Search for exchange-antisymmetric two-photon states

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Atomic two-photon $J=0 \leftrightarrow J'=1$ transitions are forbidden for photons of the same energy. This selection rule is related to the fact that photons obey Bose-Einstein statistics. We have searched for small violations of this selection rule by studying transitions in atomic Ba. We set a limit on the probability $v$ that photons are in exchange-antisymmetric states: $v < 1.2 \cdot 10^{-7}$.

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Recent experiments have explored the possibility of small violations of the usual relationship between spin and statistics [1,2]. Although such violations are impossible within conventional quantum field theory [3], there are motivations for considering them; see e.g. Ref. [4]. If photons do not obey Bose-Einstein (BE) statistics, there will be a non-zero probability that two photons are in an exchange-antisymmetric state. Here we report the results of a search for such states based on a selection rule [5,6] that forbids two-photon transitions between atomic states with $J = 0$ and $J' = 1$ for degenerate photons (i.e., photons of equal energy).

Consider the general amplitude for a $J = 0 \rightarrow J' = 1$ two-photon transition. This amplitude is a scalar which must be constructed from the polarization vectors $e_1, e_2$ of the photons and the polarization vector $e_v$ describing the $J' = 1$ state (each exactly once); and may also contain arbitrary powers of the photon propagation directions $k_{1,2}$, and some function of the photon energies $h\Omega_{1,2}$. In the specific case of E1-E1 transitions, the amplitude must be independent of $k_{1,2}$, and only one form is possible:

$$A \propto \langle (e_1 \times e_2) \cdot e_v \rangle F(\Omega_1, \Omega_2),$$

which requires orthogonally-polarized photons. If photons obey BE statistics, this amplitude must be invariant under exchange of labels 1 $\leftrightarrow$ 2. Eqn. (1) satisfies this condition only if $F(\Omega_1, \Omega_2)$ is odd under exchange. Therefore the amplitude must vanish in the degenerate case, if photons behave as normally expected. This argument can be readily generalized beyond the E1-E1 case [1]. For the case of counterpropagating degenerate photons (with $k_2 = -k_1$), the Landau-Yang theorem states that all possible amplitudes vanish [1]. We consider only the E1-E1 amplitude of Eqn. (1), since higher multipolarity transitions are too weak to be observed in the present experiment.

For ordinary photons, the atomic two-photon-resonant transition rate is [3,7,8]:

$$W_{gf}(\Omega_1, \Omega_2) \propto |e_{1a}e_{2b}(f|Q_{ab}|g)|^2 \frac{d^4\Omega_1}{d^4\Omega_2} \delta(\omega_{fg} - \Omega_1 - \Omega_2),$$

(2)

$$Q_{ab}(\Omega_1, \Omega_2) = d_a \left( \sum n \frac{n|n\rangle \langle n|}{\omega_{fg} - i\zeta_{2}} \right) d_b + d_b \left( \sum n \frac{n|n\rangle \langle n|}{\omega_{fg} - i\zeta_{1}} \right) d_a.$$  

(3)

Here indices $g, f$, and $n$ indicate ground, final, and (virtual) intermediate states of the transition; $d_{1(2)}$ are the spectral distributions of light intensity; $\omega_{ij}$ are frequencies of atomic transitions; $d$ is the dipole operator; and the subscripts $a, b$ refer to Cartesian components. Consistent with our experimental conditions, we neglect Doppler and natural widths compared to laser spectral widths. For a $J = 0 \rightarrow J' = 1$ transition, only the irreducible rank-1 component of $Q_{ab}$ can contribute to the matrix element [3,7,8].

Thus

$$Q_{ab}(\Omega_1, \Omega_2) = Q_{ab}^{(1)} = \frac{1}{2} (Q_{ab} - Q_{ba}) = \frac{1}{2} (\Omega_1 - \Omega_2) \sum_n \langle d_{a}|n\rangle \langle n|d_{b}\rangle \langle n|d_{a}\rangle \langle n|d_{b}\rangle,$$

(4)

Eqn. (4) shows explicitly that degenerate transitions are forbidden: $Q_{ab}(\Omega_1 = \Omega_2) = 0$. Also, the transition amplitude $e_{1a}e_{2b}(f|Q_{ab}|g)$ has, as expected, the form of the rotational invariant in Eqn. (3).

