The Molecular Beam Method: From Otto Stern's pioneering experiments to Rabi's Magnetic Resonance and Ramsay's Separated Oscillatory Fields

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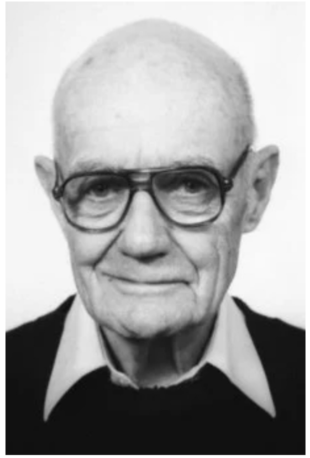
WILHELM UND ELSE HERAEUS-STIFTUNG

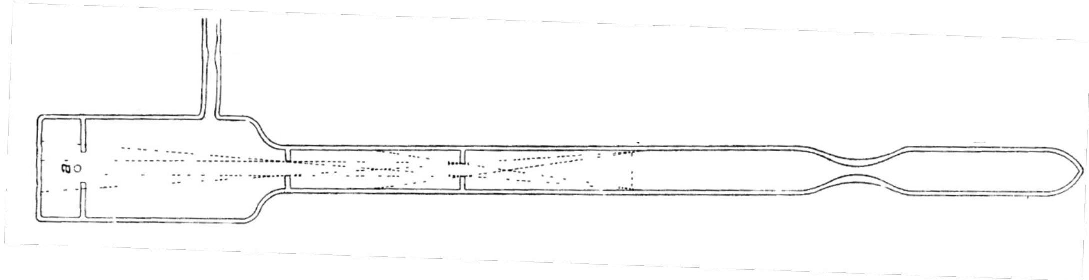
Quantum Technologies – Origins and Applications

Steinbach/Taunus, 1-4 September 2025

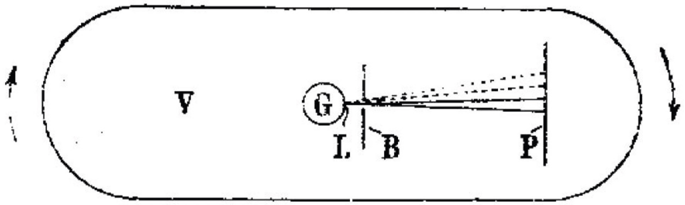
- **John Fenn (1988)**: "Born in leaks, the original sin of vacuum technology, molecular beams are collimated wisps of molecules traversing the chambered void that is their theatre."



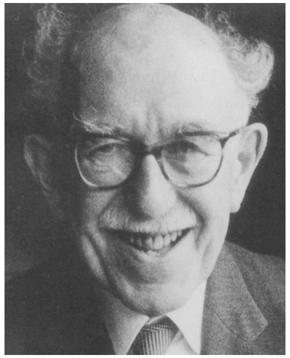
- **Louis Dunoyer (1911)**: *Do gas molecules move on straight lines between collisions?* Test of the mean free path assumption of the kinetic theory using rayonnement matériel



- **Otto Stern (1920)**: *Is there a zero-point translational energy?* Measurement of the thermal velocity distribution of molecules in an effusive molecular beam

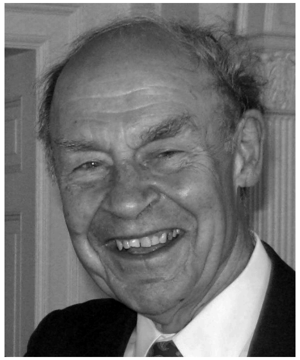


- **Otto Stern (1946)**: "The most distinctive characteristic property of the molecular [beam] method is its simplicity and directness. It enables us to make measurements on isolated neutral atoms or molecules with macroscopic tools. For this reason it is especially valuable for testing and demonstrating directly fundamental assumptions of theory."



- **Isidor Rabi (1988)**: "From Stern and from Pauli I learned what physics should be. For me it was not a matter of more knowledge … Rather, it was the development of taste and insight … Stern had this quality of taste in physics and he had it to the highest degree."

- **Dudley Herschbach (1988)**: "Concepts and techniques developed by Stern have proved remarkably durable and versatile, yet still more vital for science is his exemplary pursuit of insight and beauty."



Otto Stern's seminal Molecular Beam experiments

- The experimental verification of the mean velocity of a Maxwell-Boltzmann velocity distribution via centrifugal deflection of a molecular beam (Frankfurt, 1919-1920)
- The Stern-Gerlach experiment (SGE), carried out with Walther Gerlach (Frankfurt, 1920-1922)
- The three-stage SGE experiment, carried out together with Thomas Phipps, Otto Robert Frisch, and Emilio Segrè (Hamburg, 1932-1933)
- The experimental verification of de Broglie's relation for the wavelength of matter waves, performed with Friedrich Knauer, Immanuel Estermann, and Otto Robert Frisch (Hamburg, 1929-1933)
- The measurement of the magnetic dipole moment of the proton and deuteron, with Otto Robert Frisch, Immanuel Estermann, and Oliver Simpson (Hamburg and Pittsburgh,1933-1937)
- Experimental demonstration of momentum transfer upon absorption or emission of a photon by Otto Robert Frisch (Hamburg, 1933)
- The experimental verification of the Maxwell-Boltzmann velocity distribution via deflection of a molecular beam by gravity, with Immanuel Estermann and Oliver Simpson (Pittsburgh, 1938-1945)

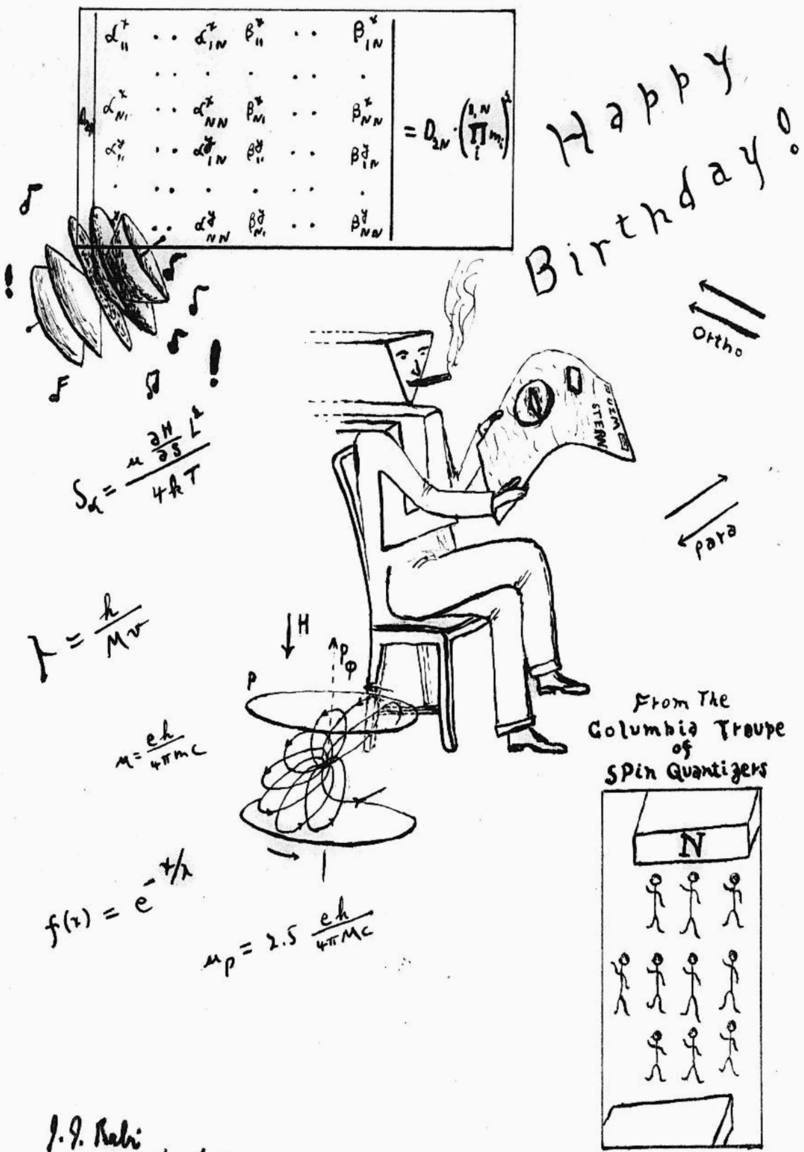
Otto's Motto

Lichtstrahlen sind zum Brechen

Molekularstrahlen sind zum Kotzen

UZM JOURNAL

Anniversary Number: $\Gamma(6) \sin^2 \frac{\pi}{4}$ February 17, 1948.



