Quantum-optical properties of polariton waves

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We develop a quantum-mechanical Hamiltonian formulation to treat the polariton in the framework of quantum optics. We exploit two specific Hamiltonians: the conventional Hopfield model, and a more general Hamiltonian. For both of these, exciton-polariton quantum states are found to be squeezed *(intrinsic polariton squeezing)* with respect to states of an intrinsic, nonpolaritonic, mixed photon-exciton boson. The amount and duration of intrinsic squeezing during the polariton period are calculated for exciton polaritons in typical I-VII and III-V semiconductors. Among the noteworthy features is the possibility of tuning the amount of intrinsic squeezing by varying the frequency–wave-vector dispersion of the polariton mode. We further analyze the photon statistics of the electromagnetic component of the polariton. Tunable non-Poissonian photon statistics and squeezing *(optical polariton squeezing)* are found in the radiative component of the exciton polariton. This entails the reduction of the fluctuations of the polariton electromagnetic field component below the limit set by the vacuum fluctuations. The Mandel Q factor for the number distribution of photons in a polariton coherent state has been evaluated. Although small, for I-VII and III-V materials in the range of modes analyzed, the Q factor could be enhanced for phonon polaritons as well as for other materials. Interpretations of the origin of squeezing in polariton states are presented.

INTRODUCTION

The present paper reports detailed results of our investigations of quantum-optical properties of the mixed polarization modes in crystals known as polaritons. In a previous brief report[1] we showed that polaritons are "intrinsically" squeezed and we also analyzed theoretically an experiment that could detect such squeezing.

The polariton, the normal mode of the coupled matter-radiation system, has been well known for at least 40 years since the independent pioneering work of Tolpygo[2] and Huang.[3] Certain properties of the polaritons have been extensively studied by a variety of spectroscopic methods. In particular, the resonant Brillouin scattering (RBS) technique proposed and theoretically analyzed by Brenig, Zeyher, and Birman[4] has permitted careful and quantitative mapping of the frequency–wave-vector dispersion $\omega(k) \equiv \omega_k$ for exciton polaritons. In addition, delicate steady-state optical reflection and transmission studies using polarized light and optical pulse (transient) studies have complemented the RBS experiments to determine ω_k with very high precision. Many details are given in several review volumes.[5,6] Phonon polaritons have been studied by related methods, which in many cases antedate the work on exciton polaritons.[5,7]

The theoretical framework for predicting and analyzing these optical properties which depend on the polariton dispersion ω_k is also of long standing and was early developed by Tolpygo,[2] Huang,[3] Pekar,[8] Agranovich and Ginzburg,[9] Hopfield,[10] and others. Striking success was achieved by using rather simple theoretical models which encompass the major relevant physics. In particular, one model commonly referred to as the Hopfield-model Hamiltonian has been useful.[10] For our present purposes this will be our "benchmark" and will be referred to below as the *conventional* model. We shall also extend this model along lines of somewhat related early work of Dexter and Heller,[11] Knox,[12] and others on excitons to give a *generalized* model Hamiltonian. Still further modifications of these basic descriptions are often used to investigate nonlinear processes, decay processes, and other higher-order effects which are not directly of interest in our work.

Despite this extensive literature, the investigation of the quantum-optical properties of these crystal mixed modes has lagged. By contrast, the study of the quantum-optical properties of the single-mode and multimode cavity electromagnetic radiation, either in isolation or in interaction with one or more atoms, has been extensively developed.[13] Part of this impetus has been the expectation that low-noise devices can be made for use, e.g., as gravity-wave detectors or in communication technology which will take advantage of "squeezed light."[14] Of course, the study of the basic statistical properties of light is by itself a major stimulus for such investigations. These effects found in optics have a certain generality and they may have analogs in condensed media.

The present work will exploit a variety of interesting physical consequences of the simple mathematical result that the linear canonical Bogoliubov transformation which diagonalizes the matter-radiation Hamiltonian to produce polaritons is a squeeze transformation. One noteworthy feature is that the amount of squeezing depends on its frequency, i.e., it is "tunable." Here, there is a quantitative difference, depending on which polariton

Hamiltonian is used: the conventional or the generalized one. Because of this, we find it useful to discuss separately (in fact, sequentially) these two cases, and to emphasize different quantum-statistical properties of the two models.

In all cases, the polariton is the true normal mode of the system. In order to study properties related to this mode, one must populate the mode, e.g., by irradiating the system with an incident laser beam. Just as with any Bose normal mode, the occupancy number of each state needs to be specified. We defer to elsewhere a detailed discussion of questions related to converting incident coherent laser light to the occupancy of squeezed polariton states. This is related to the quantum-optic extinction theorem announced elsewhere by one of us.[15]

In Sec. I of this paper, we review some useful results related to the coherent- and squeezed-state formalism. We develop modifications of the standard results in order to account for mixed modes, as will arise for polaritons.

In Sec. II we discuss the nonspatially dispersive (local) Hopfield polariton mode and the nonclassical effects that arise when that is populated. Among them, we show that the quantum statistics of the (dressed) photonic part of the polariton is nonclassical. The deviations from classical behavior are tunable and depend on the wave-vector–frequency mode that is populated. The Mandel Q factor for the number distribution of photons in a polariton coherent state is determined, and we discuss how best to measure it. In particular, *optical polariton squeezing,* that is, the reduction of the fluctuations of the *photon* field in polariton quantum states below the vacuum-state value is predicted. These considerations are illustrated by numerical calculations for exciton polaritons in typical I-VII and III-V semiconductors where the nonspatially dispersive model is relevant. For these materials, indeed, the conventional Hopfield polariton model has been shown to reproduce the correct experimental energy dispersion curves.[16] Because the polar phonon polariton is also nonspatially dispersive, a completely similar analysis applies and estimates are given for a phonon polariton also.

In Sec. III we begin with a study of a more general polariton Hamiltonian. It includes some additional coupling and it belongs more completely to the dynamical algebra Sp(8,$\mathbb{C}$). With suitable parameters this Hamiltonian gives rise to a polariton dispersion ω_k which agrees with experiments. Again, the Bogoliubov polariton transformation gives squeezed states. These are isomorphic to the conventional two-mode squeezed photon states. Here, however, squeezing is investigated not with respect to photons but to certain boson quasiparticles ($\hat{c}$), which are mixtures of photon and exciton, yet not polaritons. They are intrinsic to the polariton structure and this squeezing will be therefore referred to as *intrinsic polariton squeezing.* Within this generalized model Hamiltonian, interpretations of intrinsic squeezing in polariton states are presented. Both sudden frequency change and wave-vector mode quantum correlations are envisaged as the mechanism for producing squeezed fluctuations of the intrinsic boson $\hat{c}$. Among the nonclassical properties of the $\hat{c}$'s, we analyze the dependence on the wave-vector–frequency dispersion of the intrinsic polariton squeezing, and of the periodic reduction of their envelope of fluctuations below the vacuum value. Quantitative calculations are given for exciton polaritons in typical I-VII and III-V semiconductors. Finally, note that the theory presented here differs in one qualitative fashion from usual treatments of electromagnetic squeezing because our theory is linear. It is commonly believed that nonlinearity is needed to produce squeezed light, e.g., parametric amplification, four-wave mixing, etc. But our work shows that squeezing can occur owing to the particular polariton mode mixing in a linear resonant medium. Nonlinearities are not necessary, although nonlinearity may be sufficient in other cases. A certain similarity in viewpoint to that of Abram,[17] and Glauber and Lewenstein,[18] is pointed out.

I. MULTIMODE COHERENT AND SQUEEZED STATES

In this section we will establish notations, and we will review the definitions and some useful properties of number states, coherent states and squeezed states.

We denote by a and b two different Bose particles while 1 and 2 denote two distinct modes for each particle; thus,

$$\begin{aligned}&[\hat{a}_1,\hat{a}_1^\dagger]=[\hat{a}_2,\hat{a}_2^\dagger]=[\hat{b}_1,\hat{b}_1^\dagger]=[\hat{b}_2,\hat{b}_2^\dagger]=1\ ,\\ &[\hat{a}_1,\hat{a}_1]=[\hat{a}_1^\dagger,\hat{a}_1^\dagger]=[\hat{a}_2,\hat{a}_2]\\ &\qquad=[\hat{a}_2^\dagger,\hat{a}_2^\dagger]=[\hat{a}_1,\hat{a}_2]\\ &\qquad\qquad=[\hat{a}_2^\dagger,\hat{a}_2^\dagger]=0\ .\end{aligned} \tag{1.1}$$

Recall that for the vacuum state $|0\rangle$ of each operator, one has

$$\hat{a}_1|0\rangle=\hat{a}_2|0\rangle=\hat{b}_1|0\rangle=\hat{b}_2|0\rangle=0\ .$$

A. Number states

We denote a normalized number state for the mode 1 of particle a by

$$|n_1\rangle_a=[n_1!]^{-1/2}(\hat{a}_1^\dagger)^{n_1}|0\rangle\ .$$

Two-mode normalized number states for particle a are

$$|n_1n_2\rangle_a=|n_1\rangle_a\otimes|n_2\rangle_a$$

and likewise for particle b. The mixed-mode operator

$$\hat{\gamma}_l\equiv\alpha_l\hat{a}_l+\beta_l\hat{b}_l\quad(l=1,2) \tag{1.2}$$

is a Bose-particle destruction operator which is a linear combination of $\hat{a}_l$ and $\hat{b}_l$ with "weights" α_l and β_l. Then,

$$[\hat{\gamma}_l,\hat{\gamma}_l^\dagger]=1\ ,\quad\text{provided that }|\alpha_l|^2+|\beta_l|^2=1\ . \tag{1.3}$$

A relative phase χ_l between the two distinct particles may also be introduced by redefining (1.2) as

$$\hat{\gamma}_l=|\alpha_l|\hat{a}_l+e^{i\chi_l}|\beta_l|\hat{b}_l\ . \tag{1.4}$$

The number states of these operators can be defined in complete analogy with those for the $\hat{a}_l$'s and $\hat{b}_l$'s.

B. Coherent states

Following Glauber,[19] coherent states of the bosons $\hat{a}_l$ or $\hat{b}_l$ or $\hat{\gamma}_l$ can be defined as eigenstates of the respective destruction operators, e.g.,

$$\hat{a}_l|a_l\rangle = a_l|a_l\rangle \ , \tag{1.5}$$

where a_l (no caret) is the complex (c-number) eigenvalue. Two-mode coherent states $|a_{12}\rangle$ are defined to be simultaneous eigenstates of both annihilation operators; e.g., for $\hat{a}_1$ and $\hat{a}_2$, one has

$$\hat{a}_1|a_{12}\rangle = a_1|a_{12}\rangle \quad \text{and} \quad \hat{a}_2|a_{12}\rangle = a_2|a_{12}\rangle \ , \tag{1.6}$$

with continuous complex eigenvalues

$$a_1 \equiv |a_1|e^{i\phi_1^a} \quad \text{and} \quad a_2 \equiv |a_2|e^{i\phi_2^a} \ .$$

It is often useful to consider that two-mode coherent states are generated by applying the two-mode displacement operator onto the vacuum $|0\rangle$,

$$|a_{12}\rangle = \hat{D}(a_{12})|0\rangle$$
$$\equiv \exp[-\tfrac{1}{2}(|a_1|^2+|a_2|^2)e^{a_1\hat{a}_1^\dagger}e^{a_2\hat{a}_2^\dagger}|0\rangle \ . \tag{1.7}$$

Here, $|a_{1,2}|^2$ is the average number of bosons a in mode 1 or 2. In the configuration space, these states represent minimum-uncertainty (MU) Gaussian wave packets whose width is fixed in time. In a time-dependent picture of the e.m. field this property is referred to as time stationarity of the noise of the light field in a coherent state.[20] We can proceed similarly to introduce single-mode and two-mode coherent states for the $\hat{b}_l$'s and $\hat{\gamma}_l$'s.

C. Squeezed states

Here our particular interest is in squeezed ("low-noise") states, and especially in two-mode squeezed states. They have been discussed in the literature.[14,21] One useful definition of a two-mode squeezed state is that it is obtained by displacing the squeezed vacuum; e.g., one has

$$|a_{12}\rangle_z = \hat{D}(a_{12})\hat{S}\,^a_{12}(z)|0\rangle \ , \tag{1.8}$$

where

$$\hat{S}\,^a_{12}(z) \equiv \hat{S}\,^a_{12}(r,\varphi) = \exp[r(\hat{a}_1\hat{a}_2e^{-2i\varphi} - \hat{a}\,^\dagger_1\hat{a}\,^\dagger_2 e^{2i\varphi})] \tag{1.9}$$

is the two-mode squeeze operator, r is the squeeze factor, and φ is a phase. $z \equiv \{r,\varphi\}$ is used as an abbreviated collective notation. Of course, identical formulas hold for the $\hat{b}_l$ and $\hat{\gamma}_l$ bosons.

There is an alternate useful way to obtain the state $|a_{12}\rangle_z$, i.e., by transforming the two annihilation operators $\hat{a}_1$ and $\hat{a}_2$ into squeezed annihilation operators. For example, we may transform a_1 as

$$\hat{S}\,^a_{12}(z)\hat{a}_1\hat{S}\,^{a\dagger}_{12}(z) = \hat{a}_1\cosh r + \hat{a}\,^\dagger_2 e^{2i\varphi}\sinh r \equiv \hat{A}_1(z) \ , \tag{1.10}$$

and likewise for $\hat{a}_2$ to obtain $\hat{A}_2$. In analogy with the coherent states, they can be defined alternatively as eigenstates of the $\hat{A}$'s:

$$\hat{A}_1(z)|a_{12}\rangle_z = A_1(z)|a_{12}\rangle_z \ ,$$
$$\hat{A}_2(z)|a_{12}\rangle_z = A_2(z)|a_{12}\rangle_z \tag{1.11}$$

where $A_1(z)$ and $A_2(z)$ are complex eigenvalues. If there is no possible confusion, we may write

$$\hat{A}_{1,2}(z)|a_{12}\rangle_z = A_{1,2}(z)|a_{12}\rangle_z \ . \tag{1.12}$$

In the configuration space these states represent Gaussian wave packets. For a suitable Hamiltonian the width of these Gaussians varies periodically in time, becoming smaller and bigger than that of the vacuum state: They are not therefore minimum-uncertainty states for all times as the state evolves, but only for certain specific times during the entire oscillation cycle.[21] For the cavity photons this property reflects the nonstationarity of the noise of the e.m. field in a squeezed state.[20]

For the mixed boson $\hat{\gamma}_l$, one proceeds identically, defining single-mode ($l=1,2$) squeezed states $|\gamma_l\rangle_z$,

$$\hat{\Gamma}_l(z)|\gamma_l\rangle_z = \Gamma_l(z)|\gamma_l\rangle_z \quad (l=1,2) \tag{1.13}$$

as the eigenstates of the squeezed annihilation operator

$$\hat{\Gamma}_l(z) \equiv \hat{S}\,^\gamma_l(z)\hat{\gamma}_l\hat{S}\,^{\gamma\dagger}_l(z) = \hat{\gamma}_l\cosh r + \hat{\gamma}_l^\dagger e^{i\varphi}\sinh r \ , \tag{1.14}$$

with

$$\hat{S}\,^\gamma_l(z) = \exp[(r/2)(\hat{\gamma}_l\hat{\gamma}_l e^{-i\varphi} - \hat{\gamma}\,^\dagger_l\hat{\gamma}\,^\dagger_l e^{i\varphi})] \quad (l=1,2) \ . \tag{1.15}$$

The corresponding two-mode squeezed states $|\gamma_{1,2}\rangle_z$ for the mixed bosons $\hat{\gamma}_1$ and $\hat{\gamma}_2$ are defined as eigenstates of

$$\hat{\tilde{\Gamma}}_1(z)|\gamma_{12}\rangle_z = \tilde{\Gamma}_1(z)|\gamma_{12}\rangle_z \ ,$$
$$\hat{\tilde{\Gamma}}_2(z)|\gamma_{12}\rangle_z = \tilde{\Gamma}_2(z)|\gamma_{12}\rangle_z \ . \tag{1.16}$$

Here,

$$\hat{\tilde{\Gamma}}_1(z) \equiv \hat{S}\,^\gamma_{12}(z)\hat{\gamma}_1\hat{S}\,^{\gamma\dagger}_{12}(z) = \hat{\gamma}_1\cosh r + \hat{\gamma}\,^\dagger_2 e^{i2\varphi}\sinh r \ , \tag{1.17}$$

$$\hat{\tilde{\Gamma}}_2(z) \equiv \hat{S}\,^\gamma_{12}(z)\hat{\gamma}_2\hat{S}\,^{\gamma\dagger}_{12}(z) = \hat{\gamma}_2\cosh r + \hat{\gamma}\,^\dagger_1 e^{i2\varphi}\sinh r \ , \tag{1.18}$$

and

$$\hat{S}\,^\gamma_{12}(z) = \exp[r(-\hat{\gamma}\,^\dagger_1\hat{\gamma}\,^\dagger_2 e^{2i\varphi} + \hat{\gamma}_1\hat{\gamma}_2 e^{-2i\varphi})] \ . \tag{1.19}$$

Again, the squeezed state $|\gamma_{12}\rangle_z$ can be generated by first applying (1.19) on the vacuum followed by a displacement [cf. (1.7)]:

$$|\gamma_{12}\rangle_z = \hat{D}(\gamma_{12})\hat{S}\,^\gamma_{12}(z)|0\rangle \ . \tag{1.20}$$

