

# How we know that photons are bosons: experimental tests of spin-statistics for photons

D. DeMille,<sup>1</sup> D. Budker,<sup>2</sup> N. Derr,<sup>3\*</sup> and E. Deveney<sup>3†</sup>

<sup>1</sup> *Physics Department, Yale University, New Haven, CT 06520 USA; david.demille@yale.edu*

<sup>2</sup> *Physics Department, Univ. of California, Berkeley, CA 94720 USA; budker@socrates.berkeley.edu*

<sup>3</sup> *Physics Department, Amherst College, Amherst, MA 01002 USA*

\* *Present Address: MIT Lincoln Laboratory, Lincoln, MA 02420 USA; ndderr@ll.mit.edu*

† *Present Address: Bridgewater State College, Bridgewater, MA 02325 USA; edeveney@bridgew.edu*

**Abstract.** We discuss theoretical models and experimental data that shed light on possible small violations of the spin-statistics relation for photons. Particular emphasis is given to our recent experimental search for non-symmetric photon states, using atomic two-photon transitions.

## I. INTRODUCTION

Photons are interesting particles with which to search for small violations of the usual spin-statistics relation, for a number of reasons. The photon is of course the only *fundamental* boson that can be readily produced in states with many identical particles present. In addition, it has proven possible to formulate some explicit theoretical models for a spin-statistics violation; such models have been absent for other particles such as electrons or nuclei.\*

With this in mind, we review here the experimental data that place limits on possible small violations of the spin-statistics relation for photons. In the absence of any entirely suitable theory for guidance, it seems prudent to consider all possible means of detecting such a violation. We consider three such experimental signatures: deviations from the Planck blackbody spectrum; non-standard behavior at large single-mode occupation number; and the existence of non-permutation-symmetric states. We give particular emphasis to the latter, culminating with a description of our recent experiment to search for non-symmetric states using two-photon transitions in atoms. Before moving to the experiments, however, we begin with a brief discussion of the available models for interpreting each of these types of data.

### Theoretical Models for Spin-Statistics Violation for Photons

We know of two theoretical models that have been used to describe possible spin-statistics violations for photons; both are based on deformed commutation relations. The first of these is the "quon algebra,"<sup>1</sup> where

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\* Only recently has a description been formulated, using the quon algebra, of experiments searching for permutation symmetry-forbidden states in molecules and in atomic He. See the contribution by R. Hilborn in these proceedings.

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

with  $q$  real and  $|q| \leq 1$ . It has been shown that photons with deformation parameter  $q < 1$  can exist in non-permutation-symmetric states,<sup>2</sup> and that certain microwave cavity states are altered from their usual form.<sup>3</sup> Limits on the output power of lasers (i.e., a soft upper bound on the single-mode occupation number  $N$ ) were suggested to exist,<sup>4</sup> but the argument leading to this conclusion was shown to be invalid.<sup>5</sup> No further work on possible non-standard behavior of quons at high  $N$  has been reported, and the situation with regards to such behavior is thus unclear. The status of blackbody radiation in the quon theory is similarly unclear. Though the relevant partition function has been given,<sup>6</sup> we have not found any explicit derivation of the analogue of the Planck spectrum for quons. Even if this distribution is derived, it may be difficult to interpret: it is known that the statistical mechanics of quons suffers from the Gibbs paradox (for any value of  $|q| < 1$ ), and thus may not be tenable at all.<sup>7</sup>

The behavior of photons which obey the "Q-oscillator" algebra,<sup>8</sup> where

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

has also been considered. (We use capital  $Q$  throughout, to distinguish from the quon algebra.) Several versions of a deformed Planck distribution have been derived,<sup>9</sup> and a model has been developed in which this  $Q$ -deformation leads to a blue shift of the mode frequency at high  $N$ .<sup>10</sup> In this algebra, there is no specified relation between different modes; thus, this deformation does not necessarily lead to the existence of non-symmetric states.

## II. PREVIOUS EXPERIMENTAL LIMITS

### Deviations from the Planck Blackbody Spectrum

It seems reasonable that any deviation from the usual spin-statistics relation for photons could lead to a deformation of the usual Planck blackbody spectrum, since the derivation of this spectrum explicitly invokes Bose-Einstein (BE) statistics. To our knowledge, no analysis of blackbody spectral data has been performed to search for small, systematic deviations from the Planck formula. However, it appears that the best absolute measurements of blackbody spectra have uncertainties of no better than about 1%.<sup>11</sup>

This statement may appear to be contradicted by the spectacular data on the cosmic microwave background (CMB), which are stated to have deviations of  $< 50$  ppm from the usual Planck distribution.<sup>12</sup> However, this claim is based on comparison of the CMB with a reference blackbody that is assumed to follow the Planck distribution, rather than on absolute measurements. Since the reference is held at almost exactly the temperature of the CMB, such a comparison is quite insensitive to a general deformation of the Planck spectrum. We conclude that deviations from the Planck distribution of  $\lesssim 1\%$  cannot be ruled out by any existing blackbody data.

## Non-Standard Behavior At Large Occupation Number $N$

One obvious distinction between bosons and fermions is that bosons can have unlimited occupation number in a single mode, while fermions can have at most one particle in any given state. Thus, it seems sensible that a breakdown of the spin-statistics relation could give some upper limit on the single-mode occupation number  $N$ , or at least some unusual behavior in the limit  $N \rightarrow \infty$ . With this in mind, it is interesting to point out what is known about electromagnetic fields with large  $N$ .

