Analysis of an atomic $J=0$ to $J=1$ two-photon transition as a test of the spin-statistics connection for photons

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Using the q-deformed commutator formalism ("q mutators"), we have calculated the two-photon transition amplitude connecting a $J=0$ atomic ground state to a $J=1$ atomic excited state of the same parity. We find, in agreement with a semiclassical calculation, that this transition amplitude vanishes for two equal-frequency photons if the photons are traditional bosons with $q=1$. If $q<1$ (i.e., if the spin-statistics connection is violated for photons), then the amplitude is nonzero and is proportional to $(1-q)$. Thus such an experiment, originally proposed by Budker and DeMille, provides a sensitive test of the spin-statistics connection for photons within the q-mutator formalism.

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I. INTRODUCTION

The standard spin-statistics theorem [1–4] is predicated upon using only commutators or anticommutators for the algebra of creation and annihilation operators for quantized fields. It has been known since at least the early 1950s that more general operator algebras [5] enlarge the number of permutation symmetries of states beyond the usual symmetric and antisymmetric possibilities and thus call into question the usual spin-statistics connection. More recently, an operator algebra [6] has been introduced that allows a smooth interpolation between boson and fermion behavior. In this algebra, the usual commutators and anticommutators are replaced by a so-called q mutator. The creation $a^\dagger$ and annihilation a operators satisfy

$$a_k a_l^\dagger - q a_l^\dagger a_k = \delta_{kl}. \quad (1)$$

q is a real number lying between $+1$ and -1. For $q=+1$, we get the usual commutation relation leading to boson behavior (symmetric multiparticle states). For $q=-1$, we get the fermion anticommutation relations, which lead to antisymmetric states and the Pauli exclusion principle. Particles with $|q|\neq 1$ are called "quons." These q mutators have been proposed as a formalism to describe "small" violations of the usual spin-statistics connection (for which half-integer spin-quantum-number particles are fermions and integer spin particles are bosons). Later we shall describe more precisely what a small violation of the spin-statistics connection means in terms of observations. If $q=e^{i\theta}$ and with suitable restrictions on the product space for the particles, Eq. (1) describes the algebra of anyon fields [7].

At present, there is no formal prediction for a violation of the spin-statistics connection. However, the usual spin-statistics theorem [1,2,8] depends on several features of the quantum field theory: (a) Lorentz invariance, (b) locality, (c) four-dimensional space-time, (d) continuity of space-time, and (e) commutivity of space-time variables [9]. Both theoretical and experimental investigations of possible violations of all these properties are currently underway. In any case, given the importance of the spin-statistics connection in almost all areas of physics, it is crucial to give serious consideration to the possibility of its violation independent of any particular model.

Several recent experiments have set upper limits on the violation of the spin-statistics connection for electrons (normally regarded, of course, as fermions) [10,11], for ^{16}O nuclei [12–15] (normally regarded as bosons), and for Be atoms [16]. Tests of the boson character of photons are much less obvious. Man'ko and Tino have searched for an intensity-dependent frequency shift of the beat note between two stabilized lasers [17]. Their experiment was interpreted with a slightly different form of a so-called Q-oscillator model [18,19] in which the creation and annihilation operators are described by

$$a_k a_k^\dagger - Q s_k^\dagger a_k = Q^{-N}, \quad (2)$$

where N is the occupation number for that mode. In this model, there is no relationship among operators for different modes.

Several other tests for photons have been proposed. Fivel [20] has suggested that photons not in pure bosonic states would lead to a maximum possible laser intensity, but this prediction has been criticized [21]. In the so-called q-deformed Jaynes-Cummings model [22], the atomic-inversion oscillation revival times are slightly modified if the photons are quons. Rydberg atoms interacting with photons in a high-Q cavity would behave differently if the photon state were not purely bosonic [23,24]. Given current technology, however, none of these experiments leads to a high-precision test of the spin-statistics connection for photons. Moreover, there are substantial theoretical ambiguities [21] for those proposals that involve high-intensity fields and strong transition probabilities. We shall discuss some of those issues in Sec. V. Greenberg and Hilborn [25], using a simplified interaction Hamiltonian, showed that upper limits on the spin-statistics violation for one species of particles, electrons, for example, could be interpreted to give upper limits for another species with which the first interacts, for example, photons. In principle, such a connection means that

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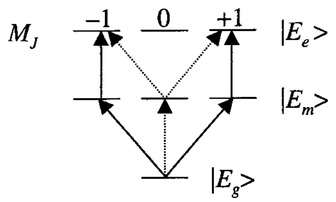

FIG. 1. The two alternative excitation routes (solid arrows versus dashed arrows) from a $J=0$ ground state to a $J=1$ final state of the same parity through a $J=1$ intermediate state for linearly polarized light. One mode is linearly polarized in the z direction, the other in the x direction. (Recall that a $J=1$, $M_J=0$ to $J=1$, $M_J=0$ transition is forbidden.)

the very low limits set on the violation probability for electrons [10] can be translated into a similar low limit for photons. However, the generality of that connection remains unexplored.

More recently, DeMille, Budker, Derr, and Deveney [26] have searched for a spin-statistics violating two-photon transition in atomic barium. A cw version of that experiment is in progress [27] and should lead to several orders of magnitude improvement in the sensitivity of the search for a possible spin-statistics connection violation for photons. This paper is devoted to analyzing that experiment using the quon formalism.