I. I. Rabi
for the troupe

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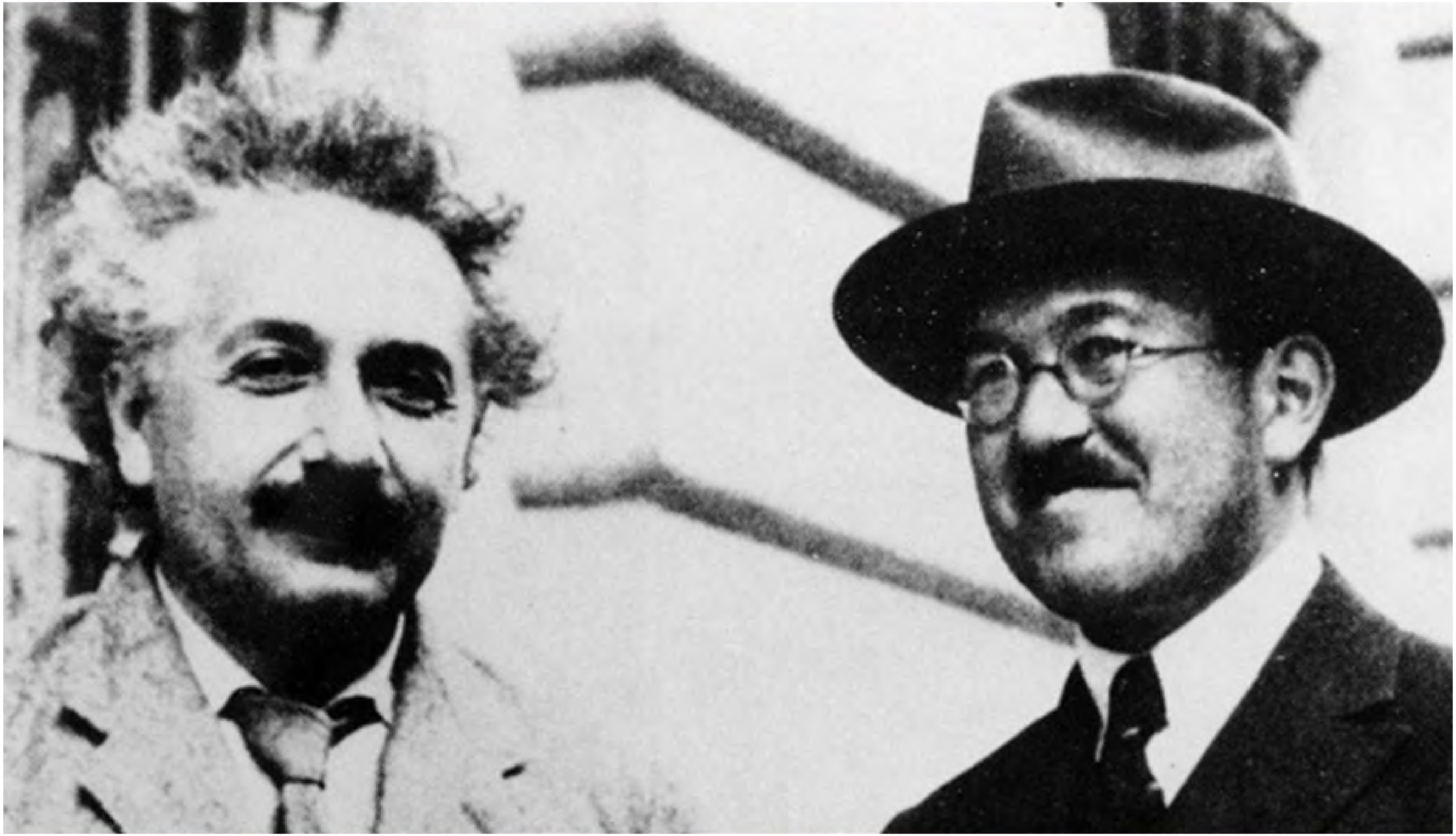
This talk:

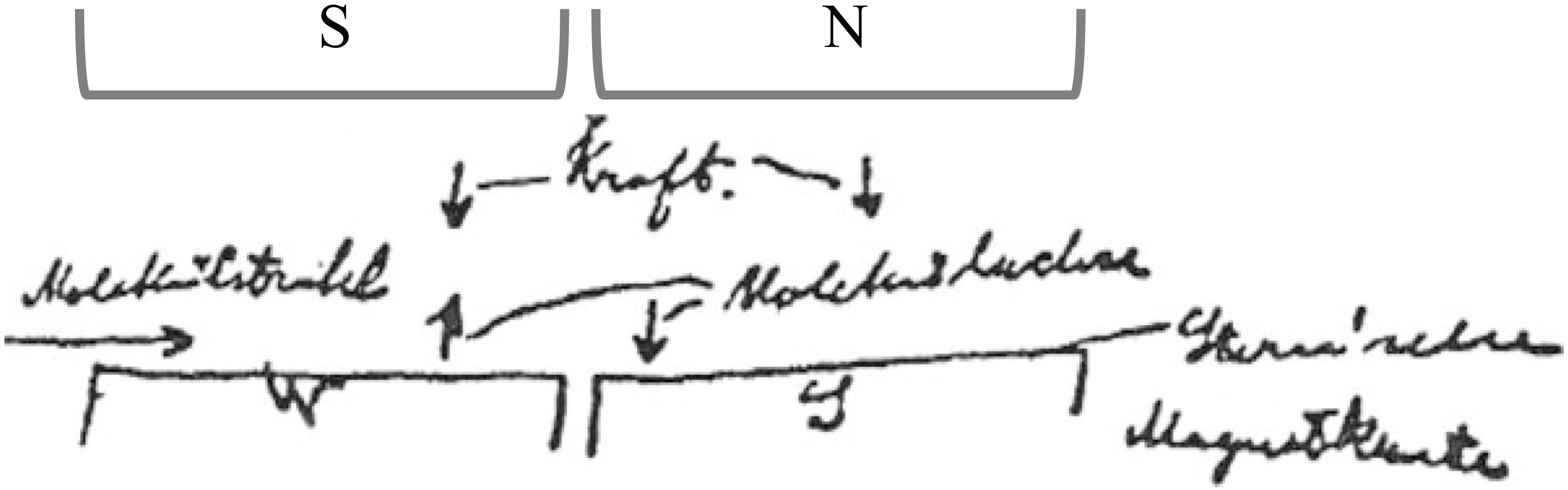
- Introduction
- The Three-Stage Stern-Gerlach experiment (Hamburg, 1933)
- Rabi's Molecular Beam Magnetic Resonance method (Columbia, 1938)
- Ramsey's Separated Oscillatory Fields method (Harvard, 1949)
- Conclusions



Albert Einstein and Otto Stern during a meeting in the mid-1920s

Einstein to Stern (1928):

"An atomic beam passes consecutively through two oppositely oriented inhomogeneous magnetic fields. Assume that an atom is oriented in such a way as to be deflected upward in the first field. If [the atom] flips its orientation [in the region between the two fields], then, because of the reversal [of the orientation] of both the [second] field and the dipole, the beam must [be deflected by the second field] as if the two magnetic fields were oriented in the same direction."



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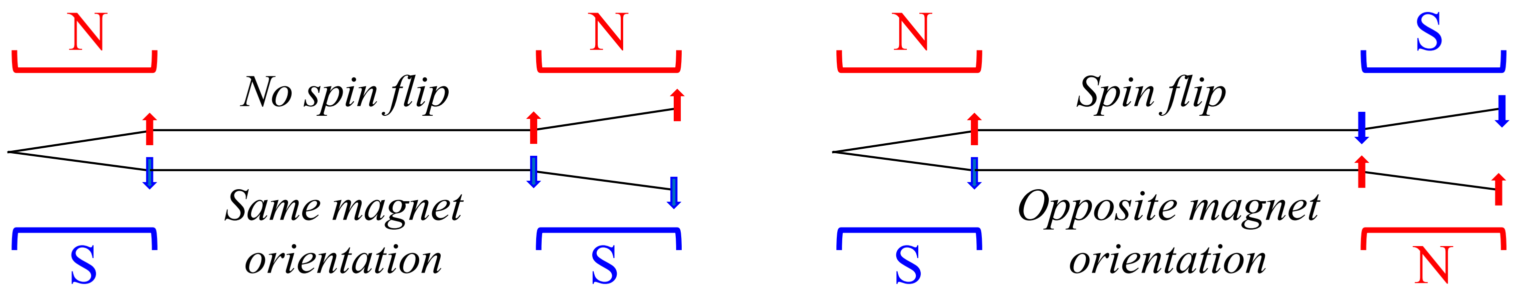

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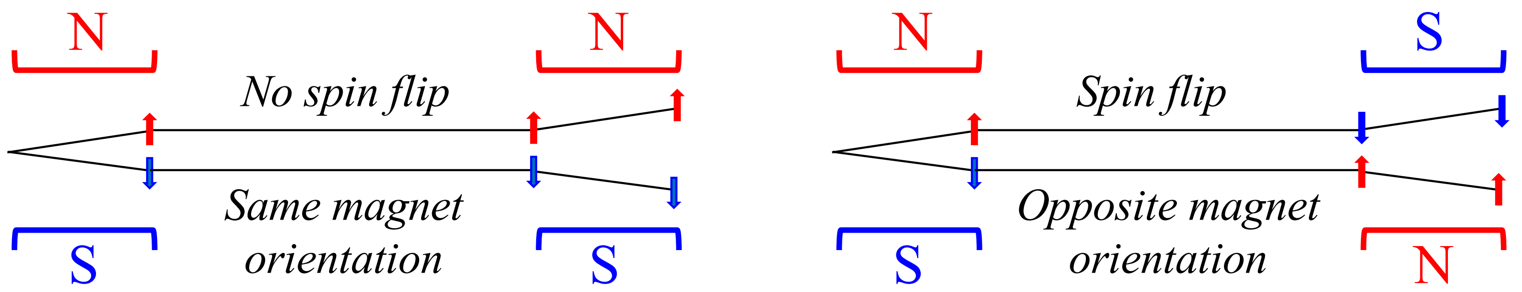
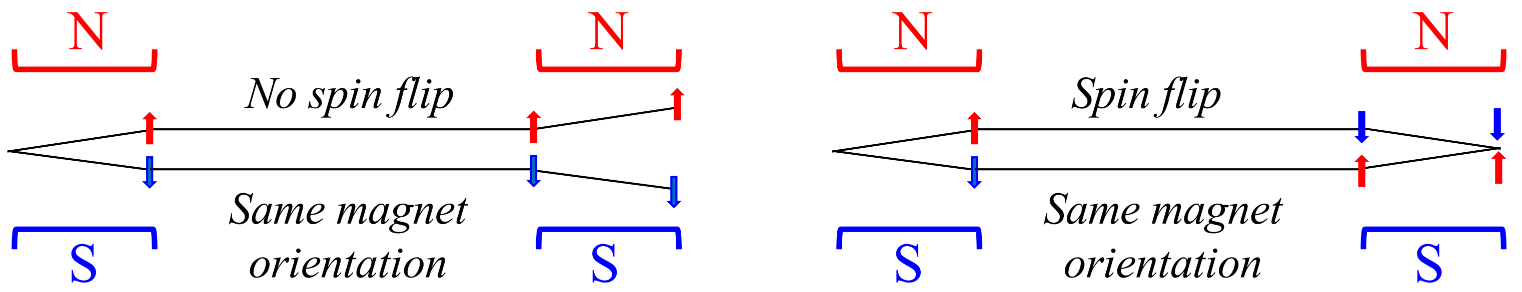


