF SOLID STATE
V. 27, No. 1 (2026) pp. 62-68

Section: Physics

DOI: 10.15330/pcss.27.1.62-68

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ФІЗИКА І ХІМІЯ ТВЕРДОГО ТІЛА
Т. 27, № 1 (2026) С. 62-68

Фізико-математичні науки

PACS: 71.36.+c;

ISSN 1729-4428 (Print)
ISSN 2309-8589 (Online)

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The photophysical properties of strongly coupled hybridized light-matter states in a classical physics perspective

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The Lorentz oscillator model is applied to a strongly coupled hybridized light-matter state. It is noted that the real part of the index undergoes rapid change in the area of the so-called “dark states.” This property of hybridized light-matter states could allow them to function as unorthodox optical materials in which their absorptive and refractive properties are selectively manipulated. In this manuscript, basic properties of hybrid light-matter states, also referred to as cavity polaritons, are reviewed. The Lorentz oscillator model is then applied to two types of light-matter states, one formed from coupling between a single exciton and a single cavity photon, and one formed from coupling two excitons to a single cavity photon. Furthermore, the simple harmonic oscillator model is used to estimate the spring constant of a diatomic molecule under strong light-matter coupling conditions. It is found that the spring constant is directly related to the photonic character of the system, with a higher photonic character resulting in a smaller spring constant.

Keywords: Polaritons, Molecular vibration, Lorentz, Oscillator.

Received 20 May 2025; Accepted 28 January 2026.

Introduction

The interaction of electromagnetic radiation (light) with matter is a fundamental process required to sustain complex life [1]. For example, the conversion of carbon dioxide and water into complex molecules via photosynthesis is hypothesized to have facilitated the evolution of multicellular organisms [1]. There are two limits to light-matter interaction. The “weak” coupling regime in which the interaction of light and matter results in a change in the radiative properties of the states involved, but does not fundamentally alter the physical properties of either state, and the “strong” coupling regime, in which the physical properties of the states, such as the potential energy surface, are altered [2,3]. Strong light-matter coupling occurs when an electromagnetic wave of a certain frequency interacts with a chromophore which resonates at the same frequency. If the chromophore is unable to dissipate the energy faster than it is being exchanged with the photon, strong light-matter coupling may occur [2]. The energy of light coupling to the molecular transition can vary from coupling to an excitonic transition involving a change of electronic states, to strong-light matter coupling of a molecular vibration and a lower energy infrared photon [2]. In order to achieve such conditions, the photon is typically trapped in a Fabry-Pérot cavity, as seen in Fig. 1A. Strong light-matter coupling between two states (one photonic and one excitonic) will result in the appearance of two polaritonic states, as seen in Fig. 2B and described in Eq. (1) [2]. As evident from Eq. (1), strong coupling hybridizes the photonic and excitonic states, imparting the characteristics of one unto the other, hence they are often simply referred to as hybrid light-matter states. The formation of “dark states” will also occur, which will occupy an area intermediate of the polaritons. The dark states are a superposition of molecular excitons of the chromophore with little optical character. The optical properties of these polariton states have been of some interest to scientists due to their possible uses such as lasing applications, as well as being used as novel tools to alter chemical reactions [4,5].