The DeMille *et al.* experiment [26] uses a two-photon atomic absorption transition from a $J=0$ atomic ground state to a $J=1$ atomic excited state (of the same parity) as a test of the spin-statistics connection for photons. The $J=0$ to $J=1$ transition is an unusual two-photon transition because it requires photons of orthogonal polarization (either linear or circular or any arbitrary orthogonal elliptical polarization), and the transition probability vanishes when the two photons have the same frequency [28,29] if photons are bosons in the usual way. The previous analysis of this experiment [26] was based on a semiclassical calculation in which the electric field is treated as a classical (c number) field. We shall see that a quantized-field analysis yields the same results. We then extend the analysis by treating the photons as quons.

The orthogonal-polarization requirement can easily be seen by considering the energy level diagram shown in Fig. 1. The two-photon transition must pass through an intermediate $J=1$ state (in the electric-dipole approximation). The usual electric-dipole selection rules then require photons of either orthogonal linear polarization or opposite circular polarization or any arbitrary orthogonal elliptical polarization. The absence of a two-equal-frequency-photon transition between $J=0$ and $J=1$ states of the same parity is analogous to the Landau-Yang theorem [30], which explains why a vector particle ($J=1$) cannot decay into two photons. Recently, Ignatiev and co-workers have used limits on the decay of the Z boson into two photons as a test for the boson character of photons [31]. It is difficult to interpret this limit simply in terms of the character of photon states because the Z particle itself does not couple to photons.

In order to show how a two-photon absorption experiment can be used as a test of the spin-statistics connection for photons, we have performed the quantized-field calculation for two-photon absorption assuming that the photon creation and annihilation operators are described by q mutators. The goal of the calculation is to see how the two-photon absorption amplitude depends on q. We find that for $q=+1$ (the usual boson condition), the transition amplitude vanishes for photons of the same frequency. We also find, and this is the crucial result reported in this paper, that for $q<1$, the two-photon transition is permitted for two equal-frequency (but orthogonal-polarization) photons with a relative transition amplitude proportional to $(1-q)$.

II. FORMULATION OF THE PROBLEM

To describe the proposed experiment, we need the probability for two-photon absorption from an atomic ground state $|g\rangle$ to an atomic excited state $|e\rangle$. One photon has frequency ω_u and the other has frequency ω_v. From general second-order perturbation theory, the amplitude for the transition from a state $|i\rangle$ (which includes the specification of both the atomic state and the photon-field state) to the final state $|f\rangle$ is given by

$$M_{if}=\sum_j \frac{\langle f|H_I|j\rangle\langle j|H_I|i\rangle}{E_i-E_j}, \tag{3}$$

where H_I is the interaction Hamiltonian and E_i and E_j are the total (atom and field) energies of the initial and intermediate states (labeled by j), respectively. [The overall energy conservation is enforced by the delta function $\delta(E_i-E_f)$ in the Fermi golden rule for the transition probability.] For the moment, we shall ignore energy-level widths and the bandwidth of the photon source. Those factors will be discussed in Sec. VI.

We will specify the initial state and final states in terms of products of atomic states and photon number states,

$$|i\rangle=|g\rangle|n_u,n_v\rangle,$$
$$|f\rangle=|e\rangle|n_u-1,n_v-1\rangle, \tag{4}$$

where the initial state has n_u photons in mode u and n_v photons in mode v. (The basic features of the results will be unchanged for other types of initial field states, say, for coherent states as we shall show explicitly in Sec. IV.)

The relevant intermediate states are of two types,

$$|j\rangle=|m\rangle|n_u-1,n_v\rangle,$$
$$|j\rangle=|m\rangle|n_u,n_v-1\rangle, \tag{5}$$

each with some intermediate atomic state $|m\rangle$ (with energy E_m) and one photon removed from one of the modes, but not the other. (Limiting the range of intermediate states in this way is the QED equivalent of the ‘‘rotating-wave approximation.’’ In the rotating-wave approximation, we keep only those parts of H_I that connect the appropriate initial, intermediate, and final states.) There will be two types of intermediate states: those with energies

$$E_j=E_m+\hbar(n_u-1)\omega_u+\hbar n_v\omega_v \tag{6}$$

and those with energies

$$E_j = E_m + \hbar n_u \omega_u + \hbar (n_v - 1)\omega_v. \tag{7}$$

Under these conditions, the energy denominators in Eq. (3) can be written as

$$E_i - E_j = E_g - E_m + \hbar \omega_{u \text{ or } v}. \tag{8}$$

We use the standard minimal-coupling interaction Hamiltonian, which in the Schrödinger picture is given by

$$H_I = \frac{e}{mc}\vec{A}\cdot\vec{p} + \frac{e^2}{2mc}A^2. \tag{9}$$

Here, $\vec{A}$ is the vector-potential operator for the electromagnetic field and $\vec{p}$ is the momentum operator for the atomic electrons. (As usual, we have assumed that the vector potential is constant across the atom. The momentum operator is actually a sum over all the momenta of the electrons.) For the experimental conditions we wish to describe, higher-order multipole transitions are completely negligible [26].

Greenberg [21] has raised the issue of the proper form of the Hamiltonian for a quon field (here, the photons) interacting with an ''external source.'' He argues that one must construct the Hamiltonian carefully to assure that all modes of the quon field contribute in equivalent ways to the transition amplitude, no matter where the mode appears in the state vector. This issue is discussed in more detail in Sec. V, where we show that for a weak transition of the type discussed here, the ''correction terms'' to the Hamiltonian are proportional to $1-q$, and hence can be ignored for our purposes.

