

History of molecular beam research: Personal reminiscences of the important evolutionary period 1919–1933

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Dr. Immanuel Estermann was engaged in writing a book on the History of the Molecular Beam Method when he died in Haifa, Israel on 31 March 1973. The original plan of the manuscript was to cover three distinct historical periods: the first, the years 1911–1933, when the center of research in molecular beams was at the University of Hamburg; the second, from 1933 to the outbreak of World War II, when the most active laboratory working in the field was at Columbia University; and the postwar period, when many laboratories on both sides of the Atlantic became actively engaged in this field. The material in this paper deals primarily with the early historical period and would have formed the essence of the first two chapters of the book. In addition to presenting interesting historical facts on a crucial period in the development of quantum physics, it contains some amusing historical sidelights on the research personalities that dominated that period.

I. IMPORTANCE OF THE MOLECULAR BEAM METHOD IN THE DEVELOPMENT OF QUANTUM PHYSICS

A. Introduction

The state of quantum physics in 1919–1920, when I was a student at the German universities, could be summarized as follows. (i) The radiation law formulated by Planck (1900) had been accepted and was no longer a subject of controversy. (ii) The theory for the specific heats of solids was generally accepted, although it was realized that the simple Einstein theory (1907) led to the wrong slope at low temperatures (predicting an exponential rather than the experimentally observed T^3 law as $T \rightarrow 0$). Debye's model (1913) gave the correct slope, but used a somewhat arbitrary high-frequency cutoff; the Born–von Karman approach appeared to be the right one, but computation for real cases was complicated. (iii) The acceptance of light quanta was less than universal; while absorption and emission of photoelectrons were well



Fig. 1. Immanuel Estermann. Photograph taken in Washington, DC, 1959. (AIP Niels Bohr Library.)

explained by this theory, propagation, interference, and diffraction were considered incompatible with it—I had the impression that the physicists with whom I was in contact shrugged their shoulders and did not seriously discuss this dilemma; after all, the photo effect was so much on the sidelines that the problem was generally swept under the rug. (iv) The quantum theory of the atom, however, was very prominent in physics discussions, but was not yet generally taught in university courses.

I became acquainted with the quantum theory of the atom in 1922 when I studied Sommerfeld's book and was greatly impressed by it. Although there were many questions left open, the hope that these would soon be answered seemed to be justified.

Others saw the situation in a less positive light. They pointed out that the relationship between the Rydberg constant and the "elementary" quantities e , m , c , and h had already been given in 1910 by Haas and was, therefore, not a unique consequence of the theory. Einstein saw the relationship $E = h\nu$ primarily as a property of light and only secondarily as one of matter or of the atom. One prominent physicist (I forgot who it was) had written to Bohr after the publication of his 1913 paper, "I congratulate you on your success in expressing the R in terms of e , h , m , c , but I do not believe in your (or any other) model of the atom." And, of course, the three postulates of Bohr's theory (stationary states, no radiation, and $\Delta E = h\nu$) were still very mysterious.

Even more controversial was the situation with respect to the hypothesis of space quantization, which had been proposed by Debye in order to explain the Zeeman effect on the basis of Bohr's model. If this hypothesis were taken at face value, it meant that atoms carrying a magnetic moment could only assume discrete orientations with re-

spect to the direction of the magnetic field, in direct contradiction to the consequences of classical theory, according to which the orientation of the atomic magnet should be distributed uniformly (with equal probability) over all directions. This was such a revolutionary idea that even Debye did not think that it had a physical reality, but that it was merely a computational recipe for the additional magnetic energy of atoms in a magnetic field. On the other hand, Bohr thought that the atoms in question would divide into two groups, with their moments either parallel or opposed to the magnetic field. Sommerfeld did not have a clear opinion; he oscillated between a choice of three possible discrete positions and a random distribution.

The situation was aggravated by two facts: (i) the results of the space quantization hypothesis with regard to the Zeeman splitting of spectral lines were identical with those of the Lorentz theory based on classical assumptions, and (ii) neither of the three assumptions (nor the classical theory) was able to explain the so-called anomalous Zeeman effect (which was actually observed in the majority of cases). Finally, nobody was able to understand how atoms entering a magnetic field with random orientations could divide into groups when their density was so low that collisions did not occur to exchange energy. Where should those that would receive higher energy get their energy from, and vice versa?

In short, the subject was intensely discussed, but I gained the impression that the majority did not believe in space quantization. In August 1921, Otto Stern published a short paper¹ with the title "Ein Weg zur experimentellen Prüfung der Richtungsquantelung in Magnetfeld," in which he proposed an experiment which should answer this question unambiguously. For the execution of this experiment, he suggested the use of molecular beams.

B. Beginnings of the molecular beam method

There is a note by Dunoyer² in the *Comptes Rendus de l'Academie des Sciences* of 6 March 1911, entitled "Sur la theorie cinetique des gaz et la realization d'un rayonnement material d'origine thermique." It was followed by three additional papers during the years 1911–1913, in which the author describes the formation of a beam of material particles emitting from a source containing a vapor at moderate pressure through a hole into a vacuum, where these particles (i.e., molecules or atoms) proceed in straight trajectories as the kinetic theory predicted.

Dunoyer's applications of these "molecular beams" dealt with problems of spectroscopy and fluorescence and were more or less forgotten during the following years when the first world war presented more urgent problems. The only exceptions were some experiments by Walther Gerlach in 1912. Gerlach, who was then in Paschen's laboratory in Tübingen, tried to repeat Dunoyer's experiments with a few different metals without success and did not publish anything about these attempts.

A revival of interest in molecular beams occurred in Frankfurt after the end of the war. Otto Stern, who had received his doctorate in physical chemistry in 1912 in Breslau and who spent the following years with Einstein in Prague and Zürich, had come to Frankfurt at the beginning of 1915, after Einstein had left Zürich to become Director of the Kaiser Wilhelm Institut für Physik. Having been called up for military service shortly thereafter,



Fig. 2. Otto Stern. Photograph in the Meggers Gallery of Nobel Laureates. (AIP Niels Bohr Library, W. F. Meggers Collection.)

he began to work in Frankfurt only in 1919 where he joined Max Born, who was then Professor for Theoretical Physics. Stern occupied himself initially with problems of molecular theory. Born, although a professional theorist, had also developed an interest in molecular beams which he and E. Bormann were attempting to use for the measurement of the mean free path, but they were beset by vacuum difficulties. Stern made a more concerted attempt to use the method for the successful measurement of molecular velocities.