It is commonly understood that the existence of powerful lasers shows that large occupation numbers are possible. Indeed, from a pulsed laser with wavelength  $\lambda \sim 1 \mu\text{m}$ ,  $N \sim 10^{19}$  has been demonstrated.<sup>13</sup> Even more striking results come from high-power microwave resonance cavities, of the type used for particle accelerators; here, for photons with  $\lambda \sim 10 \text{ cm}$ ,  $N \gtrsim 10^{23}$  has been achieved.<sup>14</sup> These bounds improve even further at lower frequencies. Indeed, the existence of *any* finite energy stored in a *static* electromagnetic field would indicate  $N \rightarrow \infty$ , since the energy per photon  $h\nu \rightarrow 0$  in this case. It is thus difficult to imagine any way to reconcile the very existence of static fields with an upper bound on  $N$ . In reality, of course, no field can ever be truly static (because of power outages, battery failure, etc.). Nevertheless, the laboratory limits from "nearly-static" fields are very strong: we estimate that  $N \gtrsim 10^{40}$  has been achieved without any special effort.<sup>†</sup>

In the Q-oscillator model, a deviation from BE statistics is manifested by a blue-shift of any mode frequency  $\omega$ , at high values of  $N$ . The fractional shift is  $\delta\omega/\omega \approx \lambda^2 N^2/2$ , where  $\lambda = \ln Q$  and  $|\lambda| \approx 1-Q \ll 1$ .<sup>10</sup> Using this principle, a strict bound has been placed on  $\lambda$ , by Man'ko and Tino.<sup>15</sup> Their experiment compared the frequency difference between beams of light from two lasers, as a function of the relative power in the beams. No frequency difference could be observed at the level  $\delta\omega/\omega \approx 10^{-14}$ , despite changing the single-mode occupation number  $N \sim 10^{10}$  by nearly its full value. This null result implies that in the Q-oscillator model,  $1-Q \lesssim 10^{-17}$ .

### Existence of non-permutation-symmetric states

According to the usual spin-statistics relation, any multi-photon wavefunction must be symmetric under interchange of particle labels. Thus, the existence of any non-symmetric state would indicate a violation of BE statistics for photons. How could such states be distinguished from the usual symmetric states? Answers to the analogous question for particles such as electrons and nuclei are familiar. Every physicist is taught that the exchange-symmetric  $(1s)^2 \ ^3S_1$  state of the two electrons in atomic helium does not exist. It is also common knowledge that half the rotational states of the  $^{16}\text{O}_2$  molecule are never observed, because they are antisymmetric under exchange of the  $^{16}\text{O}$  nuclei. By contrast, it is far less familiar that, because photons obey BE statistics, there are "missing" states among the possible wavefunctions of two photons. The result has nevertheless been known for some time, and is referred to in particle physics as the Landau-Yang (LY) theorem. Here we re-derive this theorem and discuss how it can be used to search for non-symmetric photon states.

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<sup>†</sup> This estimate is based on a large NMR magnet with magnetic field  $B \sim 5 \text{ T}$  in volume  $V \sim (10 \text{ cm})^3$ , for a time  $T \sim 1 \text{ week}$ .

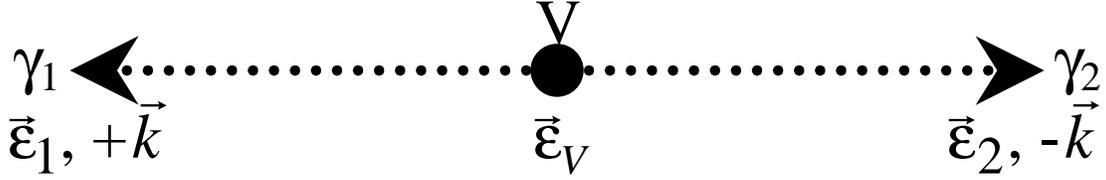


FIGURE 1. Kinematics of the decay  $V \rightarrow \gamma\gamma$ , in the rest frame of  $V$ .

The LY theorem states that it is impossible for a particle with angular momentum  $J = 1$  to decay into two photons.<sup>16</sup> A simple proof of this theorem shows that this result is simply a manifestation of the fact that the two-photon state is symmetric under interchange.<sup>17</sup> The kinematics of the hypothetical two-photon decay of a  $J=1$  (vector) particle,  $V$ , are shown in Fig. 1. It must be possible to write the decay amplitude  $\mathcal{A}$  for this process, in terms of the only quantities available: the polarizations of the two photons,  $\vec{\epsilon}_1$  and  $\vec{\epsilon}_2$ ; the polarization of  $V$ ,  $\vec{\epsilon}_V$ ; and the outgoing photon momentum,  $\vec{k} = \vec{k}_1 = -\vec{k}_2$ . Moreover, in quantum field theory the creation and annihilation operators for  $V$  and  $\gamma$  carry with them the corresponding polarization vectors. Thus, in order to destroy one  $V$  and create two  $\gamma$ 's, each of the polarization vectors must appear exactly once in the expression for  $\mathcal{A}$ . (The momentum vector  $\vec{k}$  can appear an arbitrary number of times.) With the requirement of gauge invariance on the photons (i.e.,  $\vec{\epsilon}_{1,2} \cdot \vec{k} = 0$ ), only three forms of  $\mathcal{A}$  are possible:

$$\mathcal{A}_a = (\vec{\epsilon}_1 \times \vec{\epsilon}_2) \cdot \vec{\epsilon}_V F_a(k^2); \quad (3a)$$

$$\mathcal{A}_b = (\vec{\epsilon}_1 \cdot \vec{\epsilon}_2) \cdot (\vec{\epsilon}_V \cdot \vec{k}) F_b(k^2); \quad (3b)$$

$$\mathcal{A}_c = [(\vec{\epsilon}_1 \times \vec{\epsilon}_2) \cdot \vec{k}] (\vec{\epsilon}_V \cdot \vec{k}) F_c(k^2). \quad (3c)$$

Interchange of the photons leads to the transformations  $\vec{\epsilon}_1 \leftrightarrow \vec{\epsilon}_2$  and  $\vec{k} \leftrightarrow -\vec{k}$ . It can be easily verified that each of the forms written in Eqns. (3) is *odd* under interchange. Therefore, these amplitudes vanish exactly because photons obey BE statistics.