For a quantized electromagnetic field, the vector-potential operator has the form

$$\vec{A}(\vec{r}) = \sum_{b,\beta} \sqrt{\frac{2\pi\hbar}{V\omega_b}}\,[\hat{\varepsilon}_{b\beta}a_{b\beta}e^{i\vec{k}_b\cdot\vec{r}} + \hat{\varepsilon}^*_{b\beta}a^\dagger_{b\beta}e^{-i\vec{k}_b\cdot\vec{r}}]. \tag{10}$$

V is the quantization volume. As usual, $\vec{k}_b$ is the wave vector and $\hat{\varepsilon}_{b\beta}$ is the unit polarization vector for photons in mode b. β labels the components of the polarization vector. $a_{b\beta}$ and $a^\dagger_{b\beta}$ are the annihilation and creation operators for the field mode labeled by b and β.

To put the transition amplitude into an effective operator form, which will be useful in seeing the angular momentum and frequency dependence of the amplitude, we define an operator H_{eff} so that

$$M_{if} = \langle f|H_{\text{eff}}|i\rangle. \tag{11}$$

Comparing Eqs. (11) and (3), we see that H_{eff} takes the form

$$H_{\text{eff}} = \sum_j \frac{H_I|j\rangle\langle j|H_I}{E_i - E_j}. \tag{12}$$

We will focus on the $\vec{A}\cdot\vec{p}$ part of the interaction Hamiltonian since the A^2 term does not contribute to the two-photon amplitude in the electric-dipole approximation. Ignoring constants of proportionality and assuming that the polarization vectors are real, as they are for linearly polarized light, we may write

$$H_I^{(1)} = \vec{A}\cdot\vec{p} = \sum_{b\beta} \hat{\varepsilon}_{b\beta}\cdot\vec{p}[a_{b\beta}e^{i\vec{k}_b\cdot\vec{r}} + a^\dagger_{b\beta}e^{-i\vec{k}_b\cdot\vec{r}}]. \tag{13}$$

With these assumptions, the effective interaction Hamiltonian can be written using the electric-dipole approximation (for which $e^{i\vec{k}\cdot\vec{r}}=1$) and the rotating-wave approximation (which in this case is equivalent to dropping photon-creation-operator terms) in the following form:

$$H_{\text{eff}} = \sum_{m\beta\gamma}\left\{\frac{\hat{\varepsilon}_{v\gamma}\cdot\vec{p}a_{v\gamma}|n_u-1,n_v\rangle|m\rangle\langle m|\langle n_u-1,n_v|\hat{\varepsilon}_{u\beta}\cdot\vec{p}a_{u\beta}}{E_g - E_m + \hbar\omega_u} + u\beta \Leftrightarrow v\gamma\right\}. \tag{14}$$

Note that Eq. (14) is symmetric in the labels $u\beta$ and $v\gamma$ as it must be for a quantum-mechanical operator describing identical particles.

We can consider the atomic and electromagnetic field parts of the transition amplitude separately. Let us focus on the case relevant for the actual experiment [26]: two orthogonal linearly polarized modes, one with polarization vector along z, the other along x.

The effective operator acting on the atomic states can be written with Cartesian components as

$$G_{xz} = \frac{(d_{fm})_x(d_{mg})_z}{E_g - E_m + \hbar\omega_v} + \frac{(d_{fm})_z(d_{mg})_x}{E_g - E_m + \hbar\omega_u}, \tag{15}$$

where, for the sake of simplicity, we have suppressed the sum over the intermediate states. Here, we have used d for the (dipole) operator for the atomic transitions. (The atomic dipole matrix elements are proportional to $(d_{fm})_x \propto \Sigma\langle f|er_{jx}|m\rangle$, where the sum is over all the charged particles.) The angular momentum dependence of the matrix elements of the terms in the numerators of Eq. (15) is easily evaluated using the Wigner-Eckart theorem [32]. (As an aside, we note that this is the point in the calculation where the spin of the photon enters. The ''statistics'' enters via the creation- and annihilation-operator algebra.) For a $J=0$ to $J=1$ transition (via a $J=1$ intermediate state), the product of matrix elements in one numerator of Eq. (15) is the negative of the other. In either the semiclassical field model or the standard quantized-field calculation (with commutators), as we shall see, the field part of the matrix elements is the same for both terms. Thus, when we add the amplitudes in Eq. (15), the two terms will cancel if and only if the two photons have the same frequency.

We can also see this cancellation from more general symmetry considerations [29]. The effective operator G_{xz} is a second-rank Cartesian tensor (formed from the two vectors $\vec{d}_{fm}$ and $\vec{d}_{mg}$) and can be decomposed into irreducible tensors of rank 0, 1, and 2. To connect a $J=0$ initial state to a $J=1$ final state, we need a rank-1 irreducible tensor. The rank-1 irreducible tensor is antisymmetric in the Cartesian-tensor labels [33]. (The rank-0 and rank-2 parts are symmetric in these labels.) So the rank-1 part can be isolated by writing

$$G^{(1)}=\tfrac{1}{2}(G_{xz}-g_{zx}), \tag{16}$$

which then yields

$$G^{(1)}\propto(\omega_u-\omega_v)\left\{\frac{(d_{fm})_x(d_{mg})_z}{(\omega_{gm}+\omega_u)(\omega_{gm}+\omega_v)}-\frac{(d_{fm})_z(d_{mg})_x}{(\omega_{gm}+\omega_u)(\omega_{gm}+\omega_v)}\right\}, \tag{17}$$

where $\omega_{gm}=(E_g-E_m)/\hbar$. $G^{(1)}$ obviously vanishes if the two photons have the same frequency.