In the two-mode case the meaning we assign to α,β,r,φ is the same as for the single-mode case, except for the squeeze factor. In the former case, r mediates the coupling between two *distinct* modes 1 and 2, while in the latter case, r mediates the coupling between the *same* mode. Compare, e.g., $\hat{S}$ in (1.19) with $\hat{S}$ in (1.15). This point may be made even clearer if one lets $r\to 0$ in the explicit expression for the annihilation operator $\hat{\hat{\Gamma}}_1$ (or $\hat{\hat{\Gamma}}_2$):

$$\hat{\hat{\Gamma}}_1=\hat{a}_1|\alpha_1|\cosh r+\hat{a}_2^\dagger e^{2i\varphi}|\alpha_2|\sinh r +\hat{b}_1 e^{i\chi_1}|\beta_1|\cosh r+\hat{b}_2^\dagger e^{i(2\varphi-\chi_2)}|\beta_2|\sinh r \; . \quad (1.21)$$

In this limit, $\hat{\hat{\Gamma}}_1$ only couples $\hat{a}_1$ to $\hat{b}_1$. In the states $|\gamma_{12}\rangle_z$, instead $r\neq 0$, the two distinct modes become so tightly correlated that they no longer fluctuate independently by even the small amount allowed in a coherent state.[14] As we shall see later states like $|\gamma_{12}\rangle_z$ turn out to be a good physical representation for a polariton quantum state where the two distinct bosons are excitons and photons coupling inside the crystal with pairs of opposite wave-vector modes to form a new excitation of the combined electromagnetic-radiation–dielectric-material system.[10]

D. Non-Poissonian statistics and squeezing

One useful figure of merit for a squeezed state is the departure from Poissonian statistics for the number of bosons in the state.[22–23] Consider the distribution (P) of the number of bosons $\hat{\gamma}_l$ in the state $|\gamma_{12}\rangle_z$. Results that will be used in the following are now derived. The width of P can be characterized by the square root of

$$\langle(\Delta\hat{N}_l)^2\rangle \equiv {}_z\langle\gamma_{12}|(\hat{\gamma}_l^\dagger\hat{\gamma}_l)^2|\gamma_{12}\rangle_z -\{{}_z\langle\gamma_{12}|\hat{\gamma}_l^\dagger\hat{\gamma}_l|\gamma_{12}\rangle_z\}^2 \quad (l=1,2)$$

namely, the variance of the fluctuations of the number of bosons $\hat{\gamma}_l$ in the squeezed state $|\gamma_{12}\rangle_z$. In order to evaluate, say, $\langle(\Delta\hat{N}_1)^2\rangle$, we first give

$${}_z\langle\gamma_{12}|\hat{\gamma}_1|\gamma_{12}\rangle_z=\gamma_1 \; . \quad (1.22)$$

Then,

$$\langle\hat{N}_1\rangle \equiv {}_z\langle\gamma_{12}|\hat{\gamma}_1^\dagger\hat{\gamma}_1|\gamma_{12}\rangle_z=|\gamma_1|^2+\sinh^2 r \; , \quad (1.23)$$

where

$$|\gamma_1|^2=|a_1|^2|\alpha_1|^2+|b_1|^2|\beta_1|^2 +|\alpha_1\beta_1|(a_1 b_1^* e^{-i\chi_1}+\text{c.c.}) \quad (1.24)$$

is obtained from (1.2). It is clearly seen from (1.23) and (1.24) that in the state $|\gamma_{12}\rangle_z$ the mean number has two contributions: one arising from the coherent excitation of the mixed boson $\hat{\gamma}_1$, i.e., $|\gamma_1|^2$, and the other from the squeezing. Thus, P strongly depends on the relative weight of these two contributions. Furthermore, as shown by (1.24), several elements influence the coherent contribution. A straightforward calculation yields

$$\langle(\Delta\hat{N}_1)^2\rangle \equiv |\gamma_1|^2\{e^{-2r}\cos^2[\tfrac{1}{2}(\varphi-2\phi_1)] +e^{2r}\sin^2[\tfrac{1}{2}(\varphi-2\phi_1)]\} +\tfrac{1}{2}\sinh^2(2r) \; , \quad (1.25)$$

where we set $\gamma_1\equiv|\gamma_1|e^{i\phi_1}$. For a given r, extremal values for (1.25) obtain when $2\phi_1=\varphi$ and $2\phi_1=\varphi+\pi$, that is,

$$\langle(\Delta\hat{N}_1)^2\rangle \equiv e^{\pm 2r}\langle\hat{N}_1\rangle+\sinh^2 r(1+\sinh 2r) \; . \quad (1.26)$$

The upper sign corresponds to π and the lower to 0.

For clarity of exposition in the remainder of this paper, we will treat only these two limits rather than intermediate cases. Furthermore, we will consider the limit $|\gamma_1|^2 \gg e^{2r}$, which will turn out to be of some importance to us. Under this circumstance the coherent contribution to the mean number [first term in (1.23)] greatly exceeds the squeezed contribution (second term), and $\langle\hat{N}_1\rangle\approx|\gamma_1|^2$. Also, the first term on the right-hand side of (1.26) exceeds the second one[9] and the variance can be approximated as

$$\langle(\Delta\hat{N}_1)^2\rangle=\langle\hat{N}_1\rangle e^{\pm 2r}\equiv s^{\pm 2}\langle\hat{N}_1\rangle \; , \quad \langle\hat{N}_1\rangle\cong|\gamma_1|^2 \; . \quad (1.27)$$

This result[24] should be compared with the equivalent result for a coherent state (Poisson distribution), for which $\langle(\Delta\hat{N}_1)^2\rangle=\langle\hat{N}_1\rangle$. Thus, the state $|\gamma_{12}\rangle_z$ carries a non-Poissonian counting statistics whose deviations are measured by the squeezed amplitude $s\equiv e^r$. In particular, depending on whether $2\phi_1-\varphi$ is 0 *or* π, $P(N_1)$ represents a distribution broader or narrower than a Poissonian: $s^2\to$super-Poissonian or $s^{-2}\to$sub-Poissonian. When $|\gamma_1|^2$ is not sufficiently dominant in $\langle\hat{N}_1\rangle$, the connection between $\langle(\Delta N_1)^2\rangle$ and $\langle N_1\rangle$ is not straightforward. A completely analogous discussion can be given for the other mode 2.

II. NONCLASSICAL PROPERTIES OF CONVENTIONAL POLARITONS

In this section we develop the theory of the quantum-statistical properties associated with the familiar local exciton polariton. The theory will apply also to the optical-phonon polariton, or other coupled polarization mode (photon plus magnon, for example). The Bogoliubov transformation from the coupled bare photons *plus* bare excitons to polaritons will be shown to be a two-mode squeezed transformation. This gives rise to tunable non-Poissonian photon statistics and optical polariton squeezing over the entire polariton dispersion. The Mandel Q factor that measures these effects is analyzed. Results for typical I-VII and III-V semiconductors are reported.

A. The polariton and squeezing

We briefly review the quantum mechanics of a polariton. We adopt the standard polariton model due to Hopfield and we emphasize that the local, nonspatially dispersive case will be treated here. The Hamiltonian[10]

$$\hat{H}^{\text{Hopfield}} = \sum_{\substack{k>0,\\ k<0}} [\hbar ck(\hat a_k^\dagger \hat a_k + \tfrac{1}{2}) + \hbar\omega_0(\hat b_k^\dagger \hat b_k + \tfrac{1}{2}) + B(k)(\hat a_k^\dagger \hat a_k + \hat a_k \hat a_k^\dagger + \hat a_k^\dagger \hat a_{-k}^\dagger + \hat a_k \hat a_{-k})$$
$$+ iA(k)(-\hat b_k^\dagger \hat a_k - \hat b_k^\dagger \hat a_{-k}^\dagger + \hat b_k \hat a_{-k} + \hat b_k \hat a_k^\dagger)]$$
$$\equiv \sum_{k>0} \{\hat h^{\text{pol}}_{+k} + \hat h^{\text{pol}}_{-k}\} \tag{2.1}$$

can be separated into the sum of Hamiltonians for pairs of wave-vector modes $+k$ and $-k$, so we begin by considering a polariton mode of given wave vector $+k$ in (2.1) and its relevant Hamiltonian. Here, ck and ω_0 are the bare photon and exciton frequencies, respectively, with $\hat a_k$ and $\hat b_k$ the associated annihilation operators, while

$$A(k) = \frac{(\hbar\omega_0)^2(4\pi\beta_0)^{1/2}}{2(\hbar ck\,\hbar\omega_0)^{1/2}} ,$$
$$B(k) = (\pi\beta_0)\frac{(\hbar\omega_0)^2}{\hbar}ck = \frac{A^2(k)}{\hbar\omega_0} . \tag{2.2}$$

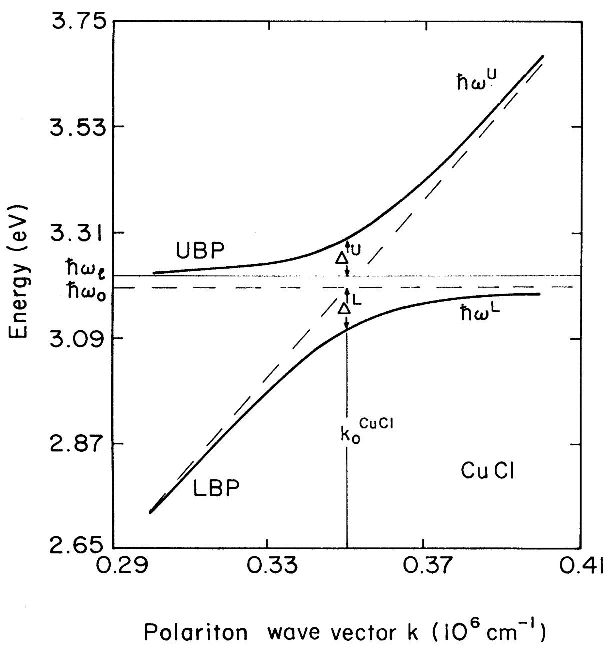

FIG. 1. Frequencies of the CuCl exciton-polariton coupled mode[16] as a function of the wave vector (solid line): two values, ω^U and ω^L, correspond to each given (real) k, one in the upper branch (UBP) and the other in the lower branch (LBP). One wave vector only corresponds to a given energy (no spatial dispersion) on each branch. The broken lines show the dispersion relations of "bare" photons and excitons. k_0 is the crossover wave vector of the bare photon and bare exciton energy dispersion curves ($\hbar ck_0 = \hbar\omega_0$). For an isotropic medium there is also a longitudinal exciton mode: $\hbar\omega_l$. $\omega_{\text{LT}} \equiv \omega_l - \omega_0$ is the transverse-longitudinal splitting frequency, ω_0 is the transverse exciton energy, and $\Delta^{U,L}$ are the distances of the upper and lower polariton energy branches from the longitudinal and transverse exciton energies, respectively.

β_0 is the photon-exciton oscillator coupling strength proportional to a dipole matrix element squared. We temporarily replace

$$\hat a_{1,2} \to \hat a_{+k,-k} , \quad \hat b_{1,2} \to \hat b_{+k,-k} ,$$
$$\hat a_{1,2}^\dagger \to \hat a_{+k,-k}^\dagger , \quad \hat b_{1,2}^\dagger \to \hat b_{+k,-k}^\dagger ,$$
$$\lambda \equiv \lambda(k) \to \hbar ck , \quad \mu \to \hbar\omega_0 , \quad \sigma \to 4\pi\beta_0 , \tag{2.3}$$

so as to make a connection with the results of Sec. I. The Hamiltonian (2.1), say, for a fixed single mode, can be represented by the generic bilinear Hamiltonian

$$\hat h_1^{\text{pol}}(\lambda,\mu,\sigma) = \lambda(\hat a_1^\dagger \hat a_1 + \tfrac{1}{2}) + \mu(\hat b_1^\dagger \hat b_1 + \tfrac{1}{2})$$
$$+ f(\hat a_1^\dagger \hat b_1 - \hat a_1 \hat b_1^\dagger + \hat a_2 \hat b_1 - \hat a_2^\dagger \hat b_1^\dagger)$$
$$+ g(\hat a_1^\dagger \hat a_1 + \hat a_1 \hat a_1^\dagger + \hat a_1 \hat a_2 + \hat a_1^\dagger \hat a_2^\dagger) , \tag{2.4}$$

with $f \equiv i\sqrt{\sigma}\mu^{3/2}/2\sqrt{\lambda}$ and $g \equiv \sigma\mu^2/4\lambda$. It can be shown that this Hamiltonian can be diagonalized by using a Bogoliubov transformation[25] in the particular form developed in Eq. (1.21). The diagonalizing transformation is implemented with the values of α_1, α_2, β_1, β_2, r, φ, χ_1, and χ_2 defined by

$$|\alpha_1|\cosh r = \frac{(\lambda+\varepsilon)(\mu^2-\varepsilon^2)}{2\sqrt{\lambda\varepsilon}}[(\mu^2-\varepsilon^2)^2+\sigma\mu^4]^{-1/2} ,$$
$$e^{+i\chi_1}|\beta_1|\cosh r$$
$$= -i\left[\frac{\sigma\mu^3}{\varepsilon}\right]^{1/2}(\mu+\varepsilon)[(\mu^2-\varepsilon^2)^2+\sigma\mu^4]^{-1/2} ,$$
$$e^{+2i\varphi}|\alpha_2|\sinh r \tag{2.5}$$
$$= -\frac{(\lambda-\varepsilon)(\mu^2-\varepsilon^2)}{2\sqrt{\lambda\varepsilon}}[(\mu^2-\varepsilon^2)^2+\sigma\mu^4]^{-1/2} ,$$
$$e^{-i(\chi_2-2\varphi)}|\beta_2|\sinh r$$
$$= -i\left[\frac{\sigma\mu^3}{\varepsilon}\right]^{1/2}(\mu-\varepsilon[(\mu^2-\varepsilon^2)^2+\sigma\mu^4]^{-1/2} ,$$

to obtain

$$\hat h_1^{\text{pol}} = \varepsilon(\hat{\Gamma}_1^\dagger \hat{\Gamma}_1 + \tfrac{1}{2}) . \tag{2.6}$$

ε in Eqs. (2.5) and (2.6) satisfies a biquadratic equation,

$$\varepsilon^4 - F(\lambda,\mu,\sigma)\varepsilon^2 + G(\mu) = 0 , \tag{2.7}$$

with F, G certain functions of λ,μ,σ.

Restoring hereafter the notation, one finds [cf. (2.3)]

$$\hat h_k^{\text{pol}} = \varepsilon[\hat{\Gamma}_{+k}^\dagger \hat{\Gamma}_{+k} + \tfrac{1}{2}] , \tag{2.8}$$

with ε solution of

$$\varepsilon^4-\varepsilon^2[(\hbar ck)^2+(\hbar\omega_0)^2+4\pi\beta_0(h\omega_0)^2]+(\hbar\omega_0\hbar ck)^2=0 . \tag{2.9}$$

This yields the correct energy dispersion for polaritons when $\varepsilon\rightarrow\varepsilon_k$ is identified as the "physical" polariton energy for a given wave-vector mode k. Eq. (2.9) has the solutions $\varepsilon^L_{+k}=\varepsilon^L_{-k}$ and $\varepsilon^U_{+k}=\varepsilon^U_{-k}$: U and L denote upper and lower branches of the energy dispersion curves. Typical curves for exciton polaritons in CuCl are plotted in Fig. 1.

Thus, Eqs. (2.4)–(2.6) demonstrate the transformation from bare photon *plus* bare exciton to a normal-mode physical polariton is a two-mode particle squeezing transformation whose generator is of the form (1.17). Hence, states of the Hamiltonian (2.8) are equivalent to two-mode squeezed states of the mixed bosons[26]

$$\begin{aligned}\hat\gamma_{+k}&=|\alpha_k^+|\hat a_{+k}+e^{i\chi_{+k}}|\beta_k^+|\hat b_{+k};\\ \hat\gamma_{-k}&=|\alpha_k^-|\hat a_{-k}+e^{i\chi_{-k}}|\beta_k^-|\hat b_{-k} .\end{aligned} \tag{2.10}$$

The vacuum, number, and coherent states of the polariton Hamiltonian (2.8) turn out to be squeezed with respect to the vacuum, number, and coherent (two-mode) states of the mixed bosons (2.10). The latter are the annihilation operators of a photon-exciton mixed particle, intrinsic to the polariton structure, yet not a polariton. We will then call the present type of squeezing *intrinsic polariton squeezing,* whereas it occurs with respect to the states of the intrinsic boson $\hat\gamma$. The physical polariton described by the operator below is a superposition of "dressed" photons and excitons,

$$\hat{\hat\Gamma}_{+k}=\hat S^{\,\mathrm{pol}}\hat\gamma_{+k}\hat S^{\,\mathrm{pol}^\dagger}=\hat{\hat\Gamma}{}^{\mathrm{ph}}_{+k}+\hat{\hat\Gamma}{}^{\mathrm{exc}}_{+k} , \tag{2.11}$$

and

$$\begin{aligned}\hat S^{\,\mathrm{pol}}&\equiv\hat S^{\,\mathrm{pol}}(r_k,\varphi_k)\\ &=e^{r_k(-\hat\gamma^\dagger_{+k}\hat\gamma^\dagger_{-k}e^{2i\varphi_k}+\hat\gamma_{-k}\hat\gamma_{+k}e^{-2i\varphi_k})} ,\end{aligned} \tag{2.12}$$

that in turn are specified linear combinations of "bare" photon and "bare" exciton given as [cf. (1.21)]

$$\hat{\hat\Gamma}{}^{\mathrm{ph}}_{+k}\equiv|\alpha_k^+|\hat a_{+k}c_{r_k}+e^{2i\varphi_k}|\alpha_k^-|\hat a^{\,\dagger}_{-k}s_{r_k} \tag{2.13a}$$

and

$$\begin{aligned}\hat{\hat\Gamma}{}^{\mathrm{exc}}_{+k}\equiv e^{i\chi_{+k}}(&|\beta_k^+|\hat b_{+k}c_{r_k}\\ &+e^{2i\varphi_k}e^{-i(\chi_{-k}+\chi_{+k})}|\beta_k^-|\hat b^{\,\dagger}_{-k}s_{r_k}) .\end{aligned} \tag{2.13b}$$

Here $c_{r_k}\equiv\cosh r_k$ and $s_{r_k}\equiv\sinh r_k$. Analogous expressions hold for the wave-vector mode $-k$. Equation (2.12) along with (1.10) further suggests that the photon-field component of a polariton may be squeezed with respect to the vacuum photon field $\hat a_{\pm k}$.[14] We will call this type of squeezing *optical polariton squeezing* as compared to the intrinsic squeezing, introduced above. The exciton-photon polariton mixture alters the statistical properties of the vacuum radiation field. This will be treated in some detail below. Similarly, we can discuss the change in the statistics of an uncoupled exciton upon polariton formation.