It is also instructive to look at the LY theorem from a slightly different point of view. Suppose we try to calculate the decay rate of a particle into two photons, using Feynman diagrams. We must take into account that the total decay amplitude is the sum of amplitudes from two diagrams, with photon labels exchanged between the two terms:  $\mathcal{A}_{\text{total}} = \mathcal{A}_{12} + \mathcal{A}_{21}$ . The LY theorem shows that the process  $V \rightarrow \gamma\gamma$  must have amplitude  $\mathcal{A}_{\text{total}} = 0$ ; i.e., it must be that  $\mathcal{A}_{12} = -\mathcal{A}_{21}$ . This simple fact leads to a way to search for the existence of exchange-*antisymmetric* two-photon states. For such states, the amplitude  $\mathcal{F}$  for decay into "fermionic photons" should be simply  $\mathcal{F} = \mathcal{A}_{12} - \mathcal{A}_{21}$ , so that in general  $\mathcal{F} \neq 0$ . If we assume that there is a probability  $\nu$  for any pair of photons to be in an antisymmetric state, then the decay rate  $\Gamma_\nu$  for the process  $V \rightarrow \gamma\gamma$  will be  $\Gamma_\nu \propto \nu |\mathcal{F}|^2$ . If  $\mathcal{F}$  can be calculated, one can set limits on  $\nu$  from experimental upper limits on the partial width  $\Gamma(V \rightarrow \gamma\gamma)$ .

We have compiled here all experimental limits known to us on the decay rates of vector particles into two photons. For each particle, we have also estimated the spin-

statistics violating decay rate  $\Gamma_{\nu}(\nu=1)$  and the corresponding branching ratio (B.R.) for  $V \rightarrow \gamma\gamma$ , in order to set limits on the Bose-statistics violation parameter  $\nu$ . The manner of obtaining estimates for  $\Gamma_{\nu}$  differs between cases, and bears some explanation.

We set  $\mathcal{F} = 0$  for decays of the form  $f\bar{f} \ ^3S_{1(-)} \rightarrow \gamma\gamma$ . Our reasoning is that these decays are forbidden not only because photons are in symmetric states, but also because charge conjugation (C) symmetry would be violated by this decay. Since there is no *a priori* reason to expect these two symmetries to be broken together, these decays could be absent even for  $\nu \sim 1$ .<sup>‡</sup> By contrast, we see no similar additional selection rules that forbid the two-photon decays of the  $\chi_{c1}$  particle or the  $Z^0$  boson.

For  $\chi_{c1} = c\bar{c} \ 1p \ ^3P_{1(+)}$ , we estimate that the partial width for decay to two photons is  $\Gamma_{\nu}(\chi_{c1} \rightarrow \gamma\gamma) \sim \nu \cdot 1 \text{ keV}$ . This value is obtained by assuming that  $\Gamma_{\nu}(\nu=1)$  will lie between the measured rates for the allowed decays  $\chi_{c0} \rightarrow \gamma\gamma$  and  $\chi_{c2} \rightarrow \gamma\gamma$ . The decay  $Z^0 \rightarrow \gamma\gamma$  could occur only through loop diagrams, since—like all neutral particles—the  $Z^0$  has no direct coupling to photons. Each such diagram is thus suppressed by two electromagnetic vertex factors (i.e., by  $\sim q^2\alpha^2$  for a fermion of charge  $q$ ), even before the cancellation due to Bose statistics. We have estimated  $\mathcal{F}$  based on the known couplings of the  $Z^0$  to all fermions (and assuming a typical, additional factor of  $\pi$  for the "geometric" suppression of the loop-diagram amplitude). We stress that in both of these cases, a careful calculation of  $\mathcal{F}$  could be of interest.

The data and our conclusions are summarized in Table 1. Remarkably, it appears that little can be learned about  $\nu$  from the existing data. We note in this context that Ignatiev *et al.* previously discussed the decay  $Z^0 \rightarrow \gamma\gamma$ , as a means for studying violation of BE statistics for photons.<sup>18</sup> They emphasize that one may naively expect that any such violation would be enhanced at high energies. However, they did not attempt to derive a value for the parameter  $\nu$  as we have done here. Unfortunately, our estimate indicates that the experimental limits on  $\Gamma_{\nu}(Z^0 \rightarrow \gamma\gamma)$  must be improved dramatically, in order to place any quantitative limit on  $\nu$ .

**TABLE 1. Limits on two-photon antisymmetric states from particle physics.**

Particle, State, $^{2S+1}L_{J(P)}$	Observed B. R. $\rightarrow \gamma\gamma$ (ppm)	B.R. $\rightarrow$ antisymmetric $\gamma\gamma$ , with $\nu=1$ (ppm)	Limit on $\nu$	References
ortho-Ps $= e^+e^- \ 1s \ ^3S_{1(-)}$	<200	0?	none?	[19]
J/ $\Psi$ $= c\bar{c} \ 1s \ ^3S_{1(-)}$	< 500	0?	none?	[20]
$\Psi(2S)$ $= c\bar{c} \ 2s \ ^3S_{1(-)}$	< 200	0?	none?	[20]
$\chi_{c1}(1P)$ $= c\bar{c} \ 1p \ ^3P_{1(+)}$	< 1500	$\lesssim 1000$	$\nu \lesssim 1$	[20]
$Z^0$	< 50	$\lesssim 3$	none	[20]

<sup>‡</sup> Note, however, that the intimate relationship between the spin-statistics connection and the discrete space-time symmetries such as C, make it entirely plausible that a violation of BE statistics could carry with it a violation of C as well. In this case, the limit on  $\nu$  from , e.g., the ortho-Ps  $\rightarrow \gamma\gamma$  B.R., could be quite strong.

### III. USING ATOMIC TWO-PHOTON TRANSITIONS TO SEARCH FOR NON-SYMMETRIC PHOTON STATES

We have recently used a generalization of the ideas outlined above, to search for photon states that are not symmetric under interchange. By searching for certain forbidden two-photon transitions in atoms, we have placed an upper limit  $\nu < 1.2 \times 10^{-7}$  on the BE statistics violation parameter for visible photons.<sup>21</sup> The remainder of this paper will discuss the principle and execution of this experiment, as well as some subtleties in its interpretation.

