Two-Photon Bound State in Self-Focusing Media

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(Received 31 May 1991)

We derive a two-photon bound-state solution to the problem of quantum propagation of light in Kerr-effect nonlinear media which classically would exhibit self-focusing and self-trapping. We propose an experiment to see this two-photon bound state.

PACS numbers: 42.50.Dv, 05.30.Jp, 42.65.Jx

Quantum effects in the propagation of light in nonlinear media have been the subject of much recent interest. New phenomena, such as squeezed-light production due to self-phase modulation, arise in quantum propagation [1,2]. The resulting nonclassical sources of light are not only of intrinsic physical interest, but are also potentially applicable to optical communications. Of particular interest is the problem of temporal soliton propagation in optical fibers, described classically by the nonlinear Schrödinger equation. The work of Lai and Haus [3] on quantum solitons in such fibers was based on earlier work [4], where it was pointed out that the corresponding quantum field theory is equivalent to that of a one-dimensional system of nonrelativistic bosons interacting through attractive δ-function potentials. Quantum solitons are intimately related to bound-state solutions of this system [3,4].

The nonlinear Schrödinger equation also describes the propagation of light in self-focusing media at the classical level [5]. *Spatial* soliton solutions, which correspond to self-trapped beams of light, are known to exist for this equation in planar geometries [6]. Such (1+1)-dimensional classical solitons (i.e., those in one transverse and one longitudinal spatial dimension) have recently been observed in electronic Kerr media with a positive nonlinear index coefficient [7]. The stability of these solitons under soliton-soliton collisions has also been observed [8].

The question then naturally arises: What are the corresponding *quantum* spatial solitons? Since the classical field theory for the spatial soliton is formally equivalent to that of the classical temporal soliton, one suspects that analogous quantum bound-state solutions should exist. How these bound states are related to the classical spatial soliton solutions in the correspondence principle limit is a question which we will not attempt to answer here (however, in the case of the temporal soliton, see Lai and Haus [3]). Here, rather, we shall concentrate on the solution corresponding to the simplest bound state. This state should be a number or Fock state of some self-replicating mode of propagation. Since there are no fluctuations in photon number, and hence in the intensity of a beam of light in a number state, there should be no fluctuations in the index of refraction arising from the Kerr nonlinearity. Therefore the compensation of diffractive losses by the nonlinear index change which occurs classically should not be destroyed by quantum fluctuations. A number state should thus lead to a stable, nondiffractive solution of the quantum propagation problem. We have confirmed this by studying the problem of paraxial quantum propagation [9] in Kerr-effect media.

This problem also reduces to that of nonrelativistic bosons interacting through attractive 1D δ-function potentials. The fundamental eigenstate consists of two bosons, here photons, bound to each other through an attractive δ-function potential in their relative transverse coordinate. One can think of this solution as being the unique two-photon bound state which, for brevity, we shall call the "diphoton." The origin of the interaction between the two photons is their exchange of a virtual excitation in an atom. In the case of fast atomic response (i.e., when the response rates are large compared with the bandwidth of the light), there are no retardation effects, and the interaction can be approximated by a static δ-function potential. In the case of a two-level model of the atom, when the light is detuned slightly above resonance, then there is a virtual decrease in the energy of the system during the exchange process. This leads to an attractive potential between the photons.

At the classical level, the effective photon-photon interaction corresponds to a Kerr, or n_2, intensity-dependent change in the index of refraction [10]. For attractive interactions, n_2 is positive. In a classical temporal soliton the self-steepening due to the nonlinear index change balances the spreading of the wave packet due to dispersion. Likewise, in a spatial soliton the self-focusing due to the nonlinear index change balances the spreading due to diffraction. Thus the classical (1+1)-dimensional spatial soliton is characterized by a hyperbolic secant transverse profile which does not change its shape with propagation [6]. At the quantum level, this attractive interaction gives rise to photon pairing, which is reminiscent of Cooper pairing in superconductors. It is the formation of *bound states* of photons which prevents the spreading of a beam of light due to diffraction. The diphoton in particular is characterized by a two-point correlation function which also does not change with propagation.

