## Search for Small Violations of the Symmetrization Postulate in an Excited State of Helium

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(Received 24 January 1995)

We have searched for the existence of the permutation symmetric 1s2s S state of helium in an atomic beam. Such a state directly violates the symmetrization postulate (SP) of quantum mechanics and implies a breakdown of the Pauli exclusion principle. Our data constrain recent SP-violating models at the 5 ppm level. This is the first experiment to look systematically for an SP-violating state with no multiple occupancy of a quantum state.

PACS numbers: 31.10.+z, 13.90.+i, 32.10.-f, 32.20.-r

It has long been known that, with ad hoc restrictions, quantum field theories admit violations of the Pauli exclusion principle (PEP) and the more general symmetrization postulate (SP) while preserving the fundamental indistinguishability of identical particles [1-4]. Even before the pioneering mathematical work of Green in 1953 [5], Pauli, Dirac, and Heisenberg were aware of this possibility, and drew attention to it explicitly [6-8]. Recent work has produced theories that allow for small but nonzero PEP violations by using variations of the trilinear commutation relation of Green and Volkov [5,9-14] or bilinear Bose and Fermi commutators [15-18]. Although the above theories, and other approaches [19-21], are fully consistent with ordinary quantum mechanics, attempts to consistently retain small violations in the general quantum field theoretical framework have been forced to relax ordinary assumptions concerning locality or the existence of states with negative squared norms. We therefore discuss the present work in the framework of ordinary quantum mechanics as analyzed by Greenberg and Mohapatra [14]. This later paper, unlike their original work [13], provides an internally consistent framework for interpreting our measurements.

Surprisingly, relatively few experiments have been performed to test PEP to high precision [14]. This is due in part to the existence of a superselection rule which makes many apparent tests of PEP insensitive [22]. Recent valid experiments have been undertaken to infer high precision limits on a PEP violation by studying the behavior of condensed matter systems [23-25] and by mass spectrometric analysis of gases [26,27]. Of particular importance is the experiment of Ramberg and Snow [23] which, under certain assumptions, bounds a violation of the exclusion principle in copper by less than  $1.7 \times 10^{-27}$ . Also, an ongoing experiment by Hilborn [28] uses a molecular spectroscopic method. Most of these experiments have reported much more severe limits on possible violations of PEP than we present here. As work on CP violation has amply demonstrated, however, the choice of which physical system to search for a discrete symmetry violation can be critical. We believe that ultrahigh precision tests on complex systems can be complemented by lower-precision tests on simple systems such as that described in this paper.

A two-fermion system such as helium represents the simplest system in which SP violation is possible. We have chosen a metastable state where one electron is in the ground state and the other electron is in a lowest excited state of its spin symmetry class, the  $1s2s^{1}S_{0}$ . Both the space and spin parts of the wave function are antisymmetric; therefore, the total wave function is symmetric, in violation of SP. We introduce the notation  $[1s2s]_{A} {}^{1}S_{0}$  (where *A* refers to the antisymmetric spatial wave function) to distinguish the PEP-forbidden state from the normal state. In the nonrelativistic picture, SP-forbidden states of helium have opposite spin symmetry from the allowed states (triplets and singlets interchanged). Thus the SP-allowed and -forbidden states are nearly degenerate.

Precise calculations of the so-called "paronic" states which violate SP have been published for the case of helium by Drake [29]. Indeed, helium is the *only* system of fermions for which detailed calculations are available. These calculations can be unambiguously carried out and tested to high precision by comparison with the wellstudied spectrum of ordinary helium, since the zerothorder spatial wave functions are identical in both cases [30]. This simplification is specific to helium because there are only two irreducible representations of the permutation group for two-fermion systems [31].

The spectral line under investigation in our work arises from the predicted paronic states shown in Fig. 1. Within the theoretical framework of Greenberg and Mohapatra [14], the magnitude of the SP-forbidden line relative to the allowed lines gives the probability  $\beta^2/2$  of violating the PEP/SP. Paronic states for half-integral spin particles, like those in our experiment, correspond to mixed or symmetric representations of the permutation group [1,14]. They are generally multidimensional, and are always orthogonal to the antisymmetric representation [31,32]. The existence of the superselection rule prevents the occurrence of a small coherent admixture of states with the "wrong" symmetry and, therefore, leads to an ensemble picture appropriately described by a density matrix rather than a pure state. In this picture, the probability of detecting an SP-violating



FIG. 1. Partial level diagram of helium indicating the relative position of the paronic states under consideration.

state in our experiment is simply a measure of the relative abundance of paronic helium atoms in the sample.

Although theoretically appealing, helium poses a number of experimental challenges. The spectral line shifts due to a violation of SP are caused only by interchanging the singlet and triplet spin wave function. This guarantees that the total,  $[1s2s]_A S_0$ , wave function is symmetric. As a result, the terms related to the nonrelativistic energies do not change, and only small shifts in energies due to spindependent terms in the Breit interaction and the anomalous magnetic moment in QED corrections will be present [29]. These shifts are so small that in the absence of Doppler-free techniques, the SP-violating lines we study here would be entirely obscured by thermal broadening at room temperature. Although the largest shifts are at the lowest n quantum states of helium [29], the experiment has to be performed on the first excited state in order to employ modern laser spectroscopic techniques.

