we offer no explanation for the shape of the curves in Fig. 2, we are able to interpret the reversal of the sign of at β_0. If temperature reduces the effectiveness of the ester, then increasing temperature has the effect of increasing β. From Fig. 2 it is clear that for $\beta<\beta_0$, increasing β decreases λ; i.e., films should shift toward blue upon heating. In the region $\beta>\beta_0$, increasing β increases λ; i.e., films should shift toward red with heat. These shifts are, in fact, those that we observed. Furthermore, the inversion of solvent effect on pitch suggests that exposure to solvent effectively changes β (in the case of $CHCl_3$ it increases β). Figure 2 shows that the effect of a change in β on color inverts at β_0.

The nonmonotonic behavior of color versus composition, combined with the sign inversion of color change with temperature and color change in response to organic vapor, suggests that pitch may not be the best choice of a fundamental parameter in phenomenological theories.[8-10] Both ingredients of the liquid crystal mixtures have large permanent electric dipoles, the orientation of which relative to the molecular axis is quite different in the two molecules. We believe this must be taken into account. It is possible that the pitch is not an independent variable and is in fact determined uniquely by some combinations of other hidden variables but no such microscopic theory exists as yet.

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MECHANISM OF ABSORPTION OF LIGHT BY FREE CONTINUUM POLARONS

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The absorption of light by free continuum polarons is treated in the framework of the Landau-Pekar approach which for this problem is not restricted to strong coupling but gives at least the qualitative behavior for intermediate coupling ($\alpha \approx 3$ to 6). The absorption curve consists of a relatively narrow zero-phonon peak and a broad sideband showing a multiphonon structure. The zero- and one-phonon contributions and the low-energy part of the two-phonon contribution have been evaluated numerically.

In the continuum polaron theory, the adiabatic strong-coupling method developed by Landau and Pekar[1] leads to two kinds of internal excited states: the "Franck-Condon"-like (FC) states and the "relaxed excited states." In the former, the polarization remains adapted to the initial electronic configuration whereas in the latter, the ionic polarization of the lattice is adapted to the final electronic configuration.

The purpose of this Letter is to explain how these two types of internal excited states which we studied in previous work[2,3] could be revealed

in the absorption spectra of free charge carriers in ionic crystals in which continuum polarons are the charge carriers. As far as we know, the only theoretical investigation of absorption of light by continuum polarons has been made in the framework of Feynman's path integral method by Feynman et al.[4] However, their formalism does not take into account the relaxed internal states, and we report entirely different results.

It should be remarked that our results are not to be compared with results on small polarons. One should be well aware of the fact that continuum polarons (even in the limit of strong coupling) behave entirely different from small polarons. Not only the transport properties are entirely different (as is well known), but the same is true for the optical properties. E.g., zero-phonon transitions are almost nonexistent in the case of small polarons.

From the physical point of view, the absorption of light by free polarons can be easily explained by a mechanism of transition in two steps. Initially, the electron oscillates in the potential well of the polarization induced by itself and at the ground-state level E_0. Under the action of the electromagnetic wave the electron undergoes first a transition from the ground state towards a FC-like excited state characterized by an energy E_{FC}. This transition occurs in such a short time $\tau \simeq \hbar(E_{FC}-E_0)$ that the shape of the polarization cloud does not change during the process.

However, the FC state is unstable: The ionic polarization relaxes and adapts itself to the final electronic configuration in a time of order $\tau \simeq 1/\omega$ and so the FC state decays in a second step to an "internal relaxed state" by emission of one or more phonons.

From the formal point of view, it is important to note that perturbation theory permits the determination of the transition probability between eigenstates of the nonperturbed Hamiltonian only, i.e., between stable states (except for an electromagnetic decay). The FC states are not stable states, as indicated above, and at least in the case of the polaron, not the most convenient final states.

We found the relaxed excited states (RES) to be, in general, far more stable than the FC states for $\alpha \gtrsim 3$. The probability of decay into the ground state by emission of a single phonon, for example, becomes very small at intermediate or strong coupling strengths. We find this to be true for $\alpha \gtrsim 3$. Indeed, the conservation of energy and momentum requires then that the wave number of the emitted phonon is beyond the quantum cutoff which is known to occur in the electron-phonon interaction. A detailed calculation of the linewidth of the RES, in the framework of the Landau-Pekar adiabatic approximation and using the Wigner-Weisskopf theory, corroborates this conclusion. For $\alpha = 5$, using one-phonon processes, we found $\frac{1}{2}\gamma \simeq 0.03\hbar\omega$. Multiphonon contributions to the decay become important at stronger coupling, and we have not yet been able to evaluate their effect; however, conservation of energy and momentum shows that a more precise calculation of the linewidth at $\alpha = 5$ does not affect the order of magnitude of our result.[5]