We now generalize these results to allow for violation of BE statistics. Permutation symmetry for photons is reflected in the plus sign between the two terms in Eqn. (3). We construct a similar "BE-Violating" two-photon operator with a minus sign between the terms:

$$Q_{ab}^B(\Omega_1, \Omega_2) = \sum_n \frac{\omega_{fg} - \omega_{12}^2}{\omega_{fg} - i\zeta_{1}} \langle d_{a}|n\rangle \langle n|d_{b}\rangle \langle n|d_{a}\rangle \langle n|d_{b}\rangle.$$  

(5)

The transition rate becomes:

$$W_{gf}(\Omega_1, \Omega_2) \propto [\langle e_{1a}e_{2b}(f|Q_{ab}|g)|^2 + v \langle e_{1a}e_{2b}|Q_{ab}^B|g\rangle]^2 \frac{d^4\Omega_1}{d^4\Omega_2} \delta(\omega_{fg} - \Omega_1 - \Omega_2),$$

(6)

where $v \ll 1$ is the BE statistics violation parameter, i.e., $v$ is the probability to find two photons in an antisymmetric state. Here we explicitly include the fact that the normal and BE-violating amplitudes cannot interfere with
each other [13]. Eqns. (1) and (3) summarize the central principle of our measurement: for monochromatic light, the degenerate $J = 0 \rightarrow J' = 1$ transition rate is due entirely to BE statistics violation; i.e., $W_{gf}(\Omega_1 = \Omega_2) \propto v$.

Recent theoretical discussion of possible small violations of the spin-statistics relation has centered on the "quon algebra," which allows continuous transformation from BE to Fermi statistics [3]. Our heuristic argument above is reproduced in the quon formalism: if creation/annihilation operators for photons obey the q-deformed commutation relations

$$a_k a_i^+ - qa_i^+ a_k = \delta_{ki},$$

then in Eqn. (1), $v = (\frac{1}{\sqrt{2}}q)^2$ [14]. Degenerate $J = 0 \rightarrow J' = 1$ two-photon transitions are allowed only to the extent that $q$ deviates from 1. However, it should be noted that application of the quon formalism to photons is questionable: relativistic quon theories exhibit nonlo-
high powers (presumably due to photoinization). We saw no evidence for line shifts or distortions even at the highest powers.

The ratio of two-photon operators $R$ is determined as follows. Atomic transition energies $\omega_{m}$ and $\omega_{nf}$ and magnitudes of dipole matrix elements $|\langle n| d|g \rangle|$ are known for all significant intermediate states $|n\rangle$ in the sums of Eqsns. (1) and (2). We measured magnitudes of dipole matrix elements $|\langle f| d|n \rangle|$ by determining the lifetime of, and branching ratios of all decays from, the state $5d6d3S_1$. The lifetime ($25\pm15\text{ ns}$) was measured by recording the time evolution of fluorescence. Branching ratios were measured by observing fluorescence through a scanning monochromator. We find that the sums over intermediate states in Eqsns. (3) and (4) are all well approximated by a single term, corresponding to the intermediate state $|n\rangle = 6s6p^1P_1$. This term has small energy denominators, and large dipole matrix elements with both the initial and final states. (This was the reason we used this particular transition.) In this approximation, the matrix elements cancel in the ratio $R$, and this quantity depends only on accurately known atomic and photonic energies. We find $R^2 = 10 \pm 2$, with the uncertainty due to the neglected terms in the sums.

Data for the degenerate transition were taken in three separate runs (Fig. 4). The laser was scanned around the nominal frequency of the degenerate transition. The signals were fit with a constant background plus a peak whose width was fixed by the dye laser spectral width. The small constant background appears to arise from Ba-He collision-assisted transitions, but is not fully understood. In all three runs, there is evidence for a statistically significant peak above the background. The center frequencies are consistent with the predicted position of the degenerate transition, and with each other. Note that these peaks correspond to extremely weak transitions: although the laser intensities were much larger for the degenerate transition than for the calibration transition ($10^4 \times 10^4$), the ratio of degenerate transition to calibration transition signals is small ($S \sim 10^{-2}$).

We believe these peaks are due to the finite bandwidth of the dye laser. For light from a single laser of finite spectral width (centered around $\omega_L = \omega_{fg}/2$), the transition probability of Eqn. (1) does not vanish for $v = 0$, even though $Q_{ab}(\Omega_1, \Omega_2) = 0$ for $\Omega_1 = \Omega_2$. From the known experimental parameters and plausible models for our laser spectra, we can predict the size of the residual signal $S$ due to this "bandwidth effect". The uncertainty in the predicted size of the fitted peaks themselves was smaller: $25 - 30\%$ for each run. Averaging over all three runs gives the result for the ratio of the observed peak, to the predicted size of bandwidth-effect peak:

$$\frac{S(\text{observed})}{S(\text{predicted})} = 1.5 \pm 0.6. \quad (9)$$

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

A violation of BE statistics would appear as a resonant signal in excess of the peak due to the bandwidth effect. (Note that from Eqn. (6), there is no mechanism for cancellation of the peak when $v \neq 0$.) Although the observed peak is consistent with our predictions, we find that the size of the bandwidth-effect peak is sensitive to details of the laser spectra. In particular, in some models of the spectra which are implausible but not a priori excluded by our data, the bandwidth-effect peak can be substantially suppressed. Thus, for determination of $v$, we 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, once again, $v \propto S$; uncertainties in $S$ and the calibration constant are as described above. This yields a limit on the BE statistics violation parameter for photons:

$$v < 1.2 \cdot 10^{-7} \text{ (90\% c.l.)}. \quad (10)$$

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 $v$ can be decreased by several orders of magnitude, with experiments based on this same principle but applying new techniques (including the use of narrowband cw lasers and highly efficient detection schemes). Such an experiment is now underway.

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[1] E. Ramberg and G. Snow, Phys. Lett. B 238, 438 (1990).
[2] K. Deilamian, J.D. Gillaspy, and D.E. Kelleher, Phys. Rev. Lett. 74, 4787 (1995).
[3] M. de Angelis, G. Gagliardi, L. Gianfrani, and G.M. Tino, Phys. Rev. Lett. 76, 2840 (1996).
[4] R.C. Hilborn and C. Yuca, Phys. Rev. Lett. 76, 2844 (1996).
[5] G. Modugno, M. Ignuscio, and G.M. Tino, Phys. Rev. Lett. 81, 4790 (1998).
[6] See e.g. R.F. Streater and A.S. Wightman, PCT, Spin and Statistics, and All That (W.A. Benjamin, New York, 1964).
[7] O.W. Greenberg and R.N. Mohapatra, Phys. Rev. D 39, 2032 (1989).
[8] K.D. Bonin and T.J. McIlrath, J. Opt. Soc. Am. B 1, 52 (1984).
[9] C.J. Bowers, D. Budker, E.D. Commins, D. DeMille, S.I. Freedman, A.-T. Nguyen, S.-Q. Shang, and M. Zolotorev, Phys. Rev. A 53, 3103 (1996).
[10] J.J. Sakurai. Invariance Principles and Elementary Particles. (Princeton University Press, Princeton, 1964), pp. 15-16.
[11] L.D. Landau, Dokl. Akad. Nauk. USSR 60, 207 (1948); C.N. Yang, Phys. Rev. 77, 242 (1950).
[12] R. D. Amado and H. Primakoff, Phys. Rev. C 22, 1338 (1980).
[13] O.W. Greenberg, Phys. Rev. Lett. 64, 709 (1990).
[14] R. Hilborn and O.W. Greenberg, to be submitted.
[15] O.W. Greenberg, Phys. Rev. D 43, 4111 (1991).
[16] R. Hilborn, private communication.
[17] D. Fivel, Phys. Rev. A 43, 4913 (1991).
[18] O. W. Greenberg, in Workshop on Harmonic Oscillators, ed. D. Han, Y.S. Kim, and W.W. Zachary (NASA Conf. Pub. 3197, NASA, Greenbelt, MD, 1993).
[19] O.W. Greenberg and R.C. Hilborn, Los Alamos e-print [hep-th/9808106], submitted to Found. Phys.
[20] A. Yu. Ignatiev, G.C. Joshi, and M. Matsuda, Mod. Phys. Lett. A 11, 871 (1996).
[21] The correction factor takes into account the ratio of cross-sections for depletion of fluorescence at 566 nm and 549 nm; this quantity was measured in a separate apparatus (D.E. Brown, D. Budker, D. DeMille, E. Deveney, and S. M. Rochester, to be published).
[22] C.E. Moore, Atomic Energy Levels, Vol. III (NBS Circular No. 467: Washington, U.S. G.P.O., 1958).
[23] B.M. Miles and W.L. Weise, Atom. Data 1, 1 (1969).
[24] P. Hafner and W. Schwarz, J. Phys. B11, 2975 (1978).

FIG. 1. Excitation and detection schemes and relevant levels in atomic barium.

FIG. 2. Schematic of the apparatus. BS=beamsplitter. For the forbidden transition, the dye laser is tuned around 549 nm, and the flip-up mirror is in place as shown, so each of the counterpropagating beams in the vapor cell originates from the dye laser. For the calibration transition, the dye laser is tuned to 566 nm and the flip-up mirror is removed. The beam entering the vapor cell from the left then originates from the dye laser (566 nm), while the beam entering from the right originates from the Nd:YAG laser (532 nm).

FIG. 3. Typical scan through the nondegenerate calibration transition (points) and fit to determine peak height and linewidth (solid line). Taken with 230 µJ/pulse at 566 nm and 0.4 µJ/pulse at 532 nm.
FIG. 4. Scans through the degenerate transition and best fits to peak plus background.