Optical "Multiexcitons": Quantum Gap Solitons in Nonlinear Bragg Reflectors

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We find a Bethe-ansatz solution for pairwise interacting quanta within the effective-mass regime of band-gap propagation in nonlinear Bragg reflectors. Our theory predicts a new kind of collective excitation of the electromagnetic field dressed by such media, namely, optical multiexciton (OME) complexes (or condensates), which are quantum states associated with gap solitary waves. Their existence should be manifested by the discrete spectrum of band-gap transmission as a function of the transmitted photon number and by the multiexponential falloff of intensity-intensity correlations on a 0.1 mm scale. OMEs should have advantageous stability properties.

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Quantum effects of light propagation in nonlinear fibers have been the subject of extensive studies in recent years [1]. These studies have established that optical solitons, which are classically described by the nonlinear Schrödinger equation (NLSE) [2], have quantum analogs in the form of superposition of mutually bound (spatially correlated) multiphoton states. The description of such bound multiphoton states is given by the Bethe-ansatz solution of the second-quantized NLSE [3].

Here we address the hitherto unexplored quantum regime of optical propagation in forbidden spectral bands (band gaps) of Kerr-nonlinear one-dimensional (1D) Bragg reflectors, wherein transmission of solitary waves (gap solitons) [4] and ultrashort pulses [5] has been predicted classically. By extending the Bethe-ansatz solution to such systems, we reveal and investigate the striking analogy between mutually bound multiphoton states of quantum gap solitons and multiexciton complexes (excitonic molecules) in semiconductors [6]. These optical multiexcitons (OME) can be formed by the combined effect of (i) photon effective masses, which are endowed by the periodic-structure dispersion; (ii) Kerr-nonlinear interband photon attraction; and (iii) their intraband attraction or repulsion, also caused by Kerr nonlinearity. The main manifestations of OMEs are predicted here to be as follows: (1) a series of discrete transmission lines in the optical band gap of the Bragg reflector, which correspond to increasing numbers of photon pairs and range from a single exciton to the "ionization" threshold; (2) multiexponential falloff of two-point intensity-intensity correlations. Both features exhibit dramatic, unparalleled dependence on the refractive-index modulation in the Bragg reflector.

Under 1D-propagation conditions in a z-periodic Kerr-nonlinear Bragg reflector, with field confinement area $\pi\rho^2$ in the transverse (x-y) plane [7], the field dependence on the transverse coordinates can be factored out. The effective Hamiltonian can then be written in terms of the negative-frequency envelope operators $D(z,t)$ and $B(z,t)$ of the electric displacement and magnetic field as

$$\hat{H} = \int dz \Big[\frac{1}{2\mu_0} B^\dagger B + \frac{1}{2\epsilon_0 n^2(z)} D^\dagger D + \frac{3\chi^{(3)}(z)}{4\pi\rho^2\epsilon_0^3 n^8(z)} D^\dagger D^\dagger D D \Big]. \quad (1)$$

This Hamiltonian is similar to that used in studies of quantum solitons in nonlinear fibers [1] except that the dielectric index $n^2(z)$ and the nonlinear susceptibility $\chi^{(3)}(z)$ are now periodic in z. We make the standard assumption that only the two bands bordering a certain band gap are involved in the nonlinear process [6], and denote the lower ("valence") and upper ("conduction") bands by $+$ and $-$, respectively. The electric displacement and magnetic field operators can be quantized as

$$D(z) = \sum_{\alpha=\pm} \sum_k a_{\alpha k} D_{\alpha k}(z),$$
$$B(z) = \sum_{\alpha=\pm} \sum_k a_{\alpha k} B_{\alpha k}(z), \quad (2)$$

where $a_{\alpha k}$ are the usual annihilation operators for the two-band photons, and $D_{\alpha k}(z)$ and $B_{\alpha k}(z)$ are the appropriate Bloch functions [8] determined by the commutator between $D^\dagger(z)$ and $B(z)$. Consequently, the quantized Hamiltonian reads

$$\hat{H} = \sum_{\alpha=\pm} \Big[\sum_k \hbar\omega_{\alpha k} a_{\alpha k}^\dagger a_{\alpha k} + L^{-1} \sum_{k,k',q} V_{\alpha q}(k,k') a_{\alpha,k'+q}^\dagger a_{\alpha,-k'}^\dagger a_{\alpha,-k} a_{\alpha,k+q} \Big] + L^{-1} \sum_{k,k',q} V_{iq}(k,k') a_{-,k'+q}^\dagger a_{+,-k'}^\dagger a_{+,-k} a_{-,k+q}. \quad (3)$$

Here L is the structure length, $V_{\alpha q}(k,k')$ are the intraband matrix elements of the Kerr-nonlinear "potential," and $V_{iq}(k,k')$ are the corresponding interband matrix elements.