Our apparatus is shown schematically in Fig. 2. The atomic beam was that used in a previous work [33,34], modified to improve the collimation to 1.7  $\mu$ sr. The beam of atoms traveled through an interrogation region where they were subjected to irradiation by photons from a frequency-doubled single-mode Ti-sapphire laser. The laser was tuned to the  $2^{3}S_{1}-3^{3}P_{1}$  transition at 389 nm to cause the atoms to fluoresce. A precision retroreflector was used in combination with spectral scans to determine that the laser and atomic beams were orthogonal to each other. The residual Doppler width was approximately 8 MHz. Emitted photons were detected with a high-efficiency, ultraquiet configuration of optics and electronics described elsewhere [34,35]. Current induced on a stainless steel plate at the end of the atomic beam line measured the total flux of metastable atoms. With a triplet/singlet ratio of 2.7, measured by flash-lamp quenching, and assuming a detector efficiency reported by Dunning, Rundel, and Stebbings [36], the estimated triplet state flux is  $3 \times 10^8$ /sec.



FIG. 2. Schematic diagram of the apparatus.

Paronic matter should have an entirely different chemistry than ordinary matter and may not therefore be mixed in with ordinary samples of a particular element. Because of the ability to accept another electron into an ordinarily filled shell, for example, the paronic helium atom (except spin and magnetic properties) will be similar to hydrogen in chemical reactivity. Thus it is important that the atoms be ionized and reformed before entering the laser beam. Prior to injection of the atoms into the beam, the excited helium atomic states are formed by ionization and recombination in a dc plasma discharge operated at a pressure of  $1.5 \times 10^4$  Pa. Calculations show that the ionization rate is such that a helium atom in the discharge will very likely be ionized before it can escape into the atomic beam. Each ion will generally recombine with a new electron; if "forbidden" symmetries can be formed at all, they would presumably be formed in this recombination process with some probability. Subsequent collisions in the plasma leading to neutral excitation will preserve symmetry. Wall collisions are more likely to adsorb ground state atoms in the polarizable forbidden symmetry, but as long as any atom is ionized before directly entering the atomic beam, the relative abundance of a symmetry in the beam will be equal to its probability of formation in recombination. This circumstance removes the requirement of a knowledge of the chemical properties and history of the SP-violating states on which other experiments have depended [14].

A continuous scan over the  $2^{3}S_{1}-3^{3}P_{1}$  region of the spectrum is shown in Fig. 3. The predicted location of the SP-forbidden line is indicated by an arrow. Note that the vertical axis is greatly magnified in the inset. This spectrum was taken with a time constant of approximately 1 sec and a scan time of 100 sec. In these data, we obtained a signal-to-noise ratio (S/N) of about



FIG. 3. Fluorescence spectrum scanned over the region of interest. The inset shows a greatly magnified view of the region of the spectrum which would produce a line if PEP were violated.

10000. An additional order of magnitude in S/N is obtained by extended counting in the region of interest, as described below.

Data were accumulated for extended periods of time by two methods. In the first method, the influence of background light from the atomic beam discharge source was eliminated by chopping the incident laser light and using lock-in detection methods. Photon counting was employed in order to maintain the noise suppression advantage of lower-level discrimination. In this case, the photon-counter time interval was set at 1 msec, and the digital counts were converted to an analog voltage which was fed into a lock-in amplifier. The lock-in time constant was set to 10 sec and the output data were recorded for 100 sec. At the end of a 100 sec data collection period, the mean value of the lock-in output was computed. The laser was then shifted to another frequency and an additional 100 sec of data were collected. The results of such a digital scan over the region of interest, with linear subtraction of the background, revealed no indication of any forbidden line. The background in this case was limited by stray light from the laser. The noise appeared to be limited by short-term laser amplitude drift. Operating the laser under a laminar flow hood was helpful in minimizing this problem.

In order to stabilize the laser for extended periods of time, it was frequency locked to the allowed  $2^{3}S_{1}-3^{3}P_{1}$  line in a low-pressure helium discharge using the FM technique [37,38] in combination with Doppler-free saturated absorption. A precise offset from the lock point

was obtained by using an acousto-optic shifter between the laser and the lock apparatus, as shown. This not only stabilizes the laser against long-term frequency drift but also provides an absolute wavelength reference point. The offset corresponding to the predicted SP-violating line searched for here is 535 MHz to the blue [29].

In the second method of collecting data, photons were simply counted for 100 sec, and the result was manually recorded before shifting the laser to a new frequency where the process was repeated. The results of such a digital scan, over and back down the region of interest, is shown in Fig. 4. A background of approximately 8000 counts/sec (primarily from stray light from the atomic beam discharge) has been subtracted with a quadratic fit as a function of time. Initially, a peak of 25000 counts/100 sec was observed at precisely the predicted location for an SP violation; but this measured value did not reproduce, and subsequent examination of the data collection records showed that the laser lock was quite weak during the initial anomalously large reading. A failing laser lock could introduce spurious amplitude noise and be the cause of the initial anomalously large reading. For this reason, we do not believe that this data point should be considered significant. We did not observe any other anomalous peak in any other region of the scans taken during this entire series of experiments.

Without including the anomalous point mentioned above, the rms noise level in our second method is  $2 \times 10^5$  times below the peak signal of the SP-allowed line. From this data set, we infer a value for the dimensionless PEP/SP violation coefficient  $1/2\beta^2 = (0.2 \pm 5.0) \times 10^{-6}$  where the uncertainty represents our background noise at the one standard deviation level.

At sensitivities 5 times below those reported here, the far-wing tail of the natural line profile of the ordinary helium atoms will begin to obscure any faint SP-violating



FIG. 4. Precise digital scan of the region of interest. The arrow indicates the predicted location of the PEP violation.

line. Even this seemingly fundamental problem can be effectively overcome by using subnatural linewidth techniques such as photon burst spectroscopy (PBS) that we originally proposed for this work [39,40]. PBS not only increases the level of discrimination, but it also provides single-atom sensitivity [41–44]. These two combined advantages could increase the limit on SPviolating states in helium presented here by 5 orders of magnitude or more.

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