#### Review of Two-Photon Transitions in Atomic Physics

We begin with a brief review of the properties of two-photon transitions in atomic physics. Imagine we shine two plane-wave beams of light on an atom, as shown in Fig. 2. The photons in beam 1(2) have polarization  $\vec{\epsilon}_{1(2)}$ , momentum  $\vec{k}_{1(2)}$ , and frequency  $\omega_{1(2)}$ . Consider a two-photon transition between initial ( $|i\rangle$ ) and final ( $|f\rangle$ ) atomic states, which have well-defined energies  $E_{i,f}$  angular momenta  $J_{i,f}$ , and parities  $P_{i,f}$ . In general, such a transition could be possible whenever energy conservation is satisfied, i.e. when  $\hbar\omega_1 + \hbar\omega_2 = E_f - E_i$ . Conceptually, the two-photon transition can be thought of as proceeding stepwise via two single-photon absorptions, with the first photon absorption resulting in a virtual excited atomic state (see Fig. 3). There are two possible paths (i.e., amplitudes) along which the two-photon transition can proceed, corresponding to the order in which the two photons are (virtually) absorbed. If photons obey BE statistics, the amplitudes for these two paths must be *added*. The amplitude for the two-photon process is enhanced when the virtual intermediate state is near in energy to a real atomic state ( $|n\rangle$ ). This means that if the two absorbed photons have different frequencies (i.e.,  $\omega_1 \neq \omega_2$ ), then the amplitudes for the two excitation paths will in general be different.

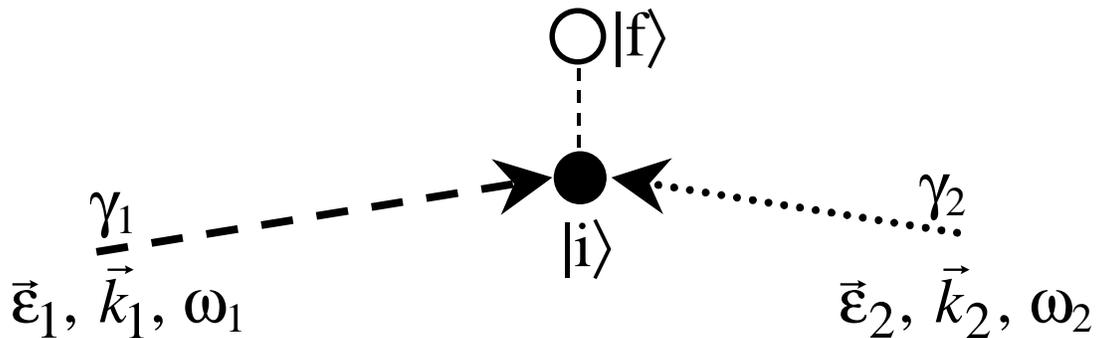
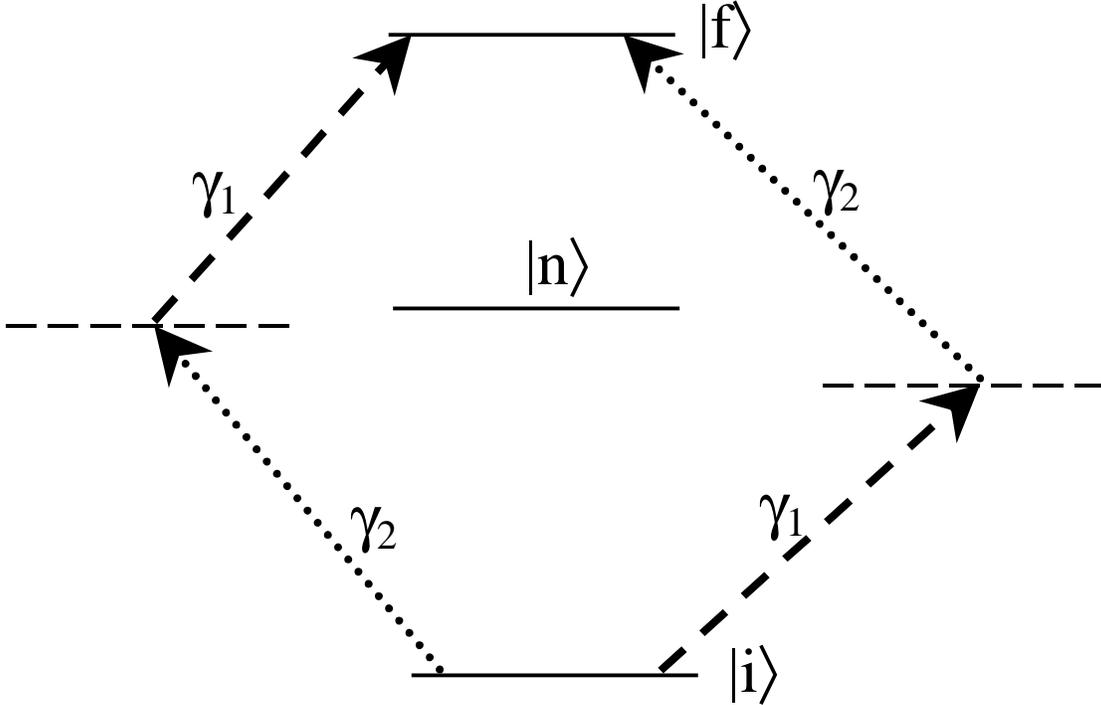


FIGURE 2. Kinematics of an atomic two-photon transition.



**FIGURE 3.** Energy level diagram for an atomic two-photon transition. The solid horizontal lines indicate real atomic states; the dashed horizontal lines are virtual intermediate states.

It will prove convenient to use an approximation that is frequently applied to atomic transitions. We expand the plane wave of each light beam in a Taylor series:

$$e^{i\vec{k}\cdot\vec{r}} = 1 + i(\vec{k}\cdot\vec{r}) - (\vec{k}\cdot\vec{r})^2 / 2 + \dots \quad (4)$$

This expansion is useful because  $(\vec{k}\cdot\vec{r})$  is small for an atomic transition between valence electron states, where  $\lambda = 2\pi/k$  is typically in the visible range and  $r \sim a_0$  (the Bohr radius):  $(\vec{k}\cdot\vec{r}) \sim 2\pi a_0 / \lambda \lesssim \alpha/2 \ll 1$ . Atomic one-photon transitions are usually classified according to their multipolarity, i.e., according to the leading term in Eqn. (4) from which they arise.