In the fall of 1920, Gerlach came to Frankfurt as assistant to Professor Wachsmuth at the Institute for Experimental Physics. Gerlach was acquainted with Dunoyer's experiments and had, as mentioned before, tried to duplicate some of his fluorescence experiments but without success. He was now interested in the problem whether the Bi atom would show the same strong diamagnetism that is exhibited by the Bi crystal and thought one could make an atomic beam experiment in which Bi atoms would be deflected in a strongly inhomogeneous field. He was occupied with an investigation on how one could make a magnetic field with the highest possible value of field gradient, $\partial H/\partial z$, and experimented with various geometrical configurations. Born thought that the experiment would not succeed; on the other hand, Gerlach quoted Edgar Meyer (Zürich) as having said, "Kein versuch ist so dumm, dass man ihn nicht probieren sollte." Before an actual experiment had been planned, Stern asked Gerlach, "Do you know what space quantization is?", to which Gerlach replied, "No." Stern then explained the controversy (which I mentioned earlier) and explained his idea of demonstrating or refuting the existence of space quantization by a definitive experiment. (Stern, incidentally, did not at that time believe in space quantization.) The critical question was whether one could make a sufficiently inhomogeneous magnetic field to obtain an observable deflection of an atom carrying a magnetic moment of the order of a Bohr magneton ($\mu_B = eh/4\pi mc$), a quantity assumed to be a kind of unit for atomic moments.

At an afternoon coffee session in a cafe, Born, Stern,

and Gerlach discussed the proposed experiment in detail and decided that it should be done. Gerlach would provide the necessary inhomogeneous field, $\partial H/\partial z$. Stern had already successfully overcome the crucial vacuum problems and had substantial experience with beams of silver atoms from his velocity measurements, while Born's work was still being delayed by vacuum problems.

C. Demonstration of space quantization

1. *The Stern–Gerlach experiment.* If an atom carrying a magnetic moment μ passes parallel to the axis of an inhomogeneous magnetic field whose inhomogeneity $\partial H/\partial z$ is normal to the beam direction x , and if θ designates the angle between μ and $\partial H/\partial z$, the atom will experience a force $\mu(\partial H/\partial z) \cos\theta$ and suffer a deflection in the z direction of $(\mu/2m)(\partial H/\partial z) \cos\theta(l^2/v^2)$ (where m is the atomic mass, v is the atomic velocity, and l is the length of the inhomogeneous field region). According to classical theory, θ could assume a continuous distribution of values; according to the space quantization theory of 1920, in terms of the azimuthal quantum number k , $\cos\theta$ could have only the values m/k , where $m = -k, -k + 1, \dots, k$. In the case of a H atom in the ground state, $k = 1$ and, therefore, $m = -1, 0, +1$ and $\cos\theta = -1, 0, +1$.

Consequently, if a narrow beam of H (or, analogously, Ag) atoms would pass through an inhomogeneous magnetic field, the classical theory predicted a broadening of the beam, while the space quantization theory predicted a splitting into discrete beams. Stern as well as Sommerfeld expected either a broadening or a split into three beams, while Bohr predicted a split into two beams—he had, apparently, some arguments to exclude $m = 0$. (The real reason for the occurrence of only two beams was to be discovered several years later.)

This is not the place to discuss the many experimental details or the difficulties that had to be overcome or the primitive state of vacuum technology that existed at that time. What would be today a suitable experiment for a student in an advanced laboratory was just then at the borderline of the possible. I will only mention a few principal requirements: (i) the necessity of defining the beam to dimensions of ~ 0.1 mm so that with $\partial H/\partial z \approx 50\,000$ G/cm the deflection would be observable; (ii) the maintenance of a sufficiently low pressure ($< 10^5$ Torr) so that the mean free path would be of the order of the beam length; (iii) the availability of a beam detector of sufficient sensitivity to reduce the time of the experiments to manageable values. Each of these items required a great amount of skill and perseverance, and the difficulties were only gradually overcome.

2. *Experimental results.* The first partly successful experiment was carried out in November 1921. It showed a broadened beam, an elliptically shaped target figure instead of a circle. This result seemed to confirm the classical approach which Stern and also Sommerfeld had expected. Gerlach then suggested replacing the circular diaphragms by slits. In the meantime, Stern had accepted a professorship for theoretical physics in Rostock, where I joined him as assistant on 1 December 1921. Gerlach continued alone, but something always happened. Early in February 1922 Stern and Gerlach met in Göttingen for a “consilium” and the result of their discussion was, “Es geht nicht” [“It won't work”].

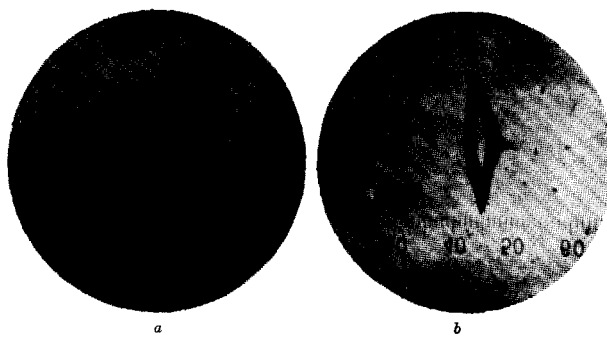


Fig. 3. Splitting of a beam of silver atoms by an inhomogeneous magnetic field. (a) Trace without the field; (b) splitting with the field.

When Gerlach was ready to return to Frankfurt, a railroad strike broke out which forced him to remain one-and-a-half days in Göttingen and travel back on a freight train. During this time he pondered again about all the details and decided to try once more. This time, everything worked and he obtained some indications of possible beam splitting. He decided that the experimental alignment was still not correct. After removing this fault, the experiment was repeated and a clear splitting into two beams was obtained, as shown in Fig. 3. A telegram was dispatched to Stern containing only four words, “Bohr hat doch recht” [“Bohr is right after all”], followed by a detailed letter. I was in Rostock when this news arrived and we celebrated accordingly.

Copies of the picture were sent to several physicists, and here are some of the replies. Paschen wrote, “This experiment confirms for the first time the existence of Bohr's stationary quantum states” (Franck and Hertz had only proven discrete energy differences). Pauli, “Jetzt wird hoffentlich auch der ungläubige Stern von der Richtungsquantelung überzeugt sein” [“This should convert even the nonbeliever Stern”]. Franck, “More important is whether this proves the correctness of the space quantization—Please add a few words of explanation.” And finally Bohr, “I would be grateful if you or Stern would inform me if you interpret your results in this way that the atoms are oriented only parallel or opposed, but not normal to the field, as one could also supply theoretical arguments for the latter assumption.”

These were all the comments that I could obtain on my



Fig. 4. Walther Gerlach giving a lecture in Tübingen, about 1925. This photograph was taken by a student in his class. (AIP Niels Bohr Library, Landé Collection.)

visit to Gerlach in 1972, almost exactly 50 years later.³ Others were destroyed during the war, but some additional communications may still exist elsewhere. At any rate, this experiment convinced many physicists of the physical reality of quantum states and advanced the theory from a "Rechenvorschrift" or "Kursbuch für Elektronen" to a description of the physical world and convinced, aside from Stern, many others that classical thinking had to be modified.