Reorientation of the atoms (i.e., a spin flip) in the intermediate region between two equally oriented magnets results in refocusing of the beam:



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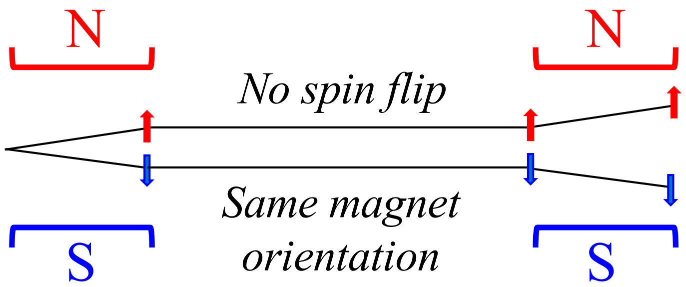

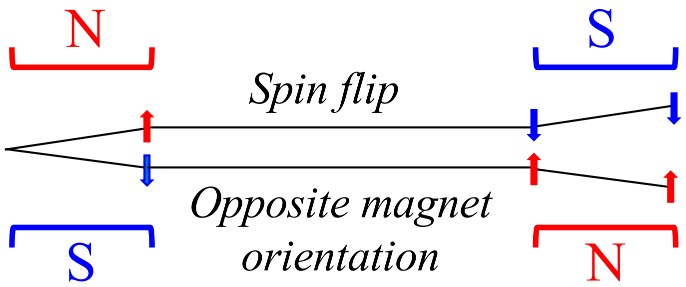

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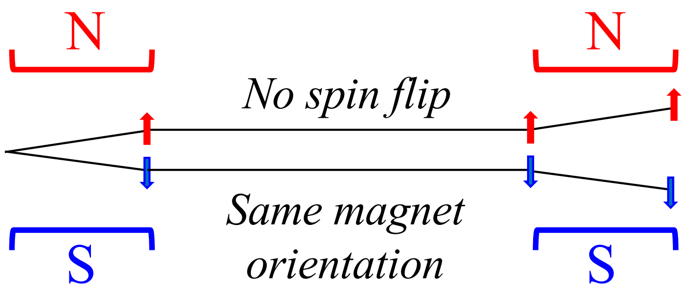
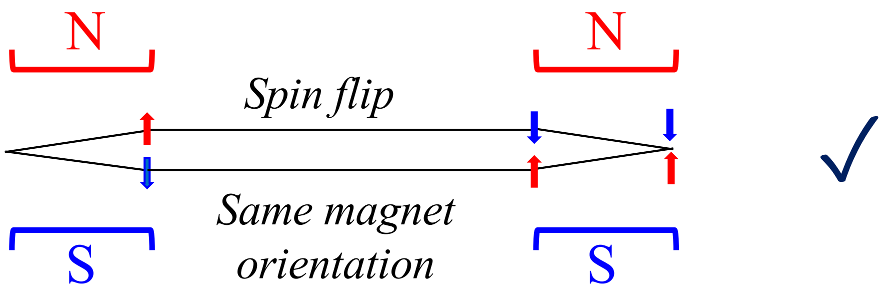




Spin flips

- Condition for spin flips was studied by a number of physicists starting in 1928

- Güttinger's adiabaticity parameter (1932): Larmor period/Interaction time

- Larmor period/Interaction time > 1 non-adiabatic behavior

  Larmor period/Interaction time ≤ 1 adiabatic behavior

Einstein (1928): "Perhaps it would be convenient to generate [a suitable] field by running [electric] current through a water-cooled pipe."

- As we'll see in a moment, Einstein's was the winning solution

The Three-Stage SGE of Phipps and Stern, 1932

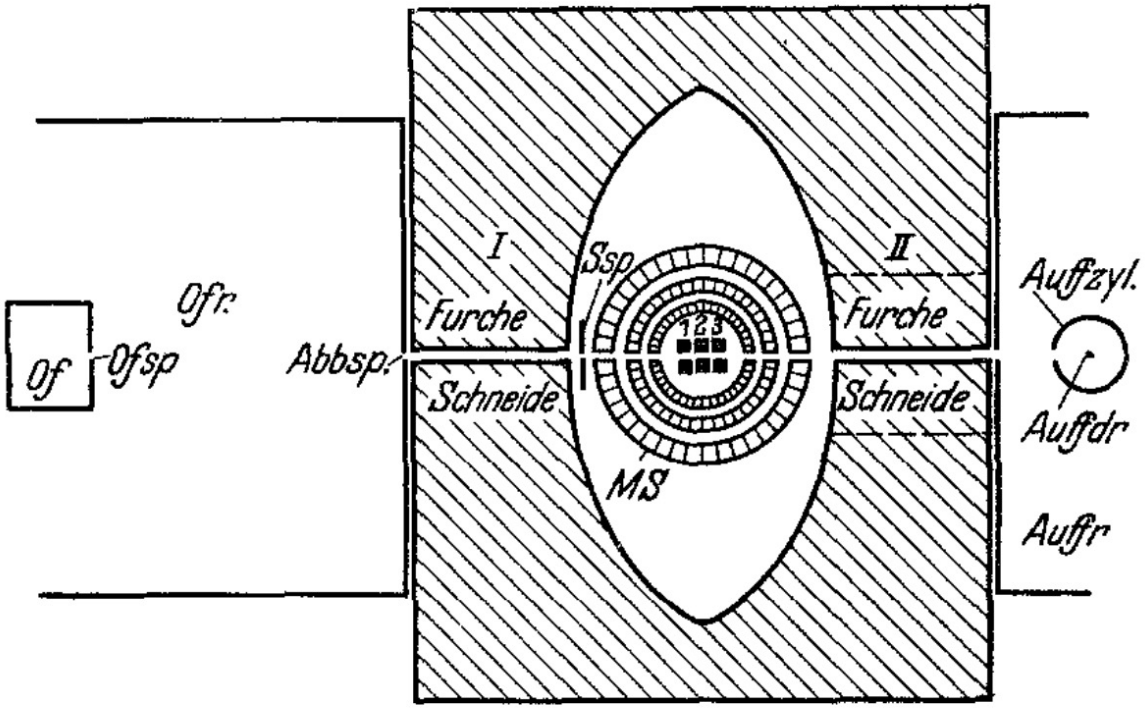


Fig. 1. *Of* Ofen; *Ofsp* Ofenspalt; *Ofr* Ofenraum: I., II. Erstes, zweites Magnetfeld; *Ssp* Selektorspalt; *MS* Magnetischer Schutz; 1, 2, 3 Kleine Magnete; *Auffzyl* Auffängerzylinder; *Auffdr* Auffängerdraht; *Auffr* Auffängerraum.

- The magnetic shield was supposed to keep the magnetic fields generated by the two Stern-Gerlach magnets (selector and analyzer) out of the region where the small magnets interacted with the spin-selected sodium beam. But, apparently, it didn't. The outcome of the Phipps-Stern experiment was negative (no spin flips observed).

The Three-Stage SGE of Frisch and Segrè, 1933

Segrè 1973: “I inherited [Phipps’s] apparatus, but could not make much headway until on reading Maxwell’s [Treatise on] Electricity [and Magnetism] I found a trick by which one could achieve a certain magnetic field configuration essential to the success of the experiment.”

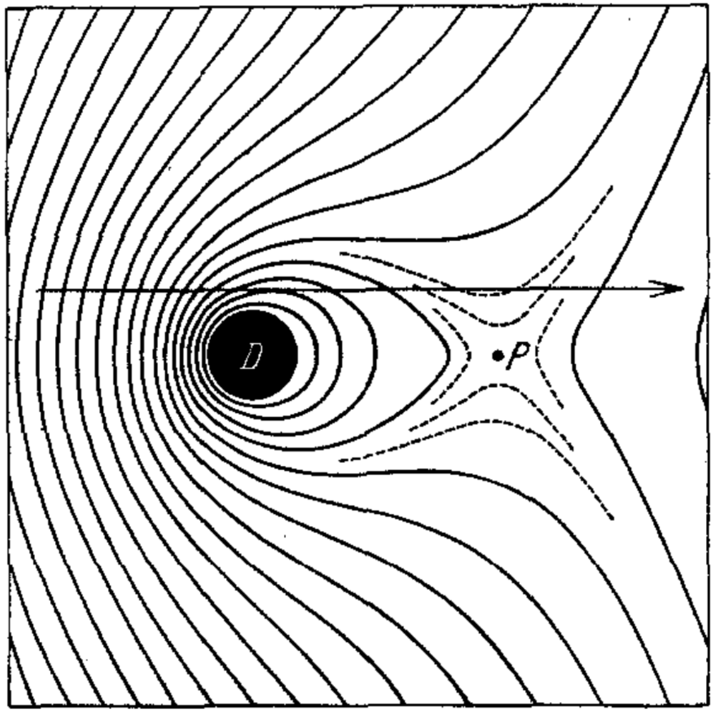


Fig. 1. Stromdurchflossener Draht im homogenen Magnetfeld: Verlauf der Kraftlinien. Der Pfeil deutet den hindurchgeschickten Molekularstrahl an.

