X rays test the Pauli exclusion principle

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Abstract
Since the publication of the models describing a small violation of the Pauli exclusion principle (PEP) there has been an explosion of world-wide interest in PEP tests and related theories.

PEP forbids an atom to have more than 2 electrons in the K-shell. If PEP is slightly violated, a third electron can occasionally join in. This would result in an anomalous X-ray emission. A high-sensitivity experiment places an upper limit of the order of \(10^{-26}\) on the PEP violating parameter.

I will outline the main theoretical and experimental ideas in this new exciting area.

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I. INTRODUCTION

The Pauli exclusion principle (PEP) is one of the pillars of physics and chemistry and we know from history that pillars can crack. The most familiar examples are parity and CP violation. For this reason alone we should be willing to look into possible PEP violations. But what are other, more specific reasons for looking into it? The main reason is that our theoretical understanding of the PEP origin is still not quite satisfactory. Perhaps the best way to illustrate it is to quote Feynman (1965): “Why is it that particles with half-integral spin are Fermi particles whereas particles with integral spin are Bose particles? We apologize for the fact that we cannot give you an elementary explanation. This probably means that we do not have a complete understanding of the fundamental principle involved.” Similar dissatisfactions were expressed by Pauli (1955) and Dirac (1981).

One way to look for more understanding is to try to think what happens if the principle is violated. The 3 main questions to be considered (of course, they are interrelated) are:

Can we make up a theory that would deviate from PEP by a small amount?
How to look for possible PEP violations experimentally?
What number characterises the accuracy with which PEP holds?

The last question in fact shows how different PEP is from other similar principles, for example CPT symmetry. For CPT symmetry there is a well defined parameter that describes possible deviations from CPT such as the mass difference between a particle and antiparticle. As for PEP, we had not had such a parameter until we started to make up theories that would be able to describe its violation and the theories then showed us how to introduce such a parameter. The purpose of this talk is to give an idea of what the answers to these 3 questions we have at the moment.

What are other differences between PEP violation and the violation of more familiar symmetries such as CP? In the case of CP violation we can write a lagrangian which is CP violating and this is a pretty straightforward procedure in QFT. In other cases we can write a lagrangian that is symmetric under some operation and then add a small part to break down the symmetry. But in the case of PEP violation we cannot write a PEP violating lagrangian. PEP can be violated only through the commutation relations and not through the lagrangian. So this was really quite a new situation to consider and for that reason it was rather challenging: “you can’t be a little bit pregnant”.

There is one question that arises almost always in discussions of PEP violation: what about the spin-statistics theorem? Doesn’t this theorem rule out the possibility of PEP violation straight away? The answer to this question is no. The spin-statistics theorem of the axiomatic quantum field theory forbids quantising the fields with half-integer spin according to Bose, that is with commutators. This theorem leaves open the question of possible alternative ways of quantising the spin one-half fields, in particular it says nothing about those new ways which could lead to small PEP violations. A recent review of attempts to derive PEP from other principles of quantum mechanics is given by Kaplan (2002).
No theoretical models of small PEP violation existed up until 1987. Although non-standard types of statistics, such as parastatistics (Green, 1953), had been known, they could be viewed as “100% violation” of PEP rather than small violation. This kind of violation for electrons and nucleons was clearly ruled out by experiment. Also, there were studies of possible small violation of electron identity (Luboshitz and Podgoretskii, 1971; Okun (1989); see also Fischbach et al., 1968 and Fermi, 1933). Although related to small PEP violation, small non-identity is a different idea.

This is a brief outline of a large and growing area. In particular, the ten-page limit does not allow for complete referencing. More detailed reviews and further references can be found in the Proceedings of the Conference “Spin-statistics connection and commutation relations” (Hilborn and Tino, 2000). To the extent allowed by the material, the present review is complementary rather than overlapping with the previous ones. An extensive bibliography with hundreds of references has been compiled by Gillaspys and Hilborn (2000).

II. THEORIES

Let us consider several formulations of the Pauli exclusion principle which we use today and which are rather different from the original formulation given by Pauli in 1925: “There can never be two or more equivalent electrons in an atom. These are defined to be electrons for which . . . the values of all quantum numbers . . . are the same.”

The second formulation came with the advent of Quantum Mechanics in our modern version. This formulation is due to Dirac and it states that the wave function of the system of electrons must be antisymmetric under exchanges and permutations of electrons. In the simplest case we have to require that the wave function of two electrons is antisymmetric with respect to the exchange of their space and spin coordinates.