3. *Some experiments that did not give expected results.* I want to mention here a few cases of experiments, based on the Stern–Gerlach experiment, which did not give the expected results. If the experiment actually verified Bohr's idea of the parallel orientation of the magnetic moments, a gas of H (or Na) atoms should have become birefringent in a homogeneous magnetic field. Gerlach came to Rostock in 1922 and tried this experiment without success.

Stern and I argued that, if the Bohr magneton had real physical significance, all atomic magnets should be multiples of this value. We examined a great many substances which had been investigated by Weiss, who had introduced a unit called the Weiss magneton, which was surprisingly equal to 1/5 Bohr magneton, and found that many of them were indeed multiples of 5 Weiss magnetons. We tried to see if the remainder could be induced by chemical treatment to assume such a value, but some like Ni were incorruptible. So we gave up.

The true reason for these failures, as well as for the somewhat mysterious absence of the third or perpendicular orientation in the Stern–Gerlach experiment, came of course several years later with the discovery of the electron spin with its associated half-integral angular momentum quantum number, and the development of quantum mechanics.

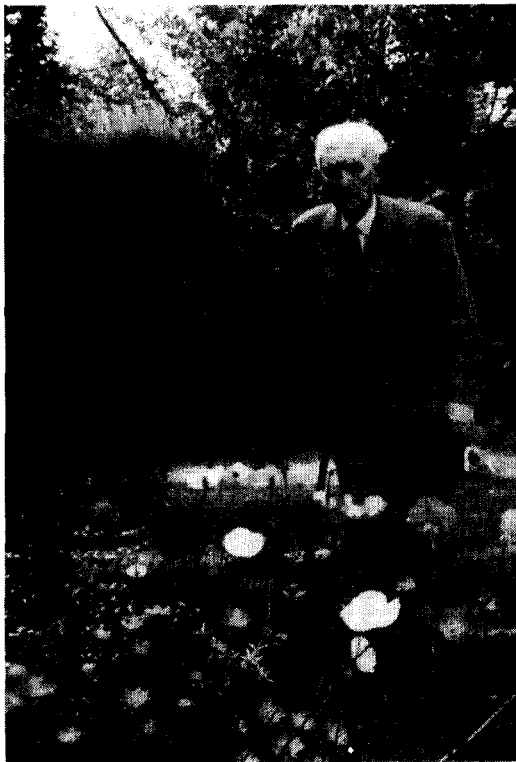


Fig. 5. Max Born in a garden in Germany, 1966. (AIP Niels Bohr Library, Stein Collection.)

D. Material waves

The dualism between corpuscles and waves, which was first introduced by Einstein in his theory of the photoelectric effect and further intensified by the discoveries of Compton (1923), Compton and Simon (1925), Bothe and Geiger (1924), and others on the Compton scattering of x rays, was extended by de Broglie (1925) to material particles with his famous formula $\lambda = h/p$, which relates the wavelength λ to the particle momentum p .

The experimental proof of the wave properties of electrons was given in 1927 by Davisson and Germer, who had observed some anomalies in the reflection of electrons from nickel targets and followed this up with experiments on single crystals of nickel in which they observed maxima and minima due to electron diffraction. Additional proof was provided later by Thompson and others from observations of the diffraction of electrons transmitted through thin metal foils.

The validity of the de Broglie relationship for heavy particles was established somewhat later by Estermann and Stern (1930), Estermann, Frisch, and Stern (1931), and T. H. Johnson (1930) by experiments in which molecular beams of He, H₂, and H were diffracted by single-crystal surfaces serving as diffraction gratings. These publications came only five and six years after the de Broglie theory had been presented and three years after Davisson and Germer's paper, and by that time the wave–particle duality for electrons was already widely accepted. There was, however, *no certainty* that this duality existed for matter in all its forms, and to quote Max Born⁴:

Here, surely, we are dealing with material particles which must be regarded as the elementary constituents not only of gases but also of liquids and solids. If we intercept the molecular ray after its diffraction at the crystal lattice and collect it in a receiving vessel, we find in the vessel a gas which still has ordinary properties.

These diffraction experiments on whole atoms show that the wave structure is not a property peculiar to beams of electrons but that there is a general principle in question: classical mechanics is replaced by a new wave mechanics. For in the case of an atom, it is clearly the centroid of all its particles (nucleus and electrons), i.e., an abstract point, which satisfies the same wave laws as the individual free electron.

I would like to add that a molecule of H₂ composed of two nuclei and two electrons behaves in the same way; this accentuates Born's statement even more, as the centroid is actually free from mass.

Immediately after we heard of de Broglie's paper, we started building an apparatus in which the cleavage surface of an alkali halide crystal would act as a diffraction grating for a molecular beam. Since the lattice spacing of the ions in these crystals is of the order of a few angstroms, they were expected to be good gratings for molecular beams of light atoms and molecules (H, He, H₂) which have de Broglie wavelengths of about 1 Å.

In the meantime, Knauer and Stern (1927) observed specular reflection of beams of molecular hydrogen and helium from highly polished metal surfaces at glancing

angles of a few minutes of arc. They also looked for diffraction from a grating ruled on speculum metal, but did not find anything, a result which was not unexpected if the molecules had the de Broglie wavelength.

The diffraction experiments with crystals were complicated and presented some special problems. For one thing, some of the crystals are quite hygroscopic so that the crystal surface could be readily spoiled. In the early experiments we tried to obtain diffraction of H_2 molecules from NaCl crystals, but got no results. We then tried He with NaCl crystals, but it was no better. For some reason, we made the detector slit longer and caught a faint trace of a diffraction maximum. It was quickly realized that, while we had been looking for a line-diffraction pattern, the crystal was acting as a two-dimensional or crossed grating. The interference pattern from such a grating consists of cones, the axes of which are the rulings on the gratings. Diffraction maxima occur where two cones intersect. Various changes were made in the apparatus and evidence of diffraction of H_2 and He from NaCl crystals was obtained.