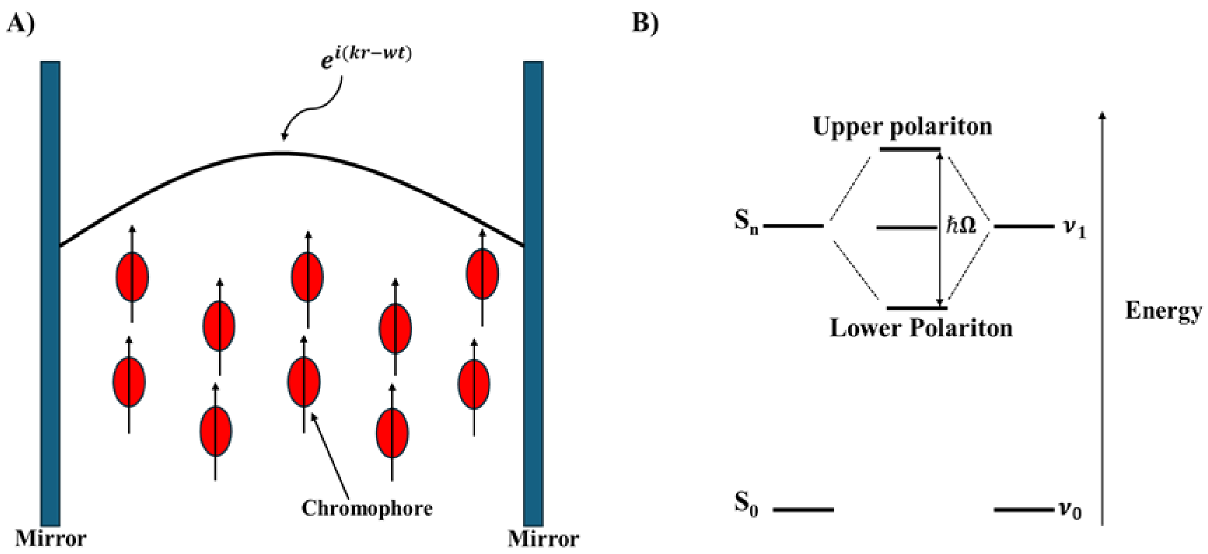

Fig. 1. A) A diagram of a model cavity polariton structure showing an electromagnetic wave being trapped inside a Fabry-Pérot cavity. The light wave is able to interact with a chromophore inside the cavity as it is reflected. B) The energy diagram of a polariton structure. Note that this is similar to the concepts of molecular orbital theory in which two molecular orbitals are created via the interaction of two atomic orbitals.

$$|{}^{UP}/{}_{LP}\rangle = \frac{1}{\sqrt{2}}[|e\rangle_e|0\rangle_c \pm |g\rangle_e|1\rangle_c] \qquad (1)$$

The two polaritonic states are denoted as the upper polariton (UP) and the lower polariton (LP), as seen in Fig. 2B. The two states are separated by an energy referred to as the Rabi splitting ($\hbar\Omega$) [2]. The strong light-matter coupling condition is satisfied by Eq. (2), where $\hbar\gamma_{ex}^2$ and $\hbar\gamma_{cav}^2$ refer to the full width half maximum (FWHM) of the exciton and the cavity photon respectively [6]. Note that the number of polaritonic states formed is equal to the number of excitonic and photonic states participating in their formation. For simplicity, in Fig. 2B only one of each state is involved. However, strong light-matter coupling involving multiple excitonic states coupled to a single cavity photon have been recorded [7,8]. In such a case, the appearance of additional “middle” polariton states are expected.

$$\hbar\Omega > \sqrt{{(\hbar\gamma_{ex}^2 + \hbar\gamma_{cav}^2)}/{2}} \qquad (2)$$

Based on the description in Eq. (1), it is clear that the wave function of a polariton state is a true hybrid of a photon and an exciton. An oscillator involving a single photon and a single cavity photon can be described by a two-level Hamiltonian, as seen in Eq. (3), in which the cross-diagonal interaction term in the matrix (*V)* can be estimated by Rabi splitting [9]. Remember, to enter the “strong coupling regime” this interaction term must be greater than the FWHM of the excitonic and photonic peaks involved. Solving for the eigenvalues of the two-level Hamiltonian produces two solutions, as seen in Eqs. (4) & (5), the energies of the UP and LP states are obtained [10]. Note that the polariton states are dispersive, the angle at which the Rabi splitting is maximized will be the resonance point of the polariton states, with both UP and LP possessing an equal amount of excitonic and photonic character [10]. It should also be noted that solving for the eigenvectors of Eq. (3) will result in determining the Hopfield coefficients of each polariton state, which describe their photonic and excitonic characteristics [11]. The Hopfield coefficients displaying the “photon-like” beahvior of each state can be estimated by Eqs. (6), and (7) [12]. Finally, the imaginary part of Eq. (3) contains information regarding the FWHM of the polaritonic states. It should be clear based on Eqs. (4) and (5) that the resulting eqigenvalues for the polariton states acquire a “light-like” character, resulting in the mass and lifetime of the states needing to be corrected for by the Hopfield coefficients [9]. To minimize these possible complications the cavity mode is typically made to have a FWHM similar to that of the exciton that is being coupled to, and experiments are typically performed near resonance, where the photonic and excitonic characters are equal [2].