B. Squeezing magnitudes

Let us then study the properties of polariton quantum states by characterizing the polariton annihilation operator $\hat{\hat\Gamma}_{\pm k}$: It depends in general on eight parameters $\{\alpha_k^\pm,\beta_k^\pm,\chi_{\pm k},\varphi_k,r_k\}$. We will now give the explicit expressions for these parameters in terms of the starting coefficients in the Hamiltonian. This in particular will permit us to exhibit the squeeze factor, from which we obtain numerical values of the amount of squeezing for some specific materials. Equations (2.3) and (2.5) give[26,27]

$$\begin{aligned}|\alpha_k^+|\cosh r_k&=\frac{\hbar ck+\varepsilon_k}{2\sqrt{\varepsilon_k\hbar ck}}\frac{N_{2,k}}{\sqrt{M_k}}\equiv C_{1,k} ,\\ |\beta_k^+|\cosh r_k&=\frac{\sqrt{4\pi\beta_0}}{2\sqrt{M_k}}\left[1+\frac{\varepsilon_k}{\hbar\omega_0}\right]\left[\frac{\hbar\omega_0}{\varepsilon_k}\right]^{1/2}\equiv C_{2,k} ,\\ |\alpha_k^-|\sinh r_k&=\frac{|\varepsilon_k-\hbar ck|}{2\sqrt{\varepsilon_k\hbar ck}}\frac{N_{2,k}}{\sqrt{M_k}}\equiv C_{3,k} ,\\ |\beta_k^-|\sinh r_k&=\frac{\sqrt{4\pi\beta_0}}{2\sqrt{M_k}}\left|1-\frac{\varepsilon_k}{\hbar\omega_0}\right|\left|\frac{\hbar\omega_0}{\varepsilon_k}\right|^{1/2}\equiv C_{4,k} ,\end{aligned} \tag{2.14}$$

where

$$N_{2,k}\equiv|1-(\varepsilon_k/\hbar\omega_0)^2|$$

$$M_k\equiv N_{2,k}^2+4\pi\beta_0 .$$

For our purposes in this paper, we only need consider absolute values in Eq. (2.5). Further, owing to the boson nature of the $\hat\gamma_{\pm k}$'s, the two parameters $|\beta_k^\pm|$ can be expressed in terms of $|\alpha_k^\pm|$, respectively, as [cf. Eq. (1.3)]

$$|\beta_k^\pm|^2=1-|\alpha_k^\pm|^2 . \tag{2.15}$$

Thus the parameters left to determine are $|\alpha_k^+|$, $|\alpha_k^-|$, and r_k. We now give $|\alpha_k^\pm|$: From the first and second two equations in (2.14), one obtains

$$\left|\frac{\alpha_k^+}{\beta_k^+}\right|=\left|\frac{C_{1,k}}{C_{2,k}}\right| \quad\text{and}\quad \left|\frac{\alpha_k^-}{\beta_k^-}\right|=\left|\frac{C_{3,k}}{C_{4,k}}\right| . \tag{2.16}$$

Thus, (2.15) and (2.16) yield

$$|\alpha_k^+|^{-1}=\left[1+\left|\frac{C_{2,k}}{C_{1,k}}\right|^2\right]^{1/2} ,\quad |\alpha_k^-|^{-1}=\left|1+\left|\frac{C_{2,k}}{C_{1,k}}\frac{\left[1-\frac{\varepsilon_k}{\hbar\omega_0}\right]\left[1+\frac{\varepsilon_k}{\hbar ck}\right]}{\left[1+\frac{\varepsilon_k}{\hbar\omega_0}\right]\left[1-\frac{\varepsilon_k}{\hbar ck}\right]}\right|^2\right|^{1/2} . \tag{2.17}$$

We last evaluate r_k from the second and fourth equations in (2.14) and the equalities (2.16). The other determination of r_k using the first and third equations in (2.14) is equivalent owing to (2.16). We obtain

$$\tanh r_k^{U,L} = \left| \frac{\alpha_k^+}{\alpha_k^-} \left[\frac{(\hbar ck - \varepsilon_k^{U,L})}{(\hbar ck + \varepsilon_k^{U,L})} \right] \right| = \left[\frac{\xi_k}{1+\xi_k} \right]^{1/2}, \quad \xi_k \equiv |C_{3,k}|^2 + |C_{4,k}|^2 . \tag{2.18}$$

From the second of these equations, $\tanh r_k < 1$ as it must, while the first one tells us that distinct squeeze factors correspond to upper and lower energy dispersion branches at fixed k (cf. Fig. 1). Further, by changing (tuning) the wave-vector–energy dispersion along a given branch, we explore all possible values of the squeeze factor relevant to that branch. With this we have connected $\alpha_k^{\pm}, \beta_k^{\pm}, r_k$, which parametrize the physical polariton transformation (2.11), to quantities subject to experimental control. Using $\varepsilon_k^{U,L}$ and $|\alpha_k^{\pm}|$ as obtained from Eqs. (2.9) and (2.17), respectively, we calculate the amount of squeezing for the upper and lower branches of exciton polaritons in CuCl and GaAs for k ranging in the vicinity of the crossover wave vector k_0. These are plotted in Fig. 2.

It is instructive to solve Eq. (2.18) for the squeeze factor in terms of the ratio $n_k^{U,L} \equiv \hbar ck / \varepsilon_k^{U,L}$, that is,

$$r_k^{U,L} = \tfrac{1}{2} \ln \left[\frac{1 + \left| \frac{\alpha_k^+}{\alpha_k^-} \left[\frac{n_k^{U,L} - 1}{n_k^{U,L} + 1} \right] \right|}{1 - \left| \frac{\alpha_k^+}{\alpha_k^-} \left[\frac{n_k^{U,L} - 1}{n_k^{U,L} + 1} \right] \right|} \right] . \tag{2.19}$$

$n_k^{U,L}$, obtained by solving Eq. (2.9),

$$[n_k^{U,L}]^2 = \left[\frac{\hbar ck}{\hbar \omega_k^{U,L}} \right]^2 = 1 + \frac{4\pi\beta_0}{1 - (\omega_k^{U,L}/\omega_0)^2} ,$$

is just the square root of the exciton-polariton dielectric function. Two points are to be noted here. The squeeze factor is directly connected to (1) the *index of refraction* of the medium and (2) the *weights* of the exciton and photon components of the polariton. This settles how squeezing in polaritons is related to intrinsic properties of the dielectric material.[28] A value of unity for the bulk dielectric constant (ϵ_b) is assumed throughout this treatment: It derives from taking ck as photon frequency in Eq. (2.1). An $\epsilon_b \neq 1$ is introduced by rescaling the speed of light in vacuum $c \to c/\sqrt{\epsilon_b}$.

A special value of r_k which will become useful in the following sections is that evaluated at the wave-vector crossover k_0 of the uncoupled light and exciton energy dispersions (see Fig. 1); from (2.17), $|\alpha_{k_0}^+| = |\alpha_{k_0}^-|$, and then

$$r_0^{U,L} \equiv r_{k_0}^{U,L} = \tfrac{1}{2} \ln [n_{k_0}^{U,L}] . \tag{2.20}$$

From (2.18) one also has

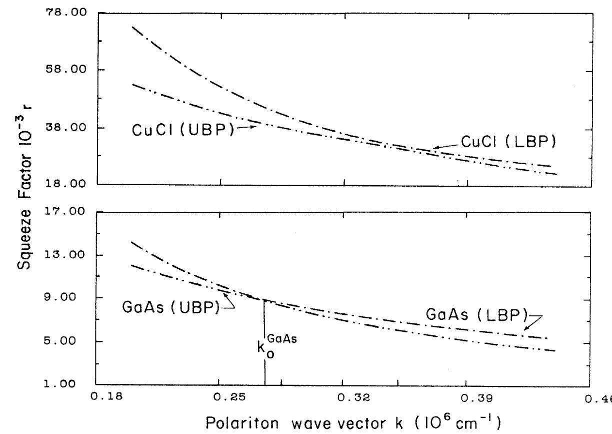

FIG. 2. Magnitude of the squeeze factor in CuCl and GaAs exciton polaritons as a function of k calculated within the *conventional* Hopfield model from the exact expressions (2.18) for LBP (—·—·—·) and UBP (—..—..—..—..) modes in the vicinity of k_0. All the numerical evaluations have been here obtained using material parameters as given in Ref. 16: $\hbar\omega_0^{\text{CuCl}} = 3.20$ eV, $\hbar\omega_0^{\text{GaAs}} = 1.51$ eV, $\epsilon_b^{\text{CuCl}} = 4.6$, $\epsilon_b^{\text{GaAs}} = 12.5$, $4\pi\beta_0^{\text{CuCl}} = 0.0158$, $4\pi\beta_0^{\text{GaAs}} = 0.0013$.

$$r_0^U \cong \tanh^{-1} \left| \frac{\Delta_U + h\omega_{\text{LT}}}{2h\omega_0} \right| , \quad r_0^L \cong \tanh^{-1} \left| \frac{\Delta_L}{2\hbar\omega_0} \right| . \tag{2.21}$$

Generally speaking, r_0 increases for small transverse exciton frequencies and large separations Δ.

C. Non-Poissonian photon statistics in a polariton coherent state: theory

In this section we discuss the fluctuations of the number of *bare* photons in a polariton quantum state. As for quantum states of the polariton field, one might consider either number states $|n_k^p\rangle$, in which a mode k is occupied by n_k^p polaritons, or coherent states. The former are a convenient basis when experiments with a fixed number of particles are involved, but they are not of direct significance, however, in a typical solid-state optical experiment where the probe exciting the polariton is usually a light source with no *definite* numbers of photons such as a laser. This more realistic situation can instead be properly described by using coherent states: in the following *polariton coherent states* will be considered as the quantum states relevant to our problem. Specifically, we will show that the Poissonian number distribution, characteristic of vacuum photons in a coherent state, does turn into non-Poissonian when referred to an exciton-polariton coherent state. According to the treatment given in Secs. I C and II A, the latter are the eigenstates $|\gamma_{\pm k}\rangle_z$ of the annihilation operator (2.11) or the two-mode states in Eq. (1.16). We first give the variance of the bare photon number $\hat{N}_{+k}^{\text{ph}} = \hat{a}_{+k}^{\dagger} \hat{a}_{+k}$ in a polariton coherent state. In Appendix A we show that[29]

$$\langle(\Delta\hat{N}_k^{\rm ph})^2\rangle = e^{\pm 2r_k}\langle\hat{N}_k^{\rm ph}\rangle - |\alpha_k^+\beta_k^+|A_{1,k} + \frac{1}{|\alpha_k^+|}(|\beta_k^+|A'_{-1,k}e^{\pm 2r_k} - |\beta_k^+|^2 A_{-1,k}) + |\beta_k^+|^2[\langle(\Delta\hat{N}_k^{\rm ph})^2\rangle - A_{0,k}] + |\alpha_k^+|^{-2}\{[1+\sinh(2r_k)]\sinh^2 r_k + e^{\pm 2r_k}|\beta_k^+|^2\langle\hat{N}_k^{\rm exc}\rangle - A_{-2,k}|\beta_k^+|^4\} , \quad (2.22a)$$

where $A_{0,k}$, $A_{\pm 1,k}$, $A_{-1,k}$, and $A_{-2,k}$ are complicated functions which, for clarity of exposition, are only given in the Appendix A. In (2.22a) the expectation values are all evaluated upon the states $|\gamma_{\pm k}\rangle_z$. Denoting by $|\text{vac}\rangle$ the vacuum photon coherent state, the corresponding number distribution is such that

$$\langle\text{vac}|(\Delta\hat{N}_k^{\rm ph})^2|\text{vac}\rangle = \langle\text{vac}|\hat{N}_k^{\rm ph}|\text{vac}\rangle ;$$

by comparison (2.22a) thus shows that bare photons in a polariton coherent state do not have a Poissonian (classical) distribution. Due to the complicated form of the general result (2.22a), we will now note some simplifications.

1. *High photon intensity*

First of all, since $\langle\hat{N}_k^{\rm ph}\rangle$ is related to the intensity of the coherent photon flux (Appendix B), one can always choose this intensity such that the first term on the right-hand side of (2.22a) dominates, the squeeze amplitude (e^{r_k}) remaining unaltered. By varying the intensity, the mode k of the polariton which is populated remains fixed and so does r_k. Thus under this circumstance [cf. also Eq. (1.27)],

$$\langle(\Delta\hat{N}_k^{\rm ph})^2\rangle \cong e^{\pm 2r_k}\langle\hat{N}_k^{\rm ph}\rangle = s_k^{\pm 2}\langle\hat{N}_k^{\rm ph}\rangle . \quad (2.22b)$$

In this case the distinction between Poissonian and non-Poissonian depends on the squeeze amplitude s_k only. The latter characterizes the noise-power reduction ratio in a semiconductor, which is nearly constant ($\cong 0.99$) for GaAs, but slightly varying (0.91–0.99) for CuCl exciton-polariton modes in the vicinity of the crossover wave vector k_0. The above result readily shows that the change of the photon statistics when an exciton polariton is created relates to intrinsic properties of the dielectric: It depends, through s_k, on the index of refraction.

2. *Selected polariton modes*

Otherwise, regardless of the coherent photon intensity, one may simplify the result (2.22a) by using the fact that the magnitudes of $|\alpha_k^+|$ and $|\beta_k^+|$ vary by tuning the energy of the polariton mode. One can conveniently excite polariton modes such that, for instance $|\beta_k^+|$ is appropriately smaller than $|\alpha_k^+|$, in which case neglecting terms quadratic and of higher order in $|\beta_k^+/\alpha_k^+|$ (2.22a) is approximated by

$$\langle(\Delta\hat{N}_k^{\rm ph})^2\rangle = e^{\pm 2r_k}\langle\hat{N}_k^{\rm ph}\rangle + \frac{\sinh^2 r_k}{|\alpha_k^+|^2}[1+\sinh(2r_k)] + |\beta_k^+|\left[\frac{e^{\pm 2r_k}}{|\alpha_k^+|}A'_{-1,k} - |\alpha_k^+|A_{1,k}\right] . \quad (2.22c)$$

More precisely, the validity of Eq. (2.22c) not only depends on the relative magnitude of $|\alpha_k^+|$ and $|\beta_k^+|$ but also on their individual magnitudes, on the index of refraction, through r_k, and on the values of $A'_{-1,k}$ and $A_{1,k}$. These are all mode-dependent quantities and the conditions under which the approximation (2.22c) is valid must be accounted for case by case.