The overall amplitude is zero for equal-frequency photons because of destructive interference between the two alternative paths: First, the atom absorbs a photon from mode u and then one from mode v or vice verse.

III. QUANTIZED-FIELD CALCULATION

Let us now turn our attention to the field part of the transition amplitude. Referring to Eq. (14), we see that the effective operator whose matrix elements we need to evaluate is

$$F=\frac{a_{u\beta}|m\rangle\langle m|a_{v\gamma}}{E_g-E_m+\hbar\omega_v}+\frac{a_{v\gamma}|m\rangle\langle m|a_{u\beta}}{E_g-E_m+\hbar\omega_u}, \tag{18}$$

where again we have suppressed the sum over intermediate states. In more detail, the matrix elements of the operators in the numerator of Eq. (18) between initial and final states will look like

$$\langle n_u-1,n_v-1|a_v|n_u-1,n_v\rangle\langle n_u-1,n_v|a_u|n_u,n_v\rangle. \tag{19}$$

(We have temporarily dropped the polarization-component indices for the sake of typographical simplicity.) The intermediate photon state can be replaced with a sum over a complete set of photon-number states (since all the other matrix elements will be zero). The sum over the complete set is equivalent to the identity; so, our task is reduced to evaluating the following matrix elements:

$$\langle n_u-1,n_v-1|a_ua_v|n_u,n_v\rangle \tag{20}$$

and the matrix elements with a_u and a_v interchanged.

The q-deformed initial (number) state is constructed by applying quon-creation operators to the photon vacuum [34],

$$|n_u,n_v\rangle=\frac{(a_u^\dagger)^{n_u}(a_v^\dagger)^{n_v}|0\rangle}{\sqrt{[n_u]_q![n_v]_q!}}, \tag{21}$$

where $[n]_q$ (a so-called q-deformed number) is defined to be

$$[n]_q=\frac{1-q^n}{1-q}, \tag{22}$$

and the q factorial is $[n]_q!=[n]_q[n-1]_q\cdots 1$. Equation (21) is just the standard two-mode number state with a normalization that takes into account the normalization of the quon states. The normalization for simple cases, such as a two-quon state, can be easily worked out by using Eq. (1) to evaluate the amplitude $\langle 0|a_va_ua_u^\dagger a_v^\dagger|0\rangle$. The q-deformed number $[n]_q$ reduces to the ordinary number n in the limit $q\to 0$.

For ordinary QED, the calculation of the matrix elements is a straightforward exercise found in many texts. (See Ref. [33], pp. 536–547; Ref. [35], pp. 335–347; or Ref. [36], for example.) However, for q mutators we must proceed cautiously because in the q-mutator formalism, we do not have algebraic relations that allow us to interchange two creation operators or two annihilation operators [37]. We have only Eq. (1), which allows us to interchange a creation operator and an annihilation operator. That is all, however, that we need to evaluate the desired matrix elements. We simply start with an annihilation operator that has a creation operator to its right and move that annihilation operator to the right using Eq. (1) until the annihilation operator hits the vacuum state, which then gives us 0.

For the simple initial state with one photon in each mode, the two amplitudes to evaluate are

$$\langle 0,0,|a_ua_v|1,1\rangle=\langle 0,0|a_ua_va_u^\dagger a_v^\dagger|0,0\rangle,$$

$$\langle 0,0|a_va_u|1,1\rangle=\langle 0,0|av_va_ua_u^\dagger a_v^\dagger|0,0\rangle. \tag{23}$$

Each interchange of creation and annihilation operators for different modes introduces a δ function and a factor of q. Since we are concerned with two different polarizations (even if the frequencies are the same), the delta functions will always be zero. Interchanging creation and annihilation operators for the same mode then forms a polynomial in q. These polynomials are similar to those used in constructing the number and energy operators for quons [38,39]. Greenberg [40] has given a simple graphical rule for determining the appropriate power of q for each term in the polynomial.

For more general initial number states, the necessary matrix elements can be expressed as

$$\langle f|a_va_u|i\rangle=g_{n_un_v}(q),$$

$$\langle f|a_ua_v|i\rangle=qg_{n_un_v}(q), \tag{24}$$

where the function g is a polynomial in q that depends on the number of photons in each of the modes in the initial state. The final states have one less photon per mode. (The results differ by the factor q because of the choice of a specific ordering of the creation operators acting on the vacuum to produce the initial state. If we change that ordering, the factor of q may move from one of Eq. (24) to the other. Only the phase difference is important.) Table I lists several specific cases for this polynomial.

We can see some general features by examining Table I. Except for the (1,1) initial state, $g(q)$ is zero when $q=-1$. This result should be expected because $q=-1$ corresponds to anticommutators for the operators and hence leads to the exclusion principle: all matrix elements that involve states with more than one fermion per mode must equal zero. For

TABLE I. The polynomial $g_{n_u n_v}(q)$ for several photon number states.

