Delayed single-photon self-interference

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It has been suggested that a single photon can interfere with itself even if the difference between the two paths in the interferometer is larger than the ''length'' of the photon [Kessel' and Moiseev, JETP Lett. **58**, 81 (1993)]. The interference is regained by detecting the photons using a photon-echo process, where the absorbing atoms will, effectively, act as narrow-band filters. Such an experiment has several unique features. For example, single photons are used to carry out what is generally regarded as a multiphoton process; the absorption of a single photon can be regarded as separated into two different moments in time; the fact that the single-photon interference is regained using atoms acting as narrow-band filters as the detector means that the control of the detection process is quite different from cases where electronic (or possibly photographic) detection is used to register the interference etc. In general, interference and absorption are combined and intertwined in the experiment, which is discussed in this paper, in a way that has not been done before. In the present paper the possibility to carry out such an experiment in practice is investigated in some detail. The signal strength is explicitly calculated and the results are compared with our experimental data for the case of many interfering photons. We imply that this result can readily be extrapolated to the single-photon situation. We analyze the material parameters that are important for carrying out the experiment and give specific examples of some suitable materials. [S1050-2947(98)07112-1]

I. INTRODUCTION

Consider a single-photon wave packet that is split into two when the photon impinges on a 50% transmission beam splitter. By letting the two paths overlap at a later location (see Fig. 1) the wave nature of the single-photon wave packet can be manifested by the observation of an interference pattern [1,2]. Such an interference pattern can generally only be observed if the time delay t_d between the two wave packets at the overlap points is less than the single-photon wave-packet coherence time t_c, i.e., $t_d = \Delta L/c < t_c$, where ΔL is the path length difference between paths 1 and 2 and c is the speed of light. Even for cases where we originally have $t_d > t_c$ (i.e., $\Delta L > l_c$, where l_c is the single-photon wave-packet coherence length) we would expect to be able to manipulate the radiation to get a revival of the interference pattern and thereby also the wave nature of the photon. This could be done by inserting narrow-band filters in the two beam paths. This would confine the wave packet to a smaller interval in frequency space and at the same time delocalize the wave packet in time, thus increasing t_c (and thereby also increasing l_c). An interesting suggestion for such a filter has been put forward by Kessel' and Moiseev, who suggest that a photon-echo material should be placed at the crossing point (C in Fig. 1) [3]. The material should be chosen such that the phase memory time (i.e., the homogeneous dephasing time T_2) of the atoms should be larger than t_d. This means that the phase of the first wave packet is remembered by the absorbing atoms at the time the second packet, taking path 2, arrives at the sample. It should then be possible for the interference to take place. Further, the absorption line of the absorber should be inhomogeneously broadened. This im-

plies that even if the absorption linewidth Γ_{hom} (where $1/\pi\Gamma_{\text{hom}} = T_2 > t_d > t_c$) of any individual atom is so small that its phase memory time is longer than t_d, the macroscopic absorption of the sample, involving a very large number of atoms, is described by a line shape with a linewidth $\Gamma_{\text{inhom}} > 1/t_c$.

With such a material placed at the crossing point C, first assume that the wave packet entering on the left hand side of Fig. 1 contains many photons. After the two parts of this wave packet taking path 1 and path 2, respectively, have passed the crossing point, path 2 is blocked. A time τ after the first packet ($\tau \gg \Delta L/c$), a second packet is sent in. Set $t = 0$ as the time when the first packet taking path 1 interacts with the sample. The sample consequently will be exposed to three wave packets at times $t = 0, \Delta L/c$, and τ, respectively. The sample may then respond to this excitation by emitting a
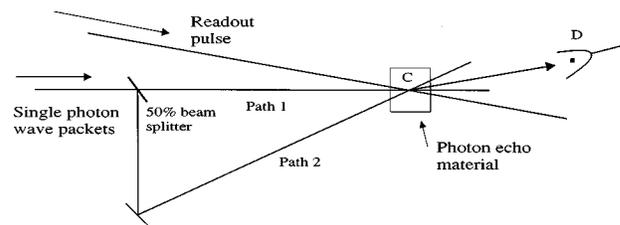

FIG. 1. Schematic illustration of the proposed experiment. Single-photon wave packets are incident on a 50% beam splitter. There are then two different paths to the crossing point C where a photon-echo material is placed. The grating resulting from an accumulation of consecutive single-photon wave-packet excitations is probed by a readout pulse. The time-delayed output signal is registered by the detector D.

coherent pulse towards the detector D at the time $t = \tau + \Delta L / c$. A sufficient condition for this to occur is that $\tau < t_l$, where t_l is the upper state lifetime of the absorbers. The experiment described above is a conventional stimulated photon-echo experiment, e.g., [4].

A reasonable description of the photon-echo process would be that the first wave packet puts the atoms in a superposition between the ground and excited states. This superposition state will emit radiation (free induction decay, e.g., [5]). The electromagnetic field in the second wave packet (reaching the sample at time $\Delta L / c$) will interact with the atoms in this superposition state and depending on the relative phase between the radiation emitted by the superposition state and the electromagnetic field of the second wave packet the upper state admixture in the absorber superposition state will either increase or decrease. This relative phase will be different for different frequency components within Γ_{inhom}. This will result in a frequency domain population grating [6]. Simply put, two pulses emitted at the same time with slightly different frequencies will give an intensity oscillation in time at the difference frequency and similarly two pulses emitted at different times will cause a modulation in frequency space (see also the Appendix). Further, a properly phased grating in frequency space will diffract pulses in time. Specifically, this diffraction in time will cause the echo output to be delayed with respect to the third pulse exactly by an amount of time that corresponds to the time difference between the first and second pulses. Thus part of the third wave packet is absorbed by the grating and later emitted towards the detector. This means that the excitation pulses do not coincide in time with the signal. From signal-to-noise point of view this can be an advantage. Since scattered excitation light does not become a problem it is only necessary that the signal is stronger than the fluorescence.

Now if the intensity in the first wave packet sent in from the left hand side in Fig. 1 is reduced to the single-photon level we mean that a frequency domain grating will still be generated. In accordance with Ref. [3] we will use the term delayed single-photon self-interference for this process. When the intensity is reduced to the single-photon level these single photons are actually carrying out what is generally regarded as a multiphoton process. In the conventional mathematical formulation using photon creation, $a^{\dagger}$, and annihilation, a, operators the interaction of each single photon contributes to an operator combination $a^{\dagger} a$. Normally one would require one photon for the $a^{\dagger}$ contribution and one photon for the a contribution. This issue is further discussed in the calculations in Sec. II C. We believe the experiment described here can stimulate a fruitful discussion of the distinction between single-photon and multiphoton processes. Using the view of the photon-atom interaction process presented here, one condition for the frequency domain grating to occur is that we should regard the interaction between the radiation and the absorber as taking place at two distinctly different times. One key issue of the experiment discussed in this paper is that it aims to demonstrate that a photon (single-photon wave packet) can not only simultaneously take two different routes in space but also interact with an absorber at two distinctly different times. This feature of single-photon wave packets has not been demonstrated before.