The Three-Stage SGE of Frisch and Segrè, 1933

Schematic of the apparatus constructed by Phipps and modified by Frisch and Segrè:

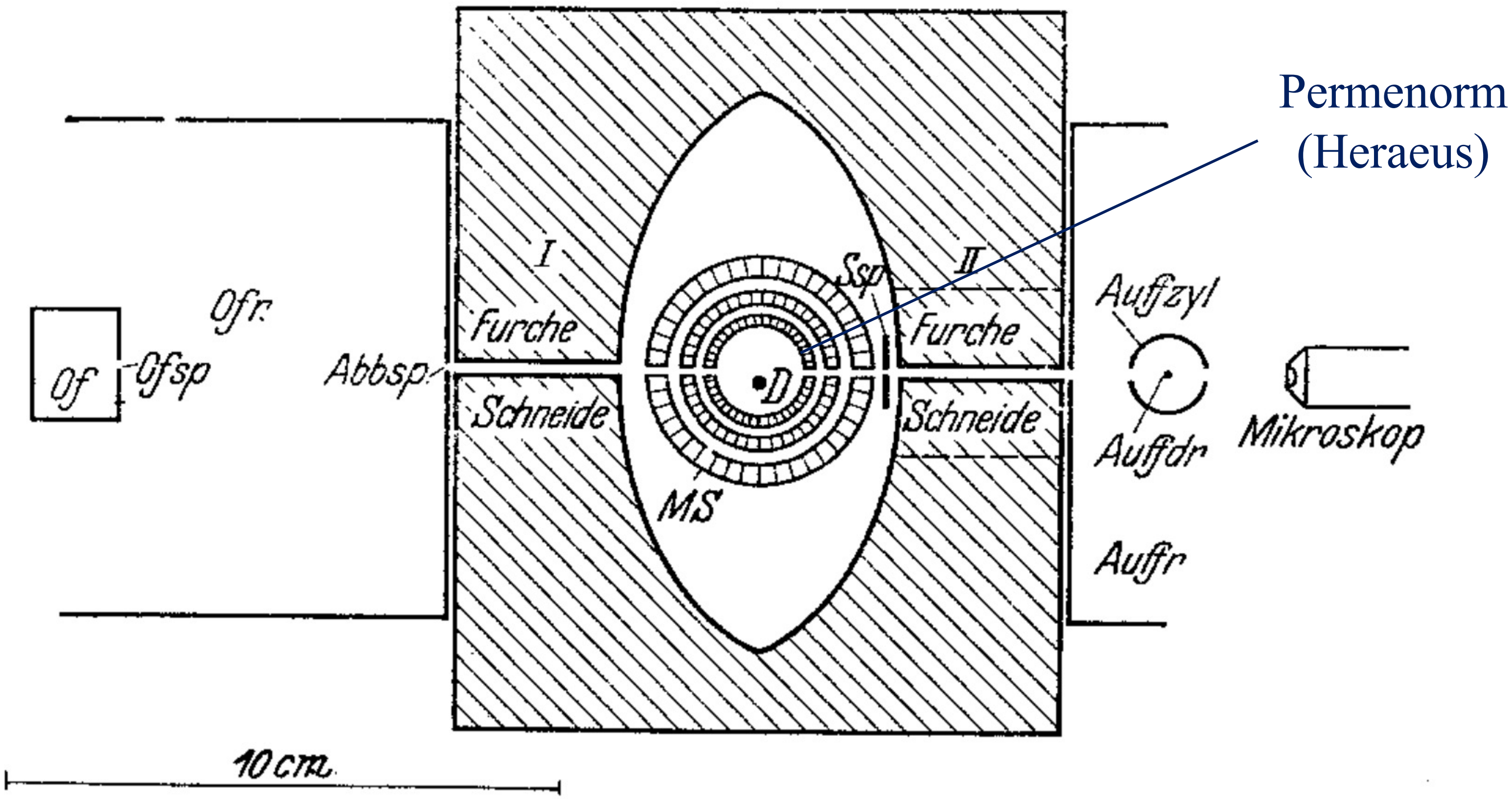


Fig. 2. *Of* Ofen; *Ofsp* Ofenspalt; *Ofr* Ofenraum: I., II. Erstes, zweites Magnetfeld; *Ssp* Selektorspalt; *MS* Magnetischer Schutz; *D* Draht zur Erzeugung des Zusatzfeldes; *Auffzyl* Auffängerzylinder; *Auffdr* Auffängerdraht; *Auffr* Auffängerraum.

The Three-Stage SGE of Frisch and Segrè, 1933

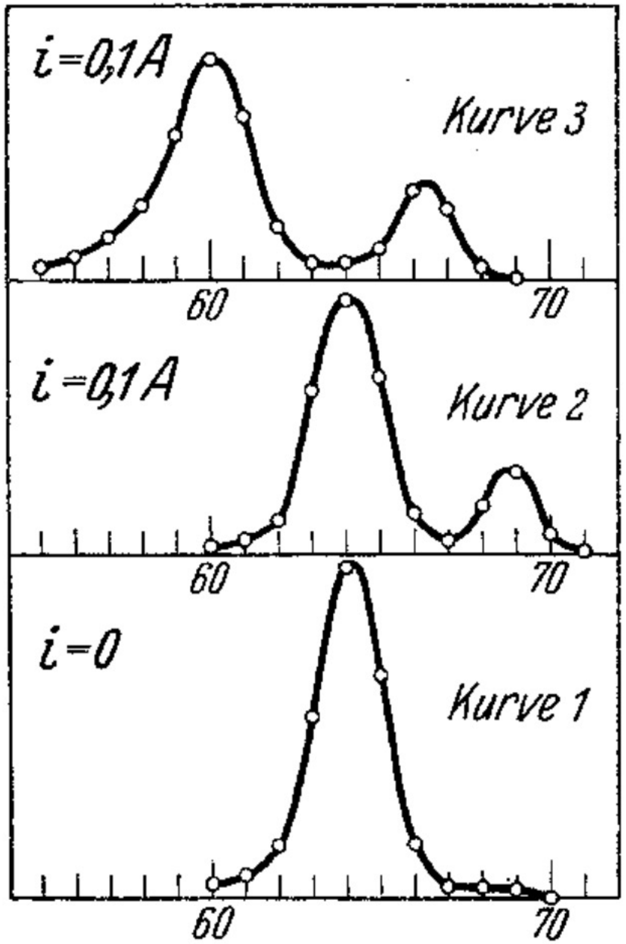


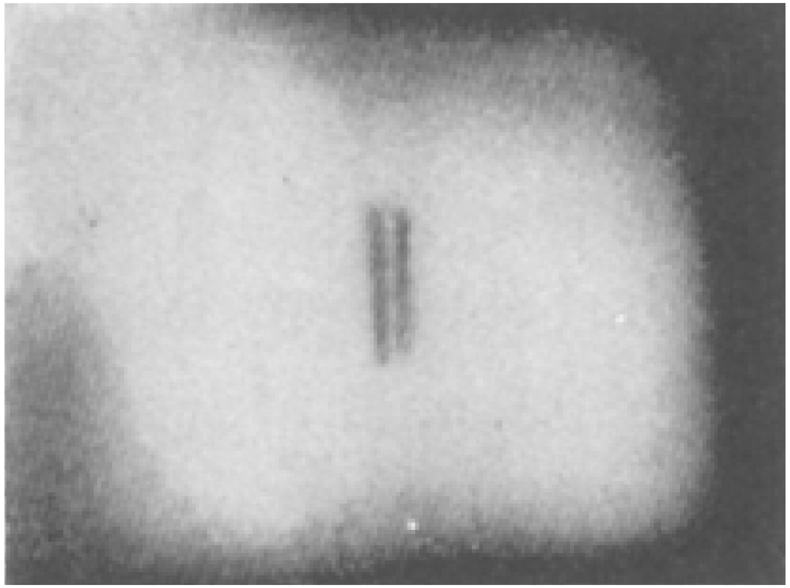
Fig. 3. Intensitätsverteilung im Strahl; das kleinere Maixmum in Kurve 2 und 3 rührt von umgeklappten Atomen her; i = Strom im Draht D.

Otto Frisch

Emilio Segrè

Isidor Rabi in Hamburg (1927-1929)

Ramsey 1993: "The Stern-Gerlach experiment … had earlier sparked Rabi's keen interest in quantum mechanics and so, while working in Hamburg with Pauli, Rabi became a frequent visitor to Stern's molecular beam laboratory. During one of these visits Rabi suggested a new form of deflecting magnetic field; Stern in characteristic fashion invited Rabi to work on it in his laboratory, and Rabi in an equally characteristic fashion accepted. Rabi's work in Stern's laboratory was decisive in turning his interest toward molecular beam research."