Finally, the most general formulation of PEP is given by the second quantisation formalism which is used in quantum field theory to describe electrons and positrons that can be created and annihilated. The formulation in terms of second quantisation concepts requires that the fields describing the electrons, positrons or any other particles with half-integer spin obey anticommutation relations of the form $AB + BA = 0$ or another c-number. The main thing is that we have here plus sign rather than minus sign when we interchange $A$ and $B$ operators. The minus sign would correspond to Bose commutation relations.

Logically, the third formulation is the most general and it implies the second formulation which in turn implies the first one. Because it is the most general, it is the best formulation to work with although it sounds a bit more abstract that the other two.

In summary, the validity of the Pauli principle follows automatically from the form of the anticommutation relations between the operators of the electron-positron fields $\psi(x)$, $\bar{\psi}(x)$ or, which is equivalent, between the creation and annihilation operators of the electrons and positrons $a_{k\sigma}, a_{k\sigma}^\dagger, b_{k\sigma}, b_{k\sigma}^\dagger$. 
Therefore in a theory, describing the violation of the principle the anticommutation relations should certainly be changed. Apriori it is not clear at all in what form one should cast new commutation relations.

Generally speaking, to solve this problem one could reason as follows: consider various sets of the commutation relations between the creation and annihilation operators $a_{k\sigma}, a_{k\sigma}^\dagger, b_{k\sigma}, b_{k\sigma}^\dagger$ or fields $\psi(x), \bar{\psi}(x)$ of the most general form. One should consider not only bilinear relations, but also trilinear ones and so on, each set containing not necessarily one but, in general, several independent relations.

Furthermore, one should require that these relations satisfy a number of general principles of the quantum field theory: positivity of energy, Lorentz invariance, relativistic causality, the electric charge (fermionic number) conservation, and C-parity. As for the last four principles, one should not require that these principles be absolutely valid, it suffices to require that their possible violations were at most of order $O(\beta)$ (where $\beta$ is the Pauli principle violating parameter) so that when $\beta \to 0$ all these violations were unobservably small.

In this way one could in principle find all the allowed commutation relations (or prove that they do not exist) containing a small parameter $\beta$ and resulting in the usual fermi-statistics of the electrons when $\beta \to 0$.

In practice, however, this way seems intractable. A simpler method consists in the following (Ignatiev and Kuzmin, 1987). Instead of searching for the algebra of the operators, let us try to construct first the representation of this algebra possessing the necessary property and after that try to find the commutation relations themselves. It is more convenient to work with the creation and annihilation operators than with the field operators. Let us further simplify the problem by discarding the momentum and spin variables and considering only electron (but not positron) operators.

Thus, we should find representation of the creation and annihilation operators $a, a^\dagger$ depending on the parameter $\beta$ so that when $\beta$ tends to zero this representation goes over into the usual Fermi representation

$$a_F = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad a_F^\dagger = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \tag{1}$$

As the orthonormal basis here we take the vacuum $|0\rangle$ and the one-particle state $|1\rangle$. Evidently, the minimal dimensionality of the state space we are looking for is three, so we choose as the basis of that space the states $|0\rangle$ (vacuum), $|1\rangle$ (one-particle state) and $|2\rangle$ (two-particle state). Suppose that the action of the creation and annihilation operators is defined as follows (the parameter $\beta$ is supposed to be real):

$$a^\dagger|0\rangle = |1\rangle \quad a|0\rangle = 0$$
$$a^\dagger|1\rangle = \beta|2\rangle \quad a|1\rangle = |0\rangle$$
$$a^\dagger|2\rangle = 0 \quad a|2\rangle = \beta|1\rangle. \tag{2}$$
Then the matrices of these operators in the chosen basis take the following form:

\[ a = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & \beta \\ 0 & 0 & 0 \end{pmatrix}, \quad a^\dagger = \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & \beta & 0 \end{pmatrix}. \quad (3) \]

The Hilbert state space \( H \) can be decomposed into the direct sum of the subspaces \( H_2 \) (built on the vectors \(|0\rangle, |1\rangle\)) and \( H_1 \) (built on the vector \(|2\rangle\)). It is clear that if \( \beta = 0 \) the transitions between the states in \( H_2 \) and \( H_1 \) become forbidden so that the space \( H_1 \) gets completely decoupled from \( H_2 \).