For concreteness, consider the following possible experimental realization (see Fig. 1). A uv laser produces tightly correlated pairs of photons by degenerate spontaneous parametric down-conversion in a $\chi^{(2)}$ crystal with collinear phase matching. Two photons traveling in the same direction are created essentially simultaneously in this process [11], providing a good source for generating the diphoton. The output of this two-photon light source is focused by lens $L1$ into a very small focal volume at the input face of a Kerr-effect medium, such as an alkali vapor possessing a resonantly enhanced $\chi^{(3)}=n_2n_0/2\pi$. Inside this medium is a slab waveguide structure consisting of two parallel, closely spaced mirrors, which provide linear confinement of the light in one transverse dimension (the y direction). The light will be confined in the other transverse dimension (the x direction) through the nonlinear Kerr effect. Analytic calculations (see below) show that the two-photon bound state should form if the pair of input photons is injected into a sufficiently small focal volume. Detection of the diphoton can be achieved by means of coincidence detection of the output light by detectors $D1$ and $D2$ as a function of their transverse separation in the far field.

We study the propagation of light in Kerr-effect media using a recently formulated quantum field version of the paraxial approximation [9]. The positive-frequency part of the electric-field operator for a given polarization $\boldsymbol{\varepsilon}$ is written as a slowly varying envelope which modulates a rapidly varying carrier wave. For (1+1)-dimensional propagation we define

$$\mathbf{E}_\omega^{(+)}(x,z,t)=\boldsymbol{\varepsilon} i\left(\frac{2\pi\hbar\omega v_g}{cn_0L_y}\right)^{1/2}\Psi(x,z,t)e^{i(kz-\omega t)}. \quad (1)$$

The envelope field Ψ is normalized with respect to the effective vacuum fluctuation energy in the medium so

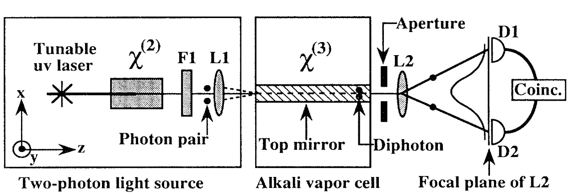

FIG. 1. Schematic (top view) of an experiment to observe a two-photon bound state in a $\chi^{(3)}$ medium. A pair of photons is generated in a $\chi^{(2)}$ crystal by degenerate parametric down-conversion. The photon pair passes through filter $F1$ and is focused by lens $L1$ into a resonantly enhanced $\chi^{(3)}$ medium. A slab waveguide consisting of two closely spaced parallel mirrors (top mirror shown) confines the light in the plane of the page. Two detectors $D1$ and $D2$ are placed in the focal plane of lens $L2$ following the cell. The diphoton is detected in coincidences between $D1$ and $D2$ as a function of their separation.

that it has units of a wave function in two dimensions. The length scale L_y is the confinement dimension of the slab waveguide, and n_0 and v_g are the linear refractive index and group velocity at the carrier frequency, respectively. To lowest order in the slowly varying envelope approximation (SVEA), the effective field theory formally resembles a nonrelativistic many-body theory for a complex scalar field Ψ, which satisfies Bose equal-time commutation relations

$$[\Psi(\mathbf{x},t),\Psi^\dagger(\mathbf{x}',t)]=\delta_c^{(2)}(\mathbf{x}-\mathbf{x}'). \quad (2)$$

Here $\delta_c^{(2)}(\mathbf{x}-\mathbf{x}')$ is a "coarse-grained" δ function which must be viewed with the caveat that it only acts as expected when integrated with slowly varying test functions. Thus, $\delta_c^{(2)}(\mathbf{x}-\mathbf{x}')=\delta_c(x-x')\delta_c(z-z')$ is not truly a singular object; its value at the origin is determined by

$$\delta_c(x=x')=1/L_x,\quad \delta_c(z=z')=1/L_z, \quad (3)$$

where L_x and L_z are the length scales of the SVEA, i.e., the characteristic lengths over which the envelope of the quasimonochromatic paraxial fields can change in the transverse and longitudinal directions, respectively [12]. For the experimental configuration in Fig. 1, these are given by the waist diameter and Rayleigh range of the focal volume as set by lens $L1$. The volume $L_xL_yL_z$ sets the minimum localization volume for the photons in paraxial fields [9].