The observation of an interference pattern in a single-photon wave-packet experiment would normally imply simultaneous detection of the wave packets. One may on the other hand debate whether excitation processes taking place on a time scale shorter than the homogeneous dephasing time $(= 1/\pi\Gamma_{\text{hom}})$ can be considered as separate. We do not know whether this issue has been discussed explicitly, though few related issues have been debated earlier in connection with quantum jumps [7]. However, two actions occurring at two distinctly different times are necessary to form the frequency domain modulation in a photon-echo process according to the description given here. The description may be too simplistic, but we are not aware of a better physical description of the photon-echo phenomenon. In the experiment discussed here the detection of interference is intertwined with the photon-atom interaction process. The control and description of the detection is therefore quite different compared to electronic registration normally used to study single-photon interference, e.g., [1,2]. Further motivation for investigation of the phenomenon of delayed single-photon self-interference and some new possibilities such an investigation could open up are given in Sec. V.

The experimental investigation as described above has many limitations. Clearly a problem with observing delayed single-photon self-interference experimentally is that a frequency domain grating written by a single photon will be very weak. However, it might be possible to overcome this problem by using the so-called accumulated photon echoes [8]. (It may be mentioned that in the nonaccumulated case new nonclassical features have been predicted for the output field [9]. These features cannot be manifested in the approach suggested in this paper.) For accumulated photon echoes two pulses are sent in to form the grating that could then diffract a third pulse. But instead of sending a third pulse to be diffracted by the grating, a new pulse pair, identical to the first one, is sent in. The excitation created by this second pulse pair then enhances the grating created by the first pair and a sizable grating can be built up by using a large number of such pulse pairs. This obviously is possible only if there are no mechanisms destroying the grating. The idea is then to accumulate a grating from a large number of single-photon absorption events and later probe the accumulated grating with a stronger pulse. As far as we understand, the physics of the absorption process should still be the same. A suitable material for demonstrating delayed single-photon self-interference should then have an inhomogeneously broadened transition and be able to form permanent or near permanent frequency and space domain gratings. In this paper we will explicitly consider the $^7F_0$-$^5D_0$ transition in Eu^{3+}-doped Y_2SiO_5 crystals (site 2). At a temperature of 4 K the relevant material parameters for this crystal are $\Gamma_{\text{inhom}} = 4\,\text{GHz}$, $\Gamma_{\text{hom}} < 1\,\text{kHz}$, $t_l = 1.6\,\text{ms}$ [10]. However, ions decaying from the upper state do not immediately return to the ground state. Instead, they are trapped in an intermediate state for many hours [11] before returning to the ground state. The minimum path length difference ΔL_{min} allowed is approximately determined by the relation $c/\Delta L_{\text{min}} < \Gamma_{\text{inhom}}$. With $\Gamma_{\text{inhom}} = 4\,\text{GHz}$ the minimum time difference allowed between the path one and path two wave packets is approximately $\Gamma_{\text{inhom}}^{-1} = 250\,\text{ps}$. Allowing the single photons to be separated by ten times ΔL_{min} implies that more than 10^{13} single photons can be accumulated in 10 h. As will

be shown later, this seems more than sufficient for observing a signal.

The paper will be organized as follows. The number of accumulated photons needed to observe the delayed single-photon self-interference phenomenon is calculated in the next section using a model based on the Dicke super-radiance [12,13]. Since this is not a very commonly used approach, we first perform the calculation using a more conventional four-wave mixing (FWM) approach. Considering that the light intensity in the experiment goes down to the single-photon level the most appropriate approach for calculating the signal would be to use quantum field operators. Although the Dicke model and FWM approach do agree well with each other we have also performed calculations for quantized input fields. These calculations give no indication that the semiclassical calculations would be in error. On the experimental side single-photon data do not exist at this time. However, the theoretical calculation can readily be extrapolated to nonaccumulated higher energy pulses. In Sec. III the theoretical calculations are compared with preliminary data on Pr-doped Y_2SiO_5 crystals. In Sec. IV we discuss in some detail what materials would be suitable for a delayed single-photon self-interference experiment. In the discussion section we briefly continue our motivation for investigating the phenomenon of delayed single-photon self-interference and mention some possibilities such an investigation could open up. The paper is concluded in a final section.

II. THEORY

To assess the viability of the proposed experiment an expression for the number of photons in a stimulated photon echo from a grating built up from a large number of accumulated single-photon absorption events is needed. We have calculated this quantity in three different ways. All these methods do have some weaknesses, but they are essentially consistent with each other. They are briefly described below. The first method is the conventional perturbation theory approach for a third-order nonlinear process. The second approach is the Dicke model of super-radiance which deals with a signal-to-noise ratio analysis based on the calculated number of photons in the photon-echo data output signal. Although we are discussing an experiment at the single-photon level neither of the two first approaches involves quantization of the input fields. A third approach with (partly) quantized fields is therefore also presented. All the three models give similar results.

A. A perturbation expansion approach

One approach is to perform a standard perturbation expansion in the incoming fields, implicitly assuming that the incident fields are weak. This, in any case, is certainly true for the first two pulses, which in principle consist of "1/2 a photon" each. Photon echoes can be considered as a degenerate FWM process and the third-order contribution to the nonlinear polarization is given by

$$P^{(3)}(t) = \varepsilon_0 \chi^{(3)} A_1^* A_2 A_R. \quad (1)$$

In the above expression A_1 and A_2 are the electromagnetic field amplitudes for the two paths of the single photon, A_R is

the field of the third pulse, and $\chi^{(3)}$ is the third-order susceptibility. Although it is not seen from the expression above there is also a strict time ordering in the photon-echo process. [The order is pulse 1 (A_1), pulse 2 (A_2), and then the read pulse (A_R). A time ordering different from this cannot generate an output in the direction of the detector (D).] The nonlinear polarization acts as a source term in reradiating the FWM signal in accordance with the wave equation

$$\nabla^2 E - \mu_0 \varepsilon_0 \varepsilon \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{nl}}{\partial t^2}. \quad (2)$$

E is the electromagnetic field, P_{nl} is the nonlinear polarization, ε_0 and μ_0 are the permittivity and permeability of free space, respectively, and ε is the index of refraction squared. This wave equation can be simplified using the slowly varying envelope approximation (e.g., [14]) yielding

$$2ik \frac{\partial A_e}{\partial z} = -\frac{\omega^2}{c^2} \chi^{(3)} A_1^* A_2 A_R. \quad (3)$$

Integrating this over the interaction length L, the amplitude of the FWM signal, in our case the photon echo, is obtained as

$$|A_e| = \frac{\pi}{n\lambda} |\chi^{(3)} A_1^* A_2 A_R| L. \quad (4)$$

λ is the excitation wavelength and n is the index of refraction. To obtain this result it is assumed that the field amplitudes were constant over the interaction length L. This is not true for the single-photon fields as they would become zero in case an interaction occurs. Nevertheless, we proceed with the calculations. However, this is a strong reason to also verify the calculations using a different approach as is done later in this section.

