Test of the Symmetrization Postulate for Spin-0 Particles

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We investigated the spectrum of the $^{16}O_2$ molecule by high-sensitivity laser spectroscopy and searched for transitions involving states which are antisymmetric under the exchange of the nuclei. These states are forbidden by the symmetrization postulate of quantum mechanics. This is the first reported experiment to test the symmetrization postulate for integer-spin particles to high precision. Our data set an upper limit of 5×10^{-7} to a possible violation of this postulate.

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Our understanding of the properties of the systems containing identical particles is based on a postulate of quantum mechanics, namely, the symmetrization postulate (SP). According to this postulate, in a system containing identical particles the only possible states are either all symmetrical or all antisymmetrical with respect to permutations of the labels of the particles. In the first case the particles are called bosons; in the second case they are called fermions. Different statistical properties result in the two cases from the postulate. Bosons follow Bose-Einstein statistics; fermions obey Fermi-Dirac statistics. Experiments indicate that particles with half-integer values of spin are fermions, while particles with integer spin are bosons. The reason why only symmetric and antisymmetric states seem to occur in nature and the connection with the spin of the particles has been a puzzle since the early days of quantum mechanics [1].

In fact, quantum mechanics would also allow symmetries different from those allowed by the SP, and theories have been studied allowing also for small deviations from conventional statistics which might have been masked in the experiments performed so far. The first attempts to go beyond Bose and Fermi statistics were made in [2] and [3]. Recently, theories have been developed in which Fermi and Bose statistics can be violated by a small amount. One approach uses trilinear commutation relations [4–11] instead of the usual bilinear Bose and Fermi commutation relations. Another approach uses deformed bilinear commutation relations [12–16]. Statistics other than Fermi and Bose have also been investigated for one- and two-dimensional systems [17] and in connection with anyon high-temperature superconducting systems [18].

A few experiments have been performed [19] which test with high accuracy the validity of the SP. In particular, a high precision test on electrons was performed by Ramberg and Snow [20] by introducing electrons into a copper sample and searching for x-ray emission accompanying transitions to the $1S$ state of the copper atom, which is already filled with two electrons. A lower-precision spectroscopic test was performed recently on helium [21], a simple two-fermion system, searching for a line involving the permutation symmetric $1s2s\,^1S_0$ state. These experiments concerned only fermions, that is, the validity of the Pauli exclusion principle (PEP). No accurate test of the SP for integer-spin particles had been reported so far. In [19], a bound to a possible violation of the generalized Bose statistics for pions was inferred assuming that the $K_L \rightarrow \pi^+\pi^-$ decay comes from such a violation. The implications of possible deviations of photons from Bose statistics have also been investigated [22–24]. The lack of precise tests of the SP for integer-spin particles is due to the fact that, while there are several systems in which a violation of the PEP would be detected as a signal on a zero background, the effects of a small violation for particles following Bose-Einstein statistics would usually manifest themselves as a small change in the properties of a many-particle system. This obviously limits the achievable accuracy. The system we studied in this work represents a rare case in which a violation of the SP for integer-spin particles would be detected on a virtually zero background with a sensitivity comparable to the one achieved in the experiments on fermions. We investigated the spectrum of the $^{16}O_2$ molecule and searched for transitions between states which are antisymmetric under the exchange of the two nuclei. If the nuclei are bosons, such states violate the SP. The basic idea of this experiment is therefore analogous to the one underlying the experiment on electrons in the helium atom reported in [21]. The possibility of such a test was discussed in [25]. A similar experiment on different molecules had been proposed in [26].

According to the Born-Oppenheimer approximation and neglecting the coupling of the nuclear spin with the rest of the molecule (which is not important for the experiment proposed here since the spin of ^{16}O nucleus is zero), the total wave function ψ_t can be written in the form