Splitting pattern of a beam of potassium atoms in a *homogeneous* magnetic field (Rabi, 1929)

Isidor Rabi at Columbia (1929-1988)

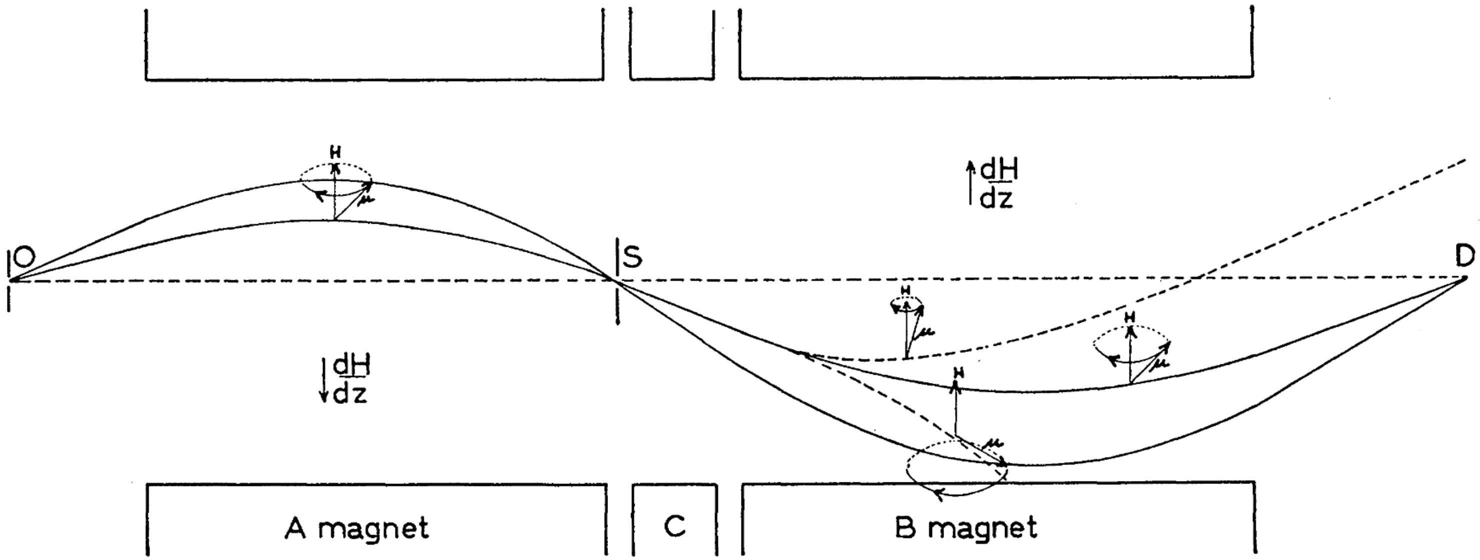
- Made spin reorientation one of the principal topics of his research
- In 1937, considered an applied field that changed its direction ("gyrated") at a fixed frequency

Ramsey 1993: "A few months after [publishing a paper about the gyrating field], following a visit by C. J. Gorter [who had a similar idea], Rabi directed the major efforts of his laboratory toward the development of the molecular beam magnetic resonance method with the magnetic fields oscillating in time."

Dan Kleppner 2012: "[Ramsey] graduated from Columbia University in 1935 with a degree in mathematics, attended Cambridge University in the UK for two years, and returned to Columbia in 1937. He joined the molecular-beams group of I. Rabi despite Rabi's admonition that molecular-beam research was pretty [much] exhausted. A few months later Rabi's invention of molecular-beam magnetic resonance triggered a revolution in atomic physics."

Isidor Rabi at Columbia (1929-1988)

- Rabi's molecular-beam magnetic resonance method

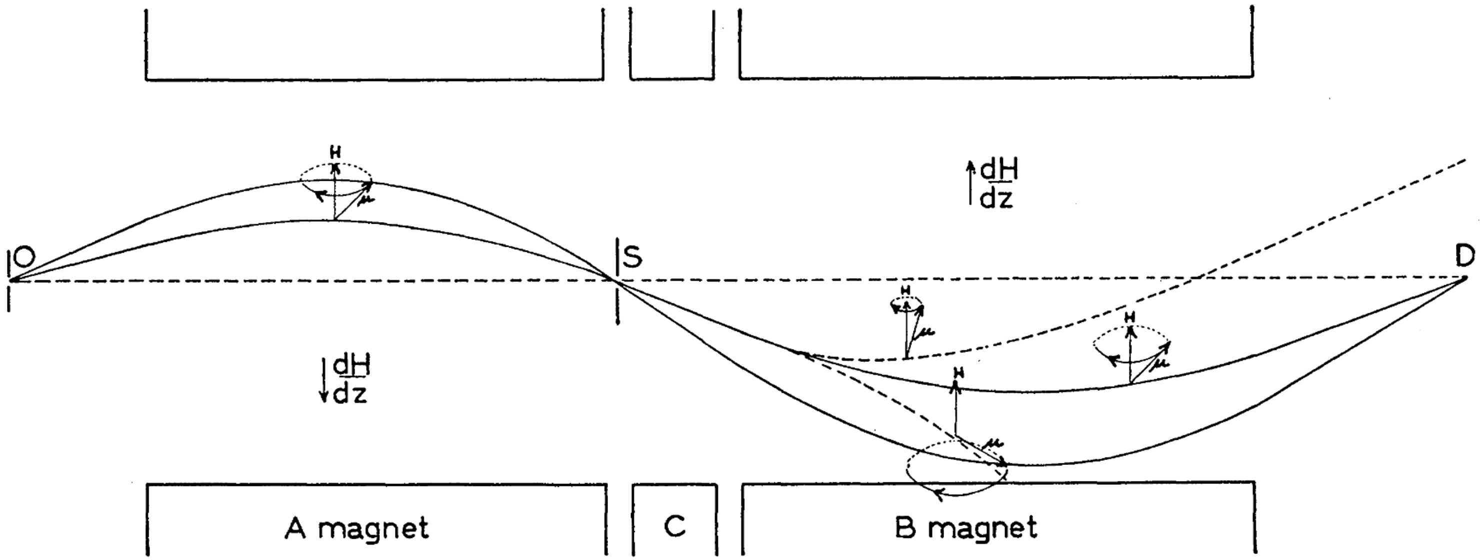


Rabi et al., Phys. Rev. 55, 526 (1939)

- A molecular beam of the substance to be studied runs along an S-shaped path through oppositely oriented inhomogeneous magnetic fields A and B and is focused on a suitable detector. A third magnet, C, which produces a strong homogeneous field H_0, is placed in the intermediate region. In its field, the nuclear moments μ are decoupled from other moments (nuclear and rotational) of a $^1\Sigma$ molecule, and precess with a Larmor frequency $\nu_0 = \mu H_0/(hJ)$. An RF field of strength H_1 perpendicular to the C field and embedded in it produces transitions to other states of space quantization when the frequency f of this field is close to ν. If such transitions take place the molecule is no longer focused onto the detector by the B magnet and the beam intensity diminishes.

Isidor Rabi at Columbia (1929-1988)

- Rabi's molecular-beam magnetic resonance method



Rabi et al., Phys. Rev. 55, 526 (1939)

- The two solid curves indicate the paths of two molecules having different magnetic moments and different velocities and whose moments are not changed during passage through the apparatus. This is indicated by the small gyroscopes drawn on one of these paths, in which the projection of the magnetic moment along the field remains fixed. The two dotted curves in the region of the B magnet indicate the paths of two molecules the projection of whose nuclear magnetic moments along the field has changed in the region of the C magnet. This is indicated by means of two gyroscopes, for one of which the projection of the magnetic moment along the field has increased and for the other decreased.

Isidor Rabi at Columbia (1929-1988)

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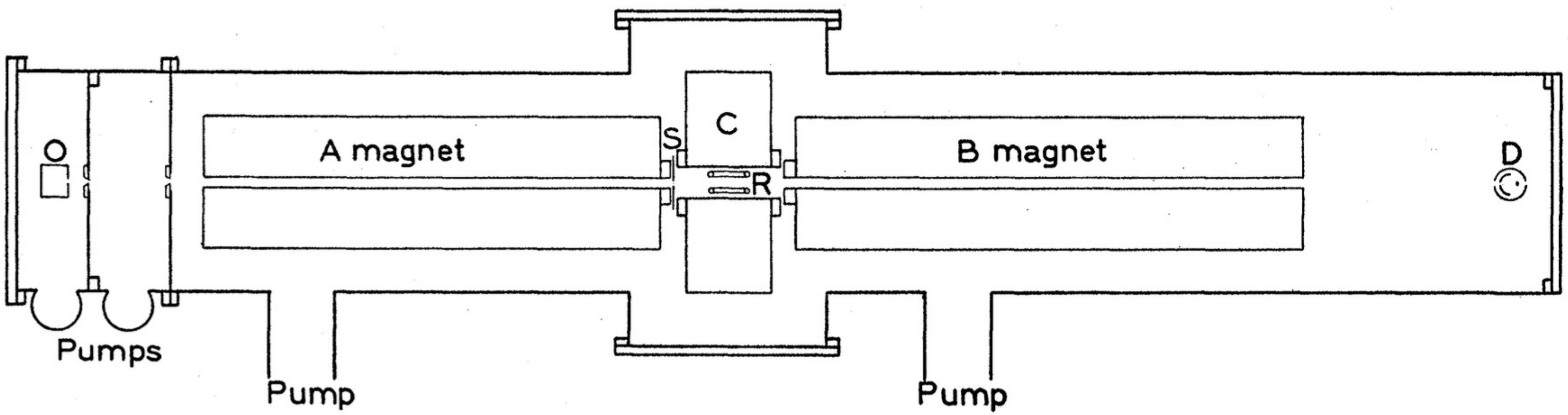


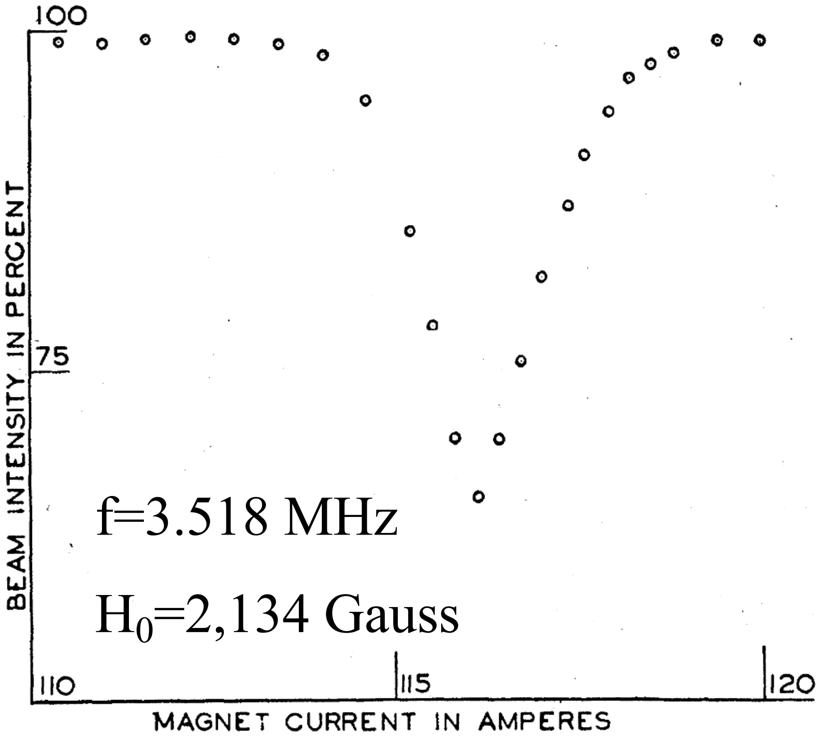
Rabi et al., Phys. Rev. 55, 526 (1939)

- If initially H_1 is perpendicular to both H_0 and the angular momentum J, the additional precession of J about H_1 will result in changing the angle between J and H_0. If the frequency f of H_1 comes close to the Larmor frequency ν, the effect will be cumulative, eventually resulting in a spin flip.