The amplitudes A_1 and A_2 are assumed to be of equal magnitude, A_0 say. In front of the beam splitter the electromagnetic field amplitude will be $\sqrt{2}A_0$. A_0 can be calculated assuming a pulse duration T, and beam cross section S.

$$I_{ph} = \frac{h\nu}{ST} = 2n\sqrt{\varepsilon_0/\mu_0}|\sqrt{2}A_0|^2 \Rightarrow A_0$$
$$= [(hc/4nST\lambda)\sqrt{\mu_0/\varepsilon_0}]^{1/2}, \quad (5)$$

where I_{ph} is the intensity and the photon energy is $h\nu = hc/\lambda$.

One straightforward method to estimate the third-order nonlinearity is to use a density matrix calculation assuming degenerate wave mixing and a resonant interaction. The third-order susceptibility in the steady state resonant excitation in the low power limit can be written as (e.g. [14], Chap. 3)

$$|\chi^{(3)}| = \frac{N}{\varepsilon_0 \hbar^3} \frac{|\mu|^4}{\gamma^3}, \quad (6)$$

where N is the concentration of active atoms/ions that lie within the spectral Fourier bandwidth of the pulses, μ is the transition dipole moment, and γ is the homogeneous transition linewidth. For transient excitations using pulses of width

T, where $T \ll 1/\gamma$, the atomic polarization created in the sample will start from zero and increase proportionally to T at rate γ. For such short times the polarization would not have had time to develop to its full steady state value. To compensate for this Eq. (6) should be multiplied by a factor γT for each transient excitation. Since this process is being viewed as a FWM process with three excitation pulses the multiplication factor becomes $(\gamma T)^3$. For the case in which we are interested, viz., the accumulated photon echo, the nonlinear susceptibility can be obtained as

$$|\chi^{(3)}| = \frac{N}{\varepsilon_0 \hbar^3} |\mu|^4 T^3 M, \tag{7}$$

where it is assumed that M pairs of pulses, each with a width T, contribute and that no shift of phase occurs between the pulses within each pulse pair.

The amplitude of the readout pulse A_R is chosen such that the pulse area equals $\pi/2$ which ensures that the output signal is maximized. Hence

$$\frac{2|\mu|A_R}{\hbar} T = \frac{\pi}{2} \Rightarrow A_R = \frac{h}{8|\mu|T}. \tag{8}$$

Substituting Eqs. (5), (7), and (8) into Eq. (4) and using the fact that $|A_1| = |A_2| = |A_0|$, the output signal amplitude can be obtained as

$$|A_e| = \frac{\pi^4}{4} \left(\frac{\mu_0}{\varepsilon_0} \right)^{1/2} \frac{Nc}{\varepsilon_0 n^2 h \lambda^2} |\mu|^3 T \frac{ML}{S}. \tag{9}$$

The intensity of the output pulse is given by $I_e = 2n\sqrt{\varepsilon_0/\mu_0}|A_e|^2$. This can be used to calculate the number of photons in the output signal given by $N_{\text{ph1}} = I_e ST\lambda/hc$. Assuming confocal focusing $(L/S = 2/\lambda)$, the number of photons in the output field using this model can be obtained as

$$N_{\text{ph1}} = \frac{\pi^8}{4} \frac{1}{(n\varepsilon_0 h\lambda)^3} |\mu|^6 N^2 T^3 M^2 \frac{L}{\lambda}. \tag{10}$$

The numerical estimates using typical experimental parameters will be shown in a later section.

B. An approach based on the Dicke super-radiance model

An expression for the number of photons in a stimulated photon echo has been derived earlier [13] based on the description by Abella, Kurnit, and Hartmann [15]. The underlying idea in this analysis is that the photon echo is emitted from an ensemble of excited atoms that together have a microscopic dipole moment (a super-radiant state). It will be sufficient to calculate the number of atoms that will emit radiation in order to determine the signal strength. The number of photons S_e emitted from such a super-radiant state during a time t is approximately

$$S_e = \frac{N_{\text{atom}}^2 p(t)}{4}. \tag{11}$$

N_{atom} is the number of excited atoms and $p(t)$ is the probability that any single atom will radiate during the time t

[12,15]. Consider a material doped with active centers of some concentration N, which has a transition with an inhomogeneous linewidth $1/2\pi T_2^*$, where T_2^* is the inhomogeneous relaxation time. Assuming that the laser linewidth is Fourier limited, the number of centers in a sample of length L, excited by a laser pulse, of cross-sectional area S and duration T, is

$$N_{\text{atom}} = \alpha_f NSL \frac{T_2^*}{T}. \tag{12}$$

T_2^*/T is the fraction of centers within the inhomogeneous linewidth that are excited by the pulse of duration T and α_f is the fraction of atoms within the frequency bandwidth and volume excited to the upper state $(\alpha_f \lesssim 1)$. The probability for any single atom to make a transition by spontaneously emitting a photon during the time T is AT, where A is the transition probability. If the first two pulses in the photon-echo process have a pulse area equal to θ $(\theta = 2\mu TE/\hbar$, E is the electromagnetic field amplitude of the excitation pulses) and the readout pulse has $\theta = \pi/2$, the number of photons emitted in a photon echo can be written [13]

$$N_e = \left(\frac{N}{2} SL \frac{T_2^*}{T} \right)^2 (TA) \left(\frac{3\lambda}{8Ln} \right) \eta^2 \sin^4 \theta. \tag{13}$$

The factor $(3\lambda/8Ln)$ arises when the photon-echo signal is integrated over all angles of propagation (assuming $S \ll 4\lambda L$ [15], appendix C). Here λ is the transition wavelength and n is the index of refraction. η is basically the fraction of the originally excited atoms that will participate in the generation of the echo signal. For a two-pulse photon echo η may equal unity since the rephasing of the individual atomic dipole moments here can be perfect. For a stimulated echo this is not the case and η can at the most take the value $1/2$. The ratio T_2^*/T can never be larger than unity. Thus in general T_2^* should be replaced by min $\{T_2^*, T\}$. However, this has been suppressed in the subsequent equations owing to notational simplicity.