We notice that Eqs. (2.22) may also be used to obtain specific results for the degree of second-order coherence and the Mandel Q factor.[14] They provide an equivalent way to describe the photon statistics. For the former we have

$$g_k^{(2)}(0) \equiv \frac{\langle\hat{a}_k^\dagger\hat{a}_k\hat{a}_k^\dagger\hat{a}_k\rangle}{\langle\hat{a}_k^\dagger\hat{a}_k\rangle^2} , \quad g_k^{(2)}(0) - 1 = \frac{Q_k}{\langle\hat{N}_k^{\rm ph}\rangle} , \quad (2.23)$$

while the latter can be given as [cf. (2.22a)]

$$Q_k \equiv \frac{\langle(\Delta\hat{N}_k^{\rm ph})^2\rangle - \langle\hat{N}_k^{\rm ph}\rangle}{\langle\hat{N}_k^{\rm ph}\rangle} \equiv e^{\pm 2r_k} - 1 + \Delta_k^{\pm} . \quad (2.24)$$

In the case of high photon intensity or when the polariton mode can be singled out so that the approximation (2.22c) holds, one has, respectively,

$$Q_k^{\rm hi} = e^{\pm 2r_k} - 1 \quad (2.25a)$$

and

$$Q_k^{\rm sm} = (e^{\pm 2r_k} - 1) + \frac{1}{\langle\hat{N}_k^{\rm ph}\rangle}\left[\frac{\sinh^2 r_k}{|\alpha_k^+|}[1+\sinh(2r_k)] + |\beta_k^\dagger|\left[\frac{e^{\pm 2r_k}}{|\alpha_k^+|}A'_{-1,k} - |\alpha_k^+|A_{1,k}\right]\right] . \quad (2.25b)$$

Recall that in the coherent state case, $Q_k = 0$, while departures of the photon statistics from this case are measured by $Q_k \neq 0$. Sub-Poissonian statistics carries a $Q_k < 0$ while the opposite holds in the super-Poissonian case.[30] Let us also recall that the lower and upper signs correspond to $\phi_k = \varphi_k/2$ and $\phi_k = (\varphi_k + \pi)/2$, respectively. ϕ_k is the phase of the complex eigenvalue γ_k

$$\phi_k = \tan^{-1}\left[\frac{|a_k|\sin\phi_k^a + \left|\frac{\beta_k^+}{\alpha_k^+} b_k\right| \sin(\phi_k^b + \chi_k)}{|a_k|\cos\phi_k^a + \left|\frac{\beta_k^+}{\alpha_k^+} b_k\right| \cos(\phi_k^b + \chi_k)}\right] . \quad (2.26)$$

All parameters are defined in Sec. I [cf (1.2)–(1.6)]. Owing to the rest $\Delta_k^{\pm}$, it is difficult to establish whether the two choices above for ϕ_k correspond to extremal values of the exact Mandel factor (2.24). They do so, however, *either* when the polariton mode is such that $|\alpha_k^+| \gg |\beta_k^+|$, in which case

$$Q_{k,\max}^{\text{sm}} = (e^{+2r_k} - 1) + \delta_k^+ \quad \text{for } \phi_k = (\varphi_k + \pi)/2 \ , \quad (2.27\text{a})$$

$$Q_{k,\min}^{\text{sm}} = (e^{-2r_k} - 1) + \delta_k^- \quad \text{for } \phi_k = \varphi_k/2 \ , \quad (2.27\text{b})$$

$$\delta_k^{\pm} \equiv \frac{|\alpha_k^+|^2}{|a_k \alpha_k^+|^2 + \sinh^2 r_k} \times \left[\frac{\sinh^2 r_k}{|\alpha_k^+|^2}[1 + \sinh(2r_k)] + |\beta_k^+| \left[\frac{e^{\pm 2r_k}}{|\alpha_k^+|} A'_{-1,k} - |\alpha_k^+| A_{1,k}\right]\right] ,$$

or clearly in the case of high photon intensity, for which one has

$$Q_{k,\max}^{\text{hi}} = e^{+2r_k} - 1 \quad \text{for } \phi_k = (\varphi_k + \pi)/2 \ , \quad (2.28\text{a})$$

$$Q_{k,\min}^{\text{hi}} = e^{-2r_k} - 1 \quad \text{for } \phi_k = \varphi_k/2 \ . \quad (2.28\text{b})$$

In both cases one simply has $\phi_k \cong \phi_k^a$. The functional dependence of ϕ_k is otherwise quite complicated.

Typical values for Q in the wave-vector region of interest to us are rather small and can be directly estimated from the results of Figs. 2, 4, and 5. For exciton polaritons in GaAs, sub-Poissonian statistics amounts to a Q^{hi} varying in the range $[-0.016, -0.020]$ and $[-0.018, -0.021]$ for UBP and LBP modes (see Fig. 1), respectively. For exciton polaritons in CuCl the deviations from Poissonian are of the same order of magnitude, though slightly bigger: Q^{hi} ranges between $[-0.079, -0.058]$ and $[-0.095, -0.06]$ for UBP and LBP modes, respectively. In each semiconductor the same values, with positive sign, apply to the case of super-Poissonian statistics, owing to the small r ($\sim 10^{-2}$) involved. On the other hand, when particular modes are selected ($|\alpha_k^+| \gg |\beta_k^+|$), one has to include the additional δ to the relevant Q^{sm}, which, however, does not sensibly modify the previous estimations for Q. In the wave-vector crossover region of the polariton spectrum, taking, e.g., $|a_k|^2 = 10$ (Appendix B), one has $\delta \approx 10^{-5}$ for GaAs and $\delta \approx 10^{-4}$ for CuCl.

D. Detection analysis

In the remainder of this section we discuss from a theoretical point of view the problem of detection of non-Poissonian features of the electromagnetic radiation associated with polaritons. According to the results above, the main requirement is the extraction of the polariton photon-number mean and variance [Eq. (2.24)]. The simplest attempt consists of a photon-statistical type of experiment in which the polariton light, carrying a number distribution P_n, falls directly into a photodetector. This would require a photodetector embedded in the medium where the bulk polariton is excited. A luminescent impurity center might be used. Here, however, we examine another scenario.

Any bulk polariton inside a crystal of finite extension has its counterpart at the surface: It is an evanescent electromagnetic wave[31] associated with a leaking bulk polariton excited in the medium.[32] A suitable photodetector device (PD) will make it possible to measure the photocurrent due to this leaking evanescent wave in the surface of the crystal, and schemes for implementing this can be developed.[33] For the present we assume this can be done and a possible schematic of the method which could be used is given in the inset of Fig. 3. Processing of the photocurrent generated by the leaking polariton radiation just above the surface provides a statistical distribution, say, $\mathcal{P}_n(T)$, of the number of photocounts recorded during repeated periods of time T. $\mathcal{P}_n(T)$ provides a record of the photodistribution $P_n(T)$. Distortions by *surface effects* (dissipations) and *nonunity quantum efficiency* of the photodetector need attention. These effects can be included in our description phenomenologically:[34] the *losses* due to the surface and nonideal detector can be modeled by the arrangement of Fig. 3 ascribing

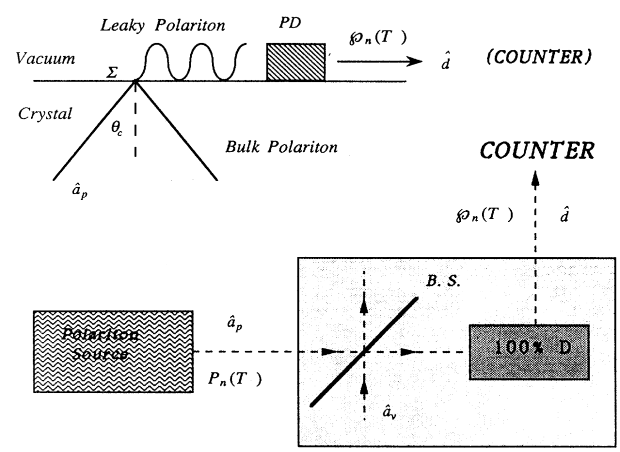

FIG. 3. Analysis of a direct-detection experiment. The electromagnetic field of a bulk polariton propagating inside the crystal at critical angle (θ_c) leaks off the surface Σ (onset), generating a running evanescent wave into the vacuum in contact with it which directly falls on to the photodetector (PD). The transmission at the beam splitter (BS) accounts for *surface losses* and the *nonunity quantum efficiency* of the photo detector. The detector D and the beam splitter model the surface-photodetector system. Practical detection schemes are more fully discussed in Ref. 33.

them to the loss of light at a beam splitter, which transmits only a fraction of the incident amplitude.[35,36] Thus, generally speaking, if $\hat{a}_p$ represents the polariton radiation field, the destruction operator of the "detected" field is

$$\hat{d}=\gamma^{1/2}\hat{a}_p+(1-\gamma)^{1/2}\hat{a}_v \ .$$

γ, the loss in photon units, accounts for both types of loss. To conserve energy (or preserve unitarity), the beam splitter must have a second input port into which a free field propagates. That accounts for the fluctuations associated with dissipation. It is assumed to be in a vacuum state ($\hat{a}_v$). The field represented by $\hat{d}$ is then assumed to be detected with 100% efficiency at the detector D. The photocount distribution produced by a similar direct detection scheme yields for the photocount mean and variance of the polariton leaking radiation

$$\langle \hat{\tilde{N}}^{\rm ph}\rangle \equiv \langle \hat{d}^{\dagger}\hat{d}\rangle=\gamma\langle \hat{N}^{\rm ph}\rangle \ , \tag{2.29}$$

and

$$\langle(\Delta\hat{\tilde{N}}^{\rm ph})^2\rangle=\gamma^2\langle(\Delta\hat{N}^{\rm ph})^2\rangle+\gamma(1-\gamma)\langle\hat{N}^{\rm ph}\rangle \ , \tag{2.30}$$

with $\langle(\Delta\hat{N}^{\rm ph})^2\rangle$ and $\langle\hat{N}^{\rm ph}\rangle$ given as in (2.22). The photon number appearing in the above expressions should be interpreted as the flux, in units of quanta per unit time, of photons associated with a given polariton mode, multiplied by the time T. The Mandel factor following from (2.29) and (2.30) is then

$$\tilde{Q}=\frac{\langle(\Delta\hat{\tilde{N}}^{\rm ph})^2\rangle-\langle\hat{\tilde{N}}^{\rm ph}\rangle}{\langle\hat{\tilde{N}}^{\rm ph}\rangle}=\gamma Q \quad (0<\gamma<1) \ . \tag{2.31}$$

Sub-Poissonian statistics, e.g., of the electromagnetic radiation associated with a bulk polariton, corresponding to negative Q's (in the crystal), thus produce sub-Poissonian statistics of the electromagnetic radiation associated with the leaking wave of a bulk polariton, indicated by negative $\tilde{Q}$'s (above the surface of the crystal). However, the magnitude of these nonclassical effects is attenuated by surface and detector losses. The attenuation is characterized by an effective efficiency $\gamma\equiv\gamma_D\gamma_S\cong 0.7$, where γ_D is the photodetector efficiency and γ_S the efficiency for propagation on the surface limited by dissipations such as, e.g., absorption, scattering, diffraction, and other extraneous dissipative effects. High detection quantum efficiency $\gamma_D>0.9$ can typically be obtained, whereas smaller values $\gamma_S\cong 0.8$ compete to surface propagation losses. In order to optimize the "useful" noise reduction in the radiation field associated with polaritons, one may excite modes with a large squeeze factor. An analogous discussion holds for the opposite case of super-Poissonian statistics.

E. Squeezing in the electromagnetic portion of the polariton

Classically,[3,5] an exciton-polariton wave has *both* the characteristics of an electromagnetic wave and of an exciton polarization wave. Quantum mechanically recall that $\hat{\Gamma}_{+k}$, the Fock space annihilation operator for a physical exciton-polariton mode k, can be separated to give the photon and exciton constituent particle operators [Eqs. (2.11) and (2.13)]. Its electromagnetic portion, described by a "dressed" photon of which we now analyze the quantum-optical properties, is for our purposes rewritten as

$$\hat{\Gamma}^{\rm ph}_{+k}\equiv|\tilde{\alpha}^{+}_{k}|c_{r_k}\hat{a}_{+k}+e^{2i\varphi_k}|\tilde{\alpha}^{-}_{k}|s_{r_k}\hat{a}^{\dagger}_{-k} \ ,$$

$$|\tilde{\alpha}^{\pm}_{k}|\equiv|\alpha^{\pm}_{k}|[|\alpha^{\pm}_{k}|^2c^2_{r_k}-|\alpha^{\mp}_{k}|^2s^2_{r_k}]^{-1/2} \tag{2.13c}$$

and an analogous expression for the mode $-k$. This equation emphasizes that the dressed photon field associated with the polariton inside the medium differs from the "bare" free-space photon field. Examining (2.13c) more closely, one notes that the photonic part of the polariton transformed operator has the form of a (two-mode) squeeze transformation from the bare photons $\hat{a}_{\pm k}$ to the dressed ones $\hat{\Gamma}^{\rm ph}_{+k}$. We emphasize once more that we reserve the term *optical polariton squeezing* for just the process converting bare $(\hat{a}_{\pm k})\rightarrow$dressed$(\hat{\Gamma}^{\rm ph}_{+k})$ photons according to Eq. (2.13c). The transformation (2.13c) is perfectly well defined for any value of k, however inspection of the expressions for $|\alpha^{+}_{k}|$ and $|\alpha^{-}_{k}|$ shows that at the crossover wave vector k_0 the squeeze transformation (2.13c) takes on a particularly simple form ($|\alpha^{+}_{k_0}|=|\alpha^{-}_{k_0}|$). In Fig. 4 we report $|\alpha^{+}_{k}|$ and $|\alpha^{-}_{k}|$ for UPB states in the vicinity of k_0. Below we will illustrate optical polariton squeezing by referring for simplicity to the particular wave-vector mode k_0.

Keeping in mind the restricted use of the term optical polariton squeezing, we then proceed to investigate the photon statistical properties connected with optical squeezing in exciton-polariton coherent states $|\gamma_{\pm k}\rangle_z$. The results of Sec. II C will then be used: namely, recall that in the case of high photon intensity one has a non-Poissonian photocount statistics characterized by [cf. Eq. (2.22b)]

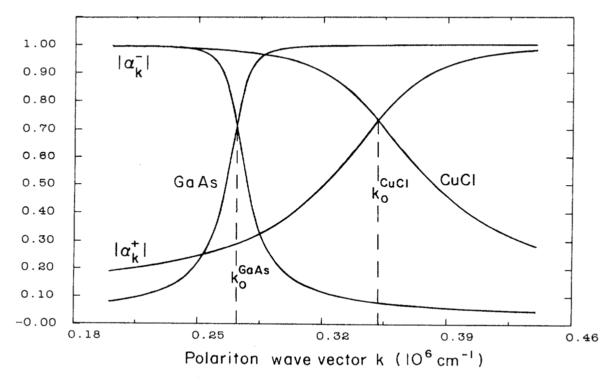

FIG. 4. Photon partial weights $|\alpha^{+}_{k}|$ and $|\alpha^{-}_{k}|$ in the vicinity of k_0 for CuCl and GaAs exciton-polariton UBP modes. Spanning the wave-vector interval from left to right, $|\alpha^{+}_{k}|$, and $|\alpha^{-}_{k}|$ assume increasing and decreasing values, respectively. A similar behavior occurs for the exciton partial weights $|\beta^{+}_{k}|$ and $|\beta^{-}_{k}|$ going in the opposite direction.

$$\langle (\Delta \hat{N}_{k_0}^{\mathrm{ph}})^2 \rangle \cong e^{\pm 2r_0} \langle \hat{N}_{k_0}^{\mathrm{ph}} \rangle \ , \quad Q_{k_0}^{\mathrm{hi}} \cong e^{\pm 2r_0} - 1 \ . \tag{2.32}$$

$+$ and $-$ respectively refer to a photon phase $\phi_{k_0}^a \cong (\varphi_{k_0}+\pi)/2$, or $\phi_{k_0}^a \cong \varphi_{k_0}/2$.

If restrictions on the photon intensity instead apply, one then should use the approximated result (2.22c). This can be done when $|\beta_k^+|$ is appropriately smaller than $|\alpha_k^+|$. This possibility is now investigated in detail and we begin by rewriting (2.16) as

$$\left| \frac{\alpha_k^+}{\beta_k^+} \right| = \kappa \left| \frac{(\varepsilon - \hbar\omega_0)(\varepsilon + \hbar ck)}{\sqrt{\hbar ck}} \right| \ . \tag{2.33}$$

$\kappa \equiv [\hbar\omega_0 \sqrt{4\pi\beta\hbar\omega_0}]^{-1}$ is a quantity characteristic of the dielectric material, while the rest of the ratio is mode dependent. At $k \cong k_0$, (2.33) takes on the form (cf. Fig. 1)

$$\left| \frac{\alpha_{k_0}^+}{\beta_{k_0}^+} \right| = \frac{1}{\sqrt{4\pi\beta_0}} \left| \frac{(\hbar\omega_{\mathrm{LT}}+\Delta_U)^2}{(\hbar\omega_0)^2} + \frac{2(\hbar\omega_{\mathrm{LT}}+\Delta_U)}{(\hbar\omega_0)} \right| \quad \text{(upper branch)} \tag{2.34}$$

$$\left| \frac{\alpha_{k_0}^+}{\beta_{k_0}^+} \right| = \frac{1}{\sqrt{4\pi\beta_0}} \left| \frac{\Delta_L^2 - 2\hbar\omega_0\Delta_L}{(\hbar\omega_0)^2} \right| \ . \quad \text{(lower branch)} \tag{2.35}$$

In principle, small oscillator strengths and transverse exciton energies along with large separations Δ are required to have satisfactorily large ratios. In Fig. 5 we report the ratio $|\alpha_k^+/\beta_k^+|$ when we sweep the wave vector across k_0. Exactly at k_0, $|\alpha_{k_0}^+|$ and $|\beta_{k_0}^+|$ are practically equal, at least for the materials here examined: therefore, approximating (2.22a) with (2.22c) may not be entirely justified by simply neglecting second- and higher-order of $|\beta_k^+/\alpha_k^+|$, but in general the values of n at k_0 and of the values of the A's must be also taken into account as discussed in Sec. II C. Interestingly enough, however, and especially in the GaAs sample, a slight detuning to the right of k_0 shows a rather fast decrease of the ratio $|\beta_k^+/\alpha_k^+|$, yet maintaining $|\tilde{\alpha}_k^+|$ and $|\tilde{\alpha}_k^-|$ satisfactorily close to one. In the LBP the use of the same result (2.22c) is more appropriate because the ratio $|\beta_k^+/\alpha_k^+|$ is slightly bigger than the one for the UBP. This is directly estimated from Eqs. (2.34) and (2.35).

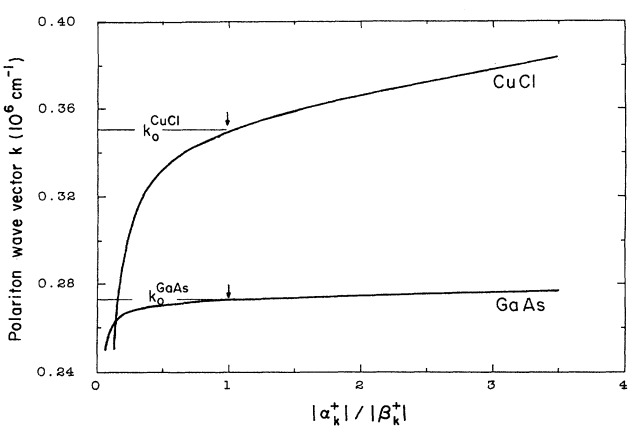

FIG. 5. Ratio of $|\alpha_k^+|$ and $|\beta_k^+|$ in the vicinity of the crossover k_0 (arrows) for UBP exciton polaritons in CuCl and GaAs.