- The smaller the ratio H_1/H_0, the sharper the feature in dependence on the frequency matching, $|f-\nu_0|$.

Isidor Rabi at Columbia (1929-1988)

- Rabi's molecular-beam magnetic resonance method



The first result: Resonance curve of the ^{7}Li nucleus observed in LiCl in January 1938

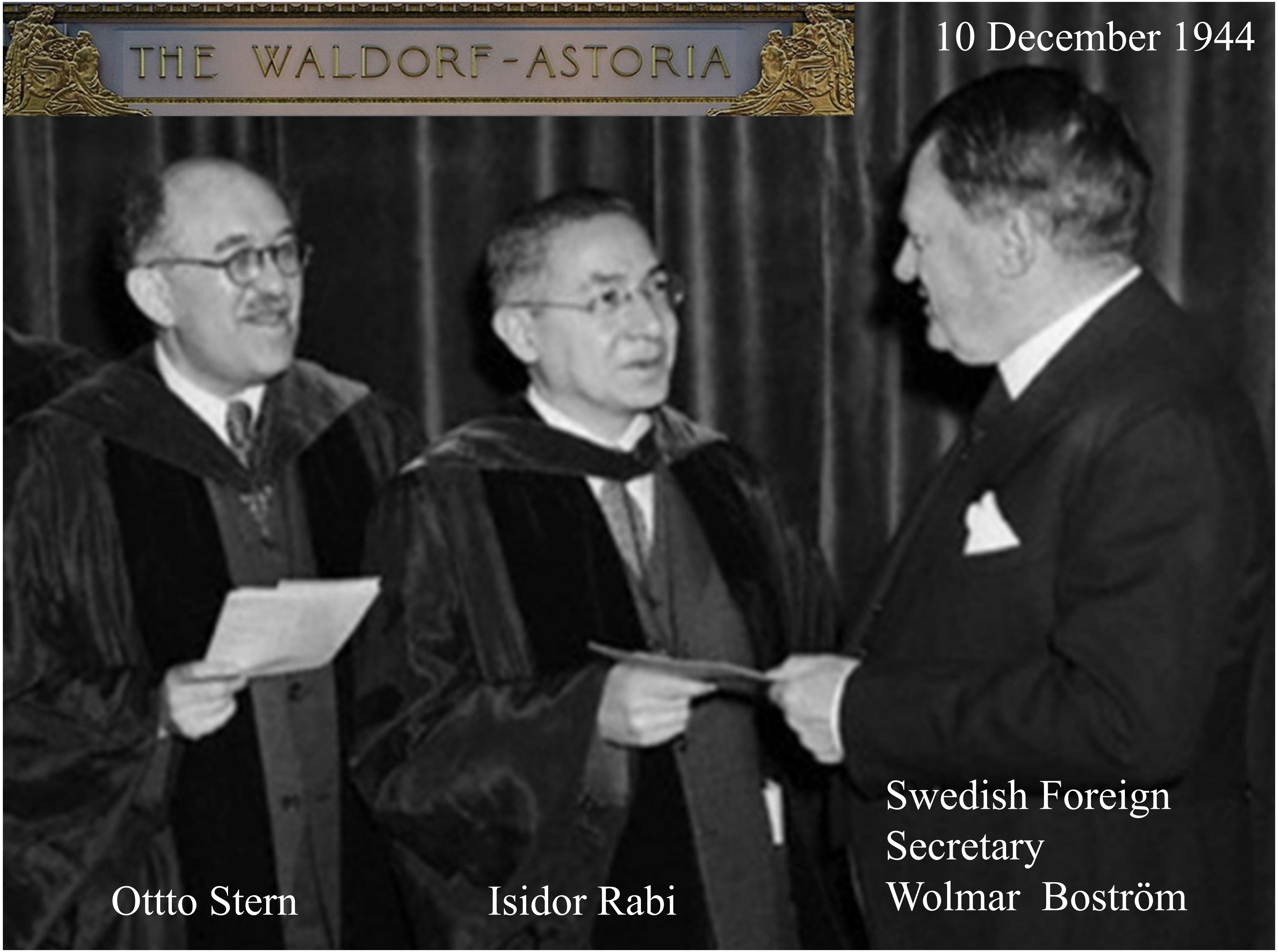
$$g=(\mu_N I)^{-1} f/H_0$$

$$\mu=g\ \mu_N\ J$$

Rabi et al., Phys. Rev. 53, 318 (1938)

Polykarp Kusch: "Rabi was beside himself."

John Rigden: "Although even at that time, January 1938, it was likely that the vents of the day might be crowned with a Nobel Prize, no one could have foreseen how important the magnetic resonance method would prove to be – for not only physics but also chemistry, biology, medicine, and the whole of science."


THE WALDORF-ASTORIA
10 December 1944
Swedish Foreign
Secretary
Wolmar Boström
Ottto Stern
Isidor Rabi

LETTRE DU PRÉSIDENT DES ÉTATS UNIS D'AMÉRIQUE

The White House, Washington. December 6, 1944.

HENRY GODDARD LEACH, Esq.
President of the Board
The American Scandinavian Foundation
116 East 64 Street
New York 22, N. Y.

Dear Mr. Leach:

It is of particular interest to me to learn that two residents of the United States, Professor OTTO STERN and Professor ISIDOR ISAAC RABI, have been selected to receive the 1943 and 1944 Nobel prizes for Physics. I am also glad to note that the 1943 and 1944 Nobel prizes for Physiology and Medicine are shared by four residents of this country, Professor HENRIK DAM and Professor EDWARD A. DOISY, Professor JOSEPH ERLANGER and Professor HERBERT S. GASSER. Here in America we have always believed that science should be a servant of the people. I am confident that after this war the scientists of all nations will again contribute their talents to a world body of knowledge, useful alike to all people. The Nobel prizes for the past 43 years have fostered this concept that science and art are builders of peace.

Very sincerely yours,
(Signed) FRANKLIN D. ROOSEVELT

Adapted from Daniel Kleppner's chapter
in *Molecular Beams in Physics and Chemistry*