$$\begin{bmatrix} E_{ex} + i\sigma_{ex} & V \\ V & E_{ph} + i\sigma_{ph} \end{bmatrix} \begin{bmatrix} \alpha \\ \beta \end{bmatrix} = \epsilon \begin{bmatrix} \alpha \\ \beta \end{bmatrix} \qquad (3)$$

$$E_{UP(\theta)} = \frac{E_{ph(\theta)} + i\sigma_{ph} + E_{ex} + i\sigma_{ex}}{2} + \frac{1}{2}\sqrt{\left[(E_{ex} + i\sigma_{ex}) - (E_{ph(\theta)} - i\sigma_{ph})\right]^2 + 4V^2} \qquad (4)$$

$$E_{LP(\theta)} = \frac{E_{ph(\theta)}+i\sigma_{ph}+E_{ex}+i\sigma_{ex}}{2} - \frac{1}{2}\sqrt{\left[(E_{ex}+i\sigma_{ex})-(E_{ph(\theta)}-i\sigma_{ph})\right]^2+4V^2} \quad (5)$$

$$c_{ph(lp)} = \frac{V^2}{V^2+(E_{LP(\theta)}-E_{ph(\theta)})^2} \quad (6)$$

$$c_{ph(up)} = \frac{V^2}{V^2+(E_{UP(\theta)}-E_{ph(\theta)})^2} \quad (7)$$

While Eq. (3) does well in describing the energy and dispersive behavior of polariton states, it is possible to closely approximate their behavior using classical optical methods. In particular, when describing the behavior of light inside a Fabry-Pérot cavity the wave-transfer matrix can be evoked [13,14]. The wave-transfer matrix, seen in Eq. (8) relies on using the Fresnel equations for reflectance and transmittance across each boundary involved in constructing the Fabry-Pérot cavity to estimate the final transmittance (or reflectance). Absorption can be simulated within the matrix by adjusting the attenuation coefficient within the layer that possesses the chromophore. This manuscript takes advantage of the fact that the behavior of light inside of a cavity is well approximated by classical methods, utilizing the Lorentz oscillator model to plot out the refractive index of the polariton states.

$$M = \begin{bmatrix} t_{12}t_{21}-r_{12}r_{21} & \frac{r_{21}}{t_{12}} \\ \frac{r_{12}}{t_{21}} & \frac{1}{t_{12}} \end{bmatrix} \quad (8)$$

While the derivation of the Lorentz oscillator model is beyond the scope of this manuscript, the basic principles of it will be explained. In summary, the electromagnetic field is modeled as, $e^{i(kr-wt)}$. When an electromagnetic field interacts with matter two assumptions are made, 1) the charges in the material will possess some inherent oscillating frequency and 2) the charges will resist the electromagnetic field in the opposite direction, in essence the model treats the charges within the material interacting with the electromagnetic field as small springs [15-17]. Already evident from this simple treatment, it should appear clear that due to the complex nature of the electromagnetic field, the Lorentz oscillator model will contain a real, and an imaginary component. The former representing refractance, while the latter attenuation. Note that the magnetic component is typically ignored due to it being exceedingly small.

I. Results and Discussion

1.1. Dielectric properties of hybridized light-matter states

$$1+\left(\frac{\omega_P}{\omega_0^2-i\gamma\omega-\omega^2}\right)_{UP}+\left(\frac{\omega_P}{\omega_0^2-i\gamma\omega-\omega^2}\right)_{LP} \quad (9)$$

The Lorentz oscillator model utilized to model the behavior of the complex index of refraction is provided in Eq. (9). The UP and LP notation denotes the upper and lower polariton. The total index can be modeled as a sum of each individual component [17]. In Eq. (9) ω_P refers to the plasma frequency, which was set at ω_P=1000γ. The excitonic resonance frequency,ω_0^2, was set to 688.9 THZ for the LP and 724.8 THz for the UP. The damping component γ was set at 7.451 THz. These values are estimates based on a polariton model formed by strong light-matter coupling between a cavity photon and a Zinc tetraphenyl porphyrin (ZnTPP) molecule described in previous literature [5]. The results of Eq. (9) are plotted in Fig. 2A. In Fig. 2A, the imaginary portion of the index represents attenuation of the electromagnetic field as it passes through a material. The real portion of the index represents the ratio of the phase velocity of the electromagnetic wave traveling through the medium to