This multipole expansion makes transparent the connection between selection rules for atomic transitions and the quantum numbers of the photon participating in the transition. This connection is not usually emphasized in atomic physics, but we will find it useful in our later discussion, so we re-derive it here.<sup>22</sup> The term with power  $|\vec{k}|^n$  in Eqn. (4) arises only from photons with orbital angular momentum  $l = n, n-2, n-4$ , etc. [This can be seen easily from the fact that the  $l$ -wave radial wavefunctions are spherical Bessel functions of order  $l$ :  $\psi_l \propto j_l(kr)$ ]. Since photons have spin  $s=1$ , the total angular momentum of the photon can only be  $j = l \pm 1$  or  $l$ . By convention, magnetic multipoles have  $l = j$ ; electric multipoles have  $l = j \pm 1$ . Photons have odd intrinsic parity, so the total parity is  $P_\gamma = -(-1)^l$ . As summarized in Table 2, these properties correspond exactly to the standard atomic transition selection rules.

**TABLE 2. Photon Quantum Numbers and Atomic Selection Rules.**

<b>Multipole Term</b>	<b>E1</b>	<b>M1; E2</b>	<b>M2; E3</b>
Power of k in Eqn. (4)	0, 2, ...	1, 3, ...	2, 4, ...
Relative Amplitude	1	$ka_0 \sim \alpha/2$	$(ka_0)^2/2 \sim \alpha^2/8$
photon $l$	0, 2	1; 1, 3	2; 2, 4
photon $s$	1	1	1
photon $j$	1	1; 2	2; 3
atom $\Delta J$	$\pm 1, 0$ ( $0 \rightarrow 0$ )	$\pm 1, 0$ ( $0 \rightarrow 0$ ); $\pm 2, \pm 1, 0$ ( $0 \rightarrow 0, 1$ )	$\pm 2, \pm 1, 0$ ( $0 \rightarrow 0, 1$ ); $\pm 3, \pm 2, \pm 1, 0$ (etc.)
photon $P = -(-1)^l$	-1	+1	-1
atom $\Delta P$	-1	+1	-1

### Analogue of the Landau-Yang Theorem in Atomic Physics

Now we discuss the analogue of the LY theorem in atomic two-photon transitions. It seems reasonable that here transitions of the form  $J=0 + 2\gamma \rightarrow J=1$  might be forbidden, as were particle decays of the form  $J=1 \rightarrow 2\gamma$ . However, the analogous result for atoms is not completely general. The kinematics of the transition have been complicated by the atomic recoil, which in general allows absorption of two photons with  $\vec{k}_1 \neq -\vec{k}_2$ ; this in turn invalidates the earlier proof. In order to recover the LY result, let us for now confine ourselves to the case where the two beams of light are both *collinear* (so that  $\vec{k}_1 \parallel \vec{k}_2$ ) and *degenerate* (so that  $\omega_1 = \omega_2$ ). This situation is almost identical to that in the earlier proof. The only difference is that there is now a possibility for the atom to absorb two photons from the *same* beam, so that  $\vec{k} = \vec{k}_1 = \vec{k}_2$  (rather than  $\vec{k}_1 = -\vec{k}_2$  as was automatically satisfied in the particle decay). Indeed, this seems to invalidate our earlier proof: for such co-propagating photons, the amplitude of Eqn. (3b) need not vanish. However, even this amplitude can be eliminated if we add one additional condition. Under a parity (P) transformation, Eqn. (3b) is odd, while (3a) and (3c) are even. Thus, the amplitude of Eqn. (3b) vanishes for transitions between atomic states of the same total parity. Therefore, our earlier proof can still be applied, to yield the following generalization of the LY theorem: *atomic transitions of the form  $J=0 + 2\gamma \rightarrow J=1$  (with  $\Delta P = 0$ ) are forbidden for degenerate, collinear photons.*

This result suggests that we can search for evidence that photons are in a non-permutation-symmetric state, by looking for these forbidden atomic transitions. Indeed, this is the principle of our experiment, which is described in detail below.

### Transition Amplitudes, Selection Rules, and Bose-Einstein Statistics

The hierarchy of atomic transition multipoles plays an important role in understanding the  $J=0 + 2\gamma \rightarrow J=1$  transitions we are considering. Under ordinary conditions, E1 amplitudes are dominant, so we will focus exclusively on these from here on. We note in passing that higher multipoles in principle can lead to very small

rates for these transitions, which might be significant in a sensitive search for transitions induced by violation of BE statistics. However, the effect of the higher multipoles can be made extremely small by good experimental design.<sup>§</sup> Likewise, atomic parity violation gives nonzero, but negligibly small, transition rates.<sup>\*\*</sup>

From the point of view of selection rules for atomic transitions, the LY result looks rather surprising. Transitions of the form  $J=0^P + 2\gamma \rightarrow J=1^P$  ( $\Delta P=0$ ) obey all selection rules for a two-step E1-E1 process: the first step is  $J=0^P + \gamma \rightarrow J=1^{(P)}$ , and the second is  $J=1^{(P)} + \gamma \rightarrow J=1^P$ . Let us look in detail at the amplitude for such a process.

