Spectroscopic Test of the Symmetrization Postulate for Spin-0 Nuclei

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We have used diode laser absorption spectroscopy of the "*A*" band of molecular oxygen near 762 nm to provide a sensitive experimental test of the symmetrization postulate of quantum mechanics. We place an upper limit of $(0.8 \pm 1.3) \times 10^{-6}$ on the probability of finding two identical ^{16}O nuclei in an antisymmetric state. This is one of the first sensitive tests of the symmetrization postulate for spin-0 particles.

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The special role of identical particles in quantum mechanics is of fundamental importance both in the application of quantum mechanics to many-particle systems (leading notably to the Pauli exclusion principle for fermions and the possibility of Bose-Einstein condensation for bosons) and in understanding the foundations of quantum mechanics (see, for example, Refs. [1–3]). The symmetrization postulate (SP) [4,5] embodies the assumption used to treat many-particle systems: The allowable wave functions must be either symmetric under the interchange (permutation) of identical particle labels or antisymmetric. If the permutation symmetric wave functions apply, the particles are bosons, and the many-body system is described by Bose-Einstein statistics. If the antisymmetric wave functions are appropriate, the particles are fermions, and the Fermi-Dirac distribution is used for the many-body system. Relativistic quantum field theory provides us with the famous spin-statistics theorem, which, under a set of appropriate assumptions [6], provides a connection between the intrinsic spin of a particle and the symmetry or antisymmetry of the states under identical particle exchange: Particles with half-integer spin quantum numbers are fermions, and those with integer spin quantum numbers are bosons.

It has been recognized since the early 1950s that the spin-statistics theorem relies on the assumption that the only permissible operator algebras involve either commutators or anticommutators for creation and annihilation operators. Green [7] showed that if more complicated algebras, such as trilinear combinations of commutators and anticommutators, are used, then field theory permits states belonging to higher-dimensional representations of the permutation group. (The symmetric and antisymmetric states belong to the one-dimensional representations of the permutation group.) Experiments indicate that nature does not seem to make use of these higher representations [8,9]. More recently, Greenberg [10] and others [11–13] have shown that so-called q-deformed commutation relations

$$a_k a_l^\dagger - q a_l^\dagger a_k = \delta_{kl}\,, \tag{1}$$

with $-1 \le q \le 1$, lead to a consistent formalism to describe the possibility of "small" violations of the SP. (As usual, $a_k^\dagger$ is the creation operator for the state labeled by k, and a_k is the corresponding annihilation operator.) To illustrate the meaning of a small violation of the SP, let us consider two identical spin-0 particles. By the SP, the states for these bosons must always be symmetric with respect to the interchange of the identical particle labels. If the deformed commutators describe reality, then there is a small probability, proportional to $1 - q$, that a state for the two particles will be antisymmetric. Without committing ourselves to any particular formalism, we must acknowledge that such a violation of the SP is theoretically possible. It is then a question for experiment to measure (or set an upper limit on) the probability of finding the two particles in a state that violates the SP.

In considering possible experiments to test the SP it is important to note the existence of a superselection rule [14]. Within the framework of standard quantum mechanics, the permutation symmetry of a system cannot change. Thus, if we wish to look for violation of the SP for two identical integer-spin particles, for example, we must look for transitions between pairs of antisymmetric states. Transitions from antisymmetric states to symmetric states are absolutely forbidden.

The superselection rule requires that the state of a two-particle system be described by a density matrix with no "coherence" terms linking the permutation symmetric sector and the antisymmetric sector. If we denote, following the now standard notation, that the probability of finding the two particles in an antisymmetric state is given by $\beta^2/2$, then the density matrix for the two identical integer-spin particles can be written as

$$\rho = (1 - \beta^2/2)\rho_s + (\beta^2/2)\rho_a\,, \tag{2}$$

where $\rho_{s(a)}$ is the density matrix for the symmetric (antisymmetric) sector. (If we adopt the q-deformed commutator formalism, then we have $\beta^2 = 1 - q$.) For a survey of experiments related to possible violations of the SP and some speculations about why the SP might be violated, see [15,16].

Two recent experiments have provided high sensitivity tests of the SP for electrons. The experiment by Ramberg and Snow [17] searched for x rays that would be emitted

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if electrons, newly introduced into a copper bar by a flow of electrical current, would go into a state that is not antisymmetric with respect to the other electrons, and consequently be free to make transitions to already-filled K shells. Because of the large number of electrons in their sample, they were able to set a stringent limit of $\beta^2/2 < 1.7 \times 10^{-26}$. In a simpler two-electron system, free of any assumptions about the nature of multiparticle states, Deilamian, Gillaspy, and Kelleher [18] looked for laser-induced fluorescence near 390 nm emitted in transitions between the symmetric states *(1s2s)*2^1S and *(1s3p)*3^1P of atomic helium. They were able to set a limit of $\beta^2/2 < 5 \times 10^{-6}$ for electrons in helium. Both of these experiments can be viewed as setting upper limits on violations of the exclusion principle for electrons.