We tried other crystals, and at last with LiF we got definitive results. An example is the diffraction of He at a glancing angle of 18.5° , shown in Fig. 6. The central peak is the specularly reflected beam and the side peaks are the diffracted beams. The broadening of the peaks is due to the Maxwellian distribution of velocities in the molecular beam. The positions of the diffracted peaks are exactly where they should be when the velocity distribu-

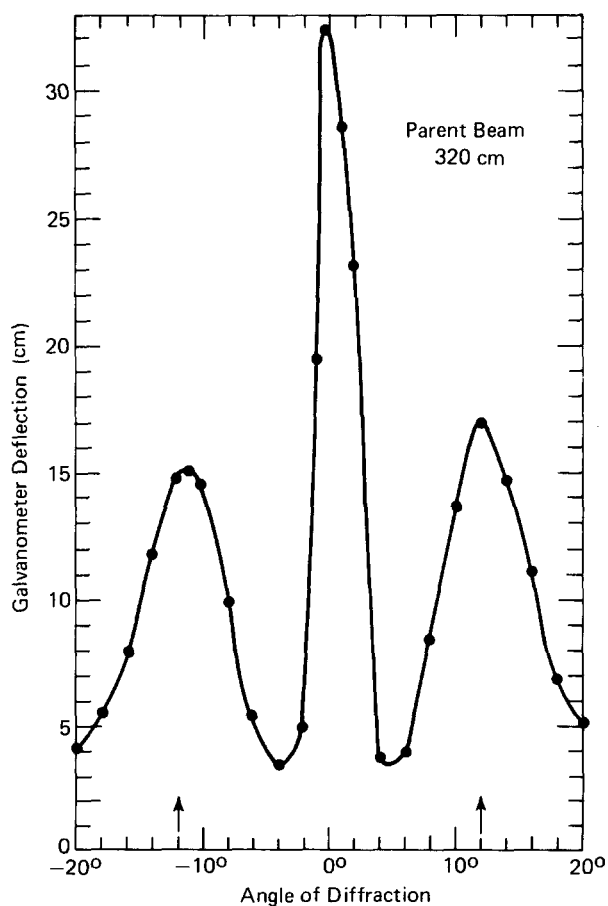


Fig. 6. Diffraction of a 290 K He beam by LiF at a glancing angle of 18.5° . The arrows indicate the calculated angles for the diffraction maxima.

tion is accounted for. These experiments are described in detail in the literature.⁵

The reflection and diffraction of monochromatic molecular beams⁶ provided both a quantitative test of the de Broglie formula and a confirmation of the relationship between wavelength and velocity. The velocity-selected beams were obtained either by passage of the beam through slits in disks rotating on a common shaft so that only a certain range of velocities would be transmitted, or by diffraction of the beam from a LiF crystal. In the latter case, a slit placed at a chosen position in the diffraction pattern selected atoms having a given wavelength and, therefore, a corresponding velocity. In each case the velocity-selected beam was then analyzed for diffraction by a second LiF crystal. In particular, the experiments with He confirmed the de Broglie formula with an accuracy of about 1%. The predicted variation of the de Broglie wavelength with molecular velocity was also verified in these experiments.

II. MOLECULAR BEAMS AND NUCLEAR PHYSICS

A. Nuclear physics in the 1920s

Toward the end of the 1920s, it became clear that molecular beams could play an important role in the development of nuclear physics. The early 1920s had achieved remarkable success in the unraveling of the features of the extranuclear electronic shell of the atom. As far as the atomic nucleus was concerned, however, very little was known.

When I announced a course on nuclear physics at the University of Hamburg in the early 1920s, one of my older colleagues asked me, "Nuclear physics—What is that? There is a chapter in Sommerfeld's book with this title, but this is not enough for a one-semester course." He was not quite right, but nearly so.

What was known was that the nucleus was a relatively massive object with a charge Ze , where Z was the atomic number. Also known were the masses of the various isotopic nuclei as determined by Aston and his successors with the then new mass spectrographs; they turned out to be almost integral multiples of the mass of the hydrogen nucleus (actually $1/16$ the mass of the oxygen isotope O^{16}), thus reviving Prout's hypothesis according to which the heavier nuclei are built up from elementary particles. Furthermore, the nuclear radius was known, at least to within an order of magnitude (10^{12} – 10^{13} cm) from Rutherford's scattering experiments, and finally, some information about the composing particles of the nuclei could be extracted from observations of radioactive decay.

Regarding the interaction of the nucleus with the extranuclear electrons, the principal quantity which stood out was the charge Ze . With improvements in spectroscopic technique, certain spectral lines which had heretofore been regarded as single, or as components of a fine-structure multiplet, showed a splitting into two or more components. This further splitting, called hyperfine structure, could not be accounted for in terms of the properties of the electronic shell, and the most likely explanation was that it was due to interactions with certain nuclear properties heretofore neglected. The detailed study of this hyperfine structure became a fascinating enterprise for spectroscopists because it required all the ingenuity and

perseverance to separate the very closely spaced lines and to measure their wavelengths. One of the first results of these investigations was that different isotopes of the same element had spectral lines of slightly different wavelength. Thus, when a mixture of isotopes was used as a light source (as was the case in most experiments), each spectral line was expected to have as many components as there were different isotopes in the mixture.

This, however, was only part of the story because a hyperfine structure also appeared in the spectra of elements having only a single isotope; secondly, many hyperfine structures had more components than there were isotopes present. The isotope shift could, therefore, not be responsible for all the aspects of the hyperfine structure. Hans Kopfermann, who was one of the most important investigators of hyperfine structure, began experiments with elements containing a single isotope or artificially separated isotopes in order to remove the isotope effect and, therefore, be better able to analyze the remainder. For the interpretation of the latter, Pauli⁷ postulated a spin angular momentum and an associated magnetic moment for the nucleus, whose interaction with the extranuclear (valence) electrons caused their energy levels to split into a number of components and thus produced the hyperfine structure.

The quantitative verification of Pauli's assumption was for a time beset with difficulties. The experimental material was frequently difficult to analyze, because the lines were very close together and often not very well defined, and measurements on single isotopes were rare. Moreover, as was later found, the electric field experienced by electrons coming very close to the nucleus deviates slightly from a Coulomb field. This effect requires a correction to the purely magnetic interaction between a nuclear magnetic moment and the extranuclear electron.

Another difficulty arose from theoretical considerations.



Fig. 7. Albert Einstein and Wolfgang Pauli. Photograph taken by P. Ehrenfest in Leiden, 1926. (CERN, Courtesy AIP Niels Bohr Library.)

At that time only two elementary particles were known, the proton and the electron. If Prout's hypothesis were valid, a nucleus of mass A should contain A protons, each carrying a positive charge numerically equal to the (negative) charge of an electron. Since the atomic number, Z , is always (except for hydrogen) less than A , it had to be assumed that the nucleus contains also $A - Z$ electrons whose negative charges account for the difference between A and Z . When S. Goudsmit and G. E. Uhlenbeck "discovered" that the electron had an inherent spin of the magnitude $\frac{1}{2}(h/2\pi)$ and an associated magnetic moment $2(e/2mc)[\frac{1}{2}(h/2\pi)] = eh/4\pi mc$ (a postulate which explained the fine structure of the spectral lines and the anomalous Zeeman effect), a new difficulty arose. If the electronic moment was responsible for the fine structure, the much smaller hyperfine structure must be caused by a much smaller moment. On the other hand, if a nucleus contained an uneven number of electrons, at least one of them would remain unpaired and its relatively large magnetic moment should manifest itself in the spectrum of the associated atom, but experiments proved that it did not. A similar difficulty became evident regarding the angular momentum. Since both the proton and the electron possess an angular momentum of $\frac{1}{2}(h/2\pi)$, a nucleus containing an *even* number of elementary particles (i.e., A protons plus $A - Z$ electrons) should have a total angular momentum $I(h/2\pi)$, where I , the angular momentum quantum number, would be an integer (including 0), while in a nucleus with an odd number of particles, I would be a half-integer, such as $1/2$, $3/2$, $5/2$, etc. This simple prediction, however, was not always confirmed by experimental results; one of the most drastic exceptions was the nitrogen nucleus of mass number 14 and charge number 7; it should contain 14 protons and $14 - 7 = 7$ electrons, or a total of 21 particles, and hence have a half-integer angular momentum quantum number, while spectroscopic measurements, which shall not be discussed here further, lead to an unambiguous assignment of $I = 1$. Similar discrepancies were found in other elements. As a result, there was no doubt that the atomic nucleus had a much more complicated structure than was previously assumed.

