Search for Small Violations of the Symmetrization Postulate for Spin-0 Particles

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An accurate test of the symmetrization postulate of quantum mechanics has been performed on the spin-0 ^{16}O nuclei, by looking for transitions in the CO_2 spectra involving states which are antisymmetric in the exchange of the nuclei. Using high-sensitivity laser spectroscopy, a bound of 2.1×10^{-9} has been set to a possible violation of such postulate, with an improvement of more than 2 orders of magnitude with respect to the previous limit. [S0031-9007(98)07778-3]

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The symmetrization postulate (SP) is at the basis of the quantum-mechanical description of the systems containing identical particles. As is well known, according to this postulate, the allowed states of such a system are only either symmetric or antisymmetric with respect to permutations of the particles labels. In the first case, the particles are called bosons and their distribution follows Bose-Einstein statistics, while in the second case they are called fermions and follow Fermi-Dirac statistics. Experiments indicate that half-integer-spin particles are fermions, and integer-spin particles are bosons. In addition, the spin-statistics theorem in relativistic quantum field theory provides a connection between spin value and statistics under appropriate assumptions.

Possible violations of the SP have been investigated both theoretically and experimentally. Theories going beyond Bose and Fermi statistics were first proposed in [1,2]. Recently, theories allowing for small violations of the SP have been developed based on deformed bilinear commutation relations [3] or trilinear commutation relations [4] instead of the usual Bose and Fermi commutation relations. In fact, experiments show that any violation of the SP would necessarily be small. In case of a SP violation, transitions between states of different permutation symmetry would be rigorously forbidden, at least within the framework of standard quantum mechanics. Therefore, a system including identical particles should be represented as an incoherent mixture of two different species, characterized by the two different permutation symmetries. As an example, in the case of a small violation of the Bose statistics, the two-particle density matrix should be written

$$\rho_2 = (1 - \beta^2/2)\rho_s + (\beta^2/2)\rho_a\,. \qquad (1)$$

A few experiments for accurate testing of the validity of the SP have been performed. In particular, the validity of the Pauli exclusion principle, that is, the consequence of SP for fermions, has been tested on electrons in copper [5] and in helium atoms [6]. In the first case, electrons were introduced in a copper sample, and x-ray emission accompanying transition to the $1S$ state of the copper atom, which is already filled with two electrons, were searched. In the second case, the test was performed on a much simpler system, looking for a transition involving the permutation symmetric $1s2s\,^1S_0$ state of electrons in helium. In these experiments, upper bounds of 1.7×10^{-26} and 0.2×10^{-6}, respectively, were set to a violation of the Pauli principle. In the case of bosons, a small violation of the SP would in general manifest itself as a small change in the properties of a multiparticle system, and therefore experimental tests are somewhat more difficult. In [7,8], the possibility of searching for lines forbidden by the SP in the spectra of molecules containing identical spin-0 nuclei was proposed. In [9,10], experiments were performed, by looking for electronic transitions in the O_2 spectrum between states which are antisymmetric in the exchange of the ^{16}O nuclei, which have nuclear spin $I = 0$. A bound to a possible SP violation was set to 5×10^{-7} in [9], and a similar, slightly less accurate result was obtained in [10]. Possible violations of the SP for photons have been also investigated [11–13].

In this Letter we report on a new accurate test of SP for spin-zero nuclei, which was performed by investigating the vibrational spectrum of the $^{12}C^{16}O_2$ molecule, searching for a nonzero population of SP-violating states. The carbon dioxide molecule has the same symmetry properties of oxygen but, since it is triatomic, it has strong active vibrational bands in the infrared, which are instead lacking for the biatomic O_2. In particular, the intensity of the combination band 12^01-00^00 around 2 μm wavelength is more than 2 orders of magnitude larger than the electronic transitions of oxygen previously investigated, and therefore the absorbance of a given population is correspondingly larger, resulting in an increased sensitivity for a possible SP violation. Although this band is weaker than the fundamental transitions in the infrared, it is particularly interesting, since it is the strongest that can be reached by available distributed-feedback (DFB) diode lasers, which are almost ideal sources for high-sensitivity spectroscopic detection.