Initial state n_u, n_v	$g_{n_u n_v}(q)$
1,1	1
2, 1	$q+q^2$
1, 2	$1+q$
2,2	$q+2q^2+q^3$
3,1	$q^2+2q^3+2q^4+q^5$
1,3	$1+2q+2q^2+q^3$
3,2	$q^2+3q^3+4q^4+3q^5+q^6$
2,3	$1+3q+4q^2+3q^3+q^4$

the (1,1) case the two matrix elements have the opposite sign when $q=-1$ as required for amplitudes that differ by the interchange of two identical fermions. On the other hand, for $q=+1$, we should expect the amplitude to be proportional to $\sqrt{n_u n_v}$ in order to have the probability be proportional to the product of the numbers of photons in each of the two modes. That result combined with the normalization in Eq. (21) requires $g(q=+1)=\sqrt{n_u(n_u!)(n_u-1)!n_v(n_v!)(n_v-1)!}$, which agrees with the listings in Table I. [The $(n-1)!$ terms come from the normalization of the final state.]

The results given in Table I are consistent with the following general result:

$$g_{n_u n_v}=q^{n_u-1}\sqrt{[n_u]_q[n_u]_q![n_u-1]_q![n_v]_q[n_v]_q![n_v-1]_q!}, \tag{25}$$

where the q-deformed numbers are defined in Eq. (22). We have not yet been able to provide a general proof of Eq. (25).

Even without knowing the general form of $g(q)$, we can draw the following conclusions: The transition amplitude can be written in the form

$$M_{if}=\frac{D_{\gamma\beta}g(q)}{E_g-E_m+\hbar\omega_u}+q\frac{D_{\beta\gamma}g(q)}{E_g-E_m+\hbar\omega_v}, \tag{26}$$

where D represents products of the atomic-dipole-matrix elements discussed previously,

$$D_{\gamma\beta}=(d_{fm})_\gamma(d_{mg})_\beta. \tag{27}$$

(Recall that β and γ represent general polarization components.) If we consider the case when the two modes have the same frequency (but not the same polarization), the amplitude takes the form

$$M_{if}=\frac{g(q)}{E_g-E_m+\hbar\omega}[D_{\gamma\beta}+qD_{\beta\gamma}]=\frac{g(q)}{E_g-E_m+\hbar\omega}\left\{\left(\frac{1+q}{2}\right)\times(D_{\gamma\beta}+D_{\beta\gamma})+\left(\frac{1-q}{2}\right)(D_{\gamma\beta}-D_{\beta\gamma})\right\}. \tag{28}$$

As we saw previously, for a $J=0$ to $J=1$ two-photon transition, the sum of the two D terms in Eq. (28) vanishes (in the electric-dipole approximation). For ordinary boson behavior $(q=1)$, the second term vanishes also. However, for $q<1$, the second term can be nonzero, thus signaling a violation of the spin-statistics connection for photons with an amplitude proportional to $(1-q)$. Equation (28) is the main result of this paper.

Equation (28) provides an interpretation of what a ‘‘small violation’’ of the spin-statistics connection means for a two-photon transition. The first term inside the braces is symmetric under the interchange of the two polarization labels—a boson-type amplitude. The second term is antisymmetric under the interchange of the two labels—a fermion-type amplitude. Hence, we say that the two photons behave like bosons with an amplitude proportional to $1+q$ and like fermions with an amplitude proportional to $1-q$. If we were dealing with more than two photons, there would be additional amplitudes for photon behavior characterized by higher-dimensional representations of the permutation group [40]. The weights for the different permutation-group representations are polynomials in q similar to the function $g(q)$ used in this paper.

In a previous paper [41], the two-photon absorption experiment was treated in terms of density matrices for the two-photon states. For two quons, the two-particle density operator can be written as

$$\rho^{(2)}=\frac{1+q}{2}\rho_s^{(2)}+\frac{1-q}{2}\rho_a^{(2)}, \tag{29}$$

where the symmetric and antisymmetric parts of the density operator are given in terms of the symmetric and antisymmetric two-photon states by

$$\rho_s^{(2)}=|\phi_s\rangle\langle\phi_s|, \quad \rho_a^{(2)}=|\phi_a\rangle\langle\phi_a|. \tag{30}$$

The symmetric and antisymmetric states are written in terms of the creation operators as

$$|\phi_{s,a}\rangle=\frac{1}{\sqrt{2(1\pm q)}}(a_1^\dagger a_2^\dagger \pm a_2^\dagger a_1^\dagger)|0\rangle. \tag{31}$$

The term $(1\pm q)$ in the denominator of Eq. (31) is necessary for normalization. Choosing the opposite order of the creation operators in Eq. (31) results in exactly the same density operator; so either order can be used for calculations.

It is tempting to use the density-operator form in Eq. (29) to argue that the spin-statistics-connection-violating *probability* should go as $(1-q)/2$ since that is the weighting factor for the antisymmetric part of the density operator. Using Eq. (31) in Eq. (30), however, we see that the weighting factor $(1+q)/2$ for the symmetric part of the density matrix is canceled by the normalization factor for the symmetric state with the analogous cancellation for the antisymmetric part. The crucial point is that we need to take into account the nonstandard normalization factors for the quon states. The net result is that the transition *amplitude* is proportional to $1-q$, which as we have seen above, comes from evaluating the matrix elements involving the photon-annihilation operators. If the probability were proportional to $(1-q)$, then the amplitude would be proportional to $\sqrt{1-q}$, and we would (apparently) miss the crucial overall minus sign in Eq. (24) in the fermion limit ($q \rightarrow -1$).

To use the density-operator approach for calculations involving states with more than two photons, it would be necessary to find the weighting factors for all the density operators associated with each of the possible irreducible representations of the permutation group. In practical terms, it is more straightforward to use the Fock-state approach (or its generalizations) employing the creation and annihilation operators directly.