The general expression for the atomic E1-E1 resonant transition rate  $W_{if}$  is:<sup>23</sup>

$$W_{if}(\Omega_1, \Omega_2) \propto |\epsilon_{1a}\epsilon_{2b}\langle f|Q_{ab}|i\rangle|^2 \frac{dI_1}{d\Omega_1} \frac{dI_2}{d\Omega_2} \delta(\omega_{if} - \Omega_1 - \Omega_2); \quad (5)$$

$$Q_{ab}(\Omega_1, \Omega_2) = d_a \left( \sum_n \frac{|n\rangle\langle n|}{\omega_{ni} - \Omega_1} \right) d_b + d_b \left( \sum_n \frac{|n\rangle\langle n|}{\omega_{ni} - \Omega_2} \right) d_a. \quad (6)$$

Here  $dI_j/d\Omega_j$  is the spectral distribution of light intensity for light beam  $j$ ;  $\omega_{jk}$  is the frequency of the atomic transition  $|j\rangle \rightarrow |k\rangle$ ;  $\vec{d} = e\vec{r}$  is the atomic dipole operator; and the subscripts  $a, b$  refer to Cartesian components. Eqns. (5) and (6) exhibit many of the features we have previously discussed qualitatively. The two terms in Eqn. (6) correspond to the two alternative absorption paths in Fig. 3, i.e., to the direct and exchange Feynman diagrams in a two-photon decay. Because photons obey BE statistics, the amplitude  $\epsilon_{1a}\epsilon_{2b}\langle f|Q_{ab}|i\rangle$  must be symmetric under interchange of the photon labels ( $1 \leftrightarrow 2$ ); this is the origin of the plus sign between the terms in Eqn. (6).

For the specific case of a  $J=0 \rightarrow J=1$  transition, only the irreducible rank-1 component of  $Q_{ab}$  can contribute to the matrix element in Eqn. (5). Thus,  $Q_{ab}$  reduces to its antisymmetric part,  $Q_{ab}^{(1)}$ :<sup>24</sup>

$$Q_{ab}(\Omega_1, \Omega_2) = Q_{ab}^{(1)} = \frac{Q_{ab} - Q_{ba}}{2} = \frac{(\Omega_1 - \Omega_2)}{2} \sum_n \frac{d_a |n\rangle\langle n| d_b - d_b |n\rangle\langle n| d_a}{(\omega_{ni} - \Omega_1)(\omega_{ni} - \Omega_2)}. \quad (7)$$

Eqn. (7) also illustrates several of our earlier points. The transition amplitude  $\epsilon_{1a}\epsilon_{2b}\langle f|Q_{ab}|i\rangle$  is antisymmetric under interchange of the polarization vectors, and in fact takes the form of the rotational invariant in Eqn. (3a):  $(\vec{\epsilon}_1 \times \vec{\epsilon}_2) \cdot \vec{\epsilon}_v$ . (That is, this amplitude requires the two light beams to have different polarizations.) The overall exchange symmetry of the amplitude is maintained because of the additional antisymmetric term  $(\Omega_1 - \Omega_2)$  that appears in  $Q_{ab}$ . This term makes it explicit that *the  $J=0^P + 2\gamma \rightarrow J=1^P$  transitions are completely forbidden if (and only if) the two photons are degenerate (with  $\Omega_1 = \Omega_2$ ).*

<sup>§</sup> Such terms can enter because of imperfect collinearity of the light beams. However, the largest components involve two M1/E2 type one-photon steps, and thus have amplitude in atomic units of  $\lesssim \theta \alpha^2$ , where  $\theta$  is a small misalignment angle.

<sup>\*\*</sup> Parity violation induces a tiny E1-E2 or E1-M1 component to the transitions of interest.

The vanishing of the transition amplitude, specifically in the degenerate case, leads to another way of understanding the role played by BE statistics for photons in these transitions. Let us consider the wavefunction of the two-photon state. For an E1-E1 transition, we take the approximation that each photon has orbital angular momentum  $l = 0$  (this is good up to a correction of order  $(\vec{k} \cdot \vec{r})^2$ ; see Table 2). Then the entire angular momentum of the photons arises from spin, and the transition  $J=0 + 2\gamma \rightarrow J=1$  requires  $S_{\text{tot}} = 1$  for the two-photon system. Since each photon has spin  $s = 1$ , the  $S_{\text{tot}} = 1$  combination is antisymmetric under interchange of the two photons (as can be seen trivially e.g. from the Clebsch-Gordon coefficients for  $1 \otimes 1 \rightarrow 1$ ). Thus, in order to satisfy BE statistics, the spatial part of the two-photon wavefunction must also be antisymmetric under interchange. The spatial part of each photon's wavefunction can be described by an s-wave:  $\psi_j(\vec{r}) \propto j_0(k_j r)$ . Of course, then, the antisymmetrized two-photon wavefunction will vanish if and only if  $|k_1| = |k_2|$  (i.e.,  $\Omega_1 = \Omega_2$ ).

As an aside, we note that it may be instructive to consider the meaning of the factor  $(\Omega_1 - \Omega_2)$  that appears in  $Q_{ab}$ . As is suggested in Fig. 3, the presence of the atomic intermediate state  $|n\rangle$  allows the atom to *distinguish* between two photons with different energies. We mean this in the same sense that one can completely distinguish between an electron in Capri and one in New Haven, because the overlap between their wavefunctions is negligible.<sup>25</sup> In our case, the relevant wavefunction overlap is best considered in momentum space. Here, the atom evidently presents an effective potential function for the photons [see Eqn. (6)]. Only when the two photons are completely indistinguishable (i.e., when they are degenerate) is there perfect cancellation of the spatial-momentum part of the wavefunction, due to its antisymmetrization.