$$\psi_t = \psi_e \psi_v \psi_r \psi_n \,, \tag{1}$$

where ψ_e, ψ_v, and ψ_r are the electronic, vibrational, and rotational functions, respectively, and ψ_n is the nuclear spin function. According to the SP, if the nuclei are bosons the total wave function ψ_t must be symmetric in the exchange of two nuclei. The $^{16}O_2$ molecule represents a particularly simple case because the nuclear spin of ^{16}O is zero and ψ_n is therefore obviously symmetric. The vibrational wave function ψ_v is also unaltered in the exchange of the nuclei because it depends only on the magnitude of the internuclear distance. Since the total wave function ψ_t must be symmetric, only the states corresponding to even (odd) rotational quantum numbers are allowed if ψ_e is symmetric (antisymmetric) [27]. In Fig. 1, a simplified scheme is shown of the levels of the $^{16}O_2$ molecule of relevance in this work. The ground state is a $^3\Sigma_g^-$ state, which is antisymmetric under the exchange of the two nuclei. The rotational states corresponding to even values of the rotational number K are therefore forbidden. Indeed, since the early work on $^{16}O_2$ spectra [28] it was observed that alternate lines are missing. This was recognized [29] as the effect of the SP for identical spin-0 nuclei. On the other hand, a violation of the SP would also allow states corresponding to even rotational numbers. Searching for transitions involving these states corresponds then to searching for violations of the SP. This is indeed the principle of the experiment reported here. In fact, the interpretation of this experiment, as of the previous experiments on fermions, is not obvious. As was pointed out in [30], in the framework of ordinary quantum mechanics transitions between states of different permutation symmetry cannot take place. This is a perfectly rigorous selection rule holding also in the presence of perturbations such as collisions or external fields. It is not possible then to consider states given by a coherent superposition of states of different permutation symmetry. The system must be described as an incoherent mixture which is represented by a density matrix. For small violations of Bose statistics, the two-particle density matrix is

$$\rho_2 = (1 - \tfrac{1}{2}\beta^2)\rho_s + \tfrac{1}{2}\beta^2\rho_a\,, \tag{2}$$

where $\rho_{s(a)}$ is the symmetric (antisymmetric) two-particle density matrix. Using the notation adopted in previous papers [9,19], we call $\beta^2/2$ the probability that a pair of identical particles is in the "wrong" symmetry state. In this sense, the purpose of this experiment is to measure or to bound the relative abundance of SP-violating molecules. Our choice of the oxygen molecule was motivated by the simplicity of this system which makes the interpretation of the results easier. A similar experiment can also be performed on different molecules, not necessarily diatomic molecules, containing identical spin-0 nuclei. We investigated the 0-0 band of the $X^3\Sigma_g^- \rightarrow b^1\Sigma_g^+$ system of $^{16}O_2$ spectra around 762 nm. The observed transitions in this region are weak magnetic-dipole transitions with an absorption coefficient of 10^{-6} cm^{-1}. However, the lines are narrow and well isolated, and high-sensitivity laser spectroscopy detection methods can be used.

A schematic diagram of the apparatus used in this experiment is shown in Fig. 2. The laser source was a distributed-feedback (DFB) diode laser emitting 5 mW cw in a single mode. The emission wavelength could be varied between 760 and 762 nm by changing the temperature of the laser. The laser was temperature stabilized and driven by a low-noise current supply. The emission linewidth was about 20 MHz. The light passed through an optical isolator and beam-forming optics to correct the astigmatism and the asymmetry of the laser spatial mode. A part of the light was sent to a 7-digit wavelength meter and to a temperature-stabilized Fabry-Pérot interferometer which provided a stable frequency marker. The absorption cell was a White-type multipass cell. The absorption path was 100 m. A 10 cm focal-length lens focused the laser light emerging from the cell into a Si photodiode preamplifier. In this experiment, we used a low-frequency

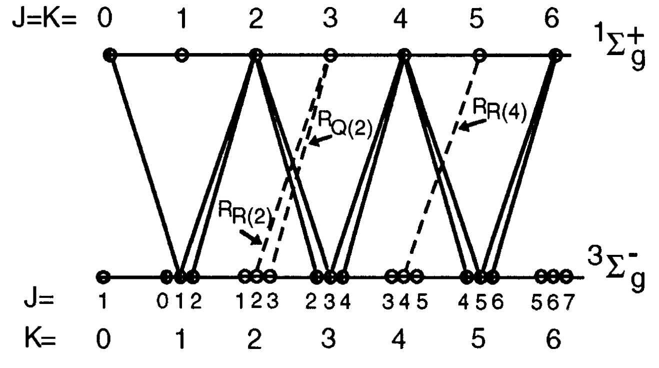

FIG. 1. Partial scheme of the energy levels and the magnetic-dipole transitions of $^{16}O_2$ of relevance in this work. There is an ^RR branch ($\Delta J = +1$, $\Delta K = +1$), a ^PP branch ($\Delta J = -1$, $\Delta K = -1$), an ^RQ branch ($\Delta J = 0$, $\Delta K = +1$) and a ^PQ branch ($\Delta J = 0$, $\Delta K = -1$). The observed transitions (full line in the figure) start from the odd-K rotational levels of the electronic ground state, which are symmetric with respect to an exchange of the nuclei. We searched for transitions (indicated by broken lines) starting from the SP-violating even numbered levels.