IV. COHERENT STATE FOR THE PHOTON (QUON) FIELD

In this section we replace the initial photon number state with a q-deformed coherent state. Since coherent states are better models of the output of a typical laser, the results of this section should be more directly applicable to the DeMille-Budker experiment. As we shall show, the results with a coherent photon state can be cast into a form analogous to those obtained with the photon number states. In particular, Eqs. (26) and (28) will still apply with suitable changes in notation.

First let us review the formalism associated with q-deformed coherent states for a single mode. In analogy to the usual coherent states [42,43], we may define the q-deformed coherent state [44,45] as a superposition of photon number states,

$$|\alpha\rangle_q = \frac{1}{\sqrt{e_q(|\alpha|^2)}} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{[n]_q!}} |n\rangle, \tag{32}$$

where the q-exponential function is given by

$$e_q(|\alpha|^2) = \sum_{n=0}^{\infty} \frac{|\alpha|^{2n}}{[n]_q!}. \tag{33}$$

As usual, the coherent state is constructed to be an eigenstate of the annihilation operator with eigenvalue α,

$$a|\alpha\rangle_q = \alpha|\alpha\rangle_q. \tag{34}$$

Since a is not a Hermitian operator, α will be complex. The states are normalized with ${}_q\langle\alpha|\alpha\rangle_q \equiv 1$. The parameter α is related to $\overline{n}$, the mean photon number for the state,

$$\overline{n} = |\alpha|^2 \frac{e_q'(|\alpha|^2)}{e_q(|\alpha|^2)}, \tag{35}$$

where the prime indicates differentiation with respect to $|\alpha|^2$. Note that two coherent states with different parameters are *not* orthogonal,

$$|\langle\beta|\alpha\rangle|^2 = e^{(-|\alpha-\beta|^2)}, \tag{36}$$

but the coherent states do form a complete set.

To describe the two-photon absorption experiment, we need to evaluate the following matrix elements: $\langle f|a_u a_v|i\rangle$ and $\langle f|a_v a_u|i\rangle$, where the initial photon state will be taken to be a coherent state for the two modes,

$$|i\rangle = |\alpha_u, \alpha_v\rangle_q. \tag{37}$$

The two-mode coherent state takes the form [42]

$$|\alpha_u, \alpha_v\rangle_q = \frac{1}{\sqrt{e^q(|\alpha_u|^2)}\sqrt{e_q(|\alpha_v|^2)}} \times \sum_{n_u, n_v=0}^{\infty} \frac{\alpha_u^{n_u}\alpha_v^{n_v}}{\sqrt{[n_u]_q![n_v]_q!}} |n_u, n_v\rangle. \tag{38}$$

We need not specify the details of the final state except to note that it generally involves a sum over photon number states as well. If we think of the operators as acting on each of the number states in Eq. (38), then only one term in the final-state sum will survive for each term in the initial sum over number states. We can then write

$$\langle f|a_v a_u|i\rangle = \sum_{n_u n_v} \langle n_u - 1, n_v - 1|a_v a_u|n_u, n_v\rangle B_{n_u n_v}(\alpha_u, \alpha_v), \tag{39}$$

where all of the numerical coefficients have been subsumed into the B term. The matrix element in Eq. (39) is the same as that given in Eq. (19). Hence we may write

$$\begin{aligned}\langle f|a_v a_u|\alpha_u, \alpha_v\rangle &= \sum_{n_u, n_v} g_{n_u n_v}(q) B_{n_u n_v}(\alpha_u, \alpha_v) \\ &\equiv K(q, \alpha_u, \alpha_v), \\ \langle f|a_u a_v|\alpha_u, \alpha_v\rangle &= qK(q, \alpha_u, \alpha_v).\end{aligned} \tag{40}$$

Comparing Eqs. (40) and (24) tells us that the overall transition-matrix element for coherent states can still be written in the form of Eqs. (26) and (28).

V. THE FORM OF THE INTERACTION HAMILTONIAN AND RELATED ISSUES

Greenberg [21,37] has noted that in constructing the Hamiltonian describing the interaction between quons and their "sources," the source terms must be represented by a generalization of Grassmann numbers in order to satisfy the conditions of (a) additivity of energies for widely separated systems and (b) equal treatment of the modes. The latter condition means that each of the mode labels for the quon fields must enter the matrix elements in the same way.

To put the quon requirements into context, we first review the situation for ordinary fermions. If the quons are ordinary fermions, then the sources are represented by the usual anticommuting Grassmann variables. The Hamiltonian representing the interaction between the fermions and their sources is of the form

$$\hat{H}_{\text{ext}} = \sum_j c_j a_j^\dagger + c_j^* a_j, \tag{41}$$

where the creation and annihilation operators for the fermions and the Grassmann variables c_j and c_j^* satisfy the following anticommutation relations:

$$c_i c_j^* + c_j^* c_i = 0,$$
$$c_i a_j^\dagger + a_j^\dagger c_i = 0,$$
$$c_i^* a_j + a_j c_i^* = 0,$$
$$a_j a_k + a_k a_j = 0. \tag{42}$$

Greenberg [21,46] has pointed out that if the creation and annihilation operators satisfy the q-mutator relation

$$a_m a_n^\dagger - q a_n^\dagger a_m = \delta_{mn}, \tag{43}$$

then the variables c and c^* representing the source, must satisfy

$$c_m c_n^* - q c_n^* c_m = 0,$$
$$c_m a_n^\dagger - q a_n^\dagger c_m = 0, \tag{44}$$
$$a_m c_n^* - q c_n^* a_m = 0.$$

Note that we do not have any specified relationship between a_m and a_n nor between c_m and a_n (or between their conjugates). For example, if we try to impose

$$a_j a_k - q a_k a_j = 0, \tag{45}$$

then by interchanging the dummy labels j and k in Eq. (45) and substituting back for $a_k a_j$, we conclude that we must have $q^2=1$. Thus, only in the pure boson case ($q=1$) or the pure fermion case ($q=-1$) do we have a simple algebraic relationship between annihilation (or creation) operators. As mentioned previously, we do not need those relations to compute transition amplitudes or expectations values.