We are interested in photon properties near the extrema of the two bands, where $k = k_0$. We can therefore use

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the well-known effective-mass approximation [6]

$$\hbar\omega_{\pm k} = \hbar\omega_{\pm} \mp \frac{\hbar^2(k - k_0)^2}{2m_{\pm}}, \tag{4}$$

where $m_{\pm} = \mp\hbar/(\partial^2\omega_{\pm k}/\partial k^2)_{k_0}$ are the photon effective masses in the two bands. It is important for what follows that m_+ and m_- are both positive and $m_+ \neq m_-$ for any nonvanishing band gap. This fact can be illustrated by a structure of alternating layers with refractive indices n_1, n_2 and thicknesses a_1, a_2 such that $n_1a_1 = n_2a_2$. For the second lowest band gap, the photon effective masses in this structure are found to satisfy

$$m_{\pm} = \mp \frac{\hbar n_1 a_1}{c(a_1 + a_2)^2}\left[1 + \frac{1}{2}\left(\frac{n_2}{n_1} + \frac{n_1}{n_2}\right)\right] \times \sin\left(\frac{2n_1a_1\omega_{\pm}}{c}\right).$$

One sees that $|m_- - m_+|$ scales with the band-gap width when $|n_2 - n_1| \ll n_{1,2}$. Consistently with the effective-mass approximation, we replace the potential matrix elements in Eq. (3) by their values $V_{\pm}$ and V_i at $q = 0$ and $k = k' = k_0$. As we shall be working in the coordinate representation, we take the Fourier transform of the annihilation operators $a_{\alpha k}$, obtaining the following position-dependent field operators:

$$\phi_{\pm}(z) = L^{-1/2}\sum_k a_{\pm k}e^{i(k-k_0)z}. \tag{5}$$

The Hamiltonian (3) can now be rewritten as

$$\hat{H} = \hbar(\omega_+\hat{N}_+ + \omega_-\hat{N}_-) + \hat{H}_s, \tag{6a}$$

where $\hat{N}_\alpha = \int \phi_\alpha^\dagger\phi_\alpha dz$ is the single-band photon-number operator, and

$$\hat{H}_s = \int dz\Big\{\sum_{\alpha=\pm}[-\alpha(\hbar^2/2m_\alpha)\partial_z\phi_\alpha^\dagger\partial_z\phi_\alpha + V_\alpha\phi_\alpha^\dagger\phi_\alpha^\dagger\phi_\alpha\phi_\alpha] + V_i\phi_-^\dagger\phi_+^\dagger\phi_+\phi_-\Big\}. \tag{6b}$$

The "kinetic energy" terms depend on the structure dispersion, whereas the "potential energy" signs are those of the Kerr nonlinearity $\chi^{(3)}$, i.e., positive (negative) for self-focusing (self-defocusing) media.

Since $\hat{H}_s$ commutes with $\hat{N}_\alpha$, we seek simultaneous eigenstates of both operators. Such eigenstates can be obtained exactly by the Bethe-ansatz method, by extending the integrable quantum-field theory in Ref. [9] to the present two-band system. We first construct the exact four-quantum eigenstate, and then generalize it to arbitrary quantum numbers $N_+ + N_-$. The resulting $(N_+ + N_-)$-quantum eigenstate is obtained from a tedious calculation according to the aforementioned procedure. It is given, in its unnormalized form, by

$$|\Phi_{N_+N_-}\rangle = \int \Psi(z_{1\pm},\ldots,z_{N_\pm\pm})\prod_{\alpha=\pm}\prod_{j=1}^{N_\alpha} \times [\phi_\alpha^\dagger(z_{j\alpha})dz_{j\alpha}]|0\rangle, \tag{7}$$

where the spatial envelope (wave function) is

$$\Psi(z_{1\pm},\ldots,z_{N_\pm\pm}) = \prod_{n=1}^{N_+}\prod_{l=1}^{N_-}\left[1 - \frac{im_-m_+V_i}{\hbar^2(m_+k_{l-} + m_-k_{n+})}\,\mathrm{sgn}\,(z_{l-} - z_{n+})\right] \times \prod_{\alpha=\pm}\left\{\prod_{j<l}\left[1 + \alpha\frac{i2m_\alpha V_\alpha}{\hbar^2(k_{j\alpha} - k_{l\alpha})}\,\mathrm{sgn}\,(z_{j\alpha} - z_{l\alpha})\right]\prod_{j=1}^{N_\alpha}e^{ik_{j\alpha}z_{j\alpha}}\right\}. \tag{8}$$