By assuming the validity of the Born-Oppenheimer approximation, the total wave function of a single molecule can be decomposed in the usual way as $\Psi = \Psi_e \Psi_n \Psi_v \Psi_r$. The ground electronic wave function of the CO_2 molecule is symmetric in the exchange of the two ^{16}O nuclei, the nuclear one is symmetric, since $I(^{16}O) = 0$, and even the ground vibrational wave function is symmetric. Therefore the rotational wave function in the ground vibrational state must be symmetric, i.e., only even values for the rotational quantum number J are allowed by the SP. A similar argument applies for the excited vibrational level, in which the rotational wave function must be antisymmetric in the exchange of the nuclei, and therefore only odd values of J are allowed. As a consequence, assuming the validity of the SP, the R branch investigated in this experiment should be composed only of $R(2J)$ transitions (Fig. 1) [14]. The appearance of weak transitions of the form $R(2J + 1)$ could thus indicate a violation of the SP. The purpose of this experiment was therefore to look for some of these transitions, by means of high sensitivity absorption spectroscopy, in order to have quantitative information on the possible SP-violating population.

The experimental setup is shown in Fig. 2. The source was an InGaAsP DFB diode laser, emitting on a single mode around 2 μm, with an output power of 7 mW. The laser was driven by a low noise current source, and its temperature could be stabilized in the range 5–35 °C, resulting in a tunability of about 600 GHz. Two optical isolators were put just in front of the laser, in order to avoid excess amplitude noise due to optical feedback. Part of the radiation was sent to a 300 MHz Fabry-Perot interferometer, which provided a frequency marker. In order to avoid possible weak secondary modes of the laser, the main beam was directed onto a diffraction grating, whose first-order diffracted beam could be aligned to the center of a slit of adjustable width. The maximum resolution of such a frequency selector was about 100 GHz. The radiation passing through the slit could be aligned along several absorption cells of different lengths. For the high sensitivity measurement it was split into two beams of adjustable power ratio, by means of a half-wave plate and a beam splitter. After that one of them had interacted with the sample gas in a White-type multipass absorption cell, on a total path length of 130 m, the two beams were eventually focused onto two identical photodetectors by parabolic mirrors. In the design of the optical path, great care was devoted to the reduction of interference effects arising from multiple reflections between optical elements, which usually are the main source of noise in this kind of experiment. For such purpose, transmissive optics were avoided where possible along the laser path. In addition, a balanced detection technique was performed, i.e., the absorption signal was extracted from the difference of the signals from the two identical photodetectors. Provided the optical power impinging onto the detectors is equal, such a technique results in a cancellation of any classical amplitude modulation common to the two beams [15,16]. It was therefore possible to cancel out most of the fringe signal due to optical elements between the laser and the last beam splitter. In addition, the optical fringes originating within the multipass cell could be averaged out by modulating the optical path length with a motorized system. The intensity of the transitions allowed by SP was preliminarly measured by the help of a direct absorption scheme, on a relatively short path length (20–80 cm) obtained with a single pass cell. For each transition the line profile was recorded for a set of CO_2 pressures and integrated, in order to extract the line intensity S [17]. The position of the missing transitions was calculated from the molecular parameters [18] with an uncertainty smaller than 50 MHz, assuming that the Hamiltonian of the SP-violating molecules was the same as that of the ordinary ones. Unfortunately, due to the crowding of the spectrum, only two of them could be investigated, the others being within 1 GHz from transitions belonging to hot bands or rare isotopes. The chosen transitions were the $R(25)$ at 2.001 790 μm (Fig. 3) and the $R(33)$ at 2.000 015 μm.

The following step consisted in the measurement of the intensity of selected weak transitions in the neighborhood of the missing lines, in order to have an accurate scaling factor to compare the intensity of allowed and missing transitions. This intermediate measurement was performed with a 20 m Herriot-type multipass cell, with the help of

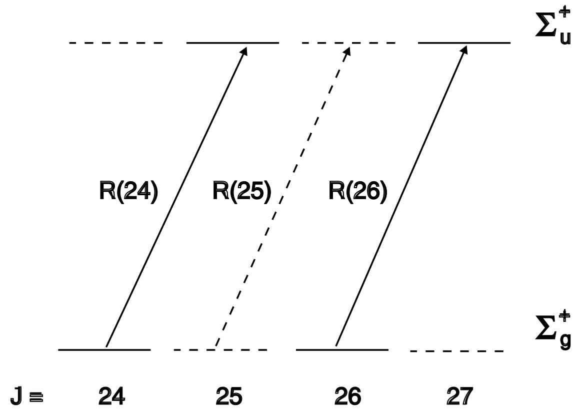

FIG. 1. Partial scheme of the energy levels involved in the R branch of the $12^01\text{-}00^00$ combination band of $^{12}C^{16}O_2$. The levels represented by dotted lines, and the corresponding transition, are those forbidden by the SP.