If the incident single photon is considered to be split at the beam splitter then the pulse area of the individual wave packets can be considered as that of half a photon, then $\theta = 2\mu TA_0/\hbar$, where A_0 is the individual wave-packet amplitude. Now the incident field corresponds to a beam of photons of wavelength λ, pulse width T, and area of cross section S. The amplitude A_0 which is being considered as that of half a photon is given by Eq. (5). For weak pulses

$$\sin^4 \theta \approx \theta^4 = \frac{16|\mu|^4 T^4}{\hbar^4} A_0^4. \tag{14}$$

The transition probability A is related to $|\mu|$ [16] through

$$A = \frac{16\pi^3 |\mu|^2}{3\varepsilon_0 h\lambda^3}. \tag{15}$$

An expression for the number of photons in the output signal can be obtained for this model, by substituting Eqs. (5), (14), and (15) into Eq. (13), as

$$N_{\text{ph2}} = \frac{\pi^7}{2} \frac{1}{(n\varepsilon_0 h\lambda)^3} |\mu|^6 (T_2^* N/T)^2 T^3 \frac{L}{\lambda} \eta^2. \qquad (16)$$

Extending this calculation for the accumulated photon-echo case, the signal strength, when M pulse pairs contribute and assuming no shift of phase between the pulses within each pulse pair, can be obtained as

$$N_{\text{ph2}} = \frac{\pi^7}{2} \frac{1}{(n\varepsilon_0 h\lambda)^3} |\mu|^6 \left(\frac{T_2^* NM}{T}\right)^2 T^3 \frac{L}{\lambda} \eta^2. \qquad (17)$$

The expression in Eq. (15) probably should be considered as being more accurate than the expression in Eq. (10). For one thing the efficiency in the process of engraving the frequency domain grating into the material has been considered and then the integration over the volume where the coherent emission takes place is also implicitly included through the treatment described in Ref. [15]. Here a cylindrically shaped area is assumed while confocal focusing is assumed in deriving Eq. (10). Significantly, the two estimates of the accumulated photon-echo signal strength in Eqs. (10) and (17) differ only by a factor of $\pi/2$, which must be considered reasonably satisfactory since the values have been derived in two quite different ways. However, since the experiment involves intensities close to a single-photon level it is, in principle, necessary to deal with a complete quantum mechanical treatment. The consistency between the above two semiclassical approaches probably indicates that no major change would occur even if the field quantization is considered. Nevertheless we also present a preliminary and somewhat simplified study on the quantum mechanical approach.

C. A quantized field approach

As a starting point we note that the stimulated photon-echo process basically can be described as a degenerate FWM process. For example, in the phase-conjugate configuration of degenerate FWM, two strong waves commonly act as pump fields and two counterpropagating weak waves get amplified. The relevant term in the interaction Hamiltonian H_{int} is proportional to $H_{\text{int}} \propto a_f a_b a_p^\dagger a_c^\dagger$, where the $a^\dagger$ are the creation operators and the a are the annihilation operators for the forward (f) and backward (b) pumps and the weak probe (p) beam and its conjugate output (c) beam. The basic process therefore involves an annihilation of one photon from mode f (at ω_f) and one from mode b (at ω_b) and a simultaneous creation of one photon at ω_p and one at ω_c. Here we consider the incident forward field, comprised of A_1 and A_2 in our case, to be generated by a single photon, the readout pulse (corresponding to one of the pump fields in the FWM configuration) is modeled as a classical intense pulse, and the echo (corresponding to the conjugate output pulse in FWM is generated at the appropriate time and in the appropriate direction (the output here is not necessarily phase conjugated, in fact other phase matching arrangements could be more appropriate).

We consider the case of three interacting fields $A_1(\mathbf{r})$, $A_2(\mathbf{r})$, and $A_R(\mathbf{r})$, at the frequency ω. If the output field is taken as $A_e(\mathbf{r})$ then the field interaction Hamiltonian density can be written as

$$U = -\chi^{(3)} \varepsilon_0 A_1 A_2 A_R A_e. \qquad (18)$$

Since we are considering a strong readout pulse, we treat $A_R(\mathbf{r})$ classically but the other fields are quantized by using their corresponding quantum mechanical operators. The representation is of the form

$$A_l(\mathbf{r},t) = -i\left(\frac{\hbar\omega}{2V\varepsilon_0}\right)^{1/2} A_l(\mathbf{r})[a_l^\dagger(t) - a_l(t)], \qquad (19)$$

where V is the mode volume.

The field interaction Hamiltonian can be written as

$$H_{\text{int}} = \int_\nu U\, d\nu = -i\varepsilon_0 \chi^{(3)} \left(\frac{\hbar\omega}{2V\varepsilon_0}\right)^{3/2} A_R'$$

$$\times \int_\nu A_1(\mathbf{r}) A_2(\mathbf{r}) A_R(\mathbf{r}) A_e(\mathbf{r}) d\nu$$

$$\times \cos\omega t (a_1^\dagger - a_1)(a_2^\dagger - a_2)(a_e^\dagger - a_e), \qquad (20)$$

where the readout field has been taken as

$$A_R(\mathbf{r},t) = A_R' A_R(\mathbf{r}) \cos\omega t \qquad (21)$$

and the integration is performed over all space. We define a parameter

$$s = -i\chi^{(3)} \hbar^{1/2} \left(\frac{\omega}{2V\varepsilon_0}\right)^{3/2} \varepsilon_0 A_R' \int_\nu A_1(\mathbf{r}) A_2(\mathbf{r}) A_R(\mathbf{r}) A_e(\mathbf{r}) d\nu, \qquad (22)$$

which can be treated as the strength of the interaction and it is proportional to the readout pulse amplitude A_R'. The functions $A_l(\mathbf{r})$ have to be so chosen such that the integral yields the interaction volume which is the region of interest,

$$\int_\nu \prod A_l(\mathbf{r}) d\nu = \mathbf{V_i}. \qquad (23)$$

We can rewrite Eq. (20) as

$$H_{\text{int}} = s\hbar \cos\omega t (a_1^\dagger - a_1)(a_2^\dagger - a_2)(a_e^\dagger - a_e). \qquad (24)$$

The total Hamiltonian is taken as the sum of the unperturbed ($s=0$) Hamiltonian, obtained by considering the field as an ensemble of independent harmonic oscillators, and the interaction Hamiltonian given above. This can be written as

$$H = \sum_{l=1,2,R,e} \hbar\omega(a_l^\dagger a_l + \tfrac{1}{2}) + s\hbar \cos\omega t (a_1^\dagger - a_1)$$

$$\times (a_2^\dagger - a_2)(a_e^\dagger - a_e). \qquad (25)$$

The equation of motion in the Heisenberg representation, for any operator A is

$$\frac{dA}{dt} = -\frac{i}{\hbar}[A,H]. \qquad (26)$$

Using this on the operator $a_e^\dagger$, we obtain

$$\frac{da_e^\dagger}{dt}=-\frac{i}{\hbar}\left[a_e^\dagger,\sum_l\hbar\omega\left(a_l^\dagger a_l+\frac{1}{2}\right)\right]$$

$$-\frac{i}{\hbar}[a_e^\dagger,s\hbar\cos\omega t(a_1^\dagger-a_1)(a_2^\dagger-a_2)(a_e^\dagger-a_e)]. \tag{27}$$