In summary for *arbitrary* photon intensity, predictions on the nonclassical behavior of photons in a polariton coherent state due to optical squeezing are not in general as straightforward as for the *high*-photon-intensity case. The validity of the approximation (2.22c) must be carefully examined case by case.

Recalling (2.21) and considering that the present model can be applied to phonon polaritons as well, a bigger order of magnitude for r_0 can be expected in general because in this case the ratio $\Delta/\hbar\omega_0$ takes on values larger than in the exciton-polariton case. Typical values of r_0 can be estimated using (2.21). For (TO-) phonon polaritons in ionic crystals such as, e.g., GaP typical values are nearly one order of magnitude bigger than in the exciton-polariton case. Optical squeezing in phonon polaritons is being studied and will be reported in Ref. 37. Elsewhere,[33] we have analyzed practical detection schemes that clearly display any squeezing and non-Poissonian behavior present in the electromagnetic component of a mixed polariton wave.

III. NONCLASSICAL PROPERTIES OF GENERALIZED POLARITONS

In this section we investigate the quantum-statistical properties of a microscopic polariton Hamiltonian that is more general than the conventional Hopfield one of Eq. (2.1). Study of the generalized version is of importance for several reasons. This Hamiltonian can be shown to be a more complete version than the approximation of Hopfield: It is shown to contain additional terms that are dropped by the Hopfield model. The Hamiltonian does give a frequency dispersion equation ω_k for polaritons which agrees with experiment—as does the previous result [Eq. (2.1)]. The added terms complete the new Hamiltonian to be an element in the dynamical algebra Sp(8,$\mathbb{C}$). Within the framework of this polariton model Hamiltonian we give an interpretation to the origin of squeezing in polaritons. Indeed, one can show that it is isomorphic to a standard two-mode squeeze Hamiltonian. Certain entities—denoted by c—are squeezed, which are themselves mixtures of exciton and photon. These Bose quasiparticles are not the physical polaritons: Their mixing coefficients are not of the polariton, and their dispersion equations also differ. From the mathematical viewpoint, the c's arise as a first step when the total new Hamiltonian is diagonalized in two steps. In the second step an effective interaction $\hat{H}_{\mathrm{Int}}$ between the c's is taken into account and the completely diagonalized Hamiltonian giving polaritons is achieved by a squeeze transformation. From the physical point of view, if the effective interaction is supposed turned off, these bosons would be the proper normal modes and would be true physical quasiparticles of that resulting Hamiltonian. They are in fact the normal modes of the propagating component of the exciton-polariton field.

Two interpretations of the appearance of squeezing in polariton states are here presented. One, within Graham's framework,[38] relates to a sudden transition from states of the boson c field to states of the physical polariton field. In this context, the boson c plays the role of an intermediate quasiparticle in the transient (of time) it takes to turn on the interaction. The other interpretation relates to quantum correlations in polariton states between quasiparticles c of opposite wave vectors. We finally present results on the magnitude of squeezing in the exciton polariton for CuCl and GaAs. These would be contrasted with the corresponding results in Sec. II for the conventional truncated Hopfield Hamiltonian. We also examine time-dependent changes in the statistics of polariton coherent states and we demonstrate a cyclic reduction and/or increase of the quadratures noise in those states: Squeezing occurs twice per cycle. A detailed quantitative illustration is given for CuCl.

A. A generalized polariton Hamiltonian

We now introduce a polariton Hamiltonian that can be obtained by a microscopic theory of the dipolar matter-radiation interaction, retaining terms in the dipole-dipole interaction which are dropped in obtaining Eq. (2.1).[11,12] In the second quantized form we have

$$\begin{aligned}\hat{H}^{\text{pol}} &= \sum_{k\geq 0}\{\hat{H}^{\text{pol}}_{+k}+\hat{H}^{\text{pol}}_{-k}\}+\text{H.c.}\ ,\\ \hat{H}^{\text{pol}}_{+k}-h_k^0 &\equiv E_k^{\text{ph}}\hat{a}^{\dagger}_{+k}\hat{a}_{+k}+E_k^{\text{exc}}\hat{b}^{\dagger}_{+k}\hat{b}_{+k}\\ &\quad +B_k'\hat{a}_{+k}\hat{a}_{-k}-C_k'\hat{b}_{+k}\hat{b}_{-k}\\ &\quad +iA_{1k}\hat{a}_{-k}\hat{b}_{+k}+iA_{2k}\hat{a}^{\dagger}_{+k}\hat{b}_{+k}\ ,\end{aligned} \tag{3.1}$$

with

$$E_k^{\text{ph}}\equiv\frac{\hbar ck}{2}+B_k\ ,$$

$$E_k^{\text{exc}}\equiv\frac{\hbar\omega_0}{2}-C_k\ ,$$

$$h_k^0\equiv\frac{\hbar ck}{4}+\frac{\hbar\omega_0}{4}+\tfrac{1}{2}(B_k-C_k)\ .$$

$\hbar ck$ and $\hbar\omega_0$ are the bare photon and transverse exciton energies, whereas $\{A_{1k},A_{2k},B_k,B_k',C_k,C_k'\}$ are coefficients that parametrize the Hamiltonian. For the present we remark that these can be determined in principle *a priori* by the microscopic theory.[39] Our approach, however, is to treat them as phenomenological parameters that we can and do determine by having recourse to the experimentally measured polariton dispersion curves. By comparison to Eq. (2.1), note that terms explicitly incorporating bare exciton-exciton interactions are now included in Eq. (3.1). Moreover, as mentioned, this Hamiltonian realizes the dynamical algebra Sp(8,ℂ); other dynamical algebraic classifications are also possible.[39]

We proceed by diagonalizing $\hat{H}^{\text{pol}}$, but in two steps. This will permit us to formally demonstrate the isomorphism between the generalized Hamiltonian (3.1) and a standard two-mode squeezing Hamiltonian. First, perform a partial diagonalization so as to introduce new mixed quasiparticles and their residual interactions: We represent them by Bose operators $\hat{c}_{\pm k}$:

$$\begin{aligned}\hat{c}_{\pm k} &\equiv e^{i\psi_k^a}\alpha_k\hat{a}_{\pm k}+e^{i\psi_k^b}\beta_k\hat{b}_{\pm k}\ ,\\ \hat{c}^{\dagger}_{\pm k} &\equiv e^{-i\psi_k^a}\alpha_k\hat{a}^{\dagger}_{\pm k}+e^{-i\psi_k^b}\beta_k\hat{b}^{\dagger}_{\pm k}\ ,\end{aligned} \tag{3.2}$$

$$[\hat{c}_k,\hat{c}_{k'}]=[\hat{c}^{\dagger}_k,\hat{c}^{\dagger}_{k'}]=0\ ,\quad [\hat{c}_k,\hat{c}^{\dagger}_{k'}]=\delta_{k,k'}\ . \tag{3.3}$$

The α_k's and β_k's are real. After some algebraic manipulations, (3.1) can be transformed to the following form:

$$\hat{H}^{\text{pol}}=\sum_{k>0}\{\hat{H}^{\text{pol}}_{+k}+\hat{H}^{\text{pol}}_{-k}\}+\text{H.c.}\equiv\hat{H}_0+\hat{H}_{\text{Int}}\ , \tag{3.4}$$

$$\hat{H}_0\equiv\tfrac{1}{2}\sum_{k>0}\Omega_k(\hat{c}^{\dagger}_{+k}\hat{c}_{+k}+\hat{c}^{\dagger}_{-k}\hat{c}_{-k}+1)+\text{H.c.}\ , \tag{3.5}$$

$$\hat{H}_{\text{Int}}\equiv 2\sum_{k>0}\zeta_k\hat{c}_{+k}\hat{c}_{-k}+\text{H.c.} \tag{3.6}$$

$\hat{H}^{\text{pol}}$ now depends on the six parameters $\Omega_k,\zeta_k,\alpha_k,\beta_k,\psi_k^a,\psi_k^b$ that are related to the old ones by the transformations

$$\begin{aligned}&\Omega_k\alpha_k^2=\hbar ck+2B_k\ ,\quad \Omega_k\beta_k^2=\hbar\omega_0-2C_k\ ,\\ &\zeta_k\alpha_k^2e^{2i\psi_k^a}=B_k'\ ,\quad \zeta_k\beta_k^2e^{2i\psi_k^b}=-C_k'\ ,\\ &2\zeta_k\alpha_k\beta_ke^{i(\psi_k^a+\psi_k^b)}=iA_{1k}\ ,\quad \Omega_k\alpha_k\beta_ke^{i(\psi_k^b-\psi_k^a)}=iA_{2k}\ .\end{aligned} \tag{3.7}$$

Here we take $\psi_k^a=0$ and $\psi_k^b=\pi/2$, $\alpha_k\equiv\cos\theta_k$, and $\beta_k\equiv\sin\theta_k$. The later identification is consistent with the commutation rules (3.3). The number of parameters in (3.4) then reduces to $\Omega_k,\zeta_k,\theta_k$. The next step of our (diagonalization) procedure proceeds by introducing a second Bose quasiparticle,

$$\hat{\eta}_{\pm k}=M_k\hat{c}_{\pm k}+N_k\hat{c}^{\dagger}_{\pm k}\ ,\quad M_k^2-N_k^2=1\ , \tag{3.8}$$

in terms of which we can rewrite $\hat{H}^{\text{pol}}$ in the fully diagonalized form

$$\hat{H}^{\text{pol}}=\sum_k\varepsilon_k(\hat{\eta}^{\dagger}_k\hat{\eta}_k+\tfrac{1}{2})\ . \tag{3.9}$$

At this stage we observe that finding the coefficients in $\hat{H}^{\text{pol}}_k$ $(A_{1k},A_{2k},B_k,B_k',C_k,C_k')$, so that for every mode: (1) the eigenenergy ε_k reproduces the experimentally measured exciton-polariton energy and (2) the parameters of $\hat{H}^{\text{pol}}_k$ satisfy consistently the isomorphism (3.7), is equivalent to showing that $\hat{H}^{\text{pol}}_k$ represents a physical polariton Hamiltonian and that $\hat{H}^{\text{pol}}_k$ implies squeezing. Equation (3.4) is indeed the usual form of a two-mode squeezing Hamiltonian,[20] and squeezing occurs with respect to the $\hat{c}_{\pm k}$'s, mixed bosons whose dispersion is given by Ω_k, and whose residual interaction is $\hat{H}_{\rm Int}$ with interaction strength measured by ζ_k. In order not to interrupt the continuity of the presentation, we will show in Appendix C that it is in fact possible to fulfill the points (1) and (2) above. Thus,

$$\hat{H}^{\rm pol}(A_{1k},A_{2k},B_k,B'_k,C_k,C'_k)=\hat{H}^{\rm sq}(\Omega_k,\zeta_k,\theta_k)\ , \quad (3.10)$$

and ε_k is the energy of the physical polariton normal mode $\hat{\eta}_{\pm k}$ which we rewrite as [cf. Eq. (3.2)]

$$\hat{\eta}_{\pm k}\equiv\hat{\eta}_{\pm k}(x,y,z,w)=x\hat{a}_{\pm k}+y\hat{b}_{\mp k}+z\hat{a}^{\dagger}_{\mp k}+w\hat{b}^{\dagger}_{\mp k}\ . \quad (3.11)$$

Now we characterize the amount of squeezing relevant to a polariton. We exploit the isomorphism (3.10). We first connect the polariton eigenenergy and eigenvectors to the parameters of the squeeze Hamiltonian (3.4), e.g., by requiring that

$$[\hat{\eta}_k,\hat{H}^{\rm sq}]=\varepsilon_k\hat{\eta}_k\ . \quad (3.12)$$

The resulting secular equation,

$$\begin{vmatrix} (\Omega_k\cos^2\theta_k-\varepsilon_k) & -\frac{i}{2}\Omega_k\sin2\theta_k & -2\zeta_k\cos^2\theta_k & -i\zeta_k\sin2\theta_k \\ \frac{i}{2}\Omega_k\sin2\theta & (\Omega_k\sin^2\theta_k-\varepsilon_k) & -i\zeta_k\sin2\theta_k & 2\zeta_k\sin^2\theta_k \\ 2\zeta_k\cos^2\theta_k & -i\zeta_k\sin2\theta_k & -(\Omega_k\cos^2\theta_k+\varepsilon_k) & -\frac{i}{2}\Omega_k\sin2\theta_k \\ -i\zeta_k\sin2\theta & -2\zeta_k\sin^2\theta_k & \frac{i}{2}\Omega_k\sin2\theta_k & -(\Omega_k\sin^2\theta_k+\varepsilon_k) \end{vmatrix}=0\ ,$$

yields the eigenvalues

$$\varepsilon_k^2=\Omega_k^2-4\zeta_k^2 \quad (3.13)$$

and the components (eigenvectors) of $\hat{\eta}_k$ as functions of $\Omega_k,\zeta_k,\theta_k,\varepsilon_k$. For our purposes it suffices to give

$$\left|\frac{y}{x}\right|=\tfrac{1}{2}\varepsilon_k\sin(2\theta_k)(\Omega_k\varepsilon_k+\Omega_k^2-4\zeta_k^2)\ ,$$
$$\left|\frac{w}{x}\right|=\sin(2\theta_k)\zeta_k\varepsilon_k^2\ . \quad (3.14)$$

Next we diagonalize $\hat{H}^{\rm sq}$ by a squeeze canonical transformation, i.e.,

$$\hat{H}^{\rm sq}=\sum_k\bar{\varepsilon}_k(\hat{\mu}^{\dagger}_k\hat{\mu}_k+\tfrac{1}{2})\ ,\quad \hat{\mu}_{\pm k}=\hat{S}(r_k,\varphi)\hat{c}_{\pm k}\hat{S}^{\dagger}(r_k,\varphi)\ . \quad (3.15)$$

Here,

$$\hat{S}(r_k,\varphi_k)=\exp[r_k(e^{-2i\varphi_k}\hat{c}_{+k}\hat{c}_{-k}-e^{2i\varphi_k}\hat{c}^{\dagger}_{+k}\hat{c}^{\dagger}_{-k})] \quad (3.16)$$

is a two-mode squeeze unitary operator,[21,40] r_k and φ_k being referred to as the squeeze factor and angle, respectively. Evidently, both $\hat{\mu}_k$ and $\hat{\eta}_k$ diagonalize the same Hamiltonian [cf (3.12) to (3.15)] so they are equivalent,[41] and the eigenergies are independent of the basis representation, i.e., $\bar{\varepsilon}_k=\varepsilon_k$. Using the properties of $\hat{S}$ and the explicit definition of $\hat{c}_{\pm k}$ in terms of the original vacuum photon and exciton operators, one can rewrite $\hat{\mu}_{\pm k}$ in (3.15):

$$\begin{aligned}\hat{\mu}_{\pm k}=\hat{a}_{\pm k}\cos\theta_k\cosh r_k+\hat{b}_{\pm k}e^{i\psi_k^b}\sin\theta_k\cosh r_k\\ +a^{\dagger}_{\mp k}e^{2i\varphi_k}\cos\theta_k\sinh r_k\\ +b^{\dagger}_{\mp k}e^{-i(\psi_k^b-2\varphi_k)}\sin\theta_k\sinh r_k\ ,\end{aligned} \quad (3.17)$$

whereas term by term comparison of (3.17) and (3.11) provides the components of $\hat{\eta}_k$ as functions of θ_k,φ_k,r_k, e.g.,

$$|y|=\sin\theta_k\cosh r_k\ ,\quad |w|=\sin\theta_k\sinh r_k\ . \quad (3.18)$$

Finally, by eliminating θ_k from (3.14) and (3.18), the expression for the polariton squeeze factor r_k follows:

$$r_k=\tanh^{-1}\left[\frac{2\zeta_k}{\varepsilon_k+\Omega_k}\right]\ . \quad (3.19)$$

The amount of intrinsic squeezing can be tuned by changing the wave-vector–frequency relation of the polariton mode that is excited. This has been calculated in Fig. 6 for UBP and LBP modes in different semiconductors.

An obvious question may arise about the interpretation of $\hat{H}^{\rm pol}$ in the form given by Eq. (3.4). Accordingly, the generation of squeezing in polariton states would be by the action of the interaction Hamiltonian $\hat{H}_{\rm Int}$. The system suggested here to generate squeezing in polaritons is similar to the system of a two-photon laser where two photons of the same frequency with wave vector $+k$ and $-k$ can be absorbed in a transition which is of second order in $\mathbf{A}\cdot\mathbf{p}$.[42] In the present case the counterpropagating terms are "spontaneously" present in the Hamiltonian, using a running wave quantization of the bare photons and excitons. See Ref. 3 for the analogous case of phonons. A degenerate parametric amplifier interpretation may as well be suitable to $\hat{H}^{\rm pol}$ in (3.4): This is in fact the

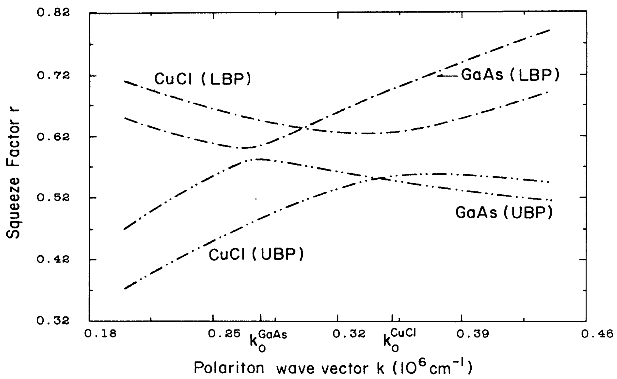

FIG. 6. Magnitude of the squeeze factor in CuCl and GaAs exciton- polaritons as a function of k calculated within the *generalized* polariton model for LBP (—·—·—·) and UBP (—..—..—..—..) modes in vicinity of the crossing wave vector k_0.