Accordingly, investigation of nuclear structure became a frontier area of physics which attracted the interest of theoretical as well as experimental physicists of the highest caliber, and measurements of nuclear spins and magnetic moments were considered to provide important clues for the development of a consistent picture of the constitution of the atomic nucleus.

At first, the only methods available for the determination of nuclear spins and magnetic moments were spectroscopic; the most prominent were hyperfine structure measurements. Regarding spin, the procedure was relatively simple because it required, in principle, only the counting of the *number* of hyperfine energy levels into which a "single" line was split by the interactions of nuclear spin with extranuclear electrons. The calculation of nuclear magnetic moments from the measurement of hyperfine structure splitting is much more complicated and required approximations which limited the validity or at least the accuracy of the results. In all cases, the resolution of hyperfine structure demanded the highest attainable refinements in spectroscopy, and here the molecular beam method provided important progress by permitting the construction of light sources and absorbers giving ex-

tremely narrow spectral lines both in emission and absorption.

B. Magnetic moment of the proton

The most challenging question was obviously presented by the hydrogen nucleus, whose designation "proton" indicated its special status as a fundamental particle. The knowledge of its magnetic moment is of decisive importance both for the theory of nuclear structure and even for the theory of elementary particles themselves. In 1928, Dirac succeeded in developing a relativistic quantum theory of the hydrogen atom which yielded without arbitrary ad hoc assumptions a value of $\frac{1}{2}(h/2\pi)$ for the spin of both the proton and electron, and this led to predictions of the associated magnetic moments $\mu_e = eh/4\pi m_e c$ and $\mu_p = eh/4\pi m_p c$ for the electron and proton, respectively, where m_e is the mass of the electron and m_p is the mass of the proton. The electronic moment had been known for some time and had been designated the Bohr magneton, μ_B (because it followed from Bohr's theory). Accordingly, the quantity given above for the proton was called a nuclear magneton (n.m.); its magnitude is 1837 times smaller than that of the Bohr magneton because the mass of the proton is 1837 times that of the electron.

Although Dirac's theory for the magnetic moment was in a strict sense valid only for the electron, it was almost universally accepted to give the correct value for the proton moment also. However, an experimental confirmation was still desirable. The optical method was unable to solve this problem because of the very small hyperfine splitting and the relative width and fuzziness of the hydrogen spectral lines.

Now, Stern had always been interested in fundamental questions, and when nuclear moments were first proposed, he began to look into the possibility of measuring them by means of the molecular beam method. Even before Dirac's theory appeared, the order of magnitude of nuclear moments was reasonably well established and experiments were carried out in the Hamburg laboratory with the aim of measuring such small moments. Success was only marginal, but after the Dirac theory predicted not only an order of magnitude but a numerical value, discussions began in earnest as to how to measure such a small moment with a respectable accuracy.

Atomic moments, about 2000 times larger, had been measured successfully in Hamburg with the aid of an improved Stern-Gerlach apparatus, and it was decided to use the same principle (i.e., the deflection of a molecular beam in an inhomogeneous magnetic field) for the proton measurement. This required, in the first place, tremendous improvements in purely apparatus details, for example, the use of much finer slits, stronger magnetic fields, and better detectors and pumping equipment. However, further problems, posed by the nature of the experiment itself, combined to make this apparently simple experiment an investigation of major proportions.

It was already evident from the beginning that the experiment could not be performed with a beam of free protons because their electric charge would produce a much larger deflection in a magnetic field than would their magnetic moment. It also could not be performed with a beam of hydrogen atoms because these undergo a strong deflection on account of their electronic moment, which would completely obscure any additional deflection due to

the small proton moment. These problems were overcome by the use of the hydrogen molecule H_2 as the beam-forming substance and the study of its deflection in an inhomogeneous magnetic field of sufficiently strong inhomogeneity and length to produce measurable effects.

All the previous measurements had been carried out with beams of atoms, but here a beam of diatomic molecules was used for the first time, and additional problems arose just from this fact.

We shall now discuss briefly the magnetic properties of the H_2 molecule at relatively low temperatures. Under this condition (which is the condition of the molecular beam deflection experiment) the two electrons contained in the molecule have no orbital moments (they are in s states), and their intrinsic magnetic moments cancel out. The remaining magnetic moment of the molecule is, however, not only the intrinsic moment of the two protons, but it contains in addition a moment of the same order of magnitude due to rotation of the two protons around their common center of mass and the reaction of this rotation on the electronic shell. This contribution, called the rotational moment, had been estimated theoretically by Majorana to about 3 n.m. per rotational quantum; that is, for the lowest rotational state, $J = 0$, there would be no rotational moment, for the next higher state (rotational quantum number $J = 1$), 3 n.m., and for higher states, $3J$ n.m.

It might appear to be a simple matter to perform the experiment with H_2 molecules at such a low temperature that all the molecules would find themselves in the $J = 0$ rotational state. However, another problem arises which precludes this simple approach. In a hydrogen molecule, the two proton spins can either be parallel to one another, resulting in a total spin of $I = 1$ and an associated magnetic moment twice the size of the proton moment, or opposed, resulting in a total spin $I = 0$ and no magnetic moment. States with the first kind of arrangement are called ortho states; the second, para states. Quantum-mechanical laws prescribe that para states can only exist in even rotational states ($J = 0, 2, 4$, etc.) and ortho states in odd rotational states ($J = 1, 3, 5$, etc.). Thus, an experiment at low temperature with the molecules in the $J = 0$ state would fail because there would be no net nuclear moment.

Moreover, transitions between ortho and para states are very slow unless certain catalytic agents are present. Thus, ordinary or "normal" hydrogen behaves like a mixture of two stable gases, containing at room temperature or higher temperatures 25% *para*- and 75% *ortho*-hydrogen. As far as the distribution of the molecules in the different rotational states is concerned, we will look first at *para*-hydrogen. At room temperature we find 52.5% in the zero rotational state, 46.1% in the second rotational state, 1.4% in the fourth, and a negligible amount in the higher states. For *ortho*-hydrogen, we find at room temperature 88.5% in the first rotational state, 11.4% in the third rotational state, and practically none in the higher states.