### Searching for Spin-Statistics Violation with $J=0 \rightarrow J=1$ Transitions

We have shown that for exactly degenerate photons, the usual transition amplitude  $Q_{ab}$  vanishes. However, the analogous amplitude for non-symmetric photons should *not* vanish. We construct this amplitude  $\mathcal{F}_{ab}$ , as before, by replacing the plus sign in Eqn. (6) with a minus sign; we then obtain

$$\mathcal{F}_{ab}(\Omega_1, \Omega_2) = \sum_n \left( \omega_{ni} - \frac{\Omega_1 + \Omega_2}{2} \right) \frac{d_a |n\rangle \langle n| d_b - d_b |n\rangle \langle n| d_a}{(\omega_{ni} - \Omega_1)(\omega_{ni} - \Omega_2)}, \quad (8)$$

which is in general non-zero for  $\Omega_1 = \Omega_2$ . We note in passing that a similar result is obtained in the quon algebra,<sup>2</sup> with the amplitude  $\mathcal{F}_{ab} \propto (1 - q)$ . Returning to the general case where  $\Omega_1 = \Omega_2$  is not required, we take the transition rate to be the sum of two probabilities:

$$W_{if}(\Omega_1, \Omega_2) \propto \left\{ |\epsilon_{1a} \epsilon_{2b} \langle f | Q_{ab}(\Omega_1, \Omega_2) | i \rangle|^2 + v |\epsilon_{1a} \epsilon_{2b} \langle f | \mathcal{F}_{ab}(\Omega_1, \Omega_2) | i \rangle|^2 \right\} \times \frac{dI_1}{d\Omega_1} \frac{dI_2}{d\Omega_2} \delta(\omega_{if} - \Omega_1 - \Omega_2). \quad (9)$$

Note that here we have assumed that the normal and BE-violating amplitudes do not interfere; this is expected as a consequence of the superselection rule for transitions between states with different representations of the permutation group.<sup>26</sup>

Eqns. (7), (8), and (9) summarize the central principle of our measurement. That is: for monochromatic light, the degenerate  $J=0 + 2\gamma \rightarrow J=1$  transition rate is due entirely to violation of BE statistics: i.e.,  $W_{if}(\Omega_1 = \Omega_2) \propto \nu$ . We can obtain a value (or upper limit) for  $\nu$  by searching for these degenerate transitions, so long as we know the amplitude  $\mathcal{F}_{ab}$  and can calibrate the sensitivity of our apparatus.

The nature of the photon states that can be detected using this principle is implicit in Eqn. (9). Note that  $W_{if} \propto I_1 I_2$ , (where  $I_j$  is the intensity in light beam  $j$ ), for both the allowed and the BE statistics-forbidden processes. The intensity  $I \propto N$ , where  $N$  is the number of photons in the beam. Thus, from the point of view of photons,  $W_{if} \propto N_1 N_2$ . This behavior is easily understood by a counting argument. Each transition requires one photon from each beam, in order to absorb two photons with orthogonal polarizations. For the allowed process, there are  $N_1$  choices for the first photon and  $N_2$  for the second, and thus a total of  $N_1 N_2$  pairs available. For the BE-statistics violating process, at least one of the photons must be antisymmetric with the rest. However, there are  $N_j$  choices for the antisymmetrized photon in beam  $j$ ; with any given choice, this photon can pair with any of the  $N_k$  photons in beam  $k$ . Therefore, again we conclude that  $N_1 N_2$  pairs are available. Note that this argument implies that we are sensitive to photon states with Young tableaux of the form

$$\begin{array}{|c|c|c|c|c|c|c|c|c|c|c|c|c|c|c|} \hline \square & \square \\ \hline \square & & & & & & & & & & & & & & \\ \hline \end{array}. \quad (10)$$

In this sense, our experiment is sensitive not only to violations of the spin-statistics relation, but also to violations of the more general permutation symmetry postulate.<sup>27</sup>

### Specific Features of Our Experiment

All relevant details of our experiment were given in Ref. [21]. Here we give just a brief outline of our specific experimental scheme. We searched for the transition  $6s^2 \ ^1S_0 + 2\gamma \rightarrow 5d6d \ ^3S_1$  in atomic Ba. This transition has an unusually large BE-violating amplitude, because an intermediate state  $|n\rangle$  ( $= 6s6p \ ^1P_1$ ) happens to lie very close to the halfway point in energy between the initial and final states.

Light from a dye laser was split into two beams with orthogonal linear polarizations. These beams counterpropagated through a Ba vapor cell. The laser was tuned around the required frequency for the degenerate two-photon transition ( $\lambda = 549$  nm). Transitions were detected by observing fluorescence at  $\lambda_{fl} = 436$  nm, accompanying the decay  $5d6d \ ^3S_1 \rightarrow 6s6p \ ^3P_2$ . Excess signal within a narrow tuning range of the laser wavelength would indicate a violation of BE statistics. The sensitivity of the experiment was calibrated with the same detection system, using non-degenerate photons ( $\lambda'_1 = 532$  nm and  $\lambda'_2 = 566$  nm) to drive the same transition. That is, we measured the ratio of signals for the degenerate and calibration transitions:

$$S \equiv \frac{W_{if}(\Omega_1 = \Omega_2 = \omega_{if}/2)}{W'_{if}(\Omega'_1, \Omega'_2)}, \quad (11)$$

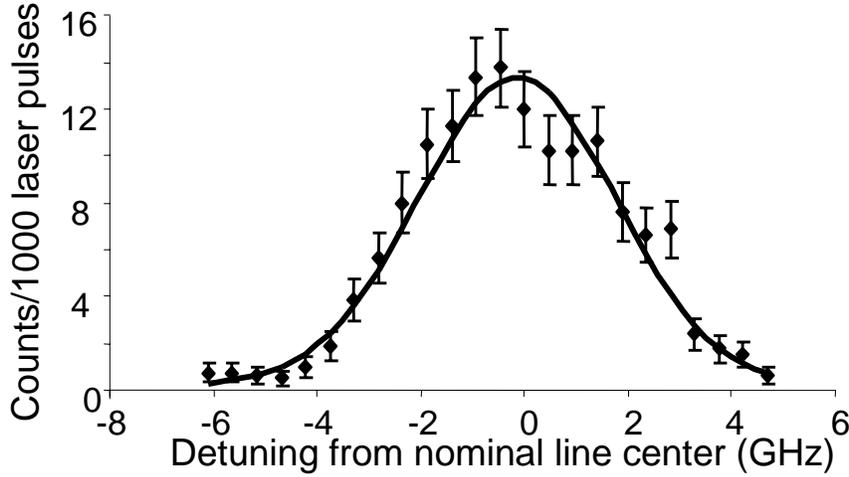
where the primed quantities correspond to the non-degenerate transition.