Hamiltonian for a typical two-photon device.[13,21]

The analogies are, however, only formal.[43] The physical scenario here is different, yet the quantum statistics is the same. The $\hat{c}$'s are actual quasiparticles of the photon-exciton system, namely, they are the quanta of the *propagating* component of the polariton field. They have been studied in a different context.[44] Thus, $\hat{H}^{\text{pol}}$ given in Eq. (3.4) can be physically interpreted. $\hat{H}_0$ is the free evolution of the quasiparticle $\hat{c}$ that is being squeezed. $\hat{H}_{\text{Int}}$, giving the squeezing, is the interaction that originates from mixing $\hat{c}$'s with opposite wave vectors. This type of interaction is intrinsic to the polariton "construction" and it does produce correlations between counterpropagating $\hat{c}$'s in a polariton state. Specifically, this correlation causes pairs $(\hat{c}_{+k},\hat{c}_{-k})$ to be created or annihilated: That is apparent in the interaction part of the Hamiltonian, which contains terms that are purely quadratic in the creation and annihilation operators. The connection between wave-vector mode correlations and squeezing will be discussed later. Since the interaction that produces squeezing in the propagating component of the polariton field is built in $\hat{H}^{\text{pol}}$, polariton squeezing will be referred to as *intrinsic* squeezing.

A time-dependent analysis can be implemented, in which one considers the finite time (transient) it takes to turn on the effective interaction (3.6). Thus, the boson $\hat{c}$ is the actual physical quasiparticle that describes the photon-exciton system during the transient. Since the latter constitutes only an intermediate stage during the process of formation of a polariton state, the $\hat{c}$'s can be referred to as *intermediate* quasiparticles. It is worthwhile stressing that before $\hat{H}_{\text{Int}}$ is turned on, the bare photon and bare exciton fields and the propagating $\hat{c}$ field of the polariton share the same vacuum state $|0\rangle$;[45] this implies that during this intermediate stage no change of the quantum statistics occurs. The connection between the turning on of the interaction $\hat{H}_{\text{Int}}$, with the subsequent energy change $\Omega_k \to \varepsilon_k$ and change in the structure (squeezing) of the vacuum state of the photon-exciton system, will be discussed in Sec. III B.

B. Time evolution from coherent to squeezed state

Here, continuing with the two-mode squeeze framework, we take advantage of the structure of polariton states as squeezed with respect to the states of the boson $\hat{c}$ field of the polariton to explore an interesting time-development scenario. In turn this will permit us to discuss some of the nonclassical statistical properties associated with exciton polaritons. Again, as in Sec. II, we will consider the polariton system in a coherent state. We introduce such a state and the properties which we will need by first recalling the separation of the generalized polariton Hamiltonian into $\hat{H}_0$ and $\hat{H}_{\text{Int}}$. Coherent states of the diagonal part $\hat{H}_0$ are defined either as the eigenstates of the (mixed) destruction operators $\hat{c}_{\pm k}$,

$$\hat{c}_{\pm k}|c_{\pm k}\rangle = c_{\pm k}|c_{\pm k}\rangle \ , \tag{3.20}$$

or by displacing the vacuum state $|0\rangle$ of the bare photon and bare exciton system,

$$|c_{\pm k}\rangle = e^{-(1/2)(|c_{+k}|^2+|c_{-k}|^2)} e^{c_{-k}c^{\dagger}_{-k}} e^{c_{+k}c^{\dagger}_{+k}}|0\rangle \equiv D|0\rangle \ . \tag{3.21}$$

It is precisely the state vector $|c_{\pm k}\rangle$ so constructed that is a two-mode coherent state[21] for the free Hamiltonian $\hat{H}_0$.

Next we introduce the squeezed state that one obtains by applying $\hat{S}$ on the $|c_{\pm k}\rangle$'s.

$$\hat{S}(r_k,\varphi)|c_{\pm k}\rangle \equiv |\mu_{\pm k}(r_k,\varphi)\rangle \equiv |\mu_{\pm k}\rangle_z \ . \tag{3.22}$$

Using the properties of $\hat{S}$ and those of the displacement operator $\hat{D}$ yields

$$|\mu_{\pm k}\rangle_z = e^{-(|\bar{\mu}_{+k}|^2|+\bar{\mu}_{-k}|^2)/2} \times \sum_{n_+,n_-=0}^{\infty} \frac{(\bar{\mu}_{+k})^{n+k}(\bar{\mu}_{-k})^{n-k}}{\sqrt{n_{+k}!n_{-k}!}} |\gamma_{\pm k}\rangle_z \ , \tag{3.23}$$

where

$$\hat{H}^{\text{pol}}|\gamma_{\pm k}\rangle_z = \varepsilon_k(\hat{n}_{\pm k}+\tfrac{1}{2})|\gamma_{\pm k}\rangle_z \ , \qquad \hat{H}^{\text{pol}} \equiv \sum_k \varepsilon_k(\hat{\mu}^{\dagger}_k\hat{\mu}_k+\tfrac{1}{2}) \ . \tag{3.24}$$

The $\bar{\mu}_{\pm}$'s are complex numbers. Since the $|\gamma_{\pm k}\rangle_z$'s are polariton energy eigenstates, from (3.23), $|\mu_{\pm k}\rangle_z$ represents a polariton coherent state.

Let us now assume it is possible to prepare a polariton cavity where the photon-exciton system inside be described for $t<0$ by the Hamiltonian $\hat{H}_0$. Let the initial state of this system be the coherent state (3.20). If at $t=0$, $\hat{H}_{\text{Int}}$ is turned on,[46] the system develops for $t>0$ under the total Hamiltonian $\hat{H}_0$ plus $\hat{H}_{\text{Int}}$, i.e., $\hat{H}^{\text{pol}}$. Conversely, if $\hat{H}_{\text{Int}}$ were always absent, the state of the system will remain (3.20) for all time. A plausible physical mechanism accounting for the switching on of this interaction is a sudden frequency change[38,47] during a Frank-Condon transition

$$|c_{\pm k}\rangle_{(t=0)} \rightarrow |\mu_{\pm k}\rangle_{z\,(t=0)} \,. \tag{3.25}$$

Thus for positive times,

$$|\mu_{\pm k}\rangle_{z\,(t>0)} = \exp(-it\hat{H}^{\text{pol}}/\hbar)|\mu_{\pm k}\rangle_{z\,(t=0)} \tag{3.26}$$

gives the temporal evolution of a polariton coherent state with energy $\varepsilon_k \neq \Omega_k$. One notes that (two-mode) coherent states of the bare photon–bare exciton field are also coherent states of the boson $\hat{c}$ field of the polariton, which, however, are not coherent states for $\hat{H}^{\text{pol}}$ [cf. (3.22)]. Therefore, the energy change $\Omega_k \rightarrow \varepsilon_k$ that ultimately produces squeezing in the (boson) $\hat{c}$ field of the polariton is responsible for the change in the statistics.

In the initial state ($t<0$) the wave function of the photon-exciton system is a wave packet whose width remains constant whereas for $t>0$ the wave packet of the corresponding wave function has a complicated time development in that its width changes periodically in time over the oscillation period of the polariton. In order to examine these time-varying features, we find it advantageous to decompose the propagating field of the polariton into two amplitude components 90° out of phase. The operators that correspond to these two *quadrature* components are

$$\hat{X}_k^q = \frac{1}{2\sqrt{\hbar}}\left[\alpha_k\sqrt{\omega_k}\,\hat{q}_k^{\text{ph}} - \frac{\beta_k \hat{p}_k^{\text{exc}}}{\sqrt{\omega_k}}\right] + (k \rightarrow -k)\,, \tag{3.27}$$

$$\hat{X}_k^p = \frac{1}{2\sqrt{\hbar}}\left[\beta_k\sqrt{\omega_k}\,\hat{q}_k^{\text{exc}} + \frac{\alpha_k \hat{p}_k^{\text{ph}}}{\sqrt{\omega_k}}\right] + (k \rightarrow -k)\,. \tag{3.28}$$

$\hat{q}_k^{\text{ph}} = \sqrt{\hbar/2\omega_k}(\hat{a}_k + \hat{a}_k^\dagger)$ and $\hat{p}_k^{\text{ph}} = -i\sqrt{\hbar\omega_k/2}(\hat{a}_k - \hat{a}_k^\dagger)$ are the canonical position and momentum operators for photons or excitons if $a_k \rightarrow b_k$. Their variances $\langle|\Delta\hat{X}_k^{q,p}|^2\rangle$ directly relate to the widths $(\Gamma_k^{q,p})$ of the wave functions that represent the states of the propagating field of the polariton in the coordinate and momentum representation, respectively.[22] Here (3.27) and (3.28) are canonical variables $[\hat{X}_k^q, \hat{X}_k^p] = i/2$; hence, their variances satisfy the Heisenberg uncertainty principle

$$\langle|\Delta\hat{X}_k^q|^2\rangle_t \langle|\Delta\hat{X}_k^p|^2\rangle_t \geq \tfrac{1}{16}$$

which must hold for both states $|c_{\pm k}\rangle$ and $|\mu_{\pm k}\rangle_z$.

1. *Quadrature noise: Coherent state of the propagating field*

In the initial state ($t<0$) the variance of $\hat{X}_k^q$ is

$$_t\langle c_{\pm k}|\Delta\hat{X}_k^q|^2|c_{\pm k}\rangle_t = \tfrac{1}{4} = \Gamma_k^{(0)q} \equiv \Gamma_k^0 \tag{3.29}$$

and that of the conjugate quadrature is

$$_t\langle c_{\pm k}|\Delta\hat{X}_k^p|^2|c_{\pm k}\rangle_t = \tfrac{1}{4} = \Gamma_k^{(0)p} \equiv \Gamma_k^0 \,. \tag{3.30}$$

As expected for a coherent state, the two variances are constant in time. Further, coherent states are minimum-uncertainty states so that the equality holds for the Heisenberg uncertainty relations, at all times.[22] Once again, let us emphasize that the states $|c_{\pm k}\rangle$ are coherent states of the *free* part $\hat{H}_0$ of the partially diagonalized Hamiltonian and that they refer to the intermediate quasiparticle $\hat{c}$, but *not* to physical polaritons.

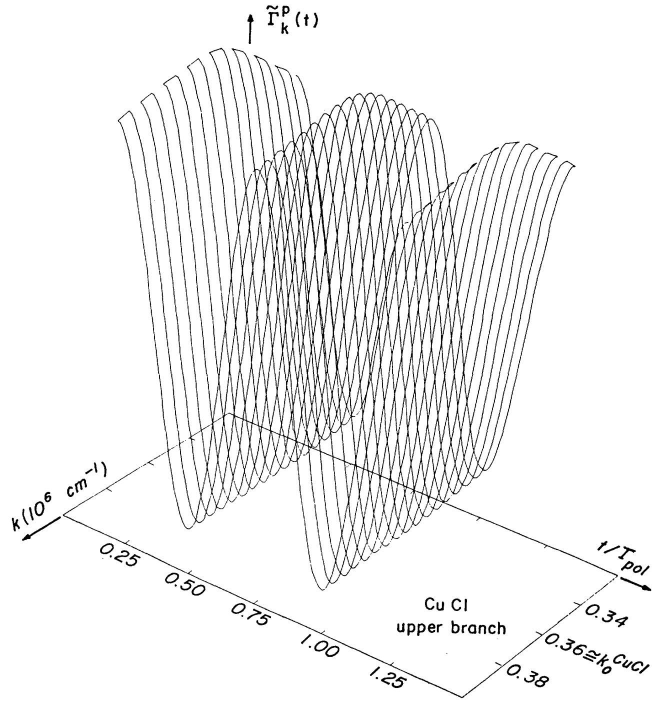

FIG. 7. Normalized envelope of fluctuations $[\tilde{\Gamma}_k^p(t) \equiv \Gamma_k^p(t)/\Gamma_k^0]$ of the propagating field of a polariton in a UBP CuCl exciton-polariton coherent state. We exhibit the time dependence over the polariton cycle while the wave vector sweeps through the crossover (k_0) region.

2. *Quadrature noise: Coherent polariton state*

In a polariton coherent state ($t>0$) the variances of the two quadratures are

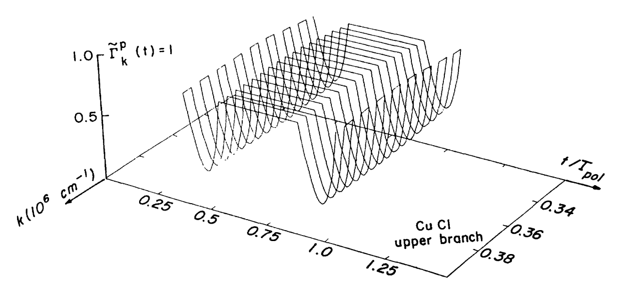

FIG. 8. Horizontal section of Fig. 7. Time and wave-vector dependence of $\tilde{\Gamma}_k^p(t)$ in the squeezing region. In a polariton coherent state the flucutations $\tilde{\Gamma}_k^p(t)$ reduce twice, half a cycle distant from one another, below the value $\frac{1}{4}$ associated with the vacuum state. The squeezing arrives a maximum $\approx 70\%$.

$$_{t,z}\langle \mu_{\pm k}|\Delta \hat{X}_k^q|^2|\mu_{\pm k}\rangle_{t,z} \equiv \Gamma_k^q(t) = \frac{e^{-2r_k}}{4} + \frac{\sinh(2r_k)}{2}\sin^2(\omega_k t) \quad (3.31)$$

and

$$_{t,z}\langle \mu_{\pm k}|\Delta \hat{X}_k^p|^2|\mu_{\pm k}\rangle_{t,z} \equiv \Gamma_k^p(t) = \frac{e^{+2r_k}}{4} - \frac{\sinh(2r_k)}{2}\sin^2(\omega_k t) \; . \quad (3.32)$$

With respect to the Heisenberg uncertainty relations, the inequality holds at each time also for these states, yet note that here the equality only holds when $\cos(2\omega_k t)=1$. As time evolves these states do *not* constantly remain minimum-uncertainty states.[22] This has been analyzed in detail over a time interval corresponding to a polariton cycle and for different modes: Results are reported in Figs. 7 and 8. For a certain interval of time (ΔT_{sq}), twice in a cycle, one of the variances becomes smaller than Γ_k^0 (coherent or vacuum value), while the conjugate one becomes larger than Γ_k^0 in order to enforce the Heisenberg principle. ΔT_{sq} can be evaluated for each energy–wave vector polariton mode, as well as the variable amount of squeezing during ΔT_{sq}. The numerical evaluation of ΔT_{sq} with relative squeezing is discussed in Appendix D: Results are reported in Fig. 9 for an illustrative example of CuCl UBP exciton polaritons, for which the typical ΔT_{sq} is approximately a fraction of femtoseconds.

In conclusion, we have shown that the probability density distribution for a polariton coherent state changes remarkably over the polariton cycle. In particular, in this state the strength of the propagating field $\hat{c}$ of the polariton exhibits an uncertainty envelope (fluctuations) that is periodically reduced far below the classical value $\frac{1}{4}$ associated with the vacuum state: This reduction, that is, squeezing, occurs during a time interval ΔT_{sq} and, within it, takes on different amounts depending on the polariton mode that is populated.

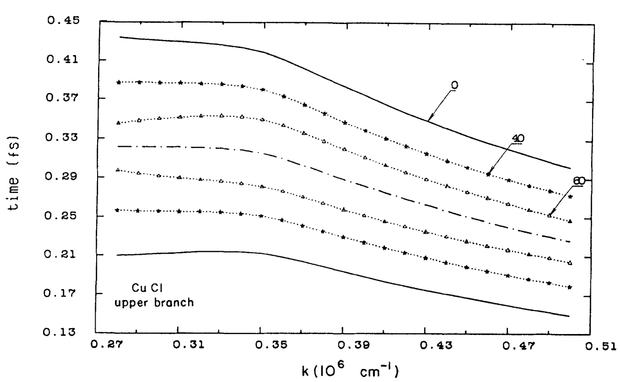

FIG. 9. Variation of the amount of squeezing in a UBP CuCl exciton polariton during ΔT_{sq}^{CuCl}. "0" refers to the case of no squeezing, i.e., $\Gamma_k^p(t)=\Gamma_k^0$; 40 (star) refers to the case in which $\Gamma_k^p(t)\le 0.6\Gamma_k^0$, while 60 (triangle) refers to the case in which $\Gamma_k^p(t)\le 0.4\Gamma_k^0$. Proceeding downwards, the above explanation repeats itself. Twice the vertical distance between, e.g., two given triangles yields the time interval during which the width of the envelope of fluctuations falls below nearly $\frac{1}{3}$ of the coherent vacuum value, whereas twice the vertical distance between the solid lines gives ΔT_{sq}^{CuCl}. Here (—·—·—·) represents the quarter period: $t=T_{pol}/4$ (cf. Figs. 7 and 8).