If hydrogen is cooled down to, say, 90 K (temperature of liquid air), the populations of the rotational states change as follows. In the *para* form, we have 98.3% in the zero state, 1.7% in the second state, and practically nothing in the higher states. The corresponding numbers for the *ortho* form are 99.98% in the first, and 0.02% in the third rotational states. In the absence of a catalyst, the

relative concentration of *ortho*- and *para*-hydrogen will still be 75% *ortho*- and 25% *para*-hydrogen.

If we were passing a beam of hydrogen molecules at liquid-air temperature with uniform translational velocity through an inhomogeneous magnetic field, we should expect the following splitting pattern. Neglecting the small contribution from rotational states of 2 or higher, we would have 25% *para*-hydrogen in the rotational state 0 and 75% *ortho*-hydrogen in the rotational state 1. The first group would not be deflected, having neither a nuclear nor a rotational magnetic moment. The second group have a rotational moment of about 3 n.m. (according to the theoretical estimate by Majorana) and a nuclear moment of 2 n.m. according to the universally accepted conclusions of Dirac's theory, keeping in mind that *ortho*-hydrogen contains two protons whose nuclear spins are parallel and whose nuclear moments add. Since the rotational moment is larger, the basic splitting would be into three components because of the rotational moment ($J = 1$) with a separation corresponding to 3 n.m. Each of these would split again into three components because of the nuclear moment ($I = 1$), but with the separation corresponding to 2 n.m. All the components would have equal intensity, that is, $1/9$ of $3/4$, or $1/12$ of that of the original beam. The central component, or the nondeflected one, would be augmented by the *para*-hydrogen having an intensity of $1/4$ of the original beam, so its total intensity would be $1/4 + 1/12 = 1/3$. The splitting pattern would therefore look as shown in Fig. 8.

Because of the finite width of the beam-forming slits and the distribution of translational velocities in a molecular beam according to Maxwell's law, each of the components is broadened into a pattern which is much wider than the distance between two components. As a result, the observable distribution would not show all nine components, but only a relatively sharp central maximum and a broad maximum on each side of the center line. The effect of the finite slit width and the Maxwellian distribution can be calculated without ad hoc assumptions, and one could hope to construct a series of patterns for assumed values of both the rotational moment and the pro-

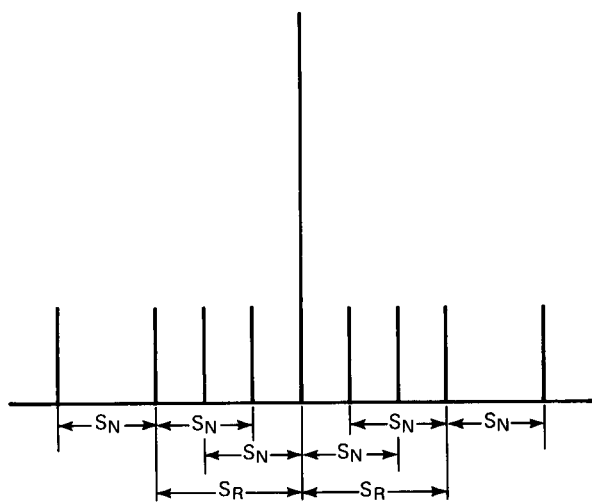


Fig. 8. Theoretically predicted intensity pattern for the splitting of a beam of hydrogen molecules at 90 K by an inhomogeneous magnetic field. The rotational splitting, S_R , and the nuclear splitting, S_N , were, respectively, assumed to be due to a rotational magnetic moment of 3 n.m. and a nuclear moment of 2 n.m.

ton moment and to find by comparison with the observed pattern the set which gave the best fit. The original confidence in the theoretically predicted values was so great that one calculated a deflection pattern based on these values as a starting point.

This was the situation when it was decided to construct an apparatus for the measurement of the proton moment. The basic requirements were the following: very narrow slits for beam formation, large inhomogeneity of the magnetic field, a long path of the beam in the field, sensitive detectors, and a good vacuum system. As always, compromises had to be made and available components had to be used even if they did not meet all the requirements. A workable apparatus was built by O. R. Frisch and Otto Stern in the spring of 1932. (I was at that time at the University of California in Berkeley, holding an International Research Fellowship of the Rockefeller Foundation.) Although technical difficulties persisted and many experiments could not be completed because of malfunctioning of one component or another, such as vacuum leaks, galvanometer noise, etc., a few succeeded well enough to show that the assumed pattern discussed above was not in agreement with experimental results. The apparatus had been given the nickname "Kreuger"—after the then famous international match king and impostor Ivar Kreuger—because it swindled all the time when one expected definite, understandable results.

When I returned from Berkeley in early November of 1932, we decided that "Kreuger" had outlived its usefulness and that a new apparatus of basically the same design but with a few changes in dimensions and certain technical improvements should be constructed. At this stage, Frisch wanted to embark on an independent effort in order to be able to submit a *Habilitationschrift* and achieve the academic *venia legendi*, the first step of the academic ladder then in force in Germany. Stern and I joined forces again and proceeded with the construction of a new apparatus. We increased the gap between the magnet pole pieces from 0.5 to 1.0 mm, sacrificing field strength and inhomogeneity, but allowing much more accurate measurement, improved the manometer detectors by making them smaller in size and obtaining a better signal-to-noise ratio, applied some new tricks in vacuum technique which I had brought back from the United States, etc. But the really important step forward was that we decided to reduce our reliance on theoretical predictions. As pointed out above, we were really confronted with two unknown quantities simultaneously, the nuclear moment and the rotational moments. We have seen already that the properties of the hydrogen molecule make it impossible to obtain molecular states which exhibit the nuclear moment free from the rotational moment, but there are states in which *only* the rotational moment appears in evidence. These are the *para*-hydrogen states at not too low temperatures, and the deflection of beams consisting of pure *para*-hydrogen yield patterns which contain only *one* unknown quantity. The evaluation of these patterns is, of course, much simpler than that of the ordinary hydrogen patterns containing two unknown moments of not very different magnitudes.