Using the commutator relations

$$[a_l,a_m]=[a_l^\dagger,a_m^\dagger]=0 \quad \text{and} \quad [a_l,a_m^\dagger]=\delta_{l,m} \tag{28}$$

from Eq. (27) we obtain

$$\frac{da_e^\dagger}{dt}=i\omega a_e^\dagger-is\cos\omega t(a_1^\dagger-a_1)(a_2^\dagger-a_2). \tag{29}$$

Since the interaction in which we are interested is of the degenerate FWM type we need to choose only those terms that would contribute to a field at ω. The phase matching constraints do also further govern the choice of the contributing terms. Thus we obtain

$$\frac{da_e^\dagger}{dt}=i\omega a_e^\dagger+\frac{is}{2}a_1^\dagger a_2 e^{i\omega t}. \tag{30a}$$

Similar equations can be obtained for the other operators as

$$\frac{da_1^\dagger}{dt}=i\omega a_1^\dagger+\frac{is}{2}a_2 a_e^\dagger e^{i\omega t}, \tag{30b}$$

$$\frac{da_2}{dt}=-i\omega a_2+\frac{is}{2}a_1 a_e e^{-i\omega t}. \tag{30c}$$

By substituting $a_e^\dagger(t)=A(t)e^{i\omega t}, a_1^\dagger(t)=B(t)e^{i\omega t}$, and $a_2(t)=C(t)e^{-i\omega t}$ in the above equations and choosing the dc terms, they can be reduced to

$$\frac{dA(t)}{dt}=\frac{is}{2}B(t)C(t), \tag{31a}$$

$$\frac{dB(t)}{dt}=\frac{is}{2}C(t)A(t), \tag{31b}$$

$$\frac{dC(t)}{dt}=\frac{is}{2}A^*(t)B^*(t). \tag{31c}$$

This set of coupled equations leads to solutions that take the form of Jacobi elliptic integrals. For now we restrict ourselves to looking at the solutions when either two or all the fields are treated classically. It is also illustrative to look at both the time and spatial evolution of the operators. This can form a logical basis for comparison with the semiclassical results described in the earlier section.

1. Case (i): One quantized field

If all the three incoming fields are treated as classical fields given in Eq. (21), then the interaction Hamiltonian can be rewritten as

$$H_{\text{int}}=s_1\hbar\cos^3\omega t(a_e^\dagger-a_e), \tag{32}$$

where the parameter s_1 is defined in terms of the field amplitudes and the interaction volume as

$$s_1=-\frac{i}{\hbar}\varepsilon_0\chi^{(3)}\left(\frac{\hbar\omega}{2\varepsilon_0 V}\right)^{1/2}A_1' A_2' A_R' \mathbf{V_i}. \tag{33}$$

Following the procedure outlined above, the equation of motion for the output signal photon creation operator, in the Heisenberg representation, can be written as

$$\frac{da_e^\dagger}{dt}=i\omega a_e^\dagger-is_1\cos^3\omega t. \tag{34}$$

choosing $a_e^\dagger(t)=A(t)e^{i\omega t}$ and choosing only the dc terms we get

$$\frac{dA(t)}{dt}=-\frac{3}{8}is_1. \tag{35}$$

Integrating and using the initial condition $a_e^\dagger(0)=A(0)$, the solution is obtained as

$$a_e^\dagger(t)=a_e^\dagger(0)-\frac{3}{8}is_1 t. \tag{36}$$

2. Case (ii): Two quantized fields

In typical FWM the pumps are generally assumed to be much more intense than the probe and conjugate beams, and can be treated classically in a nondepleted pump approximation. Consider A_1 and A_R to be classical as given in Eq. (21). Then the interaction Hamiltonian can be rewritten as

$$H_{\text{int}}=s_2\hbar\cos^2\omega t(a_2^\dagger-a_2)(a_e^\dagger-a_e), \tag{37}$$

where the parameter s_2 is defined in terms of the field amplitudes and the interaction volume as

$$s_2=-\chi^{(3)}\left(\frac{\omega}{2V}\right)A_1' A_R' \mathbf{V_i}. \tag{38}$$

Considering the total Hamiltonian, the equation of motion in the Heisenberg representation for the creation and annihilation operators for the two interacting fields can be written. Once again choosing only those terms which contribute to a signal field oscillating at the frequency ω, and neglecting the nonsynchronous terms a set of coupled equations can be obtained. These coupled equations can be solved and the complete solution for the field operators can be written in terms of $\Omega(=s_2/4)$ as

$$a_e^\dagger(t)=[a_e^\dagger(0)\cosh\Omega t-is_2 a_2(0)\sinh\Omega t]e^{i\omega t}, \tag{39a}$$

$$a_2(t)=[a_2(0)\cosh\Omega t+is_2 a_e^\dagger(0)\sinh\Omega t]e^{-i\omega t}. \tag{39b}$$

D. Calculation of the number of signal photons

Our main interest is to estimate the number of quanta in the output echo signal. This can be calculated using the number operator $a_e^\dagger(t)a_e(t)$ for the output field. The expectation value of a quantum mechanical operator A is given by $\langle A\rangle=\langle\Psi(0)|A_H(t)|\Psi(0)\rangle$ where $A_H(t)$ is the operator in the

Heisenberg representation and $\Psi(0)$ is the solution of the time-dependent Schrödinger equation taken at $t=0$. Thus the number of photons in the signal state is calculated using

$$\langle n_e\rangle = \langle\Psi(0)|a_e^\dagger(t)a_e(t)|\Psi(0)\rangle. \tag{40}$$

Let us assume for the sake of generality that the radiation field at $t=0$ has n_{2i} photons in the incident field and n_{ei} photons in the echo field. The corresponding wave functions for the two cases discussed above can be written as

$$\Psi(0) = \begin{cases} |n_{ei}\rangle & \text{for case (i)} \tag{41} \\ |n_{ei},n_{2i}\rangle = |n_{ei}\rangle|n_{2i}\rangle & \text{for case (ii),} \tag{42} \end{cases}$$

where $|n_i\rangle$, for example is the harmonic oscillator wave function that obeys the usual relations with the operators a_i and $a_i^\dagger$. The states also satisfy the orthonormality condition $\langle n_k+p|n_k+q\rangle = \delta_{pq}$.