The Hamiltonian for the system decomposes into

$$H=H_{\text{env}}+H_{\text{int}}, \quad (4)$$

where H_{env} governs the propagation of the envelope including diffraction,

$$H_{\text{env}}=\hbar v_g\int dx\,dz\,\Psi^\dagger(x,z)\left(-i\frac{\partial}{\partial z}-\frac{1}{2k}\frac{\partial^2}{\partial x^2}\right)\Psi(x,z), \quad (5)$$

and H_{int} describes the nonlinear interaction (here, the Kerr effect),

$$H_{\text{int}}=-G_2\int_A dx\,dz[\Psi^\dagger(x,z)]^2[\Psi(x,z)]^2, \quad (6)$$

where A is the area of the effective two-dimensional medium, and the 2D effective coupling constant is

$$G_2=\left(\frac{2\pi\hbar\omega v_g}{cn_0}\right)^2\frac{\chi^{(3)}}{L_y}. \quad (7)$$

We seek a solution to this dynamical system which corresponds to steady-state propagation of the optical field. According to the arguments put forth in a recent analysis [13], the steady-state-propagation solution is characterized by the stationary states of the propagation Hamiltonian,

$$(H_{\text{env}}+H_{\text{int}})|\Phi\rangle=\mathscr{E}|\Phi\rangle. \quad (8)$$

This condition leads to *spatial* evolution equations for the photon wave functions. The frequency of the light is fixed by the carrier wave and does not change as a function of propagation distance z. The bound-state solution is obtained by separating out the center-of-mass motion in z, and assuming that the remaining wave function depends only on the transverse variables, as is expected for self-trapping. Substituting the two-photon ansatz

$$|\Phi\rangle = \int dz\, dx_1\, dx_2 e^{iKz} F(x_1,x_2)\Psi^\dagger(x_1,z)\Psi^\dagger(x_2,z)|0\rangle \tag{9}$$

into Eq. (8) gives the eigenvalue equation,

$$-\frac{\hbar^2}{2m_{\rm eff}}\left[\frac{\partial^2}{\partial x_1^2}+\frac{\partial^2}{\partial x_2^2}\right]F(x_1,x_2) - G\delta(x_1-x_2)F(x_1,x_2) = (\mathcal{E}-\hbar v_g K)F(x_1,x_2)\,, \tag{10a}$$

$$G \equiv \frac{G_2}{L_z} = \left[\frac{2\pi\hbar\omega v_g}{cn_0}\right]^2 \frac{\chi^{(3)}}{L_y L_z}\,, \tag{10b}$$

where $m_{\rm eff} \equiv \hbar k/v_g$. The interpretation of $F(x_1,x_2)$ is that $|F(x_1,x_2)|^2$ is the probability density of finding one photon at x_1, and the other photon at x_2 [9]. In deriving Eq. (10), the z-dependent part of the coarse-grained δ function has been evaluated at the origin by use of Eq. (3), and the x-dependent part is approximated by a true δ function under the caveats described above [14].

The appearance of the length scale L_z in the coupling constant G solves some long-standing difficulties in the theoretical treatment of the propagation of nonclassical light [15]. The standard approach of decomposing fields into normal modes with creation and annihilation operators is well suited for dealing with temporal evolution, but is ill suited for describing *spatial* evolution [16]. Many of the previous workers have treated this problem by modeling traveling waves as a train of photon wave packets, and letting the propagation coordinate z play the role of time. The width of the packet then plays a fundamental role in the quantization. Indeed, in all previous analyses of propagation of nonclassical light in Kerr media, the effective coupling constant implicitly depends on the choice of length scale [13]. In our approach, the length scale L_z is unambiguously determined by the spatial bandwidth of the incident radiation. In addition, our use of the fundamental equal-time canonical commutator avoids possible difficulties introduced by the *ad hoc* use of the equal-z commutator postulated in previous analyses.

Using standard techniques, this two-body problem is solved by separating variables in the center-of-mass and relative transverse coordinates, $X \equiv (x_1+x_2)/2$ and $\xi \equiv x_1 - x_2$, respectively. The ansatz

$$F(x_1,x_2) = e^{iQX}u(\xi) \tag{11}$$

leads to the reduced Schrödinger equation, with $\mu \equiv m_{\rm eff}/2$,

$$-\frac{\hbar^2}{2\mu}\frac{\partial^2 u}{\partial \xi^2} - G\delta(\xi)u(\xi) = \left[\mathcal{E} - \hbar v_g K - \frac{\hbar^2 Q^2}{2m_{\rm eff}}\right]u(\xi)\,. \tag{12}$$

Equation (12) is known to have one bound-state solution,

$$u(\xi) = \sqrt{\gamma}\exp(-\gamma|\xi|)\,, \tag{13a}$$

$$\gamma = \frac{\mu G}{\hbar^2} = \frac{2\pi^2\hbar\omega^3 v_g \chi^{(3)}}{c^3 n_0 L_y L_z}\,. \tag{13b}$$

The eigenvalue fixes the propagation speed of the diphoton. Note that γ depends explicitly on $\hbar$; therefore the diphoton is unambiguously a quantum object.