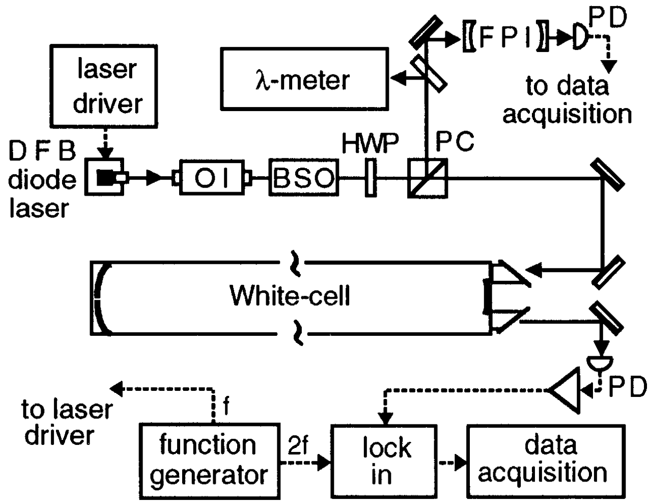

FIG. 2. Scheme of the apparatus used in this work for high-sensitivity laser spectroscopy of O_2. OI: optical isolator, BSO: beam-shaping optics, HWP: half-wave plate, PC: polarizing cube, PD: photodetector, and FPI: Fabry-Pérot interferometer.

wavelength-modulation detection technique. The laser frequency was modulated at a frequency $f = 40$ kHz by adding a modulation to the bias injection current of the diode. The output of the photodetector was sent to a lock-in amplifier and demodulated at a frequency of $2f$ in order to increase the detection sensitivity and to reduce a background signal due to the change of the laser intensity during the frequency scan. Optimum $2f$ signals were recorded with a modulation index of 1.75. The output signal of the lock-in was sent to a digital oscilloscope and to a personal computer for the data acquisition. In order to improve the signal-to-noise ratio, we averaged over several scans of the laser frequency. During the data acquisition, the laser frequency was monitored using the Fabry-Pérot interferometer. The change of the laser frequency was measured to be less than 20 MHz in a time of 100 s.

As already mentioned, the transitions we considered as a reference are weak magnetic-dipole transitions. The sensitivity achieved with the apparatus described above was high enough to observe, in addition to the lines of $^{16}O_2$, also those of the $^{16}O^{18}O$ and $^{16}O^{17}O$ molecules, with a total pressure in the cell as low as a few mbar. Figure 3 shows, for example, the signal recorded for the $^RQ(1)$ and $^RR(2)$ line of $^{16}O^{18}O$ and $^{16}O^{17}O$, respectively, in a natural abundance sample with a total pressure in the cell of 20 Torr. Historically, the detection of these weak lines in the atmospheric oxygen spectra led to the identification of the ^{18}O and ^{17}O isotopes of oxygen [28,31]. Because of the small natural abundance of these isotopes ($^{18}O = 0.2\%, ^{17}O = 0.04\%$), their weak lines provided an accurate test of the sensitivity of the apparatus.