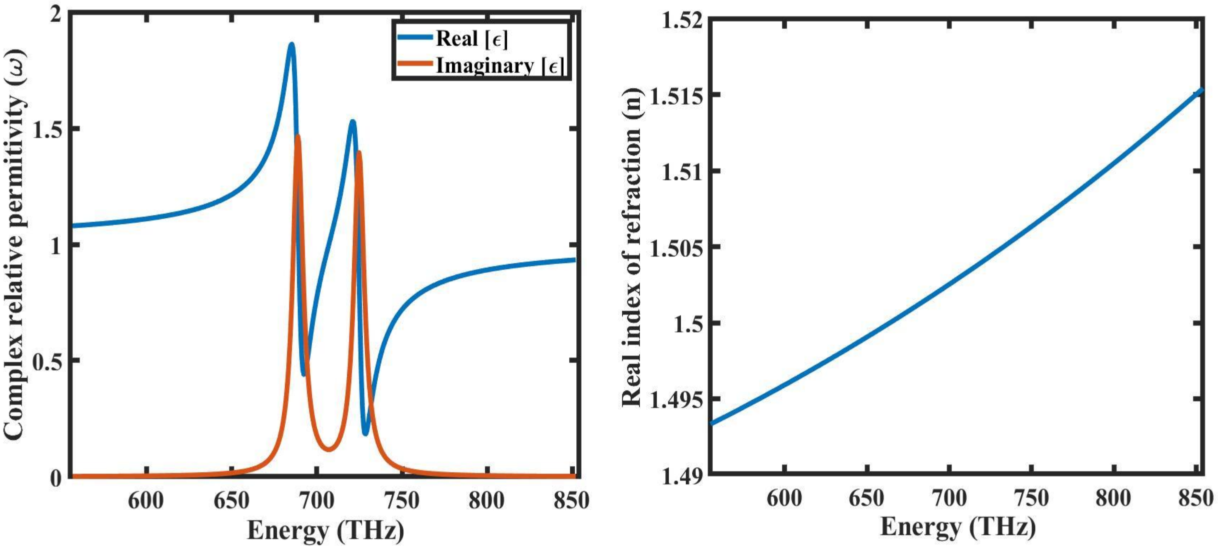

Fig. 2. A) The real and imaginary parts of the index of refraction plotted for a hybrid light-matter state. B) The refractive index of poly methyl methacrylate (PMMA), as calculated using the Sellmeier equation.

that of an electromagnetic wave in vacuum [17]. The simple Lorentz oscillator model demonstrates the large rate of change in phase velocity experienced by the real portion of the refractive index near the dark states (706.9 THz).

The Lorentz oscillator model is further applied to a system that contains two excitonic and one photonic state, requiring the modeling of three polariton states, the UP, LP, and a middle polariton (MP), as seen in Fig. 3. In order to accomplish this, Eq. (9) is simply expanded to accommodate an additional polariton energy level, as seen in Eq. (10).

$$1 + \left(\frac{\omega_P}{\omega_0^2 - i\gamma\omega - \omega^2}\right)_{UP} + \left(\frac{\omega_P}{\omega_0^2 - i\gamma\omega - \omega^2}\right)_{LP} + \left(\frac{\omega_P}{\omega_0^2 - i\gamma\omega - \omega^2}\right)_{MP} \tag{10}$$

In Eq. (10) the γ component for the LP was set at 68.9 THz, the LP at 26.3, while the MP was the average of the two values. The resonance frequency ω_0^2 was set at 773.7 for the LP, 922.1 THz for the UP and the average of the two values for the MP. These values were based on work obtained by Forrest *et al.* [7]. In the Forrest model the two excitonic states coupling to the cavity polariton are effectively degenerate, signifying that the middle polariton state, in effect, occupies an energy level near the original excitonic states. Despite this, as seen in Fig. 3, the middle polariton's complex permittivity does not mirror what one would expect from a prototypical excitonic state. Note that while simplistic, at resonance the MP will have an equal amount of contribution from both excitonic states [18]. Therefore, averaging the two excitonic contributions should provide for an estimate of the FWHM of the MP. A strongly light-coupled state formed from two excitons and a single cavity photon results in the real portion of the refractive index undergoing considerable change in the area of the middle polariton, as seen in Fig. 3.