To satisfy the two conditions noted above, the Hamiltonian describing the coupling of the quon fields to the external sources must satisfy

$$[\hat{H}_{\text{ext}}, a_k^\dagger]_- = c_k^* \tag{46}$$

and must be written as an infinite-order polynomial in the creation and annihilation operators and the source terms. Greenberg [21] has given the first few terms of the polynomial,

$$\hat{H}_{\text{ext}} = \sum_k c_k^* a_k + a_k^\dagger c_k + (1-q^2)^{-1} \sum_{k,t} (a_t^\dagger c_k^* - q c_k^* a_t^\dagger) \times (a_k a_t - q a_t a_k) + (1-q^2)^{-1} \sum_{k,t} (a_t^\dagger a_k^\dagger - q a_k^\dagger a_t^\dagger) \times (c_k a_t - q a_t a_k) + \cdots . \tag{47}$$

For the case of q photons, the source terms represent the charged-particle (electron and nucleus) currents. In the usual nonrelativistic atomic-physics electric-dipole approximation, these currents are proportional to the momentum operators for the electrons.

The prescription outlined above seems to be ignored by almost all treatments of quons in the quantum-optics literature. For example, in the q-deformed Jaynes-Cummings model [22], the radiation field is described by creation and annihilation operators that satisfy a q-mutation relation. However, the terms describing the atom to which the q photons couple are described by the standard pseudospin operators (Pauli operators) with the usual commutation relations

$$[\sigma_+, \sigma_-] = 2\sigma_3 \quad \text{and} \quad [\sigma_3, \sigma_\pm] = \pm \sigma_\pm . \tag{48}$$

This procedure seems to be valid because there is only one q-photon mode (so treating many modes identically is not relevant), and there is no spatial variable for the atom's location (so worrying about separated systems is not relevant).

For the case of the two-photon absorption experiment of interest here, we argue that the additional terms in the Hamiltonian produce a contribution to the transition amplitude that is smaller by a factor of $1-q \equiv \varepsilon$ compared to the contribution of the "ordinary" part of the Hamiltonian. Since we expect ε to be small for photons (and other ordinary boson particles, as well), we can safely ignore the corrections to the Hamiltonian for most kinds of experiments, where we are dealing with already small transition probabilities. Greenberg [21] has pointed out that corrections are necessary for experiments in which the ordinary transition probability may be saturated, for example, with high-intensity laser beams.

To see how the additional Hamiltonian terms contribute to the transition amplitude for the two-photon experiment, let us examine the correction terms in Eq. (47). For an absorption experiment, we focus our attention on the terms containing c_k^*, which corresponds to an atomic raising operator,

$$\hat{H}_{\text{ext}} = \sum_k c_k^* a_k + (1-q^2)^{-1} \sum_{k,t} (a_t^\dagger c_k^* - q c_k^* a_t^\dagger) \times (a_k a_t - q a_t a_k) \tag{49}$$

We are interested in the case of $q \approx 1$; so let us use $\varepsilon = 1 - q$ in Eq. (49). We then have

$$\hat{H}_{\text{ext}} = \sum_k c_k^* a_k + \frac{1}{2\varepsilon} \sum_{k,t} (a_t^\dagger c_k^* - c_k^* a_t^\dagger + \varepsilon c_k^* a_t^\dagger) \times (a_k a_t - a_t a_k + \varepsilon a_t a_k) \tag{50}$$

Multiplying out the terms inside the sum in the previous equation yields

$$\hat{H}_{\text{ext}} = \sum_k c_k^* a_k + \frac{1}{2\varepsilon} \sum_{k,t} \{ a_t^\dagger c_k^* a_k a_t - a_t^\dagger c_k^* a_t a_k + \varepsilon a_t^\dagger c_k^* a_t a_k - c_k^* a_t^\dagger a_k a_t + c_k^* a_t^\dagger a_t a_k - \varepsilon c_k^* a_t^\dagger a_t a_k + \varepsilon c_k^* a_t^\dagger a_k a_t - \varepsilon c_k^* a_t^\dagger a_t a_k + \varepsilon^2 c_k^* a_t^\dagger a_t a_k \}. \tag{51}$$