Now, let us construct the commutation relations (i.e., the algebra) which is satisfied by the operators \( a, a^\dagger \). To do that, one should calculate various products of the operators \( a, a^\dagger \) of the form \( a^2, a^\dagger a, aa^\dagger, a^3 \) etc. and then find the relations between them (such relations should certainly exist because there are only 9 independent \( 3 \times 3 \) matrices).

First, it can be shown that the operators \( a, a^\dagger \) and their bilinear products are linearly independent, i.e. the bilinear commutation relations are absent in the model under consideration.

Next, the trilinear relations do exist and can be written, for example, in the following form:

\[ a^2a^\dagger + \beta^2a^\dagger a^2 = \beta^2a \]
\[ a^2a^\dagger + \beta^4a^\dagger a^2 = \beta^2aa^\dagger a, \]

plus their Hermitean conjugate relations. To these relations one should add the equalities

\[ a^3 = 0, \quad (a^\dagger)^3 = 0. \]

Thus the equalities (4) - (6) complete the construction of our algebra.

Next let us find the particle number operator \( N \) in this model. In the chosen representation the operator \( N \) has the form

\[ N = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix} \]

so that the usual commutation relations hold true

\[ [N, a] = -a, \quad [N, a^\dagger] = a^\dagger. \]

Is it possible to find the bilinear expression for the operator \( N \) in terms of the creation and annihilation operators? The positive answer is given by the form

\[ N = A_1 a^\dagger a + A_2 aa^\dagger + A_3 \]

(9)
with the coefficients $A_i$ given by:

$$A_1 = \frac{-1 + 2\beta^2}{1 - \beta^2 + \beta^4},$$
$$A_2 = \frac{-2 + \beta^2}{1 - \beta^2 + \beta^4},$$
$$A_3 = \frac{2 - \beta^2}{1 - \beta^2 + \beta^4}. \quad (10)$$

Thus we have completed the construction of the algebra of the creation and annihilation operators and also have found the bilinear expression for the particle number operator (Ignatiev and Kuzmin, 1987).

Fundamental mathematical properties of the IK algebra have been studied by Biedenharn et al. (1989) (see also Ignatiev, 1990) and Cougo-Pinto (1993). These studies revealed interesting connections with such concepts as Jordan pairs, $C^*$-algebras, and quantum groups.

The problem of a realistic generalization of the one-level IK model can be formulated as the following question: how to write down the commutation relations if we ascribe the momentum and spin indices to the creation and annihilation operators? One way to do this was suggested by Greenberg and Mohapatra (1987, 1989a). The main problem is to make sure that no more than 2 electrons can occupy the same state. In the 1-level model that was achieved by requiring $a_3^3 = 0$. However, in the multi-level model we have infinitely many possibilities to put 3 electrons in the same state because we can “sandwich” other electrons between them. These sandwiched states turn out to have negative norms (Govorkov, 1989, 1983; Greenberg and Mohapatra, 1989b). No way out of this difficulty has been found and it is believed to be a fatal flaw. In principle, the possibility of curing this theory cannot be completely ruled out. A well-known example is the ordinary QED which involves negative-norm states that are harmless.

Another generalisation was attempted by Okun (1987) who assumed that the operators from different levels obey the usual anticommutation relations. This model also ran into serious difficulties discussed by the author.

We would like to stress that the “small PEP violation” is an intuitive concept and one can try to formalise it in many different ways leading to different theories with their specific experimental predictions.

In particular, the theory called “quons” was proposed in (Greenberg, 1990, 1991; Mohapatra, 1990; Fivel, 1990, see also Biedenharn, 1989 and Macfarlane, 1989). This theory is based on bilinear rather than trilinear commutation relations:

$$a_k a_i^\dagger - qa_j^\dagger a_k = \delta_{kl}. \quad (11)$$

The main physical feature of the model is that there is no limit on the number of particle that can occupy the same state, i.e., all types of Young tables are allowed for a system of quons. A review of other properties of quons have been given in (Greenberg, 2000, see also Chow and Greenberg, 2001).
Cosmological consequences of possible PEP violation for neutrinos have recently been discussed by Dolgov and Smirnov (2005); Dolgov et al. (2005); see also Cucurull et al. (1996).