Thus for case (i) we obtain the number of output photons as

$$\langle n_e(t)\rangle = n_{ei} + \frac{9s_1^2t^2}{64}. \tag{43}$$

For case (ii) the time evolution of the number of photons in the signal field is given by

$$\langle n_e(t)\rangle = n_{ei}\cosh^2\Omega t + (n_{2i}+1)\sinh^2\Omega t. \tag{44}$$

E. Spatial evolution

It has been shown that for the standard FWM geometry with counterpropagating beams in a nonlinear medium of length L and a third-order susceptibility $\chi^{(3)}$ the following relation holds [17]:

$$a_e^\dagger(0) = \frac{a_e^\dagger(L)}{\cos(|\kappa|L)} + i\frac{\kappa}{|\kappa|}a_1(0)\tan(|\kappa|L), \tag{45}$$

where the operator $a_e^\dagger(t)$ denotes the creation operator for the output beam and $a_1(t)$ is the annihilation operator for the probe beam. κ, the coupling constant, is defined in terms of nonlinear susceptibility and the pump amplitudes as $|\kappa| = (\omega/2)\sqrt{\mu/\varepsilon}|\chi^{(3)}A_2A_R|$.

Using this to construct the number operator and also using an appropriate wave function it can be shown that the number of output photons is given by

$$\langle n_e(l)\rangle = \frac{n_{ei}}{\cos^2(|\kappa|L)} + (n_{2i}+1)\tan^2(|\kappa|L), \tag{46}$$

where n_{ei} and n_{2i} are the initial photon occupation numbers in the two propagation modes. These operators can be used to calculate the number of photons in the output field and a comparison with the semiclassical models can be made.

F. Comparison with semiclassical models

Let us consider the spatial evolution to start with. Ignoring the noise terms in Eq. (46) and with $n_{ei}=0$, the number of photons in the output field at low coupling strengths is

$$N_{\text{ph}} \approx |\kappa|^2L^2n_{2i}. \tag{47}$$

Using the coupling constant $|\kappa| = (\omega/2)\sqrt{\mu/\varepsilon}|\chi^{(3)}A_2A_R|$, and expressing the photon numbers in terms of intensity and subsequently the field amplitude ($A_i = \sqrt{2I_1/\varepsilon_0 nc}$), the following expression can be arrived at:

$$A_e = i\frac{\pi}{n\lambda}\chi^{(3)}A_1^*A_2A_RL, \tag{48}$$

which is the same as Eq. (4).

The time-dependent expressions also reproduced the semiclassical results (under reasonable choices of interaction volume and interaction time). For example, Eq. (43), with no input photons in the echo mode ($n_{ei}=0$), can be rewritten in terms of the amplitudes and the interaction parameter s given in Eq. (33). With $n_e = (2STc\varepsilon_0 A_e^2)/h\nu$ (choosing the index of refraction to be unity) we obtain

$$A_e = i\frac{3}{8}\chi^{(3)}A_1A_2A_R\frac{\omega t}{2n}\frac{\mathbf{V_i}}{V}. \tag{49}$$

Since the volume of interaction $\mathbf{V_i}=SL$ and the mode volume $V=STc$, where S is the spot area, T is the pulse width, and c is the velocity light, this expression is equivalent to Eq. (4) or Eq. (48) apart from a factor 3/8. A similar result can be obtained for the case with two quantized fields [Eq. (44)], for weak interaction strengths, $\Omega T\ll 1$.

G. Conclusion of the theoretical considerations

As two different semiclassical approaches largely agree and they concur with the calculations with one and two quantized fields it is reasonable to believe (but clearly not explicitly shown) that the expressions that have been derived would hold also when all input fields are quantized. We therefore feel that we are on reasonably solid ground in the next section when we use the derived equations to estimate the signal strength in real experiments.

III. NUMERICAL ESTIMATION OF SIGNAL STRENGTH AND COMPARISON WITH EXPERIMENT

Prior to the design of the experimental setup it is essential to have a numerical estimate of the signal strength based on the above analyses. It is enough to consider just one of the semiclassical expressions to have an estimation of the output signal strength.

A. Numerical estimate

We choose the approach described in Model 2. Equation (16) can be further modified to a more suitable form by expressing $|\mu|$ in terms of the oscillator strengths f. This relation assuming two singly degenerate levels is given by

$$|\mu^2| = \frac{3he^2\lambda}{8\pi^2cm}f. \tag{50}$$

Then Eq. (17) can be modified to give the number of output photons N_{ph}, for M incident pulse pairs, as

$$N_{\text{ph}} = \frac{27\pi}{1024}\left(\frac{e^2}{mcn\varepsilon_0}\right)^3(MT_2^*N)^2T\frac{L}{\lambda}\eta^2f^3. \tag{51}$$

Before going to the single-photon case it is illustrative to see the estimates when pulses with a larger number of photons are used. This is a multiphoton case and the number of output photons can be calculated from Eq. (51) under the assumption of small pulse areas. The number of output photons is proportional to the product of the number of photons in beam 1 and beam 2 (Fig. 1). Equation (51) is derived assuming amplitudes corresponding to 1/2 photon in each beam. With the same number of excitation photons, N_{exc}, in beam 1 and beam 2 one obtains for a single exciting pulse pair that

$$N_{ph}^m = \frac{27\pi}{1024}\left(\frac{e^2}{mcn\varepsilon_0}\right)^3 (T_2^* N)^2 T\frac{L}{\lambda}\,\eta^2 f^3 \left(\frac{N_{exc}}{1/2}\right)^2. \quad (52)$$

With $\Gamma = (2\pi T_2^*)^{-1}$ we obtain

$$N_{ph}^m = 1\times 10^{-17}\frac{1}{n^3\lambda}LT\left(\frac{N\eta}{\Gamma}\right)^2 f^3 N_{exc}^2. \quad (53)$$

This equation expresses the number of output photons as a function of material parameters and the input pulse duration and intensity.