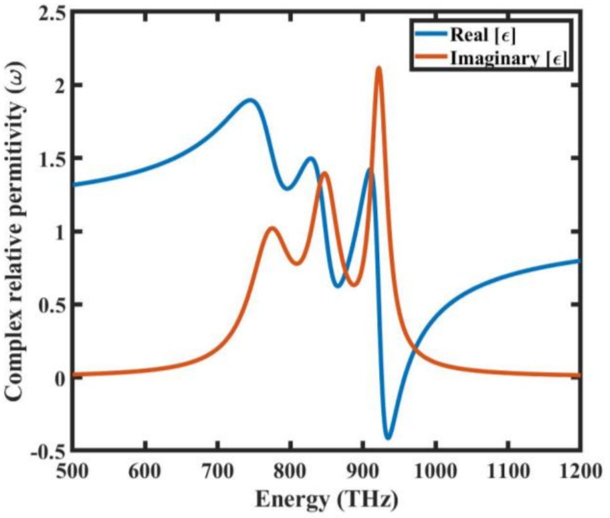

Fig. 3. The real and imaginary parts of the index of refraction plotted for a hybrid light-matter state formed from two excitons and a single cavity photon.

$$n^2 - 1 = \frac{0.99654\lambda^2}{\lambda^2 - 0.00787} + \frac{0.18964\lambda^2}{\lambda^2 - 0.02191} + \frac{0.0.00411\lambda^2}{\lambda^2 - 3.85727} \tag{11}$$

The refractive index is dependent on temperature and wavelength, however, the change in the index is often small, or inherent to the physical structure of the material [15,19]. This is visualized in Fig. 2B where the Sellmeier equation is used to estimate the real part of the refractive index for poly methyl methacrylate (PMMA). This polymeric material has been used as a medium to form polaritons due to the precision to which the coating can be made, the Sellmeier equation for PMMA is provided in Eq. (11) [5,20]. Creating materials whose index can be selectively controlled has been of some interest to scientists [21]. One such path is via the use of acousto-optic devices, in which a sound (acoustic) wave is used to generate a gradient within an optical material. In turn, the electromagnetic field traveling through such a gradient will experience a different refractive index then one without a gradient. In summary, the new transmittance must be calculated using the wave-transfer matrix seen in Eq. (8). One limitation of using acousto-optic modulators is the need to carefully focus the light unto the crystal in order to get sufficient modulation, while at the same time requiring a source of acoustic waves [22]. Another way the index of refraction of a material can be manipulated is via the photorefractive effect [20]. Unlike sound waves the photorefractive effect relies on utilizing light to manipulate the refractive index of a material. The rapidly changing index of refraction in polaritonic media between the UP and LP states would appear to make them a suitable candidate for a modular optical material. Moreover, unlike acoustic or photorefractive materials currently in use which may require material restrictions, polaritons have been formed from a wide range of materials, such as quantum well semiconductors, as well as molecular and vibronic excitations [23,24]. Finally, polaritons do not require an outside energy source, that is, once strong light-matter hybridization is achieved, a presence of a single photon trapped in the cavity maintains the polariton states [25]. This property is illustrated by Eq. (12) which defines Rabi splitting [25-28]. From Eq. (12), Rabi splitting is based on the resonance energy (ħω), the permittivity of vacuum and the volume of the electromagnetic mode ($\epsilon_0 v$) as well as the dipole moment of the chromophores involved in strong light-matter coupling (*d*). Also, due to the $n_{photon} + 1$ factor, a photon will always be present within the cavity upon polariton formation.

$$\hbar\Omega = 2d\left(\frac{\hbar\omega}{2\epsilon_0 v}\right)^{1/2}\left(n_{photon} + 1\right)^{1/2} \tag{12}$$