At the temperature of liquid air, practically all the *para*-hydrogen molecules are in the rotational state with the quantum number $J = 0$, and these are undeflected in a magnetic field. At room temperature, however, 46% are

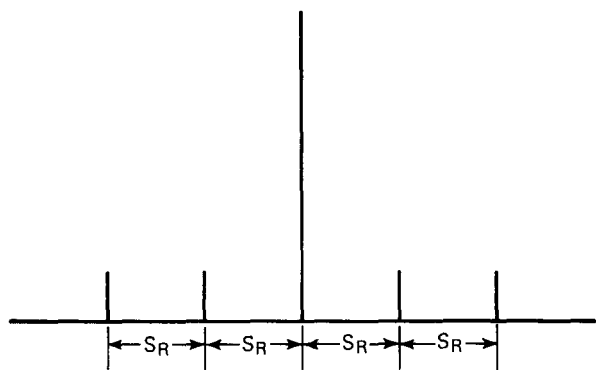


Fig. 9. Theoretically predicted splitting pattern for a beam of *para*-hydrogen molecules at room temperature. The rotational splitting, S_R , affects only those molecules with $J = 2$, which comprise 46% of the beam molecules.

in the state with $J = 2$, and these carry a magnetic moment of two rotational moments, $\mu = 2\mu_R$. The laws of space quantization permit these molecules with $J = 2$ to have five orientations with respect to the axis of the magnetic field, which are so arranged that the effective magnetic moments are $0, \pm \mu_R$, and $\pm 2\mu_R$. One would therefore expect at room temperature a deflection pattern such as is shown in Fig. 9, again assuming infinitely narrow slits and uniform translational velocity of the beam molecules. In the realistic case of small but finite slits and a Maxwellian distribution of velocities, each line in Fig. 9 is broadened, but the intensity distribution can be calculated for different values for μ_R .

To obtain the rotational moment, deflection experiments with pure *para*-hydrogen can be carried out at different temperatures. The obtained pattern can then be compared with computed patterns for various assumed values of μ_R . The assumed value which produces a pattern giving the best agreement with experiment is then considered to represent the actual value of the rotational moment. Actually, it was sufficient to calculate the intensity for only a few points and to vary the assumed value of μ_R until agreement was reached. Since the beam width is also a parameter that enters the calculation, the apparatus was so constructed that the beam width could be changed by varying the slit width during an experiment; in this way, the internal consistency of the obtained values could be checked.

The only hitch was where could we get the pure *para*-hydrogen? Bonnhöfer had shown a few years earlier that ordinary hydrogen could be converted into *para*-hydrogen by cooling it to the temperature of liquid hydrogen (20 K) in the presence of a suitable catalyst, for example, active charcoal. Liquid hydrogen (which was needed for this process) was then available in Germany only in Berlin. None of our group in Hamburg had ever handled liquid hydrogen, but I had watched experiments with liquid hydrogen in Berkeley, where Giauque had installed a low-temperature laboratory.

On the basis of this very limited experience, I undertook to provide the *para*-hydrogen for the measurements. To transport liquid hydrogen or gaseous *para*-hydrogen from Berlin to Hamburg was out of the question, but fortunately the chief technician of the low-temperature group there had designed a small hydrogen liquefier which could be operated without the need of a hydrogen com-

pressor, using factory-delivered compressed gaseous hydrogen as "raw material." I spent several days in Berlin becoming acquainted with liquid hydrogen handling and liquefiers, and we ordered one of these miniature liquefiers; a few were in the process of construction. After a while this liquefier was shipped to Hamburg and I embarked on the task of liquefying hydrogen and transforming it into *para*-hydrogen. Although the little liquefier had been working satisfactorily when I tested it in Berlin, it did not produce liquid hydrogen in Hamburg during the first tests. Unfortunately, it was built in such a way that the internal parts were not accessible without a complete disassembly, and when I finally decided to take it apart, I found that the internal Dewar vessel, in which the liquid hydrogen was supposed to accumulate and which had been made of soft glass (Pyrex was not yet available in Germany), was broken. After replacing the Dewar and reassembling the liquefier, and finally finding a source of supply for the high-priority compressed hydrogen that was needed for the operation of the liquefier, we finally succeeded in producing about a liter of liquid hydrogen and in transforming a few liters of gaseous hydrogen into pure *para*-hydrogen (we had been lucky in the choice of our charcoal catalyst because, as it turned out later, not all brands of activated charcoal are effective in promoting the ortho-*para* transformation). This *para*-hydrogen was stored in clean glass flasks at liquid air temperature in order to prevent its reconversion into ordinary hydrogen and was finally used for the first series of deflection experiments.

The surprising result of these experiments was that the experimentally obtained pattern did not agree at all with a pattern computed on the basis of the theoretically predicted value of a rotational moment of about 3 n.m. per rotational quantum, but with a much smaller value of about 0.8–0.9 n.m. A reevaluation of the previously obtained deflection patterns of normal hydrogen, introducing the above value for the rotational moment, indicated that the value of the proton moment itself must be about 2–3 times as large as the value assumed on the basis of Dirac's theory. This meant that the splitting pattern (again on the assumption of infinitely narrow slits and uniform translational velocity of the beam molecules) would not

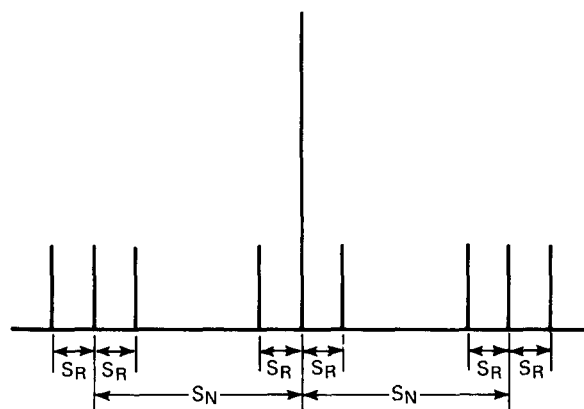


Fig. 10. Theoretically predicted intensity pattern for a beam of hydrogen molecules at 90 K split by an inhomogeneous magnetic field, but now (in contrast to Fig. 8) using the experimentally determined rotational splitting, S_R , corresponding to a rotational magnetic moment of 0.85 n.m., and a nuclear splitting, S_N , corresponding to a proton moment of 2.5 n.m. (and a total nuclear magnetic moment of 5 n.m.).

look like that given in Fig. 8 for the normal hydrogen molecule, but since the nuclear moment of the *ortho*-hydrogen would be about 5 n.m. against a rotational moment of less than 1 n.m., the primary splitting would show three components due to the nuclear moments, and each of these would show a much narrower splitting because of the rotational moment, as shown in Fig. 10.

