# From CEA/DN2 to ILL/D5 : the birth of a vocation

# Francis to-day

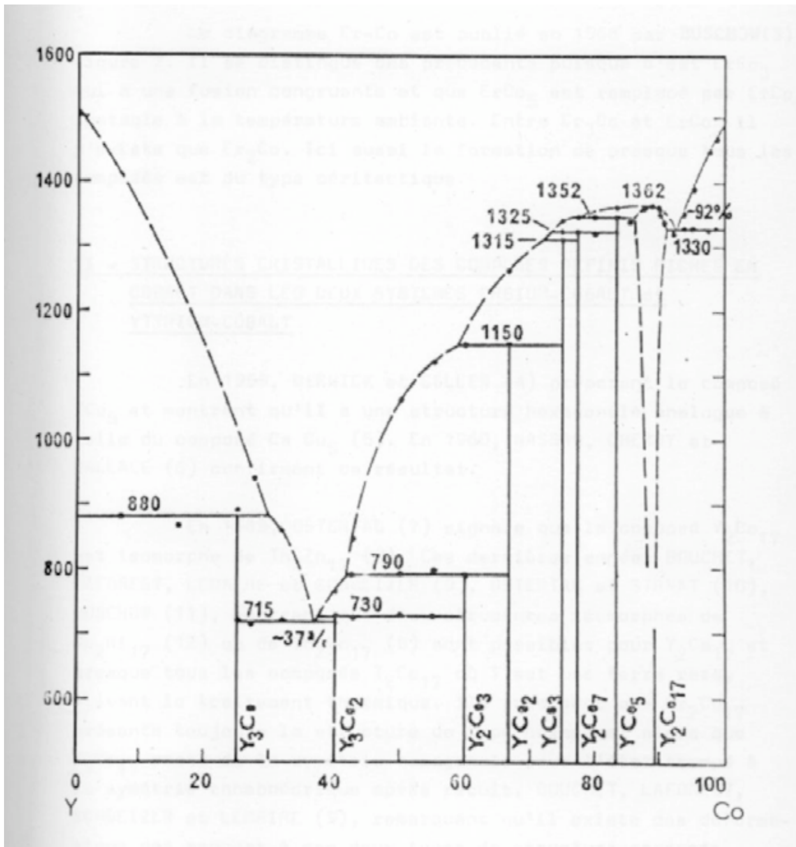
- Francis

  when he appeared in CENG, in Spring 1967

  in the laboratory of Prof Bertaut (Laboratoire de Diffraction Neutronique)

  to prepare a «thèse de 3eme cycle »

# Bertaut decided Francis will work with me…




- I was working on the RE-Co intermetallic compounds,
- in the Co rich part of the diagramme

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## Magnetic Moment Measurements in Single-Crystal Dy, Ho, Er, and Gd in dc Fields up to 170 kG

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Magnetic-moment measurements are presented for single-crystal[1] Dy, Ho, Er and Gd with applied fields up to 170 kG. These results, obtained for various crystallographic orientations with a low frequency VSM developed for use in the NML dc solenoids, extend and augment a number of earlier measurements, including the recent pulsed field data of Flippen.[2] The magnetic data are complex as is expected from the known magnetic structures at low fields. Briefly, at 4.2°K and with $B_0 \parallel C$ we find: (a) Dy–M increases slowly up to 170 kG; (b) Ho–M increases rapidly up to 12 kG, then more slowly, and there is a further rapid increase (transition) at about 130 kG; (c) Er–M increases rapidly up to 30 kG then approaches saturation more slowly; (d) Gd–M increases rapidly up to 25 kG and then is almost constant. For $B_0 \perp C$ we find: (a) Dy–M increases rapidly at low fields and approaches saturation at high fields; (b) Ho–M increases rapidly at low fields and approaches saturation at high fields; (c) Er—broad transitions occur at about 16 and 120 kG; (d) Gd—the results are similar to those for $B_0 \parallel C$. Except for Gd, the magnetic moments generally show an appreciable increase up to our maximum fields. Magnetic-moment measurements have also been completed for these materials at 77°K. The results will be published elsewhere.

* Supported by the U.S. Air Force Office of Scientific Research.
[1] Dr. J. L. Moriarty of the Lunex Co. kindly furnished some of these single crystals.
[2] R. B. Flippen, J. Appl. Phys. 35, 1047 (1964).

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## Magnetocrystalline Anisotropy of Two Yttrium–Cobalt Compounds

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The magnetocrystalline constants of two ferromagnetic rare-earth–cobalt intermetallic compounds, $YCo_5$ and $Y_2Co_{17}$, have been measured in the approximate temperature range 20°–300°K. The anisotropy constants were obtained from magnetization vs applied-field curves measured on single-crystal spheres in the principal crystallographic directions of the hexagonal structure. $YCo_5$ has a magnetically easy *c* axis and no anisotropy in the hard basal plane. $K_1$ increases from $7.03\times10^6$ erg/g at 300°K to $10.03\times10^6$ erg/g at 24°K. $Y_2Co_{17}$ has an easy basal plane with no anisotropy in the plane. At 307°K, $K_1=-3.61\times10^5$ erg/g, and $K_2=0.22\times10^5$ erg/g while at 21°K, $K_1=-6.99\times10^5$ erg/g and $K_2=-0.69\times10^5$ erg/g.