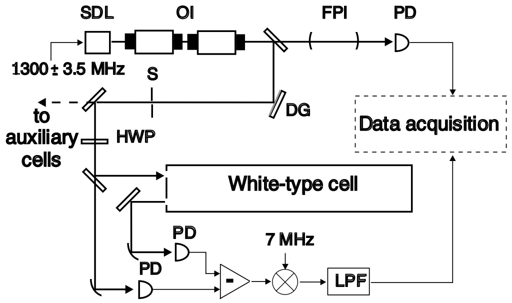

FIG. 2. Experimental apparatus used for the high sensitivity investigation. SDL: DFB diode laser; OI: optical isolator; FPI: Fabry-Perot interferometer; PD: photodiode; DG: diffraction grating; HWP: half-wave plate; LPF: low-pass filter.

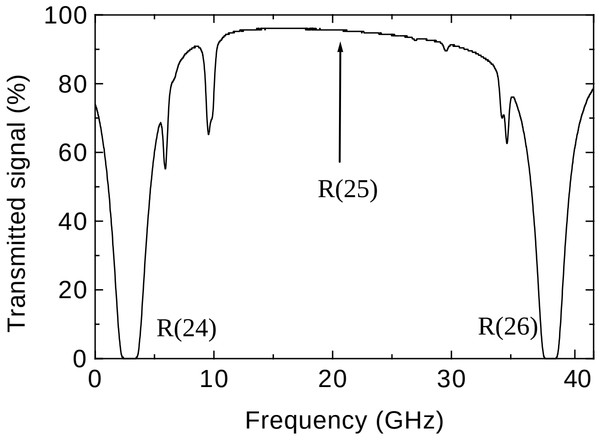

FIG. 3. Direct absorption spectrum of one of the portions of the 12^01-00^00 combination band of $^{12}C^{16}O_2$ investigated in this experiment. The weak lines belong to hot bands or other isotopes of CO_2. The calculated position of the SP-violating $R(25)$ is indicated by an arrow. The spectrum was obtained with 2.5 Torr of gas on a 130-m path length.

both direct absorption and two-tone frequency modulation (TTFM) techniques. As a result, the intensity of weak transitions, such as the two shown in Fig. 4a could be determined with a relatively low uncertainty. For example, for the $P(56)$ $^{18}O^{12}C^{16}O$ transition we obtained $S = (7.5 \pm 1.6) \times 10^{-27}\ \mathrm{cm\,mol^{-1}}$, which is more than 5 orders of magnitude weaker than the $R(24)$ transition of $^{12}C^{16}O_2$, for which $S = (1.1 \pm 0.1) \times 10^{-21}\ \mathrm{cm\,mol^{-1}}$. In exploring the whole spectral range covered by the laser, several transitions of medium intensity ($S = 10^{-28}$–10^{-25}) not assigned in literature were observed. Most of them could be grouped in three different bands, and all the experimental observations about the Doppler width, pressure broadening coefficient, and the spacing between the lines seemed to assign them to a symmetric isotope of CO_2. Although none of them were in coincidence with the calculated SP-violating transitions, an additional investigation was carried out, by measuring the scaling of the intensity of such lines for decreasing temperatures of the sample. Actually, the intensity scaled by a factor larger than 5 by changing the temperature from 300 to 220 K, indicating that the ground state of the unassigned bands is not the vibrational ground state. Although we could not identify these lines as any of the known CO_2 bands, our tests excluded any connection with possible SP-violating lines.