C. Polariton squeezing and wave-vector correlations

In this section we examine quantum correlations as a different mechanism to implement squeezing in polaritons. The definition of correlation to be used is briefly introduced. Let us consider two subsystems associated with opposite directions of propagation i.e., $+\mathbf{k}$ and $-\mathbf{k}$. Quantum correlations between this pair of subsystems can give rise to squeezeed fluctuations in the quadratures (3.27) and (3.28). If the state in which the pair of sybsystems is prepared can be described by a density matrix ρ, then the reduced density matrices that describe the properties of the subsystems relevant to each direction of propagation are $\rho_{\pm k}=\mathrm{tr}_{\pm k}\rho$. A standard approach can then be used to analyze these correlations, namely, if $\rho=\rho_{+k}\otimes\rho_{-k}$, the subsystems under consideration are uncorrelated, while if $\rho\neq\rho_{+k}\otimes\rho_{-k}$, then they are correlated. In order to develop the discussion to the (squeezing) properties of observables, we consider the two sets of operators $\{\hat{c}_{+k},\hat{c}_{+k}^\dagger\}$ and $\{\hat{c}_{-k},\hat{c}_{-k}^\dagger\}$ defined in each of the two subsystems and acting on the space of the $+\mathbf{k}$ subsystem and $-\mathbf{k}$ subsystem, respectively. A measure of the quantum fluctuations of $\hat{c}_{+k}$ or $\hat{c}_{-k}$ using the mean-square uncertainty gives

$$\langle|\Delta\hat{c}_{\pm k}^2|\rangle = \mathrm{tr}(\rho_{\pm k}\hat{c}_{\pm k}^2) - [\mathrm{tr}(\rho_{\pm k}\hat{c}_{\pm k})]^2 \; . \quad (3.33)$$

Depending only on the reduced densities $\rho_{\pm k}$, $\langle|\Delta\hat{c}_{\pm k}^2|\rangle$ are independent of whether or not in the state $|\;\rangle$ $\hat{c}_{+k}$ and $\hat{c}_{-k}$ are correlated. The variances above do not tell us anything about the correlations between the two subsystems. The latter can instead be studied by investigating the properties of operators acting on "both" subsystems. Consider, for instance, the operator $\hat{V}\equiv\hat{c}_{+k}+\hat{c}_{-k}$ whose variance is

$$\langle|\Delta\hat{V}|^2\rangle = \sum_{l=\pm k}\langle|\Delta\hat{c}_l|^2\rangle + \tilde{c} \; , \quad (3.34)$$

with

$$\langle|\Delta\hat{c}_{\pm k}|^2\rangle \equiv \tfrac{1}{2}\langle\hat{c}_{\pm k}^\dagger\hat{c}_{\pm k}+\hat{c}_{\pm k}\hat{c}_{\pm k}^\dagger\rangle - |\langle\hat{c}_{\pm k}\rangle|^2$$

and

$$\tilde{c} = \sum_{l,m=\pm k}[\langle\hat{c}_l^\dagger\hat{c}_m\rangle - \langle\hat{c}_l^\dagger\rangle\langle\hat{c}_m\rangle]$$

(real). In principle, the variance of such an operator indicates whether or not in the state $|\;\rangle$ the two subsystems are correlated.

For the purpose of the present paper let us recall the quadratures $\hat{X}_k^{q,p}$ by which we characterized the polariton squeezing in Sec. III B. $\hat{X}_k^q$ is the sum of two operators, one acting on each of the two subsystems $+\mathbf{k}$ and $-\mathbf{k}$, and it is related to $\hat{V}$. The relevant variance is

$$\langle|\Delta\hat{X}_k^q|^2\rangle=\tfrac{1}{4}(\langle|\Delta\hat{c}_{+k}|^2\rangle+\langle|\Delta\hat{c}_{-k}|^2\rangle+2\,\mathrm{Re}d_k+\tilde{c}+\mathrm{Re}\delta)$$
$$=\tfrac{1}{4}(\langle|\Delta\hat{V}|^2\rangle+2\,\mathrm{Re}d_k+\mathrm{Re}\delta)\;, \tag{3.35}$$

where $\delta\equiv\sum_k\langle\hat{c}_k^2\rangle=\langle\hat{c}_k\rangle^2$.

$$d_k\equiv\langle\hat{c}_{+k}\hat{c}_{-k}\rangle-\langle\hat{c}_{+k}\rangle\langle\hat{c}_{-k}\rangle$$

is a measure of the *correlation* between the quasiparticles $\hat{c}_{+k}$ and $\hat{c}_{-k}$. Squeezing, that is, the reduction of the variance (3.35) below $\frac{1}{4}$, can now be investigated in terms of correlations. For a polariton coherent state (3.23) it is easy to show that $\tilde{c}$ and δ vanish, leaving

$$\langle|\Delta\hat{X}_k^q|^2|\rangle=\tfrac{1}{4}[\langle|\Delta\hat{c}_{+k}|^2\rangle+\langle|\Delta\hat{c}_{-k}|^2\rangle]+\tfrac{1}{2}\,\mathrm{Re}d_k\;. \tag{3.36}$$

Further,

$$\mathrm{Re}d_k=-\cos(2\varphi_k)\cosh r_k\sinh r_k \tag{3.37}$$

and

$$\langle|\Delta\hat{c}_{+k}|^2\rangle+\langle|\Delta\hat{c}_{-k}|^2\rangle=\cosh(2r_k)\ge 1\;, \tag{3.38}$$

so that squeezing in the q quadrature depends in general on the relative magnitude of the terms on the right-hand side of Eq. (3.36). In the range of polariton modes examined in this paper, correlations contribute to the fluctuations in the quadrature $\hat{X}^q$ in CuCl and GaAs exciton polaritons, respectively, with (maximum) amounts of 0.6 and 0.66 for UBP modes, and slightly higher amounts, i.e., 0.95 and 1.18, for LBP modes. An analogous discussion holds for the conjugate quadrature $\hat{X}_k^p$. Squeezing is achieved provided that

$$\mathrm{ctgh}2r_k-\mathrm{sech}2r_k<\cos 2\varphi_k\;, \tag{3.39}$$

which is satisfied for phases such that $|2\varphi_k|<\pi/2$, with $r_k\ge 0$ in the present treatment (cf. Fig. 1). However, the main result of this section is indeed (3.37): It connects the correlations in a polariton coherent state between $\hat{c}$'s with opposite wave vectors to squeezing. Absence of correlations $d_k=0$, i.e., $r_k=0$ from Eq. (3.37), implies an equal sign in (Eq. (3.38) and $\langle|\Delta X_k^q|^2\rangle\to\frac{1}{4}$, i.e., no squeezing. The converse is also true. In order to have correlations between $\hat{c}_{+k}$ and $\hat{c}_{-k}$, the requirement of squeezing is necessary and sufficient.

In our polariton system is the polarization of the exciton that produces the correlation between the $\hat{c}$'s. In addition to our exciton-polariton system, the viewpoint that correlations can give rise to squeezing has been shown to be true for multimode squeezed states of light,[48] and for dipole fluctuations in multiatom squeezed states.[49]

Another parameter one could analyze in the present context is the *number correlation*

$$D_{n_k}\equiv\langle\hat{n}_{+k}\hat{n}_{-k}\rangle-\langle\hat{n}_{+k}\rangle\langle\hat{n}_{-k}\rangle\;,\quad \hat{n}_{\pm k}\equiv\hat{c}^\dagger_{\pm k}\hat{c}_{\pm k}. \tag{3.40}$$

This can be evaluated noting that

$$4D_{n_k}\equiv\langle|\Delta\hat{H}_0|^2\rangle-\langle|\hat{n}_{+k}-\hat{n}_{-k}|^2\rangle\;.$$

The number difference is a constant of motion with respect to $\hat{H}^{\mathrm{pol}}$ and vanishes if we take the initial number of quasiparticles $\hat{c}$ to be equal in the two modes of opposite wave vector. The other term can be calculated, and in a polariton coherent state one has ($\mu_{\pm k}$ real)

$$D_{n_k}=\frac{\mu_{+k}^2+\mu_{-k}^2}{4}\left[\cosh(2r_k)-\frac{\sinh^2(2r_k)}{\mu_{+k}^2+\mu_{-k}^2}+\frac{2\mu_{+k}\mu_{-k}\sinh(2r_k)\cos(2\phi_k)}{\mu_{+k}^2+\mu_{-k}^2}\right]. \tag{3.41}$$

D_{n_k} depends in a complicated way in general not only on the squeeze parameters, but also on the displacements $|\mu_{\pm k}|^2$, and it is not simply related to squeezing as d is.

SUMMARY AND CONCLUSIONS

There has been deep interest in nonclassical aspects of the electromagnetic field. Squeezing of light has been demonstrated, following theoretical predictions.[13,14] These new effects found in optics have a certain generality and it seems certain that many important ideas are likewise applicable to the phenomenology of condensed matter, and are yet to be discovered. In this paper we extended the effects of squeezing to the condensed-matter system of an exciton polariton,[5] a coupled mixed mode of photon and exciton. Among the interesting conclusions that we obtain, one certainly concerns the intrinsic squeezed structure of the polariton. Precisely, exciton-polariton quantum states are found to be squeezed *(polariton intrinsic squeezing)* with respect to states of certain *intrinsic* photon-exciton mixed bosons, yet not polaritons. Squeezing is time periodic during a polariton period. The polariton problem has been analyzed within a Hamiltonian formulation. Two different Hamiltonians were investigated: One is the well-known conventional Hopfield Hamiltonian $\hat{H}^{\mathrm{Hopfield}}$, whereas the generalized $\hat{H}^{\mathrm{pol}}$ is an enlarged version of the previous one, mainly because it includes the *exciton-exciton* interaction term. The former has been derived within a macroscopic approach where the macroscopic electromagnetic and exciton polarization fields were quantized.[10] The latter is a polariton Hamiltonian depending upon parameters which are derived from fitting of the experimentally measured exciton-polariton energies. The amount of squeezing is measured by a squeeze factor that depends on the model Hamiltonian that one uses, and within each model it depends on the frequency of the polariton that is excited.

The parametrization of $\hat{H}^{\text{pol}}$ which we considered yields a considerably bigger squeezing than that relevant to $\hat{H}^{\text{Hopfield}}$.

These nonclassical effects in polaritons are studied using a representation through which a physical photon inside a dielectric is dressed, as a certain linear combination of vacuum annihilation and creation operators, and similarly for the exciton. This gives us straightforward means to analyze the interaction of coherent electromagnetic radiation with a resonant polarization (exciton) and to study the influence of this resonant medium on the photon statistics of the radiation field. Squeezing in the electromagnetic component associated with a polariton is found *(polariton optical squeezing)* as well as non-Poissonian photon statistics. Since this entails the reduction of the fluctuations of the polariton electromagnetic field component below the limit set by the vacuum fluctuations, our predictions would be most likely of great interest to demonstrate that the electromagnetic field may be driven into a nonclassical state through the resonant coupling with another boson, i.e., the exciton, as occurs in a polariton. Recently, the problem of examining squeezing effects in a homogeneous dielectric medium has independently been studied by Abram[17] and Glauber and Lewenstein.[18] However, to our knowledge, nonclassical optical effects in resonant dielectrics in interaction with the electromagnetic field are for the first time studied and proposed here. The question of measurability of these effects in polaritons is therefore of considerable importance and elsewhere we analyzed certain proposed novel experiments to examine this effect.[33]

Two interpretations of the origin of squeezing in polariton states are here presented, one within the framework of a sudden transition and the other relating to quantum correlations in polariton states between *intrinsic* photon-exciton mixed bosons of opposite wave vectors. This correlation approach to polariton squeezing seems to be quite attractive in that as a possible mechanism that justifies the intrinsic squeezed structure of polaritons, it may also lead to further understanding of basic processes in the dynamics of radiation and matter mixing as in a polariton.

ACKNOWLEDGMENTS

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APPENDIX A: PHOTON FLUCTUATIONS (VARIANCE)

We start from the result (1.26). We rewrite variance and mean for $\hat{N}_1 \to \hat{\gamma}^\dagger_{+k}\hat{\gamma}_{+k}$ in terms of the bare operators $\hat{a}_{+k}$ and $\hat{b}_{+k}$ [cf. Eq. (1.4)]

$$\begin{aligned}\langle \hat{N}_1 \rangle \to \langle \hat{\gamma}^\dagger_{+k}\hat{\gamma}_{+k} \rangle = |\alpha_k^+|^2 \langle \hat{a}^\dagger_{+k}\hat{a}_{+k} \rangle & \\ + |\alpha_k^+\beta_k^+| (e^{-i\chi_{+k}} \langle \hat{b}^\dagger_{+k}\hat{a}_{+k} \rangle & \\ + e^{+i\chi_{+k}} \langle \hat{a}^\dagger_{+k}\hat{b}_{+k} \rangle) & \\ + |\beta_k^+|^2 \langle \hat{b}^\dagger_{+k}\hat{b}_{+k} \rangle & \qquad \text{(A1)}\end{aligned}$$

and

$$\begin{aligned}\langle (\Delta\hat{N}_1)^2 \rangle \to \langle [\Delta(\hat{\gamma}^\dagger_{+k}\hat{\gamma}_{+k})]^2 \rangle & \\ \equiv \langle \hat{\gamma}^\dagger_{+k}\hat{\gamma}_{+k}\hat{\gamma}^\dagger_{+k}\hat{\gamma}_{+k} \rangle - \langle \hat{\gamma}^\dagger_{+k}\hat{\gamma}_{+k} \rangle^2 \; .\end{aligned}$$

The expectation values $\langle \; \rangle$ are all evaluated on the polariton coherent states $|\gamma_{\pm k}\rangle_z$. Thus,[29]

$$\begin{aligned}\langle \hat{\gamma}^\dagger_k\hat{\gamma}_k\hat{\gamma}^\dagger_k\hat{\gamma}_k \rangle = {} & |\alpha_k^+|^4 \langle \hat{a}^\dagger_k\hat{a}_k\hat{a}^\dagger_k\hat{a}_k \rangle + |\hat{\beta}^+_{k_k}|^4 \langle \hat{b}^\dagger_k\hat{b}_k\hat{b}^\dagger_k\hat{b}_k \rangle + |\alpha_k^+|^3|\beta_k^+| [(\langle \hat{a}_k\hat{b}^\dagger_k \rangle + 2\langle \hat{a}^\dagger_k\hat{a}_k\hat{a}_k\hat{b}^\dagger_k \rangle) e^{-i\chi_k} + \text{c.c.}] \\ & + |\alpha_k^+||\beta_k^+|^3 [(\langle \hat{a}_k\hat{b}^\dagger_k + 2\langle \hat{b}^+_k\hat{b}^+_k\hat{b}_k\hat{a}_k \rangle) e^{-i\chi_k} + \text{c.c.}] \\ & + |\alpha_k^+|^2|\beta_k^+|^2 [(2\langle \hat{a}^\dagger_k\hat{a}_k\hat{b}^\dagger_k\hat{b}_k \rangle + e^{-i2\chi_k} \langle \hat{a}_k\hat{a}_k\hat{b}^\dagger_k\hat{b}^\dagger_k \rangle) + \text{c.c.}] \; ,\end{aligned}$$

and from (A1),

$$\begin{aligned}\langle \hat{\gamma}^\dagger_k\hat{\gamma}_k \rangle^2 = {} & |\alpha_k^+|^4 \langle \hat{a}^\dagger_k\hat{a}_k \rangle^2 + |\beta_k^+|^4 \langle \hat{b}^\dagger_k\hat{b}_k \rangle^2 + 2|\alpha_k^+|^3|\beta_k^+| (\langle \hat{a}^\dagger_k\hat{a}_k \rangle \langle \hat{a}_k\hat{b}^\dagger_k \rangle e^{-i\chi_k} + \text{c.c.}) \\ & + 2|\alpha_k^+||\beta_k^+|^3 (\langle \hat{b}^\dagger_k\hat{b}_k \rangle \langle \hat{a}_k\hat{b}^\dagger_k \rangle e^{-i\chi_k} + \text{c.c.}) \\ & + |\alpha_k^+|^2|\beta_k^+|^2 (\langle \hat{a}^\dagger_k\hat{a}_k \rangle \langle \hat{b}^\dagger_k\hat{b}_k \rangle + \langle \hat{a}_k\hat{b}^\dagger_k \rangle \langle \hat{b}_k\hat{a}^\dagger_k \rangle + \langle \hat{a}_k\hat{b}^\dagger_k \rangle^2 e^{-i2\chi_k} + \text{c.c.}) \; .\end{aligned}$$

Therefore,

$$\begin{aligned}\langle [\Delta(\hat{\gamma}^\dagger_k\hat{\gamma}_k)]^2 \rangle = {} & |\alpha_k^+|^4 \langle [\Delta(a_k^+ a_k)]^2 \rangle + |\alpha_k^+|^3|\beta_k^+| \{ [\langle \hat{d}_k \rangle + 2C(\hat{N}^{\text{ph}}_k, \hat{d}_k)] e^{-i\chi_k} + \text{c.c.} \} \\ & + |\alpha_k^+|^2|\beta_k^+|^2 [e^{-i2\chi_k} \langle (\Delta\hat{d}_k)^2 \rangle + C(\hat{N}^{\text{ph}}_k, \hat{N}^{\text{exc}}_k) + C(\hat{d}_k, \hat{d}^\dagger_k) - \langle \hat{N}^{\text{exc}}_k \rangle + \text{c.c.}] \\ & + |\alpha_k^+||\beta_k^+|^3 \{ [-\langle \hat{d}_k \rangle + 2C(\hat{N}^{\text{exc}}_k, \hat{d}_k)] e^{-i\chi_k} + \text{c.c.} \} + |\beta_k^+|^4 \langle (\Delta\hat{N}^{\text{exc}}_k)^2 \rangle \; , \qquad \text{(A2)}\end{aligned}$$

where we have set

$$\hat{N}^{\text{ph}}_k \equiv \hat{a}^\dagger_k\hat{a}_k \; , \quad \hat{N}^{\text{exc}}_k \equiv \hat{b}^\dagger_k\hat{b}_k \; , \quad \hat{d}_k \equiv \hat{a}_k\hat{b}^\dagger_k$$

and

$$C(\hat{X},\hat{Y})\equiv\langle\hat{X}\hat{Y}\rangle-\langle\hat{X}\rangle\langle\hat{Y}\rangle\ ,\quad \langle(\Delta\hat{X})^2\rangle\equiv\langle\hat{X}^2\rangle-\langle\hat{X}\rangle^2\ . \tag{A3}$$