The purpose of this work was to set an upper limit to the intensity of possible absorption lines starting from the wrong permutation antisymmetric states, with respect to the intensity of the normal transitions of $^{16}O_2$ involving symmetric states. The expected position of the transitions was calculated, with an uncertainty of less than 300 MHz, using the data available in the literature for the relevant molecular parameters [32]. In this work, we searched for the following transitions: $^RR(2)$ at 761.6740 nm, $^RQ(2)$ at 761.5626 nm, $^RR(4)$ at 761.3891 nm (vacuum wavelengths). Figure 4(a) shows the signal recorded as the laser frequency was scanned across the expected position (indicated by the arrow) of the missing $^RR(4)$ line. The total O_2 pressure in the cell was 200 Torr. The lock-in time constant was 100 ms, and we averaged over 10 frequency scans. In Fig. 4(b), the absorption signal for the observed $^RR(3)$ line of $^{16}O_2$ is shown for comparison, recorded in a single scan with a lock-in time constant of 1 ms. In this case, the pressure in the cell was reduced to 5 Torr in order to avoid a broadening of the line due to the optical depth of the sample. The laser modulation width was adjusted in order to keep the same value of modulation index. In Fig. 4(a) the vertical scale was expanded by a factor of 10^4 with respect to Fig. 4(b). The noise observed in Fig. 4(a) is due to the laser intensity noise and to small interference fringes caused by the residual reflectivity of the AR-coated optical components. The spectra recorded in this work showed no evidence of anomalous lines. By comparing the noise level with the intensity of the observed lines, an upper limit can then be deduced for the intensity of possible SP-violating lines with respect to the intensity of observed lines. After scaling the recorded signals for the different values of the pressure in the cell, and taking into account the correction due to the pressure broadening of the lines,

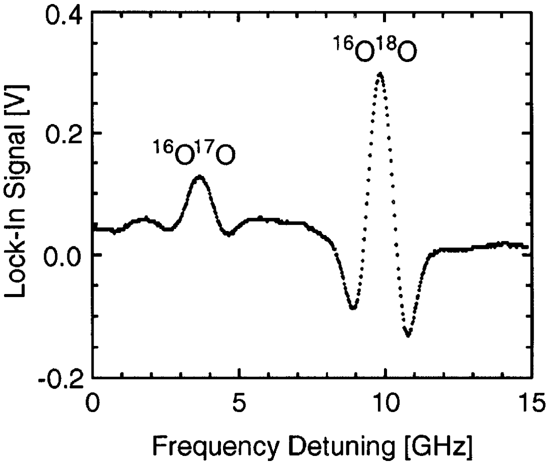

FIG. 3. Example of the detection of rare isotopes of oxygen with the apparatus used in this work. The signals shown correspond to the $^RR(2)$ line of $^{16}O^{17}O$ and to the $^RQ(1)$ line of $^{16}O^{18}O$ as observed in a natural abundance sample with a total pressure of 20 Torr in the cell.

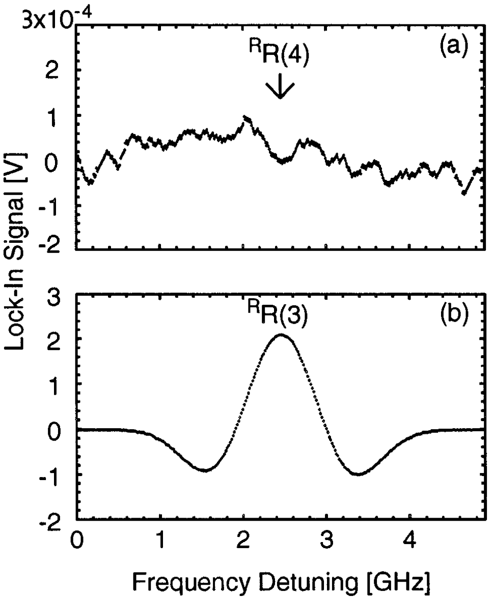

FIG. 4. (a) The $2f$ absorption signal recorded as the laser frequency was scanned over the region (indicated by the arrow) where the $^RR(4)$ SP-violating line would be observed (O_2 pressure $P = 200$ Torr). (b) Signal corresponding to the $^RR(3)$ transition of $^{16}O_2$ ($P = 5$ Torr). The vertical scale in (a) was expanded by a factor of 10^4 with respect to (b).

we deduce an upper limit of $\beta^2/2 \leq (5 \pm 2) \times 10^{-7}$ on a possible violation of the SP in this system. This can be considered the most stringent test of the SP for integer-spin particles reported so far. Reinterpreting the previous data on O_2 spectra as a test of the SP, the present result represents an improvement of at least 2 orders of magnitude.

As mentioned above, the possibility of violations of the SP and the interpretation of the experimental tests still pose problems from the theoretical point of view. However, accurate experimental data, as the one obtained in this work, are important to test the consistency of the present theory and to constrain new theoretical models. The accuracy of the data obtained in this work can be further improved by using higher sensitivity detection methods. A similar test can also be performed on different, not necessarily diatomic, molecular systems containing identical spin-0 nuclei.

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