One might object to the Landau-Pekar wave functions being used at intermediate coupling. However, although the Landau-Pekar approximation is not very accurate for an energy calculation, say for $\alpha = 5$, this approximation, which is similar to the Born-Oppenheimer approximation, is definitely a reasonable one as long as the frequency of the electron in both the ground state and the first internal excited state is larger than $\omega/2\pi$. According to our calculations, this holds true for $\alpha \gtrsim 5$. However, the RES remains stable down to $\alpha \approx 2.5$ (except possibly at the threshold value of α at which emission of two phonons becomes possible), and therefore, the general shape for the absorption coefficient which we obtain might be valid down to $\alpha \approx 2.5$. In fact, $\alpha \approx 2.5$ corresponds to a critical coupling region where the concept of the RES breaks down.

It is thus a good approximation to assume that the RES and the states describing scattering of phonons on the latter constitute eigenstates of the Hamiltonian in the absence of electromagnetic perturbation for $\alpha \gtrsim 3$ (except for some critical "threshold" values of α).

The well-known time-dependent perturbation theory can now be applied to calculate the transition probability. We assume that the transitions are produced only by electric dipolar interaction between the electron and the electromagnetic field, and we use for wave functions of the unperturbed states those given by the Landau-Pekar approximation.

Concerning the calculations, the details of which will be published elsewhere, we might indicate that the overlap integrals, including the field wave functions, are straightforward to calculate after the Fourier transforms of the charge density are calculated for the different states. These Fourier transforms allow for an appropriate treatment of the field relaxation.

Let us point out here that we predict qualitatively an absorption curve consisting of a narrow peak at the energy of RES followed on the high-energy side by a large asymmetrical band. This band has a kind of "saw tooth" structure: The curve presents angular points spaced by a quantum $\hbar\omega$, i.e., the energy of a LO phonon. The superposition of the one-phonon and multiphonon contributions is responsible for this structure. As the absorption is due to a transition towards the FC excited state, which is immediately followed by a decay, one is led to the conclusion that the whole broad band describes the actual FC state. This level cannot be described, therefore, by a theory of natural linewidth such as the Wigner-Weisskopf theory. Therefore, the concept of the Franck-Condon state commonly used in color center theory is to be modified for continuum polarons.

We have been able to evaluate quantitatively the probability $\Gamma(1)$ of transition with emission of a single phonon, and the low-energy side of the two-phonon contribution. In reduced units we find the following expression for $\Gamma(1)$:

$$\Gamma(1)=\frac{1}{\pi}\left(\frac{m_1^*}{m}\right)^{\frac{1}{2}}\left(\frac{\Delta\Omega}{\omega}\right)^{-\frac{1}{2}}\frac{e^{-0.104\alpha^2}}{\alpha^2}\left[\exp\left(-\frac{1}{2}\frac{m_1^*}{m}\frac{\Delta\Omega}{\Omega_0}\right)-\exp\left(-\frac{1}{2}\frac{m_1^*}{m}\frac{\Delta\Omega}{\Omega_1}\right)\right]^2$$

$$+\frac{2}{3}\frac{m_1^*}{m}\frac{\Delta\Omega}{\Omega_1}\left[\exp\left(-\frac{1}{2}\frac{m_1^*}{m}\frac{\Omega_1+\Omega_0}{\Omega_0}\frac{\Delta\Omega}{\Omega_1}\right)-\exp\left(-\frac{m_1^*}{m}\frac{\Delta\Omega}{\Omega_1}\right)\right]+\frac{1}{5}\frac{m_1^*}{m}\left(\frac{\Delta\Omega}{\Omega_1}\right)^2\exp\left(-\frac{m_1^*}{m}\frac{\Delta\Omega}{\Omega_1}\right),$$

where $\hbar\Delta\Omega=\hbar\Omega-(\hbar\omega+E_1-E_0)$. $\Gamma(1)$ depends essentially on the frequency Ω of incident light, the long-wavelength longitudinal optical frequency ω, the coupling constant α, and quantities which characterize the RES (Ω_1, the frequency of the electron in the RES; m_1^*, the effective mass of the electron in the RES; E_1, the energy of the RES) and the ground state (Ω_0, the frequency of the electron in the ground state; E_0, the energy of the ground state); $\Gamma(1)$ is shown in Fig. 1 for different values of the coupling constant α.