B. Experimental verification

In order to make a comparison between experimental data and the theoretical calculations, a stimulated photon-echo experiment is performed on the site 2, ${}^3H_4$-${}^1D_2$ transition of a $Pr^{3+}:Y_2SiO_5$ crystal. The sample has the following parameters: crystal length $L=5$ mm, Pr ion number density $N=5\times 10^{24}/m^3$ (there are Pr ions in 0.05% of the Y_2SiO_5 cells), the oscillator strength $f=2\times 10^{-8}$, and the inhomogeneous linewidth $\Gamma_{inhom}=3.5\times 10^9$ Hz [18]. The other relevant parameters are the excitation pulse duration $T=1$ μs which corresponds to a $\pi/2$ pulse at the experimental excitation power of 22.4 mW, $\lambda=607.9$ nm which is at the line center of the inhomogeneous line profile, and $\eta=1/2$ for stimulated photon echo with three $\pi/2$ pulses. Using these values and Eq. (53), we obtain

$$N_{ph}^m = 9.9\times 10^{-14}N_{exc}^2. \quad (54)$$

In the experiment, a ring dye laser (Coherent-699-21) pumped by an Ar^+ ion laser (Spectra-Physics 171) is used for the excitation. The excitation pulses are generated by gating the dye laser radiation using two Isomet 1205C acousto-optic modulators in series. The experimental setup is shown in Fig. 2. The loss in cryostat windows etc. when the laser is tuned off resonance was measured to be about 80% and the absorption at the inhomogeneous line center is about 60%. The excitation power at the front of the crystal is 22.4 mW which corresponds to 6.8×10^{10} photons. The echo photon number measured in the experiment is 7.9×10^8, while the theoretical echo photon number calculated using Eq. (54) is 4.6×10^8. Thus the experimental data essentially are in agreement with the theoretical calculation and it is reasonable to believe that the signal strength estimations can be trusted to within an order of magnitude or so.
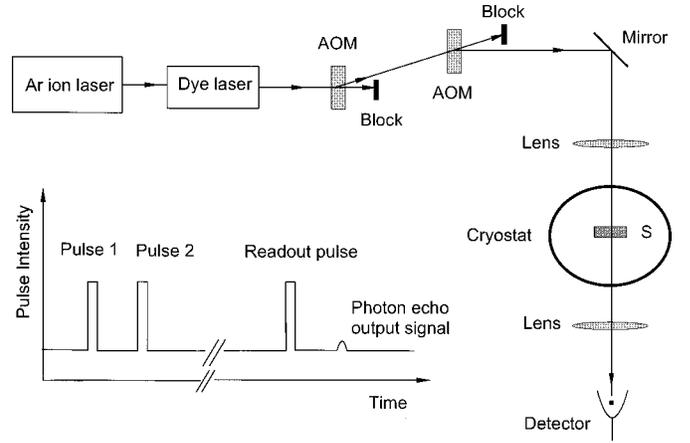

FIG. 2. Schematic view of the experimental arrangement used to compare the theoretical signal strength calculations with experimental data. The excitation pulses are switched on using two acousto-optic modulators (AOM). S is the photon-echo sample, which is placed in a cryostat to eliminate line broadening from thermal phonons. The inset schematically shows the relative timing of the excitation pulses and the echo signal.

IV. DESIGN OF THE EXPERIMENT, CHOICE OF MATERIALS

The experiment described above indicates that the signal strength calculations are reasonable. The next step would be to find good materials for the single-photon wave-packet experiments. We will consider the groups of inorganic and organic solid state materials commonly used in spectral hole burning and photon-echo experiments at liquid helium temperatures. In choosing samples for the present experiment we note that the optimum absorption in a sample used for photon echoes [19] (as well as the optimum absorption for a sample used in any other typical FWM experiment) is of the order of unity. Thus we would want

$$\alpha L = \frac{1}{\pi}\left(\frac{2}{\pi}\right)^{1/2}\frac{N\sigma_0 L}{\Gamma_{inhom}} \approx 1 \Rightarrow NfL \approx 4\times 10^5\Gamma_{inhom}. \quad (55)$$

α is the absorption coefficient at resonance, σ_0 is the peak absorption cross section, and f is the oscillator strength. A distinction can be made between the organic and inorganic materials based on the difference in the inhomogeneous linewidths. For inorganic crystals $\Gamma_{inhom}\approx 10^9$ Hz, and a typical crystal thickness could be $L\approx 10^{-3}-10^{-2}$ m. For such a sample the optimum value for Nf would be $Nf \approx 10^{17}-10^{18}$. Since the oscillator strengths of the inorganic materials are low, i.e., $f\approx 10^{-9}-10^{-6}$, the optimum concentration or number density of the absorbing centers would then be $N\approx (10^{23}-10^{27})/m^3$.

Organic materials have been used in the form of dopants in polymer thin films or glasses and have much shorter length. Hence the material parameters of interest can be taken as $L\approx 10^{-5}-10^{-3}$ m, $\Gamma_{inhom}\approx 10^{12}$ Hz, and $f \approx 10^{-3}-10^{-2}$. The optimum number density of the absorbing centers in this case would be $N\approx (10^{23}-10^{26})/m^3$. This indicates that both inorganic and organic materials can be tailored to yield optimum absorption. Another criterion which governs the choice of the material is the dephasing

[both homogeneous (T_2) and inhomogeneous (T_2^*)] time. A necessary condition for the pulse width T, as has been discussed earlier in the Introduction, is $T_2^* < T < T_2$. This sets a limit on the pulse width of the laser source and a suitable choice has to be made depending on the material.

A. Inorganic materials

The number of output photons is given by Eq. (19) with $N_{exc} = 1/2$,

$$N_{ph} = 2.5 \times 10^{-18} \frac{1}{n^3 \lambda} (MT)^2 \left\{ \frac{NfL}{\Gamma} \right\}^2 \left[\eta^2 \frac{f}{TL} \right]. \quad (56)$$

The term in the curly brackets has roughly the value 4×10^5 for the optimum case as discussed in connection with Eq. (55). The quantity MT which is the product of number of pulse pairs used in the experiment and the duration (T) of a single pulse (basically the coherence time of the single-photon wave packet) indicates the accumulation time in order to detect a signal. The index of refraction and the excitation wavelength do not change too much between different samples, thus the term in square brackets can be construed as the material optimization parameter. It can be seen that a material with larger oscillator strength and preferable thinner but, most importantly, with a high η (high efficiency for burning a spectral grating) should be chosen. The lifetime of this spectral grating puts an upper limit on the product MT. A short laser pulse width should be chosen, typically $T \approx T_2^*$. For inorganic rare-earth-ion-doped crystals the typical inhomogeneous dephasing times are in the range $10^{-9} - 10^{-11}$ s.

We consider a typical numerical example for the single-photon field case [Eq. (56)], and work out the accumulation density necessary for an observable output signal. A reasonable choice would be a Eu-doped Y_2SiO_5, (site 2) crystal. For such a crystal the inhomogeneous linewidth $\Gamma_{inhom} = 5 \times 10^9$, which gives $T_2^* \approx 30$ ps, $f = 3 \times 10^{-8}$, and assuming Eu in site 2 in 0.1% of the Y_2SiO_5 cells, $N \approx 10^{25}$. From Eq. (55) we then obtain that $L = 0.007$ m. A reasonable value for the writing efficiency is $\eta = 1/4$. Choosing $T = T_2^*$, $\lambda = 582$ nm, $n = 1.5$, we get

$$N_{ph}^M = 1.75 \times 10^{-18} M^2. \quad (57)$$

In order to get an output signal of 100 photons, i.e., $N_{ph} = 100$, the number of single photons that must be accumulated will be $M = 7.7 \times 10^9$. At 80 MHz repetition rate, and with one photon in every tenth shot this would imply that the duration of such an experiment would approximately be about 15 min. Since the grating lifetime is about 20 h the experiment clearly should be feasible. There is an obvious scope for improvement in the duration time. For example, a material with two orders of magnitude increase in the oscillator strength, and with a corresponding decrease in the number of concentration centers, would decrease the accumulation time by one order of magnitude.