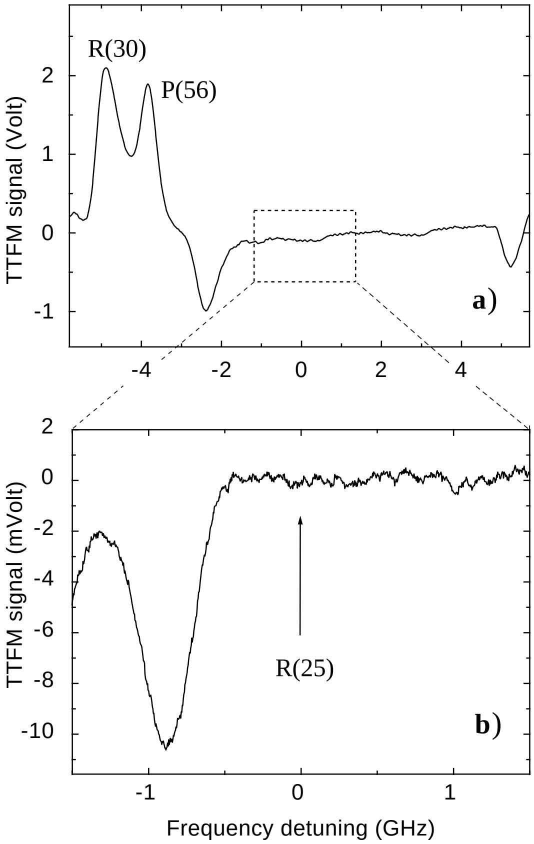

FIG. 4. TTFM spectra recorded around the position of the SP-violating $R(25)$ transition; the region highlighted in (a) is shown in (b) with increased sensitivity (note the different ordinate scales). The $P(56)$ transition, which has been used as a reference for the intensity, is a factor of 1.5×10^5 weaker than the $R(24)$. The spectral feature appearing in (b) is the second sideband of the $P(56)$ line, not visible in (a).

The central part of the experiment was a high-sensitivity search for the missing lines, using a 130 m path length. A balanced two-tone frequency modulation technique was used, according to the scheme described in [16]. The modulation frequencies were (1300 ± 3.5) MHz, and the detection was performed at 7 MHz. While the laser was scanning over a certain frequency range with a repetition rate of 2 Hz, the demodulated absorption signal was filtered in a 30 Hz band, amplified, and sent to a digital oscilloscope. The effective detection bandwidth could be reduced by averaging a large number of scans. The maximum acquisition time was 10 min, corresponding to approximately 500 scans, limited by a slow drift of the laser emission frequency with respect to the absorption profiles. The sensitivity of the spectrometer, i.e., the minimum detectable absorbance was estimated to be $(5 \pm 1) \times 10^{-7}$, a factor of 50 above the shot-noise limit, calculated for a laser power of 100 μW on each detector (the reduction of the available laser power was mostly due to losses in the coupling of the radiation to the multipass cell). The CO_2 pressure in the cell was set to 30 Torr, which, taking into account the pressure broadening of these lines, gives the maximum sensitivity for our detection method [16]. The TTFM signal consisted of various replicas of the absorption profile, spaced by 1300 MHz, with intensities scaling as a function of the modulation index. Such replicas were used to further scale the intensity measurement, down to the noise level. The result of this procedure is shown in Fig. 4, in which two recordings of a frequency scan around the expected position of the $R(25)$ line, obtained with increasing sensitivity, are reported. As is apparent in Fig. 4b, no signal could be distinguished from the noise at the expected position. A bound to the intensity of the $R(25)$ transition was therefore set by the rms noise, which was estimated to be a factor 3.1×10^3 smaller than the intensity of the $P(56)$ transition. By scaling back to the intensity of the allowed $R(24)$ transition and taking into account the small corrections due to the Boltzmann distribution of population and the degeneracy of the levels, a bound of $\beta^2/2 \leq (2.1 \pm 0.7) \times 10^{-9}$ to the relative population of the SP-violating state could be deduced. The measurement was repeated for the $R(33)$ transition and no signal was detected at the expected position. The achieved sensitivity was in this case a factor of 2 lower, due to the reduced statistical weight of the lower state. In conclusion, the experiment described here provided a stringent test of the SP (and/or of the spin-statistics connection) for ^{16}O nuclei with an improvement of more than 2 orders of magnitude with respect to previously existing data. Such an improvement was possible because a different molecule, with much stronger absorption lines, was chosen as the test system.

Further improvements are possible: the sensitivity could be increased by reducing the noise to the quantum level, by increasing the absorption path length, or by selecting stronger transitions. The fundamental vibrational band of CO_2 around 4.3 μm, which is at least a factor of 1000 stronger than the one at 2 μm, could be investigated with lead salt diode lasers, as originally proposed in [7], although the presence of a larger number of weak bands could place a severe limitation to the achievable sensitivity. A dramatic increase in the detection sensitivity can be obtained by combining the long absorption paths in high-finesse Fabry-Perot cavities and high-sensitivity detection schemes [19]; an experiment on O_2 is in progress [20]. A similar test can be performed on other spin-0 nuclei; a straightforward extension is the search of SP-violating lines in the spectrum of $^{18}O_2$ [21]. An interesting prospect is the investigation of spectra of molecules containing more than two identical nuclei, such as OsO_4, that would allow one to test the validity of the SP in cases where more complex symmetry relations can be involved. Recent experiments on Bose-Einstein condensation in dilute gases of trapped atoms [22] might also give new insight into possible deviations from conventional statistics [23].

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