The value of  $S$  determines  $\nu$  (assuming no other sources of background signal):

$$\nu = S \frac{\delta\Omega_1}{\delta\Omega'_1} \frac{\delta\Omega_2}{\delta\Omega'_2} \frac{I'_1 I'_2}{I_1 I_2} \frac{1}{R^2}, \quad (12)$$

where  $\delta\Omega_j$  and  $I_j$  are the spectral width and total intensity, respectively, of the laser at frequency  $\Omega_j$ ; and  $R^2$  is the ratio of matrix elements of two-photon operators:

$R^2 \equiv |\mathcal{P}_{if}(\Omega_1 = \Omega_2 = \omega_{if}/2)/Q_{if}(\Omega'_1, \Omega'_2)|^2$ .  $R^2 = 10 \pm 2$  was determined from known atomic transition energies and dipole matrix elements. All other quantities in Eqn. (12) were measured individually. Uncertainties from each of these measurements led to an overall uncertainty of  $\sim 70\%$  in the calibration constant used to determine the value of  $\nu$  from the measured value of  $S$ . Fig. 4 shows a typical scan of the laser through the nondegenerate calibration transition.



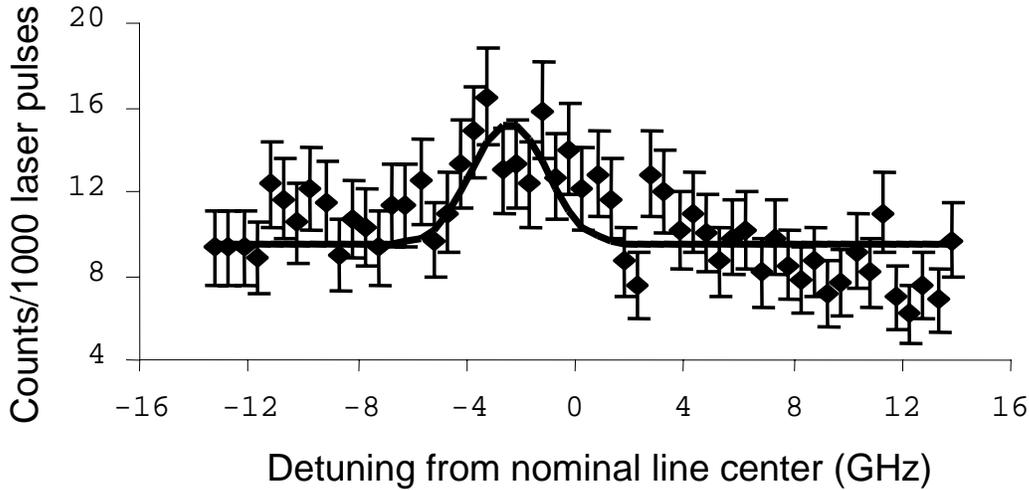
**FIGURE 4.** Typical scan through the calibration transition and fit to peak height and linewidth. Taken with  $\sim 0.4 \mu\text{J}/\text{pulse}$  at  $\lambda'_1 = 532 \text{ nm}$  and  $\sim 200 \mu\text{J}/\text{pulse}$  at  $\lambda'_2 = 566 \text{ nm}$ .

A typical scan through the degenerate transition is shown in Fig. 5. These signals were fit to a constant background plus a peak. In all data sets, there is evidence for a statistically significant peak above the background. We believe these peaks are due to the finite bandwidth of the laser. For light from a laser of *finite* spectral width, the transition probability of Eqn. (9) does not vanish for  $\nu=0$ , even though  $Q_{ab}(\Omega_1 = \Omega_2) = 0$ . This is because an atom can absorb two photons from opposite sides of the laser spectral distribution; then, from the atom's point of view,  $\Omega_1 = \Omega_2$  is not strictly satisfied. From a crude model for our laser spectra, we predicted the size of the residual signal  $S$  due to this "bandwidth effect." Averaging over all data gave the result for the ratio of the observed to predicted signal due to the bandwidth effect:

$$\frac{S(\text{observed})}{S(\text{predicted})} = 1.5 \pm 0.6. \quad (13)$$

That is, the observed resonances were consistent with those expected for purely bosonic photons, due to the finite bandwidth of the dye laser.

It should be noted that these residual peaks are extremely small. This can be verified simply by looking at the data in Figs. 4 and 5. For the calibration transition, with  $I_1 I_2' \sim 10^{-4}$  (arb. units), the signal is  $W_{if} \sim 0.1$ . By contrast, for the degenerate transition, we typically saw signals  $W_{if} \sim 3 \times 10^{-3}$ , with  $I_1 I_2 \sim 3$ . Using Eqn. (12) (with all  $\delta\Omega_j$  approximately the same), we see that the bandwidth-effect peaks have a size similar to that which would occur for a BE-statistics violation, with  $\nu \sim 10^{-7}$ .



**FIGURE 5.** A typical scan through the degenerate transition, with the fit to peak plus background. Taken with  $\sim 1.5$  mJ/pulse in each beam, at  $\lambda_1 = \lambda_2 = 549$  nm.

A violation of BE statistics would of course appear as a resonant signal *in excess* of the peak due to the bandwidth effect [see Eqn. (9)]. Thus, in order to determine an upper limit on  $\nu$ , we would ideally subtract this background. However, although it is true that the observed peak is consistent with our crude predictions, we also find that the size of the bandwidth-effect peak is quite sensitive to details of the laser spectra. Thus, for determination of  $\nu$ , we choose to take the most conservative approach and assume that the *entire* observed peak could in principle be due to violation of BE statistics. In this case, we can use Eqn. (12) to find  $\nu$ . This allows us to set a limit on the BE statistics violation parameter for photons:

$$\nu < 1.2 \times 10^{-7} \text{ (90\% c.l.)}. \quad (14)$$

This represents the first result based on a new principle, which in the ideal case gives a background-free signal arising from violation of BE statistics for photons. We believe that the limit on  $\nu$  can be decreased by several orders of magnitude, with experiments based on this same principle but applying new techniques. Such an experiment is now underway, and is described in the contribution to these proceedings by D. Budker.<sup>28</sup>

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