III. EXPERIMENTS

The physics community, both theorists and experimentalists, took up the idea of small PEP violation with great enthusiasm. *Scientific American* published an article (Kinoshita, 1988) entitled “Roll Over, Wolfgang?” that described our work and the subsequent theoretical development by Greenberg and Mohapatra as well as the plans to look for small PEP violations experimentally. These experimental efforts were ongoing soon after the appearance of the theory papers.

It is interesting to compare the number of papers published on the subject of small statistics violation before and after 1987. During 30 years prior to 1987 there were about a dozen of papers on the topic including pioneering works by Reines and Sobel (1974), Logan and Ljubicic (1979), Amado and Primakoff (1980) and Kuzmin (1984) while since 1987 the number of papers has grown to several hundreds.

We are now coming to our main Question 2: How to look for small PEP violation in experiment? There are plenty of experiments of different kinds which have tried to look for deviations from PEP and it is hard to mention all of them, but most of them are based on one of two broad ideas.

The first idea, originated by Reines and Sobel (1974) (see also Goldhaber and Scharff-Goldhaber, 1948), is to look for forbidden transitions to the levels occupied by particles such as electrons or nucleons. So we can look for anomalous transitions to filled shells. As a result, the energy would be emitted in the form of X rays, gamma rays or something else.

A more recent idea is to look for these anomalous states themselves (“the integral method”): for example, we can try to look for atoms with 3 electrons in the K shell. The first experiment of this kind was proposed in (Ignatiev and Kuzmin, 1988; Gavrin et al., 1988). The integral method was further developed by Novikov and Pomansky (1989), Novikov et al. (1990), and Nolte et al. (1991). Similar ideas were discussed by Okun (1987, 1988).

Let us sketch the first type of experiments. We have here 2 levels and one electron is on the upper level while another is on the lower level. Of course, the upper electron can go down to the lower level provided the proper selection rules are observed and to emit radiation, e.g. X rays when the lower level is the K shell of an atom. This is an allowed transition.

Now, if there are 2 electrons with opposite spins on the lower level then all states on this level are occupied if this is a K shell. Then the upper level electron cannot go down to the lower level and this is a forbidden transition in the standard theory.

Next let’s take a look at the theory where PEP is slightly violated. Then this same
transition to the fully occupied K shell can now be allowed and the emitted radiation can be a signature of this process so that we can try to look for X rays, for example, as a sign of PEP violation.

The concrete realisation of a type 1 experiment (which takes into account the Amado-Primakoff (1980) arguments) is the experiment performed by Ramberg and Snow (1990) at Maryland University. This is a remarkably simple, table-top kind of experiment. It has a thin copper plate through which a large electric current is sent and close to this strip of copper there is an X ray detector whose task is to detect the emission of X rays when the transition occurs to the K shell of a copper atom. The experiment shows that there are no anomalous X rays in this case and the upper limit can be deduced on the PEP violating parameter \( \beta^2 \). This limit turns out to be exceptionally low: \( \beta^2 \leq 1.7 \times 10^{-26} \).

An improvement of this limit by 4 orders of magnitude is currently being planned by the VIP collaboration (2005) which builds on the success of DAFNE Exotic Atom Research program (DEAR) completed in 2003. The plan is to utilise the excellent X-ray detector involved in that program for the new purpose of the PEP violation search. The VIP setup will be first transported and installed at the Gran Sasso low-background laboratory (LNGS); then (in 2005-2006) the data taking will proceed, alternating between periods of current on ("signal") and off ("background").

So far we were considering type 1 experiments that were looking for anomalous transitions accompanied by radiation. Now let’s have a look at the alternative scheme where the search is for the anomalous states themselves such as, for example, anomalous elements. The first question that we have here is: what elements could make good candidates for such a search?

One approach to answering is to consider chemical differences between the ordinary element with the atomic number \( Z \) and the anomalous one with the same atomic number which we denote by a prime \( Z' \). We know that the chemical properties of the elements are controlled by the number of valence electrons, i.e. the number of electrons in the outermost shell of the atom.

Now, the element \( Z' \) has one valence electron less than the ordinary element \( Z \) because one electron in \( Z' \) goes down to the K shell. For light elements (roughly, \( Z < 20 \)) the preceding element \( Z - 1 \) in the periodic table also has one valence electron less than \( Z \). From that we conclude that the chemical properties of the light elements with the anomalous K shell structure (i.e., having 3 electrons in it) are similar to the chemical properties of the ordinary element with the atomic number \( Z - 1 \), the previous element in the Mendeleev table.