The wave function $\Psi(z_{1\pm},\ldots,z_{N_\pm\pm})$ is symmetric with respect to the exchange of any two subscripts j and l for photonic coordinates, consistently with the bosonic nature of photons. The application of the Hamiltonian $\hat{H}$ to the eigenstate $|\Phi_{N_+N_-}\rangle$ yields the eigenenergy

$$E_{N_+N_-} = \hbar(\omega_+N_+ + \omega_-N_-) + \sum_{\alpha=\pm}\sum_{j=1}^{N_\alpha}\left(-\alpha\frac{\hbar^2k_{j\alpha}^2}{2m_\alpha}\right). \tag{9}$$

In the above, all the wave vectors are measured relative to the band-edge wave vector k_0.

We are looking for bound-state solutions of (8), wherein the exponential factors are real, and fall off with the separation of photons from different bands $|z_{j+} - z_{j-}|$ or from the same band, $|z_{j\alpha} - z_{j'\alpha}|$. The bound-state solutions require $N_+ = N_- = N$. It can be checked that the wave vectors $k_{j\alpha}$ in (8) must then satisfy

$$k_{j\pm} = \mp\frac{m_\pm}{m_- - m_+}K \pm i\frac{m_-m_+V_i}{(m_- - m_+)\hbar^2} \mp i\frac{m_\pm V_\alpha}{\hbar^2}(N - 2j + 1), \tag{10}$$

where $\alpha = \pm$ is chosen according to whether the Kerr nonlinearity is positive (self-focusing) or negative (self-defocusing), and NK is the total composite momentum, which can be expressed as

$$NK = \sum_{j=1}^{N}(k_{j+} + k_{j-}). \tag{11}$$

Equation (11) indicates that bound states consist of N "excitons," i.e., pairs of conduction- and valence-band photons. On using Eqs. (10) and (11) in the wave function

expression (8), we can bring it to the symmetrized form

$$\Psi(z_{1\pm},\ldots,z_{N\pm}) \propto \exp\left(\sum_{j=1}^{N} iK\,\frac{m_- z_{j-} - m_+ z_{j+}}{m_- - m_+}\right) \times \exp\left[-\sum_{j=1}^{N}\frac{m_- m_+ V_i}{(m_- - m_+)\hbar^2}\,|z_{j+} - z_{j-}| - \frac{|V_\alpha|}{\hbar^2}\sum_{j<l}(m_-|z_{j-} - z_{l-}| + m_+|z_{j+} - z_{l+}|)\right]. \quad (12)$$

In this expression ordering of $z_{j\pm}$'s is immaterial. The wave function (12) falls off exponentially with the distance $|z_{j+} - z_{j-}|$, under the binding condition

$$m_- > m_+,\ V_i > 0,\quad m_- < m_+,\ V_i < 0. \quad (13)$$

Clearly, such a wave function characterizes a bound state of N excitons, as evident from the real exponential factors and an overall translation (the first factor), analogously to a delocalized Wannier excitonic complex [6].

The exponential factors in the bound state $|\Phi_{2N}\rangle$ can be revealed by the dependence of the intensity-intensity correlation function [3] $G^{(2)}$ on the separation η of two photon counters detecting the field in the structure

$$G^{(2)}(\eta) = \int \langle\Phi_{2N}|\mathcal{E}^+(z)\mathcal{E}^-(z)\mathcal{E}^+(z+\eta) \times \mathcal{E}^-(z+\eta)|\Phi_{2N}\rangle dz, \quad (14)$$