Experimental tests of the SP for integer-spin particles are somewhat more difficult since they can no longer rely on the exclusion principle. For example, Greenberg and Mohapatra [19] have found that if one interprets $K_L \rightarrow \pi^+\pi^-$ as indicating a violation of the SP (rather than the customary *CP* violation) for pions, then one needs $\beta^2/2 \leq 10^{-6}$. Possible tests for the SP for photons have been discussed in [20–22], but these all predict photon-number dependent effects that are small under realizable conditions.

In order to provide a sensitive test of the SP for bosons, we have chosen to investigate the spectroscopy of molecules containing two identical spin-0 nuclei [23]. ^{16}O is an attractive nucleus for this type of investigation because its relatively light mass leads to widely spaced molecular rotational levels. In addition the other stable isotopes ^{18}O and ^{17}O have relatively low natural abundances, 0.2% and 0.04%, respectively. Both facts lead to relatively open spectroscopic bands. The diatomic oxygen "*A*" band near 762 nm is well characterized [24,25], allowing us to predict with high accuracy the location and line shape of SP-forbidden transitions. Although the *A* band $X^3\Sigma_g^- \leftrightarrow b^1\Sigma_g^+$ transition is a magnetic dipole transition, it is relatively straightforward to produce long absorption paths to compensate for the weakness of the transition.

Since diode lasers provide a stable, tunable light source for high sensitivity spectroscopy (see Refs. [26–28] for recent work on molecular oxygen absorption), we are able to provide a sensitive test of the SP for spin-0 ^{16}O [29]. As is well known [30], the permutation symmetry of the state of a diatomic molecule is closely tied to the rotational quantum number *K*. For example, in the ground

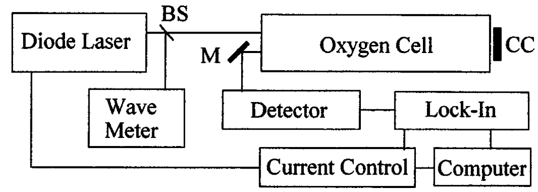

FIG. 1. A schematic diagram (not to scale) of the apparatus. The laser beam passes twice through the 2 m long absorption cell. CC is a corner cube reflector. M is a mirror and BS is a beam splitter.

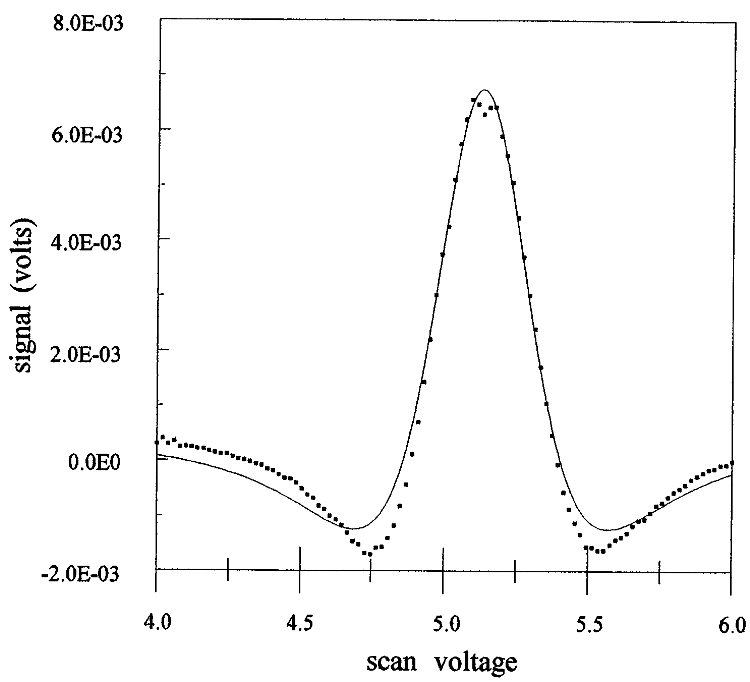

FIG. 2. A plot of the 2*f* demodulated signal (filled squares) from the lock-in detector as the diode laser frequency is scanned once through the $^PQ(21)$ line of $^{16}O_2$. The lock-in time constant was 0.1 sec. The solid line is a fit to the data using the line shape function described in the text. The abscissa corresponds to a frequency scan of approximately 13 GHz. The observed linewidth is greater than the intrinsic, collision-broadened linewidth due to the modulation imposed on the laser drive current.

state of $^{16}O_2$, the rotation levels with K'' odd are permutation symmetric and hence allowed by the SP, while those with K'' even are forbidden by the SP. For the $b^1\Sigma_g^+$ state, the roles of odd and even are reversed. Hence, our experiment consists of a high sensitivity search for transitions from K'' even levels of the ground state to K' odd levels of the b state of $^{16}O_2$.