It was now possible to repeat the measurements of the deflection of normal hydrogen with beams of different width and at different temperatures. The results obtained with the improved apparatus were compatible with a rotational moment of 0.85 n.m. per quantum number and a proton moment of 2.5 n.m., the estimated error being about 10%, which was mostly due to the difficulty of accurate measurement of the inhomogeneity of the magnetic field.⁸

The surprising result mentioned above had a strong impact because it showed without doubt that the Dirac theory of the electron could not be applied to the proton simply by substituting the mass of the proton for the mass of the electron. Theoretical physicists had been so completely convinced of the validity of the result that they regarded the difficult measurements as a waste of time. As Pauli remarked, "It is very laudable that you are making such an effort to measure the magnetic moment of the proton. Don't you think that is really superfluous, since we know perfectly well what the result will be?" Even while later experiments led to a more accurate value for the proton moment of 2.79 n.m., the deflection experiments by Estermann, Frisch, and Stern were the first ones to reveal the anomalous magnetic moment of the proton, and up to today no satisfactory theory explaining this anomaly has yet been established.⁹ Secondly, they remain the only measurements of a nuclear moment in which the interpretation is independent of the gyromagnetic ratio.¹⁰ The quantity measured is simply the force exerted on a neutral particle by an inhomogeneous magnetic field, which is proportional to the classical defined magnetic dipole moment.

The impact of this measurement was recognized in the award of the Nobel Prize for Physics to Otto Stern ten years later, when this work was specifically emphasized in the citation.

C. Magnetic moment of the neutron and deuteron

While the experiments with ordinary *para*- and *ortho*-hydrogen were under way, heavy hydrogen, later called deuterium (Urey, Brickwedde, and Murphy, 1932), and the neutron (Chadwick, 1932) were discovered. After the success of the proton measurement, it was challenging to attempt to measure the magnetic moment of the deuterium nucleus, then called "deuton" (now deuteron). At this time, the political situation in Germany had gone from bad to worse and we were not sure from one day to the next how long we would be able to continue our work at the University. Fortunately, the nazification of the State of Hamburg proceeded at a much slower rate than in other parts of Germany. However, in June 1933 I was notified (together with other Jewish faculty members) that my tenure would be terminated as of 30 September 1933, and Stern, who had not believed until then in the seriousness of the situation and who, having served on active military duty during World War I, was as yet exempt from the Nuremberg laws, tendered his resignation. While

in other German universities a notification like the one I received carried an immediate prohibition against even entering property, the good commercial sense of the Hamburg authorities decided that as long as they were paying our salaries we might as well continue to work, not so much because of our research activities, but primarily because of our teaching obligations.

We decided, therefore, to gamble on being able to continue to work for a few months longer. Now, heavy hydrogen (or heavy water) was not yet available in Germany. A few American researchers had succeeded in producing heavy water, among them Gilbert Lewis, Dean of Chemistry at the University of California in Berkeley, whom Stern as well as I knew well from visits to that university in 1930 and 1931/32, respectively. We sent a long telegram to Lewis asking him for the gift of a small amount of heavy water, and received after a few weeks a small glass capsule containing about 0.5 cm³ of water with about 85% concentration of heavy water. The process of decomposing the water into hydrogen was first tried out with ordinary water in order to gain the necessary experience, and then performed without difficulties with the precious heavy water. This was then run through the apparatus, and gave some quite unexpected results.

After the discovery of the neutron, the proton-electron hypothesis of the structure of atomic nuclei had been replaced by the proton-neutron theory, according to which a nucleus contained Z protons and $A - Z$ neutrons. In order to explain experimental values of nuclear spins of heavier nuclei, it was necessary to assign to the neutron a spin quantum number of $\frac{1}{2}$, just like that of the proton. The deuteron with $Z = 1$ and $A = 2$ thus contained one proton and one neutron, and therefore possessed a total nuclear spin of $I = 0$ or $I = 1$. Optical measurements favored the value $I = 1$. Since it was difficult to assume that a neutral particle like the neutron should possess a magnetic moment, it was generally assumed that the deuteron would have the same magnetic moment as the proton. As far as the rotational moment was concerned, a straightforward calculation for the deuterium molecule assigned it a value of one-half that of the hydrogen molecule.

The results of the experiments, which were carried out with the sword of Nazism hanging over our heads and usually continued until late into the night, were quite surprising. The magnetic moment of the deuteron was only about one-third that of the proton. The admixture of H₂ and HD molecules to the heavy hydrogen D₂ made it impossible to make a very accurate measurement, but we were sure that it was between 0.8 and 0.9 n.m.

The only possible interpretation of these results was that the neutron must possess an intrinsic magnetic moment. Furthermore, this moment must be oriented in the opposite direction to that of the angular momentum of the proton, because while the angular moments of the proton and neutron in the deuteron were additive, the magnetic moments were subtractive. The simplest conclusion was that the magnetic moment of the neutron was -1.6 n.m. (the present accepted value is -1.91 n.m.).

This is the end of the story of the measurements of nuclear magnetic moments by molecular beam deflection experiments in the Hamburg laboratory. A few weeks after the completion of these experiments, most of the staff members had to leave the laboratory and this kind of work ceased. The center of effort on nuclear magnetic

moments moved to Columbia University in New York where I. I. Rabi, who had spent a postdoctoral year in Hamburg in 1929, introduced new and powerful molecular beam methods and founded a school which was able to achieve spectacular results in the coming years.

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¹O. Stern, *Z. Phys.* **7**, 249 (1921).

²L. Dunoyer, *C. R. Acad. Sci. (Paris)* **152**, 594 (1911).

³The author visited Professor Gerlach in Munich in October 1972, to discuss their recollections of these early experiments.

⁴M. Born, *Atomic Physics*, translated by J. Dougall (Blackie, London, 1951), 5th ed., p. 92.

⁵I. Estermann and O. Stern, *Z. Phys.* **61**, 95 (1930).

⁶I. Estermann, R. Frisch, and O. Stern, *Z. Phys.* **73**, 348 (1931).

⁷W. Pauli, *Naturwissenschaften* **12**, 741 (1924). Pauli remarked frequently in seminar discussions that he doubted that nuclear moments could really explain hyperfine structure.

⁸The discovery that the rotational moment was only about one-fourth the theoretically calculated value led to a new calculation by Wick [*Z. Phys.* **85**, 25 (1933)], which included the reaction on the electronic shell imposed by the rotation of the nuclei around their common center of mass. The calculation leads to limits on the rotational moments between 0.36 and 0.93 n.m. The experimental value lies between these limits, but close to the upper limit.

⁹Certain explanations have been proposed, but they do not permit quantitative predictions.

¹⁰R. D. Evans, *The Atomic Nucleus* (McGraw-Hill, New York, 1955), p. 152.

A POSITIVE LETTER OF RECOMMENDATION

It was in 1946, or possibly in 1947, and Professor Bethe was the chief theorist at Cornell. It came time to read the recommendation letters that were sent in for new graduate students, to decide who to take and so on, and Bethe opened a letter from England, which he passed to me without comment. The letter was from R.H. Fowler, the very well known English theoretical physicist, and it went something like this: "Dear Bethe, I'd like to recommend enthusiastically that you take Freeman Dyson as a graduate student in physics. He has not studied much physics but I can say this: Although he is only 19 years old, he is, in my opinion, the best mathematician in England. Very sincerely yours, R.H. Fowler." That's all it said. And he was right.

—Philip Morrison