In order to test the results of this analysis, consider the proposed experiment shown in Fig. 1. The two-photon light source determines the initial wave function at the input face of the Kerr medium. This wave function can then be decomposed into the bound and free eigenstates of the Schrödinger equation. The probability of the formation of bound states is determined by the overlap of the initial wave function with the diphoton wave function. The unbound amplitudes will undergo wave-packet spreading (diffraction), while the bound state will maintain the width dictated by γ in Eq. (13b). Thus the diphoton survives spreading by diffraction even after propagating a distance much larger than the Rayleigh range associated with the diphoton width. After leaving the Kerr medium the aperture filters out the diffracted (unbound) component, and the lens $L2$ performs a Fourier transform of Eq. (13a) in the far field. A Lorentzian two-point correlation function in coincidence detection by $D1$ and $D2$ thus provides an unambiguous signature for the detection of the diphoton.

An important experimental consideration is the requirement of sufficient binding strength. If the diphoton width is too large, it can be unambiguously detected only after a very long propagation distance. If this distance is too large, absorption becomes important and the diphoton will decay. For concreteness, we consider a resonantly enhanced Kerr medium such as sodium vapor previously shown to exhibit self-focusing and self-trapping [17] and recently used to produce spatial dark solitons [18]. The diphoton (power) width is obtained from Eq. (13b) in terms of the experimentally relevant parameters,

$$d \equiv \frac{1}{2\gamma} = \frac{1}{8\pi^3}\left[\frac{c}{n_0 v_g}\right]\left[\frac{\lambda^3 L_y L_z}{\hbar c^2 n_2^l}\right]\times 10^7\ {\rm cm}\,, \tag{14}$$

where n_2^l is the nonlinear refractive index measured in units of cm^2/W, and all other quantities are measured in cgs units. If n_2^l is wavelength independent, and if L_z and $L_y \propto \lambda$, the diphoton width will scale as λ^5. Thus, the lighter alkali vapors should be more favorable for creating diphotons. To estimate the diphoton width d, we take numerical values obtained from a measurement of n_2^l in sodium vapor on the self-defocusing side of the D-line resonance at $\lambda = 590$ nm (the nonlinearity should be of the same magnitude with the same detuning on the self-focusing side). Detuning 2 GHz from resonance in a sodium vapor at $T = 195\,^\circ$C, Swartzlander, Yin, and Kaplan [19] measured $|n_2^l| = 1.7\times10^{-7}$ cm^{2}W^{-1}. For this temperature, we calculate the Doppler width to be $\Delta\nu_D = 1.64$ GHz. For these parameters, the linear absorption length is 3.8 cm. The vapor is sufficiently dispersionless at this detuning that the ratio between phase and group velocities is essentially unity; also $n_0 \cong 1$. We take the longitudinal localization length $L_z = 10$ μm as set by the length of the focal volume of lens $L1$, and the slab confinement thickness $L_y = \lambda$. This gives a diphoton width $d = 30$ μm. Its Rayleigh range $\pi d^2/\lambda$ is 0.48 cm (this is a slight overestimate since we have assumed a minimum-uncertainty wave packet in making this calculation). This distance is well below the absorption length of the sodium vapor given above.

In conclusion, we have shown that a two-photon bound state in self-focusing media should exist theoretically, and that it should be detectable experimentally. Such bound states are important for our understanding of quantum solitons and their associated quantum field theories.

We thank Y. Aharonov, P. Meystre, and D. S. Rokhsar for helpful discussions. One of us (R.Y.C.) would like to thank the Alexander von Humboldt-Stiftung for support, and Professor Dr. H. Walther for hospitality. This work was also supported by ONR under Grant No. N00014-90-J-1259, by a Department of Education Fellowship, and by the U.S. Department of Energy at the Lawrence Livermore Laboratory under Contract No. W-7405-Eng-48.

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