1.2. Harmonic oscillator model involving hybridized light-matter states

It is well established within the chemical sciences that molecular vibrations can be approximated using the harmonic oscillator model, as shown in Eq. 13 [29]. In Eq. 13 the potential energy *V* is estimated in terms of the spring constant (*k*) and the amount a hypothetical spring distorts (x). The spring constant can itself be defined in terms of a reduced mass of an object as well as the vibration in question, as seen in Eq. (14) [12,29]. When this basic, classical treatment of a hormonic spring is applied to a molecule, it does well in estimating its vibrational behavior. In particular, the vibrational frequency *nu* can be directly estimated in a diatomic molecular system based off Eq. (14): $2\pi v = \sqrt{k/m_r}$ [30]. While the simple harmonic oscillator is a powerful tool to estimate the frequencies of diatomic molecules, an astute observer will immediately note that during light-matter strong coupling a vibrational level will assume a "light-like" characteristic. Therefore, the question arises, since light is by definition, massless, how would the vibrations of a hybridized light-matter state be treated within the harmonic oscillator model?

$$V = \frac{1}{2}kx^2 \tag{13}$$

$$k = \omega^2 m_r \tag{14}$$

$$E = mc^2 \tag{15}$$

$$m = \frac{E}{c^2} = \frac{hv}{c^2} \tag{16}$$

$$m_r = \frac{(c_{ph}m_1+c_{ex}m_2)_1(c_{ph}m_1+c_{ex}m_2)_2}{(c_{ph}m_1+c_{ex}m_2)_1+(c_{ph}m_1+c_{ex}m_2)_2} \tag{17}$$

Luckily, Poincaré and Einstein have already discussed this topic in great detail, with the famous Eqs. (15-16) providing the relativistic mass of light [31,32]. Utilizing the formalism in Eqs. (13-16) it is possible to estimate the spring constant of a vibrating diatomic molecule, as well as a "massless" diatomic hybrid light-matter state. Furthermore, recall that the Hopfield coefficients determine the "light-like" characteristic of a polariton state, as described earlier in Eqs. (6-7). By plugging the photonic Hopfield coefficient c_{ph} into the formula for reduced mass, as seen in Eq. (17), it is then possible to estimate the spring constant of hybridized light matter states at various coupling levels. In Eq. (17) the excitonic fraction c_{ex} can be estimated as: 1-c_{ph} [12]. Also note that since the exciton and photon are hybridized under strong light-matter coupling conditions, it is necessary to treat the two masses as a mixture of both states, where m_1 is the "photon" mass as defined by Eq. (16) and m_2 is the molecular mass. The spring constant of a simple ethylene molecule is estimated at Hopfield coefficients of 1, 0.9, 0.75, 0.5, 0.25, and 0 in Fig. 4. When c_{ph} is equal to 0 Eq. (17) simplifies to the standard reduced mass formula where $m_r=(m_1m_2)/(m_1+m_2)$. In the estimation, vibrational energies between 0.05 and 0.5 eV, or approximately 400-4000 cm^{-1}, were considered as they represent the most commonly used FTIR spectral range, as well as because construction of cavity polaritons has been demonstrated to be feasible within this energy range [30,33]. A Hopfield coefficient where c_{ph}=0 represents a completely excitonic system with no "light-like" character, while 1 represents a fully "light-like" system with no excitonic character. Note that the spring constant is considerably lower for the fully photonic system at all energy levels. Intuitively, the spring constant is directly related to the shape of a molecule's potential energy surface, as seen in Eq. (13) [30]. A molecule that is difficult to displace from equilibrium, will have a steep potential energy surface and a high spring constant. On the contrary, a molecule which is easily displaced will have a shallow potential energy surface with a small spring constant. As of this paper's writing, direct observations of the potential surface energy of polariton states have not be made, however, it is estimated that a "light-like" polaritonic states will be highly dispersive, with the potential energy surface modified for easier chemical reactions [34,35]. Indeed, the results of this simple calculation seem to suggest that as the photonic character of a polaritonic state increases, the spring constant decreases, which may result in a flattened geometry of the potential energy surface of a diatomic molecule.