where the operator $\mathcal{E}^-(z) = \phi_+(z) + \phi_-(z)$ is the position-dependent negative-frequency field envelope. A laborious extension of the treatment in Ref. [3] to the two-band system yields $G^{(2)}(\eta)$ as a linear combination of exponential factors of the form $\exp[-(c_i + c_\pm)|\eta|]$, $\exp[-(c_+ + c_-)|\eta|]$, and $\exp[-(c_i + c_+/2 + c_-/2)|\eta|]$, where $c_i = 2m_- m_+ V_i/(m_- - m_+)\hbar^2$, $c_\pm = 2|V_\alpha|m_\pm/\hbar^2$, and $\alpha = +$ or $-$ according to the Kerr nonlinearity sign. We have evaluated $G^{(2)}(\eta)$ for $N = 2$ (a biexciton) in a periodic structure consisting of Kerr-nonlinear GaAs layers alternating with linear dielectric layers. The parameters concerned are as follows: $\rho = 0.5\ \mu$m, a_1(GaAs) $= 0.2338\ \mu$m, $a_2 = 0.3044\ \mu$m, and $n_1 = 3.60$. The refractive index n_2 of linear dielectric varies from 1.00 to 3.574. GaAs has a nonlinear susceptibility $\chi_1^{(3)} = -2.5482 \times 10^{-10}$ (cm/V)^2 for light frequencies below the band gap $E_g = 2.1573 \times 10^{15}\ \mathrm{s}^{-1}$ [10]. The potentials V_i, $V_\pm$ and effective masses $m_\pm$ have been evaluated for the conduction and valence bands bordering the second lowest band gap, for which the binding condition in Eq. (13) holds (as opposed to the lowest band gap in this structure, for which this condition fails). Figure 1 clearly manifests the nonmonotonic dependence of the multiexponential falloff of $G^{(2)}_{N=2}$ with η upon the refractive index ratio n_1/n_2. This dependence is a unique feature of the present solutions, whereas in quantum solitons traveling through Kerr-nonlinear fibers [3] the exponential falloff of $G^{(2)}(\eta)$ depends only on the photon number N.

The bound-state energy eigenvalues are found from Eqs. (9) and (10) to have the following form:

$$E_{2N} = 2N\hbar(\omega_0 \pm \omega_{2N}),$$
$$2\hbar\omega_{2N} = \frac{\hbar^2K^2}{2|m_- - m_+|} + \frac{m_- m_+ V_i^2}{2|m_- - m_+|\hbar^2} - \frac{|m_- - m_+|V_\pm^2}{6\hbar^2}(N^2 - 1), \quad (15)$$

where $\omega_0 = (\omega_+ + \omega_-)/2$ is the center of the band gap and the upper (lower) sign is chosen according to that of the Kerr nonlinearity. As seen from Eq. (15), bound states are associated with discrete transmission lines at $\Omega_{2N} = \omega_0 \pm \omega_{2N}$ in the band gap, whose progression scales quadratically with half the transmitted photon number $2N$. Remarkably, the discreteness of the transmission spectrum will separate out different Fock states in the incident coherent state according to their relative weights, i.e., the spectrum will reflect the superposition of optical multiexciton states with different photon number. This discreteness will be revealed only if $|m_- - m_+|V_\pm^2/\hbar^2$ is spectrally resolvable, otherwise the quasiclassical limit of gap solitons will prevail. The lowest line $N = 1$, which marks a single exciton, is the farthest removed from the appropriate band edge, and the lines are drawn nearer to the edge as N^2 increases, as in the case of ordinary Wannier excitons [6]. However, whereas Wannier excitons have a Rydberg spectrum, with an infinite number of levels (which scale as $1/N^2$), the present bound spectrum is terminated at $N_{\max}$, for which Eq. (15) yields $\omega_0 \pm \omega_{2N} = \omega_\pm$, marking what can be dubbed the "ionization" threshold. Since the wave vector K of excitons is

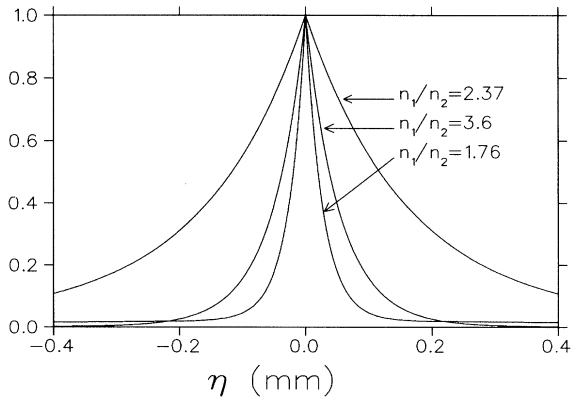

FIG. 1. Dependence of the intensity correlation function $G^{(2)}(\eta)$ on the detector separation η, for the photon number $2N = 4$.