Substituting Eqs. (A1) and (A2) in (1.26) and using Eq. (2.15), the exact expression follows:

$$\begin{aligned}\langle(\Delta\hat{N}_k^{\text{ph}})^2\rangle=&e^{\pm 2r_k}\langle\hat{N}_k^{\text{ph}}\rangle-|\alpha_k^+\beta_k^+|A_{1,k}+|\beta_k^+|^2[\langle(\Delta\hat{N}_k^{\text{ph}})^2\rangle-A_{0,k}]\\&+\frac{1}{|\alpha_k^+|}(e^{\pm 2r_k}A'_{-1,k}|\beta_k^+|-A_{-1,k}|\beta_k^+|^3)\\&+\frac{1}{|\alpha_k^+|^2}\{\sinh^2 r_k[1+\sinh(2r_k)]+e^{\pm 2r_k}|\beta_k^+|^2\langle\hat{N}_k^{\text{exc}}\rangle-A_{-2,k}|\beta_k^+|^4\}\ .\end{aligned}\tag{A4}$$

Here we defined

$$\begin{aligned}&A_{-2,k}\equiv\langle(\Delta\hat{N}_k^{\text{exc}})^2\rangle\ ,\\&A_{-1,k}\equiv[-\langle\hat{d}_k\rangle+2C(\hat{N}_k^{\text{exc}},\hat{d}_k)]e^{-i\chi_k}+\text{c.c.}\ ,\\&A'_{-1,k}\equiv\langle\hat{d}_k\rangle e^{-i\chi_k}+\langle\hat{d}_k^\dagger\rangle e^{+i\chi_k}\ ,\\&A_{0,k}\equiv e^{-i2\chi_k}\langle(\Delta\hat{d}_k)^2\rangle+C(\hat{N}_k^{\text{ph}},\hat{N}_k^{\text{exc}})\\&\qquad+C(\hat{d}_k,\hat{d}_k^\dagger)-\langle\hat{N}_k^{\text{exc}}\rangle+\text{c.c.}\ ,\\&A_{1,k}\equiv[\langle\hat{d}_k\rangle+2C(\hat{N}_k^{\text{ph}},\hat{d}_k)]e^{-i\chi_k}+\text{c.c.}\end{aligned}\tag{A5}$$

APPENDIX B: PHOTO NUMBER (MEAN)

We evaluate the expectation value of the number of photons in a polariton coherent state. We start from Eq. (1.23) where we substitute the expression (A1) on the left-hand side and the expression (1.24) on the right-hand side:[29]

$$\langle\hat{N}_1\rangle\to\langle\hat{\gamma}_k^\dagger\hat{\gamma}_k\rangle=\sinh^2 r_k+|\gamma_k|^2 \tag{B1}$$

or

$$\begin{aligned}&|\alpha_k^+|^2\langle\hat{a}_k^\dagger\hat{a}_k\rangle+|\alpha_k^+\beta_k^+|(\langle\hat{d}_k\rangle e^{-i\chi_k}+\text{c.c.})+|\beta_k^+|^2\langle\hat{b}_k^\dagger\hat{b}_k\rangle\\&\quad=\sinh^2 r_k+|a_k|^2|\alpha_k^+|^2+|b_k|^2|\beta_k^+|^2\\&\qquad+a_kb_k^*|\alpha_k^+\beta_k^+|e^{-i\chi_k}+b_ka_k^*|\alpha_k^+\beta_k^+|e^{+i\chi_k}\ .\end{aligned}$$

All expectation values are evaluated on the states $|\gamma_{\pm k}\rangle_z$. Dividing by $|\alpha_k^+|^2$ and ordering in the powers of $|\beta_k^+|^2$, one gets

$$\begin{aligned}\langle\hat{N}_k^{\text{ph}}\rangle&\equiv\langle\hat{a}_k^\dagger\hat{a}_k\rangle\\&=|a_k|^2+\frac{\sinh^2 r_k}{|\alpha_k^+|^2}\\&\quad+|\beta_k^+|\left[\frac{e^{-i\chi_k}}{|\alpha_k^+|}(a_kb_k^*-\langle\hat{d}_k\rangle)+\text{c.c.}\right]\\&\quad+|\beta_k^+|^2(|b_k|^2-\langle\hat{b}_k^\dagger\hat{b}_k\rangle)\ .\end{aligned}\tag{B2}$$

Within the context of the detection experiment of Sec. II C $|a_k|^2$ is interpreted as the coherent photon flux (coherent intensity) multiplied by the integration time period Γ.

APPENDIX C: POLARITON–TWO-MODE SQUEEZE ISOMORPHISM

The polariton Hamiltonian $\hat{H}^{\text{pol}}$ in Eq. (3.1) is characterized by the six parameters $A_{1k},A_{2k},B_k,B'_k,C_k,C'_k$, whereas $\hat{H}^{\text{pol}}$ in Eq. (3.4), a two-mode squeeze Hamiltonian in form, is characterized by the six parameters $\Omega_k,\zeta_k,\alpha_k,\beta_k,\Psi_k^a,\Psi_k^b$. The isomorphism between these two Hamiltonians is effective provided the transformations in Eq. (3.7) are fulfilled. Consistency of Eqs. (3.7) results in a functional relation, e.g., of the kind $B'_k=B'_k(B_k,C_k,C'_k)$ (first four equations), and again allows A_{1k} and A_{2k} to be expressed in terms of B_k,B'_k,C_k,C'_k (last two Equations). At last, only the three parameters B_k,C_k,C'_k are left free in the Hamiltonian of Eq. (3.1). On the other hand, by taking $\Psi_k^a=0$ and $\Psi_k^b=\pi/2$, $\alpha_k\equiv\cos\theta_k$, and $\beta_k\equiv\sin\theta_k$ as done in Sec. III A, only the parameters $\theta_k,\Omega_k,\zeta_k$ are left free in the Hamiltonian of Eq. (3.4). The former and the latter sets of three parameters are connected by

$$\begin{aligned}&\zeta_k=B'_k+C'_k\ ,\\&\theta_k=\sin^{-1}\left[\frac{C'_k}{B'_k+C'_k}\right]^{1/2}\ ,\\&\Omega_k=(\hbar ck+2B_k)+(\hbar\omega_0-2C_k)=2(E_k^{\text{ph}}+E_k^{\text{exc}})\ .\end{aligned}\tag{C1}$$

Now, using these expressions for Ω_k and ζ_k and taking ε_k as the experimental polariton energy (see Sec. III A), it is possible, with a three-parameter fitting of the dispersion relation (3.13), to reproduce the energy dispersion branches (UBP and LBP) of a measured exciton-polariton spectrum. Next, substituting back the numerical evaluations for B_k,C_k,C'_k in (C1), the energies for the intermediate quasiparticles $\hat{c}$ and the squeeze factor r_k, given by Eq. (3.19), can be derived. It is a self-consistent procedure in which the fitting of experimental polariton energies for each branch is optimized through the use of Eqs. (C1) and (3.19).[50] The existence of parameters that consistently satisfy the transformation (3.7) and reproduce the experimental polariton energies for dielectric materials such as, e.g., CuCl and GaAs settles the isomorphism between the squeeze Hamiltonian (3.4) and a Hamiltonian that represents a "physical" polariton system.

APPENDIX D. TIME-DEPENDENT SQUEEZED FLUCTUATIONS

What follows is relevant to the quadrature $\hat{X}^p_k$.[51] First, define the ratio

$$R_k(t) \equiv {}_t\langle \mu_{\pm k} | \, |\Delta \hat{X}^p_k|^2 \, |\mu_{\pm k}\rangle_t / \Gamma^0_k \tag{D1}$$

and use Eq. (3.32) to obtain the squeeze time interval ΔT_{sq} as a solution of

$$R_k(t) \le q \quad \text{for } q=1$$

or (D2)

$$\cos(2\omega_k t) < \mathrm{sech}(2r_k) - \coth(2r_k) \; .$$

For exciton-polaritons in CuCl we evaluate in Fig. 9 $\Delta T^{\rm CuCl}_{\rm sq}$ for a spectrum of wave vectors in the vicinity of k_0. $\Delta T^{\rm CuCl}_{\rm sq}$ for a given k is precisely twice the vertical distance between the two solid lines. This figure also shows how the amount of squeezing varies during $\Delta T^{\rm CuCl}_{\rm sq}$ (dotted lines); intermediate values of the squeezing with their respective durations are here explicitly reported.

The maximum squeezing $\Gamma^0_k e^{-2r_k}$ occurs twice during the period ($T^{\rm pol}$) of the polariton. Its value depends on the mode that is excited: approaching k_0, the squeezing becomes more pronounced.

According to Eq. (D2) the results of Fig. 9 are mainly determined by the magnitude of r_k and the polariton frequency ω_k. Since r_k in the range of wave vectors of relevance to us takes only slightly different values on the two materials examined (cf. Fig. 6) and since their crossover frequencies are not significantly apart, we may expect that for exciton polaritons in GaAs $\Delta T^{\rm GaAs}_{\rm sq}$ is quantitatively similar to that for CuCl for the spectrum of wave vectors we examined. The same is true for the results on the envelope of fluctuations presented in Figs. 7 and 8.

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[13]*Squeezed and Nonclassical Light,* Vol. 190 of *NATO Advances Study Institute Series B: Physics,* edited by P. Tombesi and R. P. Pike (Plenum, New York, 1989); for a review on squeezing, see, for example, J. Opt. Soc. Am. B **4**, No. 10 (1987); J. Mod. Opt. **34**, No. 6, (1987).

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[23]To detect the effects of squeezing, one usually refers to two-quadrature observables by measuring their fluctuations. These oscillate rapidly in time between a very low and a very high value, and for the purpose of detection a procedure of conversion of an oscillating quantity to a constant quantity has to be employed. In "optical squeezing" such a procedure is experimentally possible and has been realized in vacuum.[13,14] For a polariton two-quadrature operators may also be defined[1] but the measurability of their fluctuations inside the medium where the polariton is excited may be rather difficult. However, in some cases it may be relatively simpler to measure the non-Poissonian statistics connected with squeezing.

[24]This is certainly true for any value of r_k, when we take the upper sign; for the lower sign, this is true for $0 \lesssim r_k \lesssim 0.8$. Since the values of r_k relevant to us fall in this range, Eq. (1.27) is valid.

[25]N. N. Bogoliubov, J. Phys. (Moscow) **11**, 23 (1947); Dokl. Akad. Nauk. (SSSR **119**, 244 (1958) [Sov. Phys.—Dokl. **3**, 292 (1958)]; *ibid.* **124**, 1011 C1959 [*ibid.* **4**, 143, (1959)].

[26]We restore the notation accordingly: $\hat{\gamma}_1 \to \hat{\gamma}_{+k}$, $\hat{\gamma}_2 \to \hat{\gamma}_{-k}$, $\alpha_1 \to \alpha_k^+$, $\alpha_2 \to \alpha_k^-$, $\beta_1 \to \beta_k^+$, $\beta_2 \to \beta_k^-$. See also Eq. (2.17).

[27]For different wave vector magnitudes we include the k dependence on the squeeze factor and angle r and φ.

[28]Although in different contexts, results that relate the squeezing to the dielectric properties of a medium have independently been obtained by R. Glauber and M. Lewenstein, Ref. 18, and by I. Abram, Ref. 17. However, unlike these works, in this paper we analyze nonclassical properties in a *resonant* material—essential to create a polariton.

[29]To simplify the notation, we suppress the sign "+" in the $+k$ subindexes where it does not create confusion.

[30] L. Mandel, Opt. Lett. **4**, 205 (1979).

[31] It is important to stress that the electromagnetic field leaking off the surface, unlike the free transmitted field propagating in vacuum, is described by the radiative component of a polariton mixed wave excited *inside* the crystal. See, e.g., A. D. Boardman, *Electromagnetic Surface Modes* (Wiley, New York, 1982). The appropriate conceptual framework that permits one to understand this distinction is the extinction theorem [P.P. Ewald, Ann. Phys. (Leipzig) **49**, 1 (1916)] extended to the quantum optics regime: Accordingly, the evanescent field is in our case the portion (dipole field) of the inside field that does not extinguish the vacuum incident field. This is discussed in detail in Ref. 15.

[32] These leaky waves must not be confused with surface polariton waves that are discussed, e.g., in V. M. Agranovich and D. L. Mills, *Surface Polaritons* (North-Holland, Amsterdam, 1982); see also Ref. 33.

[33] M. Artoni, and J. L. Birman, Opt. Commun. (to be published).

[34] Only the principal causes of distortion in the detection of polariton light statistics by this direct method are considered here, whereas a more detailed discussion is given in Ref. 33.

[35] H. P. Yuen and J. H. Shapiro, IEEE Trans. Inf. Theory **26**, 78 (1980).

[36] D. F. Walls *et al.*, Opt. Acta **29**, 1179 (1982); **29**, 1453 (1982).

[37] M. Artoni, and J. L. Birman (unpublished).

[38] R. Graham, J. Mod. Opt. **34**, 873 (1987); J. Janszky, Opt. Commun. **59**, 151 (1986).

[39] The two distinct coefficients A_{1k}, A_{2k} manifestly designate the resonant and nonresonant parts of the interaction that couples photons and excitons in a polariton. The coefficient B_k relates to the contribution of the nonresonant part of the "photon-photon" coupling $(\hat{a}_k \hat{a}_k^\dagger)$ to the single-photon dressed energy, whereas B_k', distinct from B_k, relates to the resonant part $(\hat{a}_k \hat{a}_{-k})$ of the same interaction. These terms and H.c. physically originate from the interaction between the electromagnetic field and polarizable electrons.[12] Similarly, for C_k and C_k': They are associated to the nonresonant $(\hat{b}_k \hat{b}_k^\dagger)$ and resonant $(\hat{b}_k \hat{b}_{-k})$ parts of the "exciton-exciton" coupling. These terms and H.c. arise from the exciton dispersion relation. For *Frankel* excitons, the transfer of excitation from site to site gives rise to such a term. For *Wannier* excitons, they are related to the effective mass of the exciton, although we do not account for this type of excitons in this paper. The physical origin of these resonant processes, involving the simultaneous creation (annihilation) of two bosons clarified, e.g., in the extreme-tight-binding exciton-plus-photon model, as discussed in Peierls's early work on the absorption of light in solids [Ann. Phys. (Leipzig) **13**, 905 (1932)]; in L. Apker and Taft [Phys. Rev. **79**, 964 (1950); **87**, 814 (1951)]; or in F. Seitz [*The Modern Theory of Solids* (McGraw-Hill, New York, 1940)]. Also see M. Artoni, Ph.D. thesis, City University of New York, 1991.

[40] A discussion on the group structure of other polariton Hamiltonians may be found, e.g., in S. Kim and J. Birman [Phys. Rev. B **38**, 6, 4291 (1988)].

[41] The $\hat{\mu}$'s and $\hat{\eta}$'s represent the same operator: We introduce two different symbols purely for clarity.

[42] H. P. Yuen, Phys. Rev. A **13**, 2226 (1976).

[43] Especially, e.g., in the second case where for a paramplifier, ζ contains the appropriate pump field which has apparntly no counterparts in the present physical system.

[44] The propagating field component of the polariton is discussed, e.g., by T. Skettrup, Phys. Rev. B **24**, 884 (1981). A photon in the crystal can transform directly into an exciton, provided that they both have the same energy and wave vector. If the exciton is not scattered by a phonon or some other defects, energy "oscillates" back and forth between exciton and photon owing to energy–wave-vector conservation as discussed, e.g., in Ref. 10. The propagating field of the polariton describes this "intrinsic phase" of the polariton, and the oscillations above can be ascribed to the oscillations of the mixed boson $\hat{c}$. The latter are estimated directly from Ω [cf. Eq. (C1)] and for an exciton polariton in CdS at $k = k_0$, $\hbar^{-1}\Omega \sim 10^{15}$ sec^{-1}. This rate of energy exchange has been evaluated in other contexts and it is of the same order of magnitude [cf. W.C. Tait and R. L. Weheir, Phys. Rev. **166**, 769 (1968) and *Dynamical Processes in Solid State Optics,* edited by R. Kubo (Benjamin, New York, 1967), Part I].

[45] If $|0\rangle \equiv |0_{\rm ph}\rangle |0_{\rm exc}\rangle$, then $\hat{a}_{\pm k}|0\rangle = \hat{b}_{\pm k}|0\rangle = 0$ and from Eq. (3.2), $\hat{c}_{\pm k}|0\rangle = 0$.

[46] Here $t = 0$ is a reference time.

[47] J. A. Wheeler, Lett. Math. Phys. **10**, 201 (1985).

[48] S. M. Barnett & M. A. Dupertuis, J. Opt. Soc. Am. B **4**, 505 (1987).

[49] G. J. Milburn, J. Phys. A **17**, 737 (1984).

[50] Actually, we use trial functions so as to express the three free c functions in terms of one of them, say, e.g., B_k (C_k) and C_k' (C_k), and we then construct C_k by inverting Eq. (3.13)—now a function of C_k only—taking ε_k as the experimental energy. This fitting procedure by which we give the parametrization of $\hat{H}^{\rm pol}$ in (3.1) is not unique.

[51] An analogous treatment holds for the conjugate quadrature $\hat{X}_k^q$.