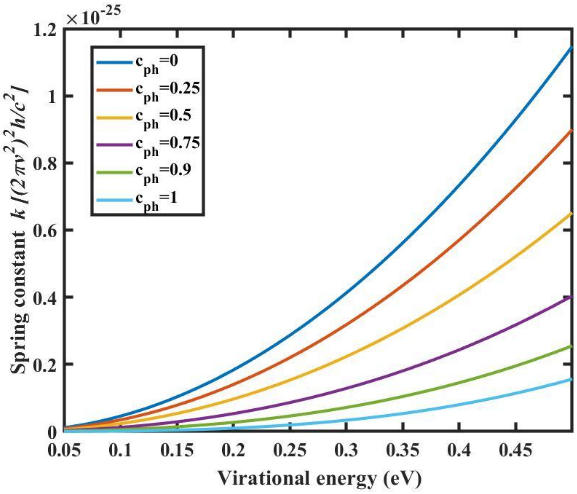

Fig. 4. Estimated spring constant of an ethylene molecule under various strong light-matter coupling conditions, with a photonic Hopfield coefficient c_{ph} of 0, 0.25, 0.5, 0.75, 0.9 and 1.

Conclusion

The complex index of refraction of two different materials strongly coupling to a cavity photon was modeled using the classical Lorentz oscillator model. The model shows a rapid change of the real index of refraction between the polariton states. This property of dark states may allow polaritons to function as a new type of photorefractive materials. For example, polaritons can be incorporated into coatings in order to alter the complex refractive index of the material, allowing the coating to be highly absorbent in areas it would typically lack such a characteristic, or selectively increase transparency in another area of the spectrum. While an obvious limitation of such a material would be the need to construct two mirrors to maintain a cavity photon, recent work has been conducted in which polaritons are created by coupling to phonon resonances, which do not require an optical cavity structure [27]. Moreover, the middle polariton, while occupying an energy state that is nearly degenerate to that of the original excitons, appears to have optical properties markedly different than expected from a typical excitonic state, in particular, the real part of the index undergoes rapid change in the area of the middle polariton as it moves between the LP to the UP. Finally, it should be mentioned that because the matrix which holds the chromophores that couple with the cavity photons to form polariton states would expand and contract with temperature changes, the strength of the optical response could be controlled by manipulating the coating's thickness, allowing for a selective manipulation of refractance and absorbance [28].

Furthermore, the simple harmonic oscillator model was used to model the spring constant of a diatomic molecule strongly coupled to a cavity photon. It was estimated that the spring constant decreases as the photonic character of a system increases. This, in turn, suggests a flattening of the potential energy surface. Because the shape of the potential energy surface is directly responsible for the reactivity of a molecule, it is demonstrated via a classical mathematical model that a hybrid light-matter interactions are likely to result in changes to a molecule's reactivity. It is hopeful that this work can further aid scientists and engineers in creating optical devices involving polariton states. Moreover, while a true picture of light-matter interaction requires quantum treatment, optical materials such as distributed Bragg reflectors and Fabry-Pérot cavity, which are often integral parts used to form polariton states, are often estimated using classical techniques. It is hopeful that estimating the behavior of polariton states using more easily accessible classical methods can lead to further receptiveness of these structures among the scientific community.

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Фотонфізичні властивості сильно зв’язаних гібридизованих світло-матеріальних станів з позицій класичної фізики

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Модель осцилятора Лоренца застосовано до сильно зв’язаного гібридизованого світло-матеріального стану. Показано, що дійсна частина показника заломлення зазнає різкої зміни в області так званих «темних станів». Ця властивість гібридизованих світло-матеріальних станів може дозволити використовувати їх як нетрадиційні оптичні матеріали, в яких абсорбційні та заломлювальні властивості можуть бути селективно керовані. У цій роботі розглянуто основні властивості гібридних світло-матеріальних станів, також відомих як порожнинні поляритони. Модель осцилятора Лоренца застосовано до двох типів світло-матеріальних станів: першого, утвореного внаслідок зв’язку між одним екситоном і одним фотоном порожнини, та другого, утвореного внаслідок зв’язку двох екситонів із фотоном порожнини. Крім того, для оцінки жорсткості пружини двоатомної молекули в умовах сильного світло-матеріального зв’язку використано модель простого гармонічного осцилятора. Встановлено, що жорсткість пружини безпосередньо пов’язана з фотонною складовою системи: вищий фотонний характер відповідає меншій жорсткості пружини.

Ключові слова: поляритони, молекулярні коливання, Лоренц, осцилятор.