We now argue that all of the terms inside the curly brackets of Eq. (51) vanish as ε^2 and hence the overall correction term is proportional to ε. To see how this works, consider the first and second terms inside the curly brackets. They can be written as

$$a_t^\dagger c_k^*(a_k a_t - a_t a_k). \tag{52}$$

The term in parentheses in Eq. (52) vanishes when $q=1$ according to the usual boson commutation relations. Thus, we must be able to write that factor as

$$(a_k a_t - a_t a_k) = \varepsilon g(a_k, a_t) + \cdots, \tag{53}$$

where g represents some unknown (but irrelevant) function of the operators and the ellipsis indicates terms proportional to higher powers of ε. The fourth and fifth terms can be combined in a similar fashion. Then those two results can be joined to give

$$(a_t^\dagger c_k^* - c_k^* a_t^\dagger)\varepsilon g(a_k, a_t). \tag{54}$$

We now apply a similar argument to the parenthetical factor in Eq. (54): it must vanish when $q=1$, and hence we can write

$$(a_t^\dagger c_k^* - c_k^* a_t^\dagger)\varepsilon g(a_k, a_t) = \varepsilon^2 f(a_t^\dagger, c_k^*) g(a_k, a_t) + \cdots. \tag{55}$$

Identical arguments show that the third, sixth, seventh, and eighth terms can be combined to give a term proportional to ε^2. We thus see that the entire curly-bracket expression in Eq. (51) is proportional to ε^2. We conclude that the first correction term to the Hamiltonian is proportional to $1-q$ and hence can be neglected compared to the ''ordinary'' Hamiltonian in computing the forbidden transition amplitude, which itself is proportional to $1-q$.

To see the effects of the ''correction terms'' in the Hamiltonian, let us assume that the reduction carried out in Eq. (55) leads to the following piece to be added to the interaction Hamiltonian for the two-photon absorption calculation:

$$\hat{H}' = \varepsilon \sum_{k=1}^{2} C_k a_k, \tag{56}$$

where C_k is an operator function involving c_k^*, a_t, $a_t^\dagger$, and perhaps a_k itself (with a sum over t). Note that we have not yet proved that the operator must be in this form, but given the previous arguments, this seems to be a reasonable form. There may be other parts that involve products of two (or more) annihilation operators (without any corresponding creation operators). We assume that we can neglect such terms at this level of approximation.

The transition amplitude without the correction term can be written in the form

$$M_{if} = \sum_m \frac{D_{\beta\gamma}\langle f|a_u a_v|i\rangle}{E_g - E_m + \hbar\omega_v} + \frac{D_{\gamma\beta}\langle f|a_v a_u|i\rangle}{E_g - E_m + \hbar\omega_u}, \tag{57}$$

where, again, D contains all of the atomic-dipole-matrix-element information. With the correction term in the Hamiltonian, we make the replacement

$$a_u \rightarrow (1 + \varepsilon C_u) a_u \tag{58}$$

with an analogous expression for a_v. When we compute the matrix elements in Eq. (57), we see that the numerator of the first term is related to the numerator of the second term simply by interchanging the mode labels u and v. Equation (24) shows that interchanging the mode labels produces a factor of q difference between the two sets of matrix elements for the annihilation operators. Assuming that a similar condition holds here, we may write

$$\langle f|a_u C_v a_v|i\rangle = qR(q)\langle f|C_v a_v a_u|i\rangle, \tag{59}$$

where $R(q)$ allows for the possibility of other factors of q when the operators C_v are taken into account. (Similar powers of q appear in expressions for the number and energy operators for quons [37–39].) The crucial point is that the two sets of matrix elements are still related by a factor of q. That means that the construction leading to Eq. (26) still obtains, and in the case of equal-frequency modes we once again arrive at Eq. (28), the equation that shows that the two-photon absorption experiment is indeed a test of the quon form of a possible spin-statistics violation.

In a recent paper [47], Chow and Greenberg have argued that in a quon theory with antiparticles (so that crossing relations can be employed) and with trilinear interactions among the quons, requiring that the interaction be an effective Bose operator (that is, it commutes with all creation and annihilation operators) leads to the conclusion that the parameter q must satisfy $q=\pm 1$. (Their argument, however, does not lead to a specific spin-statistics connection.) The Chow-Greenberg argument is *sufficient* to establish that quons satisfy energy additivity for well-separated systems. However, we believe that it is not a *necessary* condition. So the possibility of $q \neq \pm 1$ is still open. Moreover, it is not obvious that their argument applies to the situation described in this paper in which only the photons are assumed to be quons.

VI. EXPERIMENTAL CONSIDERATIONS AND CONCLUSIONS

In the two-photon absorption experiment, the transition is observed by detecting fluorescence emitted in an allowed transition from the excited state to some lower state or by ionizing the excited atoms and detecting the resulting charged particles. In principle, this test might be very sensitive because one can look for a signal against a zero background while appropriate laser sources can provide a large flux of photons. DeMille *et al.* [26] have considered a number of experimental factors that might mimic the q-mutator effect. These factors include the effects of atomic linewidths, laser bandwidth, higher multipole transitions, and so on. None of these appear to make a significant contribution to the experimental signal. They conclude that the experiment is capable of achieving a sensitivity of $<10^{-14}$ for a 1 sec averaging time. Thus, the Budker-DeMille experiment would provide a test for the spin-statistics connection for photons many orders of magnitude more sensitive than other recent proposals [23,24]. In a pulsed-laser version of the experiment, DeMille *et al.* [26] have achieved an upper limit of 1.2×10^{-7} for the probability of the forbidden degenerate two-photon transition compared to the allowed nondegenerate two-photon transition in atomic barium. In terms of the q-mutator formalism, this result can be expressed as $[(1-q)/2]^2<1.2\times10^{-7}$. They also showed experimentally that the two-photon transition did occur with small probability even with light beams of nominally the same frequency, if one takes into account the finite laser bandwidth and atomic linewidth. In other words, the atom can still interact with two different-frequency photons because of the finite laser linewidth. Observing such a signal is an important test of the experiment's sensitivity.

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