To make this idea more transparent, let’s consider a specific example and let’s take neon as our \( Z \) element (\( Z = 10 \)), then our \( Z - 1 \) element will be fluorine (\( Z = 9 \)). Neon has 1s, 2s, 2p shells all completely filled and if one electron from 2p shell goes down to 1s shell violating the Pauli principle, then we’ll have one electron less in the 2p shell. Let’s now take a look at the electronic structure of the ordinary fluorine which precedes neon in the periodic table. The outer, i.e. 2p shell of fluorine has exactly the same structure as that of anomalous neon, \( Ne' \). We can therefore conclude that chemically the anomalous neon
would behave similarly to the ordinary fluorine. Symbolically, we can write it as $Ne' \sim F$.

Why do these elements make good candidates for experiments? Our task is to find element $Z$ so that there is a sharp contrast between the chemical properties of the elements with atomic numbers $Z$ and $Z-1$. One solution to this problem is that we take noble gases as our element $Z$, such as $Ne$ and $Ar$, and therefore $Z-1$ element will be a halogen: fluorine or chlorine. The difference between these 2 groups of elements (noble gases and halogens) is great: the noble gases are very inert elements while halogens are very active ones. So we have 2 solutions to our problem: $Z = 10$, i.e. $Ne-F$ pair, with $Ne'$ behaving as $F$, and $Z = 18$, $Ar-Cl$ pair, with $Ar'$ behaving as $Cl$.

The experiments based on the idea that we have just discussed have been performed by Novikov et al. (1990) and Nolte et al. (1991) who looked for anomalous neon in a sample of fluorine and anomalous argon in a sample of chlorine. Schematically, their procedure is as follows: they first take a sample containing fluorine. Then they turn neutral fluorine atoms into negative ions. Finally, they use atomic mass spectroscopy to look for ions with atomic mass 20.

If these $A = 20$ ions are found, one can interpret it as anomalous $Ne$ atoms because of 2 reasons: first, fluorine does not have stable isotopes with $A=20$, so it cannot be a fluorine atom. Neon, on the other hand, cannot form negative ions, so these atoms cannot be neons either. Therefore, they are to be interpreted as anomalous neon atoms. This experiment obtained an upper limit on the concentration of $A = 20$ and $A = 36$ ions (Nolte et al., 1991):

$$N(^{20}Ne')/N(^{20}Ne) < 2 \times 10^{-21}, \quad N(^{36}Ar')/N(^{36}Ar) < 4 \times 10^{-17}. \quad (12)$$

Another method was used in a search for anomalous helium, $He'$ (Deilamian et al., 1995). While ordinary He has a wave function that is antisymmetric with respect to space/spin exchange, the anomalous helium would have a symmetric wave function under this exchange. Because of this difference in symmetry the energy level of $He'$ would be shifted by a tiny amount. Drake (1989) calculated these tiny energy shifts with high precision. Therefore we know the exact position of an anomalous spectral line. The idea of the experiment is to look for the anomalous spectral line ($1s3p^1P_1 \rightarrow 1s2s^1S_0$) using the modern Doppler-free laser spectroscopy technique. The absence of this spectral line allows one to obtain an upper limit on the PEP violating parameter. The final result is that this parameter must be less than $5 \times 10^{-6}$.

Other experiments are described in a review by Gillaspy (2000). An incomplete list of additional references includes Barabash et al., 1998; Javorsek II et al., 2000, 2002; NEMO Collaboration (2000); Back et al. (Borexino), 2004; VIP Collaboration (2005).

The validity of Bose statistics for photons has also been investigated (Ignatiev et al., 1996; DeMille et al., 1999).
IV. CONCLUSION AND OUTLOOK

To summarise, remarkable progress has been made since 1987 in the area of testing the Pauli exclusion principle and Bose statistics, both theoretically and experimentally. While essentially no measures of quantitative nature existed before 1987 on the accuracy of these principles, now the limits on possible statistics violation are quite impressive. For instance, the limit is $\sim 10^{-26}$ for electrons and even better for nucleons. Theoretically we have much better understanding of why PEP and Bose statistics are so accurate. We know what can go wrong when we try to violate them. And we have important and interesting directions to pursue, both in theory and experiment. Finally, numerous links with other areas of physics and mathematics have been revealed. So, a lot of exciting work is ahead.

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