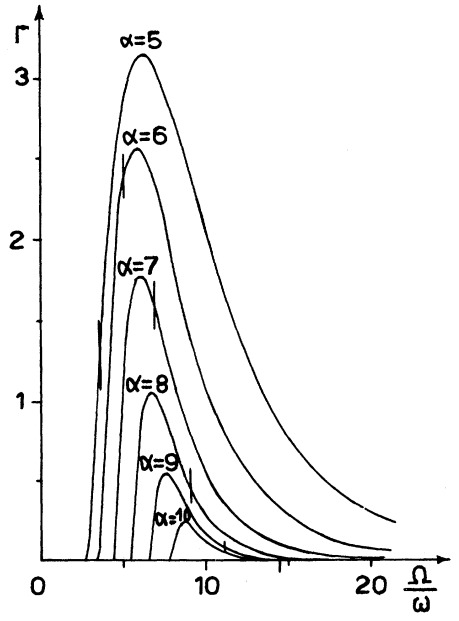

FIG. 1. Absorption (arbitrary units) with emission of a single phonon as a function of the photon frequency Ω and at different coupling strengths.

The curves are, in fact, tangential to the Ω axis at $\Delta\Omega=0$, but their slopes increase quite rapidly afterwards. The maximum is reached at a few $\hbar\omega$, and the decrease thereafter in the absorption for increasing Ω is essentially due to the quantum cutoff. The vertical lines on the diagram indicate the position of the FC energies E_{FC} as predicted by the Landau-Pekar approximation. The curves show that the transition mechanism with emission of one optical phonon becomes less probable when the coupling constant increases and the emission of several phonons takes place. Unfortunately, the determination of the probability of transition accompanied by a multiphonon emission is analytically involved. For this reason, we have limited our calculations to photon energies just beyond the threshold at which two phonons are emitted. Fig-

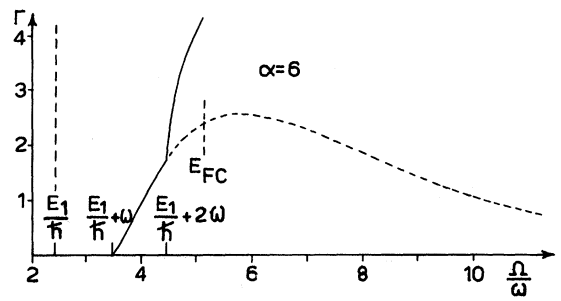

FIG. 2. Low-energy part of the absorption (arbitrary units) by free polarons at $\alpha=6$.

ure 2 shows the part of the absorption curve we are able to calculate for $\alpha = 6$. The dashed vertical line indicates the position of the zero-phonon peak, which has not been drawn. The dashed curve is the one-phonon contribution.

The general behavior predicted here is valid for $\alpha \gtrsim 5$ and is expected to remain qualitatively the same for $\alpha \gtrsim 3$ (except for some "threshold" values of α). The most important conclusion of this Letter is the appearance of a complex dependence of the optical absorption on Ω, entirely different from the peaks predicted by FHIP[4] which are just at the FC levels. In fact, we show that the concept of an FC state breaks down for continuum polarons certainly if $\alpha > 3$.

We finally wish to point out that the present results are of great importance for the possible experimental confirmation of the existence of internal excited polaron states and also of the whole polaron theory.

Unfortunately, no experimental result on the absorption spectra of free continuum polarons is available, and a comparison with experiment is not possible yet. The optical experiments by Mühlstroh and Reik[6] on $LaCO_3$ and those by Baer[7] on $SrTiO_3$ are not relevant for the present calculations, the former because small polarons are involved, the latter because one gets in a complex situation and, e.g., continuum polaron theory is not adequate to describe transport phenomena. We will discuss this situation in detail in a forthcoming publication.

However, some systems involving continuum polarons, such as color centers or excitons, have absorption spectra showing a structure similar to that described here.[8,9] We show in a subsequent paper that those systems, and especially the F center in AgBr, can be described by a continuum polaron approximation. This gives qualitative evidence of the validity of our conclusions.

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IS THE STRUCTURE OF O^{16} UNDERSTOOD?

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We argue that the structure of O^{16} cannot be completely understood unless the position of the first 0^+ state can be calculated without using adjustable parameters. We further demonstrate that this state cannot be calculated by Hartree-Fock or related methods using the present generation of nuclear forces.

Following the proposal that the low-lying positive-parity states in O^{16} might be rotational,[1] a great amount of effort has gone into an attempt at a unified description of the O^{16} energy spectrum. After the initial investigations based upon SU(3) symmetry,[2] increasingly more complex calculations, either variational[3] or shell model,[4] have been undertaken to gain understanding of the low-lying states and especially the 0^+ state at 6.06 MeV.

It became increasingly clear following the Hartree-Fock (HF) calculation of Bassichis and Ripka[3] that the four-particle, four-hole (4p-4h) state played an important role in the structure of these states. In fact because in the above calculation the 4p-4h state was well isolated, it was conjectured that it should give rise to a simple rotational band, such as that beginning at 6.06