Aspect … Dalibard→**Cohen-Tannoudji→Kastler … Stern/Rabi**

Bloch … Stern/Rabi

Bloembergen→Purcell→Stern/Rabi

Chu→Commins→**Stern/Rabi**

Clauser→Thaddeus→ **Townes … Stern/Rabi**

Cohen-Tannoudji→Kastler … Stern/Rabi

Cornell→ Pritchard → Kleppner →**Ramsey→Stern/Rabi**

Glauber→ Schwinger … Stern/Rabi

Hänsch →Schawlow→Townes … Stern/Rabi

Haroche→Cohen-Tannoudji →Kastler

Cornell→Pritchard → Kleppner/**Ramsey →Stern/Rabi**

Herschbach … Ramsey→Stern/Rabi

Kastler … Stern/Rabi

Ketterle→ Pritchard → Kleppner →**Ramsey→Stern/Rabi**

Kusch→Stern/Rabi

Lamb … Stern/Rabi

Phillips→Kleppner→**Ramsey→Stern/Rabi**

Purcell … Stern/Rabi

Ramsey→Stern/Rabi

Schawlow→Townes … Stern/Rabi

Schwinger … Stern/Rabi

Townes … Stern/Rabi

Weiss→ Zacharias→**Stern/Rabi**

Wieman→Hänsch→Schawlow→Townes … Stern/Rabi

Wineland→Ramsey→Stern/Rabi

Zeilinger→Rauch→Bonse→Kappler→Gerlach

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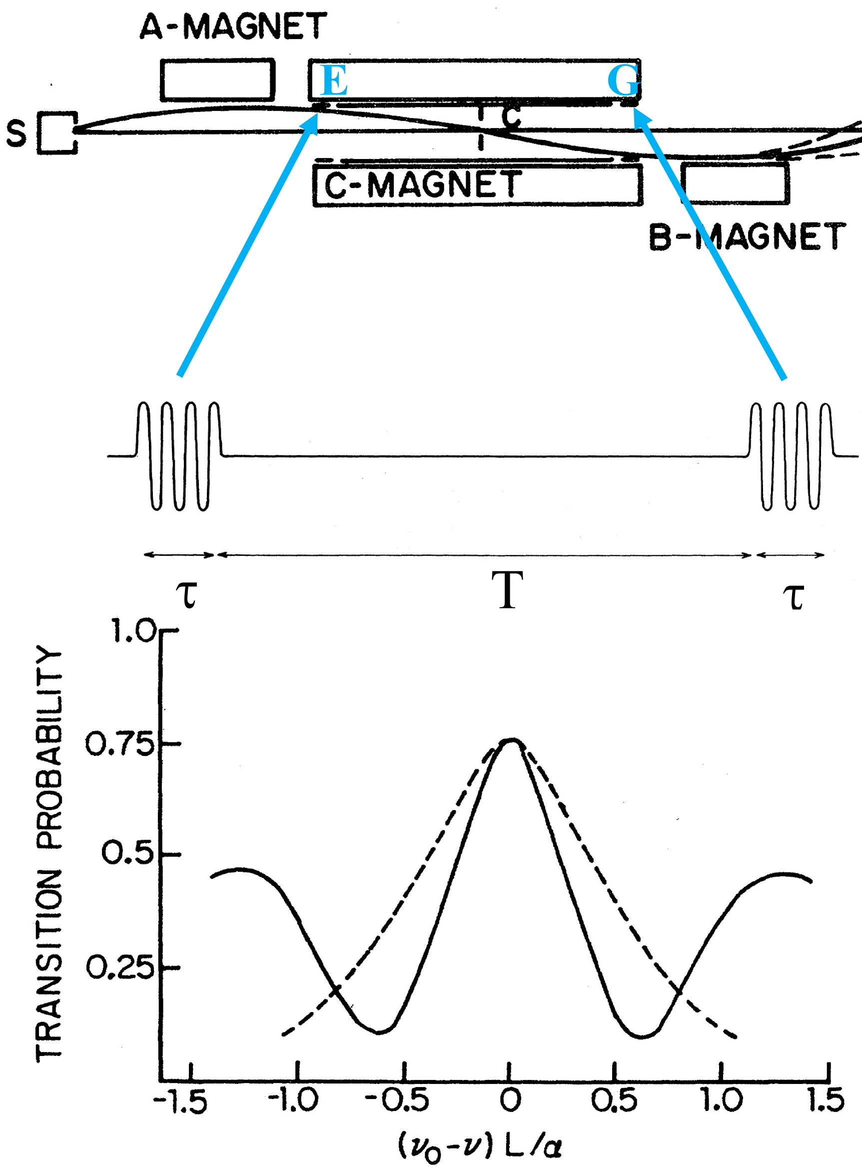
Townes … Stern/Rabi
Weiss→ Zacharias→**Stern/Rabi**
Wieman→Hänsch→Schawlow→Townes … Stern/Rabi
Wineland→Ramsey→Stern/Rabi
Zeilinger→Rauch→Bonse→Kappler→Gerlach

Norman Ramsey at Harvard (1947-2011)

- Ramsey's Separated Oscillatory Fields method, 1949



$\alpha = \langle v \rangle$

$v = L/T$

$\nu_0 = \mu H_0/(hJ)$

Ramsey, Rev. Modern Phys. 62, 541 (1990)

Norman Ramsey at Harvard (1947-2011)

- Ramsey's Separated Oscillatory Fields method, 1949

Dan Kleppner: "Today the factor of two in intrinsic resolution seems negligible compared with the factors of hundreds or thousands that have been achieved [in the meantime] using the Ramsey method … The Ramsey method continues to greatly influence atomic physics and is found in one form or another in most high-precision measurements, advanced atomic clocks, and experiments on atom entanglement and cavity quantum electrodynamics."

Princess Lilian of Sweden and Norman F. Ramsey at the Nobel Prize banquet, 10 December 1989.

Molecular Beams opened up novel perspectives and brought about wide-ranging and far-reaching applications of Quantum Science

- NMR/MRI, optical pumping, the laser, atomic clocks
- Magnetic moment of the proton, deuteron, and neutron; Lamb shift and the anomalous increment in the magnetic moment of the electron → QED
- Chemical reaction dynamics in the gas phase and on surfaces
- Electrospray and spectroscopy in superfuid He nanodroplets
- Laser cooling of atoms and now of molecules; quantum degenerate gases
- Examination and quantum simulation of condensed-matter systems, prototype quantum computers
- Metrology, including tests of fundamental symmetries and searches for dark matter

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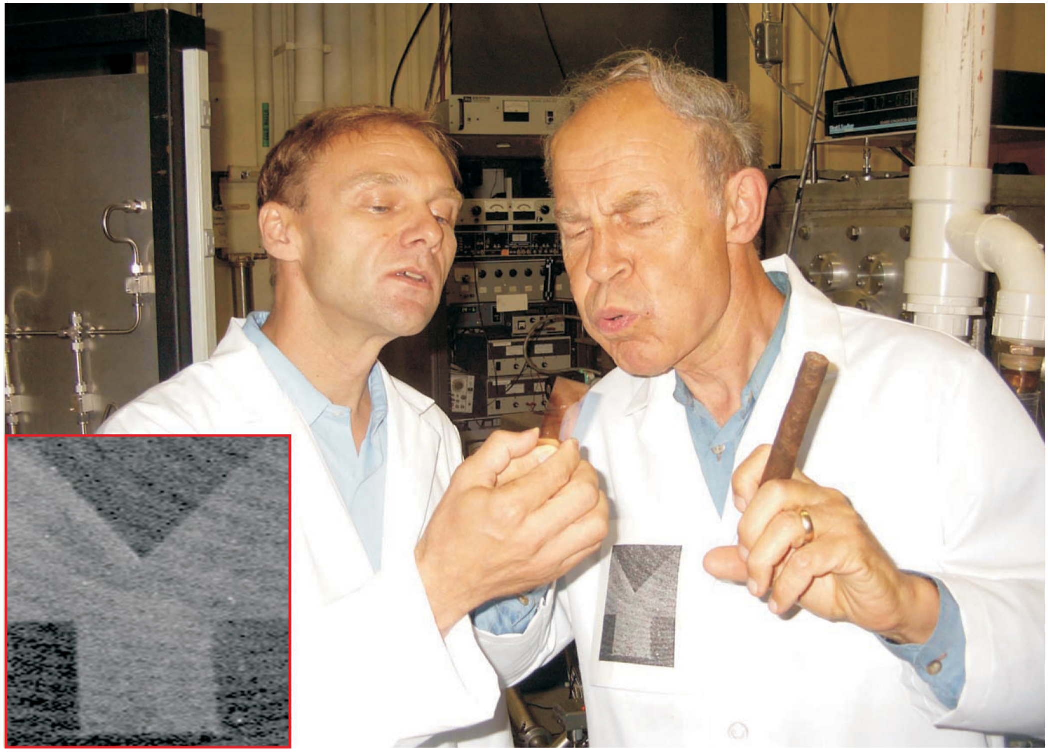
Quantum Science owes more to molecular beams than molecular beams to Quantum Science …


THE END IS NEAR

From the treasure trove of stories about how experiments work

Otto Stern as told to Dudley Herschbach in about 1960: “After venting to release the vacuum, Gerlach removed the collector plate. But he could see no trace of the silver atom beam and handed the plate to me. With Gerlach looking over my shoulder as I peered closely at the plate, we were surprised to see gradually emerge the trace of the beam … Finally, we realized what [had happened]. I was then the equivalent of an assistant professor. My salary was too low to afford good cigars, so I smoked bad cigars. These had a lot of sulfur in them, so my breath on the plate turned the silver into silver sulfide, which is jet black, so easily visible. It was like developing a photographic film.”

Edwin Land, 1980s: “I don’t believe it!”



Reenactment of the cigar episode, 2002:

Reenactment of the cigar episode, 2002:

- using partly masked glass slides with a monolayer of vacuum-deposited silver

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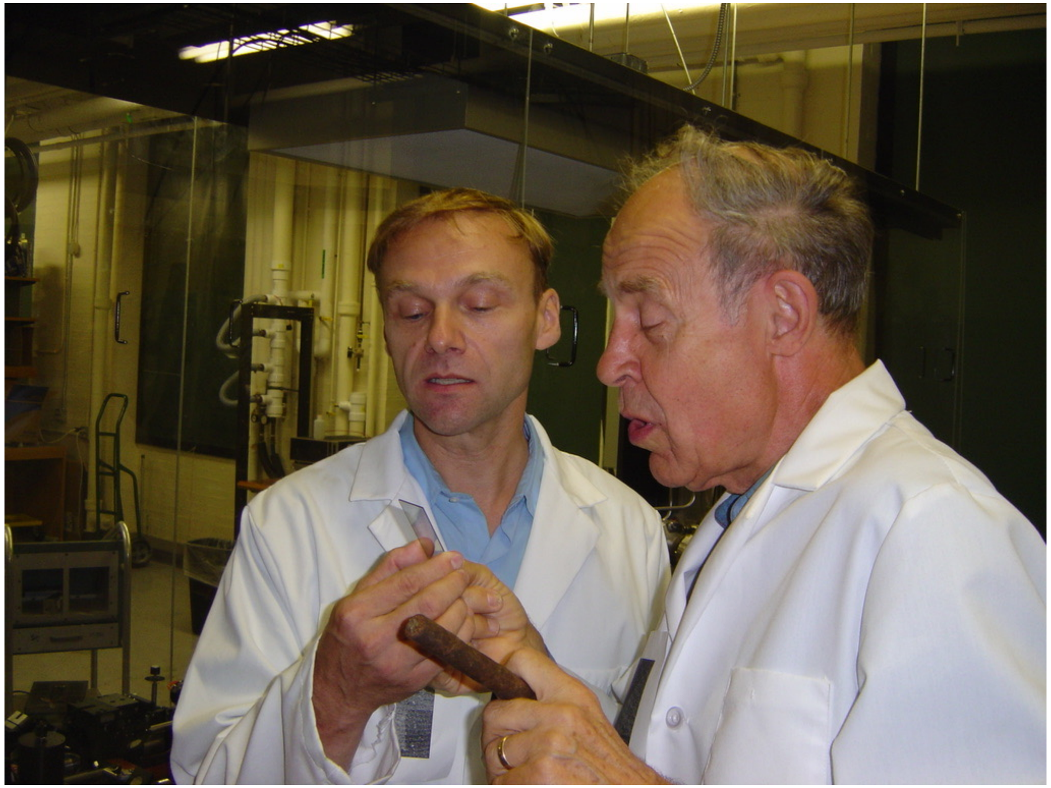
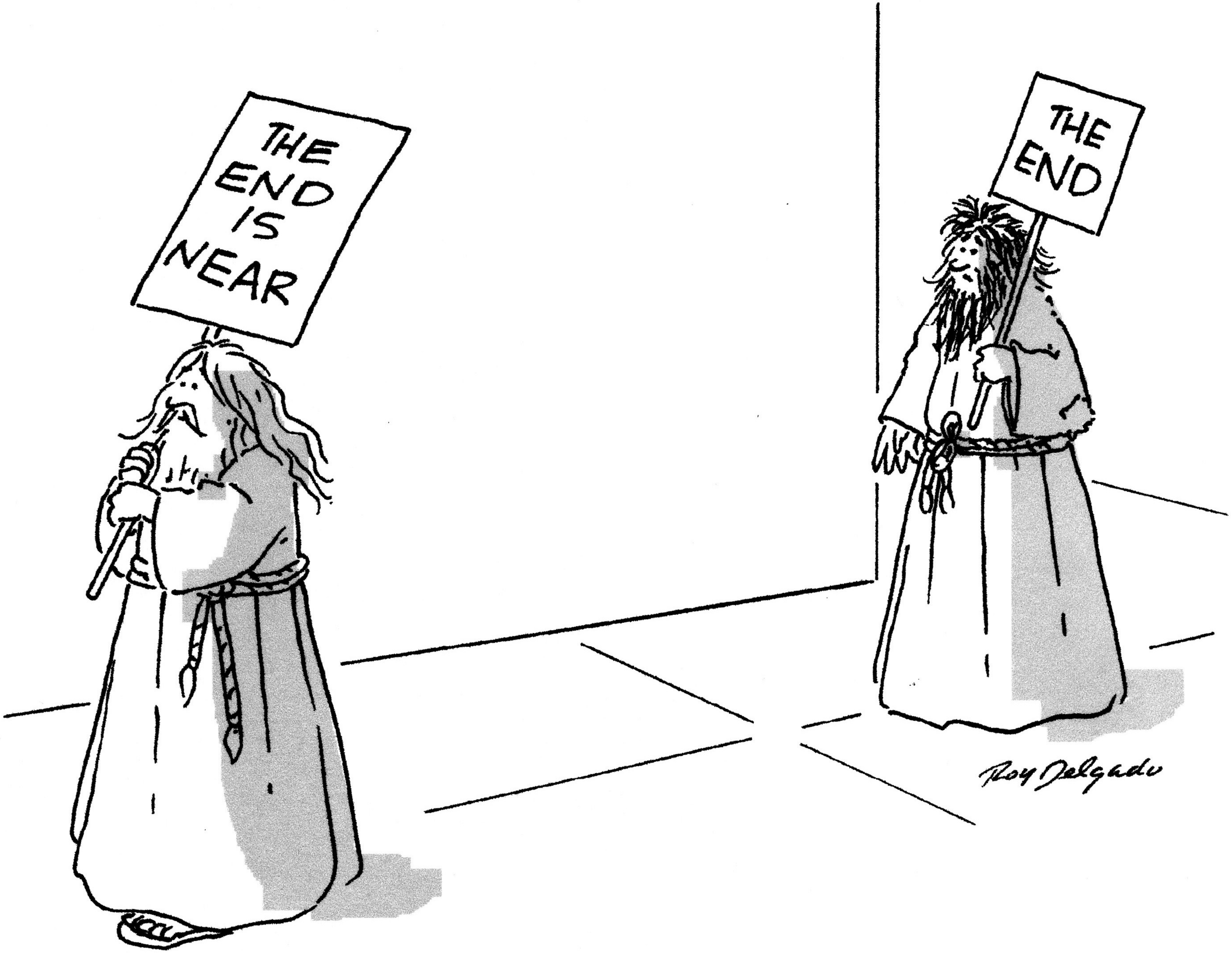


Reenactment of the cigar episode, 2002:

- using partly masked glass slides with a monolayer of vacuum-deposited silver
- merely exhaling cheap-cigar-tainted breath on the slide had no discernible effect
- direct exposure to cigar smoke quickly blackened the slide, within a few seconds to minutes


Dudley
Herschbach


THE END IS NEAR
THE END
Roy Delgado

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I'm grateful for discussions, past or present, about the history of molecular beams to Dudley Herschbach, Horst Schmidt-Böcking, Peter Toennies, and Daniel Kleppner.

A century ago the Stern-Gerlach experiment ruled unequivocally in favor of Quantum Mechanics

BF Bretislav Friedrich

Abstract

In 1921, Otto Stern conceived the idea for an experiment that would decide between a classical and a quantum description of atomic behavior, as epitomized by the Bohr-Sommerfeld-Debye model of the atom. This model entailed not only the quantization of the magnitude of the orbital electronic angular momentum but also of the projection of the angular momentum on an external magnetic field-the so-called space quantization. Stern recognized that space quantization would have observable consequences: namely, that the magnetic dipole moment due to the orbital angular momentum would be space quantized as well, taking two opposite values for atoms whose only unpaired electron has just one quantum of orbital angular momentum. When acted upon by a suitable inhomogeneous magnetic field, a beam of such atoms would be split into two beams consisting of deflected atoms with opposite projections of the orbital angular momentum on the magnetic field. In contradistinction, if atoms behaved classically, the atomic beam would only broaden along the field gradient and have maximum intensity at zero deflection, i.e., where there would be a minimum or no intensity for a beam split due to space quantization. Stern anticipated that, although simple in principle, the experiment would be difficult to carry out-and invited Walther Gerlach to team up with him.

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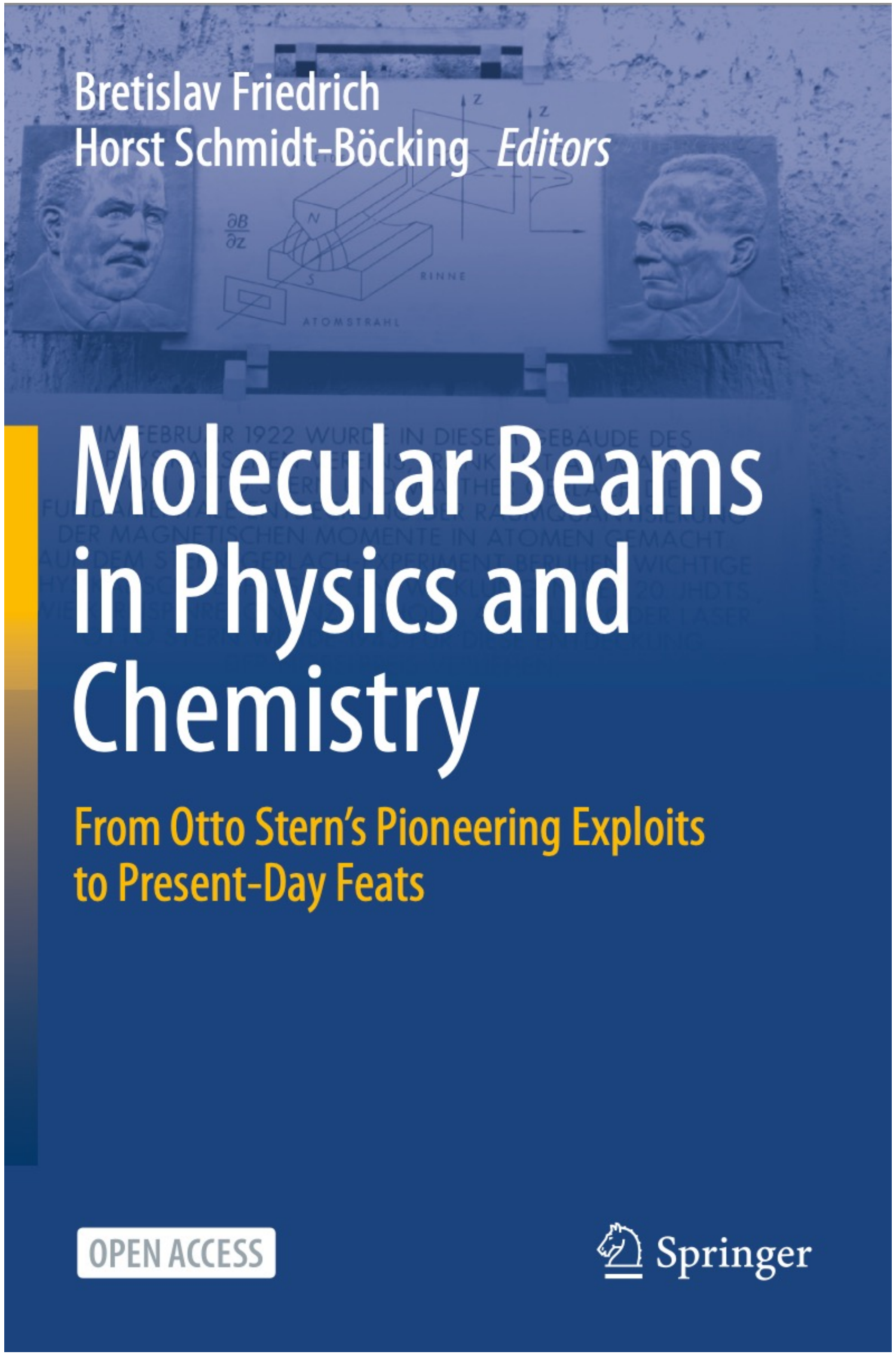

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