B. Organic materials

Typical homogeneous dephasing times of organic materials are in the range $10^{-10} - 10^{-9}$ s. Thus a source with a pulse width of a few picoseconds will be necessary. It has been mentioned earlier that the material thickness is quite small. This implies that the assumption $S \ll 4\lambda L$, where S is the spot size, used to obtain Eq. (13), does not hold. Equation (13) can then be modified by multiplying the left hand side by the quantity ($\lambda L / \pi n S$) (appendix C in Ref. [15]). The modified expression for the output signal strength [Eq. (24)] can then be written as

$$N_{ph} = 2.5 \times 10^{-18} \frac{1}{\pi n^4} (MT)^2 \left\{ \frac{NfL}{\Gamma} \right\}^2 \left[\eta^2 \frac{f}{TS} \right]. \quad (58)$$

It can be seen from this expression that shorter pulse widths, high oscillator strengths, as well as large η are necessary for a large output signal. In addition focusing of the laser pulse to a small area would also improve the signal strength. For the materials that are currently available and may be suitable for this experiment, some typical values are [20] $\eta = 0.002$, $\Gamma = 7 \times 10^{12}$, $f = 0.01$. With the number density of the absorption centers chosen as $N = 10^{25}$ the sample thickness L will have to be about 30 μm to ensure that the sample absorption (αL) is about unity. Using a source of wavelength 600 nm focused down to a 30 μm diameter spot and a material refractive index of 1.5, the accumulation factor M can be obtained.

Requiring a signal of 100 photons at the output, for a mode-locked picosecond system with $T = 3 \times 10^{-11}$ one obtains $M = 1.5 \times 10^9$. At 80 MHz repetition rate, and with one photon in every tenth shot this would correspond to an accumulation time of 180 s. This is a very encouraging result and suggests that the experiment is highly feasible. Once again an organic material doped into a thicker host would reduce the accumulation time. An order of magnitude improvement in the efficiency η would lead to an order of magnitude decrease in the accumulation time.

V. DISCUSSION

Though our main motivation for carrying out the experiment is to investigate the ideas that what normally is regarded as a multiphoton process is carried out by single photons and that an absorption process is separated into two events occurring at two different times, it is also possible to formulate other at least seemingly intriguing questions. For example, assume that a single photon is incident from the left in Fig. 1 and that the part of the wave packet taking path 1 interacts with a certain atom. A question that arises is, will the second wave packet interact with the same atom? Such a question could, for example, be motivated by the fact that there would normally be millions of atoms to choose between. A plausible reason as to why the second wave packet would choose the same atom as the first wave packet would be that there is some memory from the first wave packet.

Suppose that the part of the wave taking path 1 interacts with a specific atom. When all the other atoms see the part of the wave packet that has taken route 2, would they know that this is a second part of a single-photon wave packet and that the first part has already passed by (and interacted with some other atom) and if so at what moment in time do they obtain this knowledge? Alternatively one could ask whether one atom is excited or there exists a superposition of excited atoms and whether this choice is made at a later stage. Im-

portantly, why should both parts of the single-photon wave packet choose the same atom, i.e., why should the photon wave function not just reduce to a single path? We know that for a standard Young's double slit experiment the fringe visibility on a photographic plate placed behind the slits can be very high. From this observation it seems one needs to conclude that the wave function does not reduce to a single path unless it is forced to do so. Thus in the limit of weak overall absorption there is no reason why the wave function for the single-photon wave packet should reduce to a single path here either. A reasonable point of view could be to say that each single atom in the absorber has an observation window that is so narrow in frequency that the two wave packets from their perspective do overlap in time. In this way the issue of two interactions separated in time can be avoided.

On the other hand, one may consider a Ramsey fringe experiment on an atomic beam [21]. After the excitation in the first interaction zone the dipole moment is precessing while the atom travels in the field free space into the next interaction zone. Here it is certainly difficult to envision that the atoms due to their narrow-band absorption lines experience a narrow-band frequency component in the field with a duration longer than or equal to the time it takes to travel between the interaction zones, since obviously there is no field at the physical location between the interaction zones.

As photon-echo generation is a coherent FWM process there is not necessarily any spontaneous emission process occurring. Therefore the wave function actually does not have to collapse at any time during the process. (A first experiment would allow the upper state atoms to decay in order to form a semipermanent population grating that can be read out but, in principle, no collapse of the atomic wave function is required to read out the signal, because the readout pulse may be applied after a time which is much shorter than the upper state lifetime, turning the interaction process into a truly parametric process.) In the absence of a collapse of the atom wave function there may also be other unsolved issues within this type of experiment. It is not clear here how many photons have to be used to describe this four-wave mixing process where the first two waves are substituted by an accumulation of split single-photon wave packets and what the energy conservation and phase matching would be, in such a process. Further, how would the energy conservation and phase matching diagrams look in this case?

VI. SUMMARY

In 1993 Kessel and Moiseev suggested [3] that a single photon can interfere with itself even if the difference between the two paths in the interferometer is larger than the ''length'' of the photon. We have shown that the interpretation of such an experiment raises several interesting questions related to atom-photon interactions and single-photon interference. In this paper we have made detailed calculations on the signal-to-noise and detection limits for such an experiment. We have verified our theoretical calculations with experimental data and discussed how to select a material for the experiment. We have emphasized the point of view that the absorption of a single-photon wave packet occurs at two distinctly different moments in time and that this single photon effectively carries out a multiphoton interaction process. We believe that this experiment can stimulate a discussion at a fundamental level regarding the absorption of a photon and the subsequent atom-photon interaction.

APPENDIX

For the case when the electromagnetic field radiated by the atoms in the superposition state created by the first wave packet is in phase with the field in the second wave packet, the interaction with the second pulse basically has the effect of inducing stimulated emission in the absorber atoms. This then leads to a decrease of upper state admixture in the absorber wave function. For absorbers radiating with the opposite phase, the upper state admixture in the superposition will instead increase during the interaction with the second wave packet. The phase of the radiation emitted by the superposition state is determined by the interaction of the first wave packet. The absorbers start to oscillate at their eigenfrequency at time $t = 0$. Thus two absorbers within the inhomogeneous line separated in frequency by $\Delta \nu = c/2\Delta L$ will have opposite phase when the second pulse enters the sample at time $\Delta L/c$. In this way the upper state admixture in the superposition state will vary in frequency with a period equal to the reciprocal of the time separation between the first two excitation pulses.

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