Figure 1 shows the general layout of the experiment. Tunable radiation near 760 nm was produced by dry-ice cooling of a room-temperature 780 nm diode (Sharp LT024MD). A thermoelectric stage regulated the diode temperature to better than 0.01 °C. An external cavity using a diffraction grating guaranteed tunable single mode operation for the diode laser [28]. By a combination of temperature tuning and operation of the external cavity, we could tune the laser to essentially any desired wavelength in the 0-0 branch of the A band. The laser light was frequency modulated by modulating the diode drive current at 10 kHz. The laser beam then passed twice through a 2-m-long sample cell containing pure (99.993%) diatomic oxygen. The transmitted light was detected by an ultralow-noise silicon photodiode-amplifier combination (New Focus model 2001). A lock-in amplifier provided a $2f$ demodulated signal that is approximately described by the second derivative of a Lorentzian line shape function for the conditions of this experiment. (A detailed treatment of line shapes for the O_2 A band is given in [25].) Using $2f$ demodulation removes most of the diode laser intensity modulation that accompanies the frequency modulation. A computer recorded the signal from the lock-in amplifier and controlled the scan of the diode drive current to sweep the laser frequency through the desired frequency region. We used a wave meter with an accuracy of ± 0.3 GHz to tune the laser.

At room temperature the Doppler width (FWHM) of the O_2 absorption lines is about 0.9 GHz. At atmospheric pressure, used in this experiment, the self-collision broadening leads to linewidths of about 3 GHz. The pressure-induced line shifts are negligible compared to the linewidth. Given the known spectroscopic constants [24] for the A band, we are able to calculate the positions of the SP-forbidden lines with an uncertainty less than 0.3 GHz, about one-tenth of the observed linewidth.

Figure 2 shows the signal from the $^PQ(21)$ SP-allowed line [31] at 391 017.4 GHz with a lock-in time constant of 0.1 sec. The solid line is the result of fitting the second derivative of a Lorentzian function (plus a baseline) to the data. The amplitude is the only adjustable parameter. Figure 3 shows the signal from ten repeated scans in the region in which the $^PQ(20)$ SP-forbidden line at 391 159.3 GHz should occur. Note the expanded vertical scale. By fitting the function describing the expected line shape and position to the data shown in Fig. 3, we find that the intensity of the SP-forbidden line compared to the SP-allowed line is less than $(0.8 \pm 1.3) \times 10^{-6}$ (95%

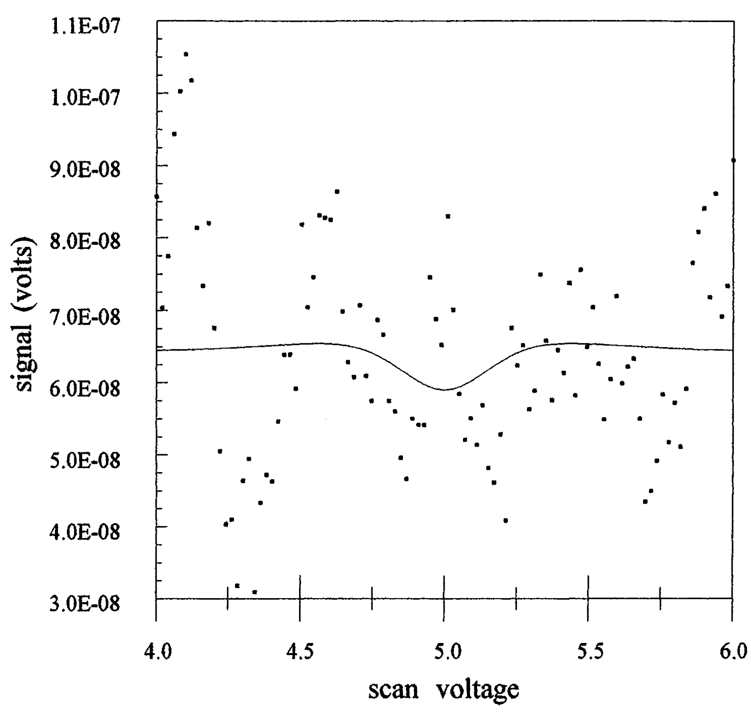

FIG. 3. A plot of the $2f$ demodulated signal (filled squares) from the lock-in detector as the diode laser frequency is scanned ten times through the location of the SP-violating $^PQ(20)$ line of $^{16}O_2$. The time constant was 2 sec. The solid line is a fit to the data using the line shape function described in the text. The uncertainty in the forbidden line position is about one-tenth of the linewidth. The fitting routine allowed for both positive and negative amplitudes, and the best fit gave a line shape inverted compared to that shown in Fig. 2.

confidence limit). This value is interpreted as the experimental upper limit on the parameter $\beta^2/2$, the probability of finding two ^{16}O nuclei in an antisymmetric state. We verified that this limit is insensitive (within a factor of 2) to changes in the line position and the linewidth compatible with the experimental and theoretical uncertainties in those parameters.

Several relatively straightforward improvements in the experiment should enable us to decrease this upper limit substantially. More complex modulation schemes are known to produce higher sensitivity in diode laser absorption spectroscopy [32]. Using a reference light beam derived from the same laser with a double-balanced detector [33] can bring the sensitivity close to the shot noise limit (a few parts in 10^9 for a 1 sec integration time for our system). We note that molecular spectroscopy can be used to provide a test of the SP for a wide variety of spin-0 nuclei.

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