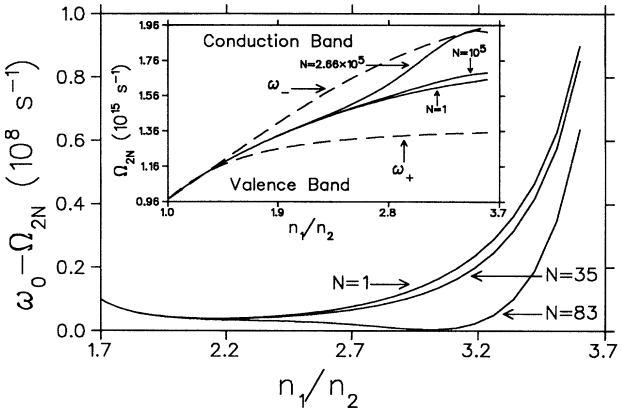

FIG. 2. Variation of the optical exciton frequency Ω_{2N} with the relative refractive index n_1/n_2. The inset displays the variation of the band edges ω_+ and ω_- with n_1/n_2, and $\omega_0 = (\omega_+ + \omega_-)/2$ is the center of the band gap.

measured relative to $2k_0$, the effective-mass approximation requires $|K| \ll 2k_0$, so that we can let $K = 0$ in Eq. (15).

The spectral features discussed above are shown in Fig. 2 for the same structure as in Fig. 1. Here too the dependence on n_1/n_2 is strongly nonmonotonic. A rather restricted range of n_1/n_2 allows the resolution of levels associated with different N on a 0.1 GHz scale. The intrinsic intensity of a $2N$-photon bound state has an approximate expression $I_{2N} = 4N\hbar\Omega_{2N}/\pi\rho^2\tau$, where τ is the pulse duration. Using a relative refractive index $n_1/n_2 = 3.4221$ and a pulse duration $\tau = 1.0$ μs, we have obtained the intrinsic intensities $I_{2N} = 8.77 \times 10^{-5}$, 3.07×10^{-3}, 7.28×10^{-3}, 8.99, and 27.41 W/cm^2, respectively, corresponding to the quantum numbers $N = 1$, 35, 83, 10^5, and 2.66×10^5, which is $N_{\max}$. For $N > N_{\max}$ the spectrum (15) is replaced by continuous bands of energy eigenvalues, embedded in the allowed conduction and valence bands. These bands are analogous to the classical "out-gap" solutions obtained above a certain intensity threshold [11].

To sum up, we have accomplished the Bethe-ansatz solution for pairwise interacting quanta within the effective-mass regime of band-gap propagation in 1D-periodic Kerr-nonlinear dielectric structures. Our theory predicts a new kind of collective excitation of the electromagnetic field dressed by such media, namely, optical multiexciton complexes (or condensates), which are the quantum states associated with gap solitary waves. Their existence should be manifested (i) by the discrete spectrum of band-gap transmission, which allows us to spectrally distinguish between OMEs with different photon number typically on a MHz scale, and (ii) by the multiexponential falloff of intensity-intensity correlations typically on a 0.1 mm scale. Another distinct signature of OMEs is the strong nonmonotonic dependence of the above properties on the periodic modulation of the refractive index in the structure.

OMEs should have advantageous stability properties, provided both linear and nonlinear absorption are negligible. The stability properties of OMEs stem from their being the exact bound eigenstates of the nonlinear field Hamiltonian which accounts for single-photon and two-photon processes in the medium. In nonabsorbing media, OMEs should be stable against (a) spontaneous radiative recombination, which is the main decay mechanism of electron-hole excitons in semiconductors [6], (b) self-focusing (self-defocusing) by the Kerr nonlinearity, (c) pulse (wave-packet) dispersion and diffraction in the dielectric structure, and (d) quantum fluctuations in the electromagnetic field. On the other hand, in order to ensure the OME stability against thermal scattering due to thermal fluctuations in the dielectric function, one needs low temperatures. If the dielectric structure is Raman inactive, i.e., optical phonon scattering is absent (as in GaAs), then thermal scattering does not affect the resolution of OME lines on a 0.1 GHz scale. The ability to resolve OME lines corresponding to much lower N would necessitate band-structure design aimed at increasing the effective-mass difference $|m_- - m_+|$ and the intraband nonlinear potentials $|V_\pm|$.

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