### EXPERIMENTAL

SINGLE crystals were grown by a zone-melting technique[1] and were ground into spheres 1–2 mm diameter. Location and identification of the crystallographic axes and alignment of the spheres was accomplished magnetically and checked by x-ray diffraction. In extension of work previously reported,[2] magnetization curves were measured in the principal crystallographic directions over the approximate temperature range 20°–300°K with a continuously recording magnetometer[3] used in conjunction with a 50 kOe superconducting solenoid. The anisotropy constants (in the expression $E_k=K_1\sin^2\theta+K_2\sin^4\theta+K_3\sin^6\theta+K_4\sin^6\theta\cos6\alpha$, appropriate to a material with a hexagonal crystal structure) were determined by finding the area between magnetization curves for different crystallographic directions, by the method of Sucksmith and Thompson,[4] and, in the case of $YCo_5$, by

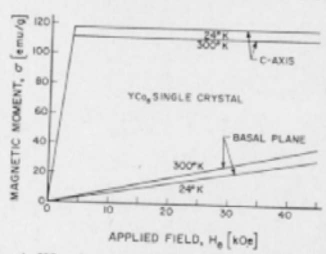



FIG. 1. $YCo_5$ single crystal: magnetization vs field curves measured in easiest and hardest directions.

a comparison of measured with theoretical magnetization curves.

[1] Prepared by Perkin-Elmer Corporation. See: J. F. Nester and J. B. Schroeder, Trans. AIME 233, 249 (1965).
[2] G. Hoffer and K. Strnat, IEEE Trans. Magnetics 2, 487 (1966).
[3] K. Strnat and L. Bartimay, J. Appl. Phys. 38, 1305 (1967).
[4] W. Sucksmith and J. E. Thompson, Proc. Roy. Soc. (London) 225, 362 (1954).

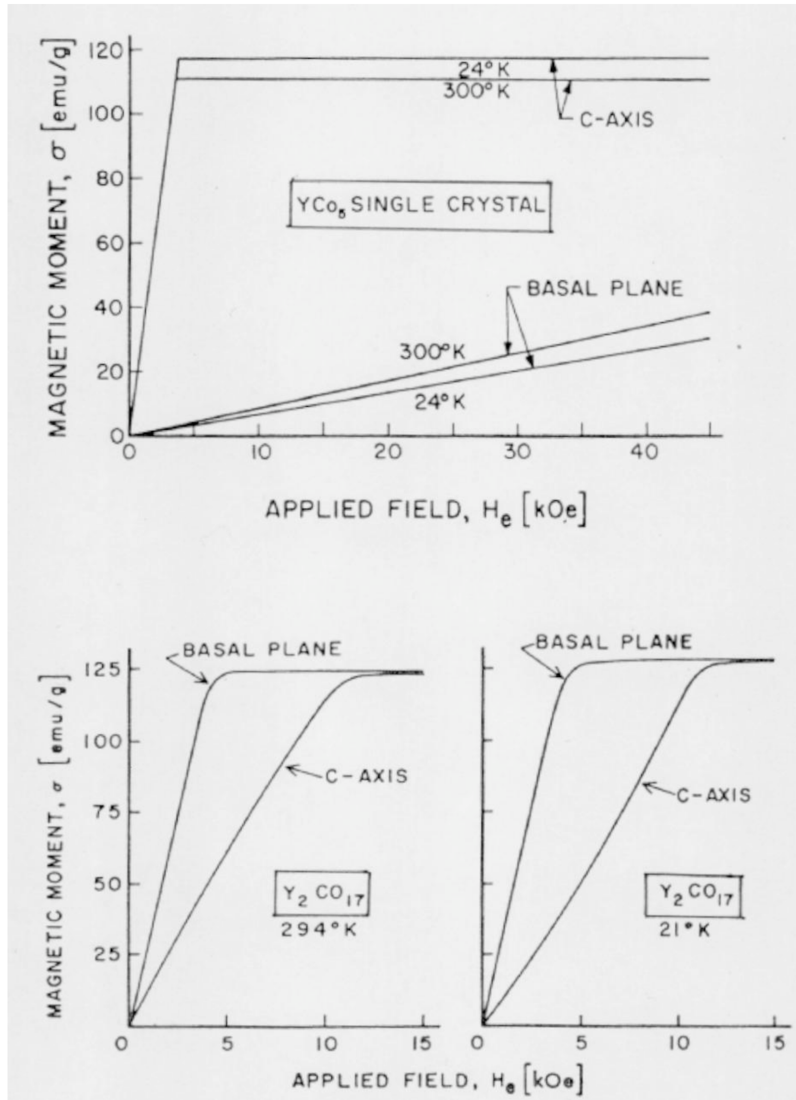


- This year, 1967, appeared the paper of Hoffer and Strnat, comparing the magnetic anisotropy of $YCo_5$ and $Y_2Co_{17}$, two ferromagnetic intermetallic compounds
where the cobalt only is magnetic

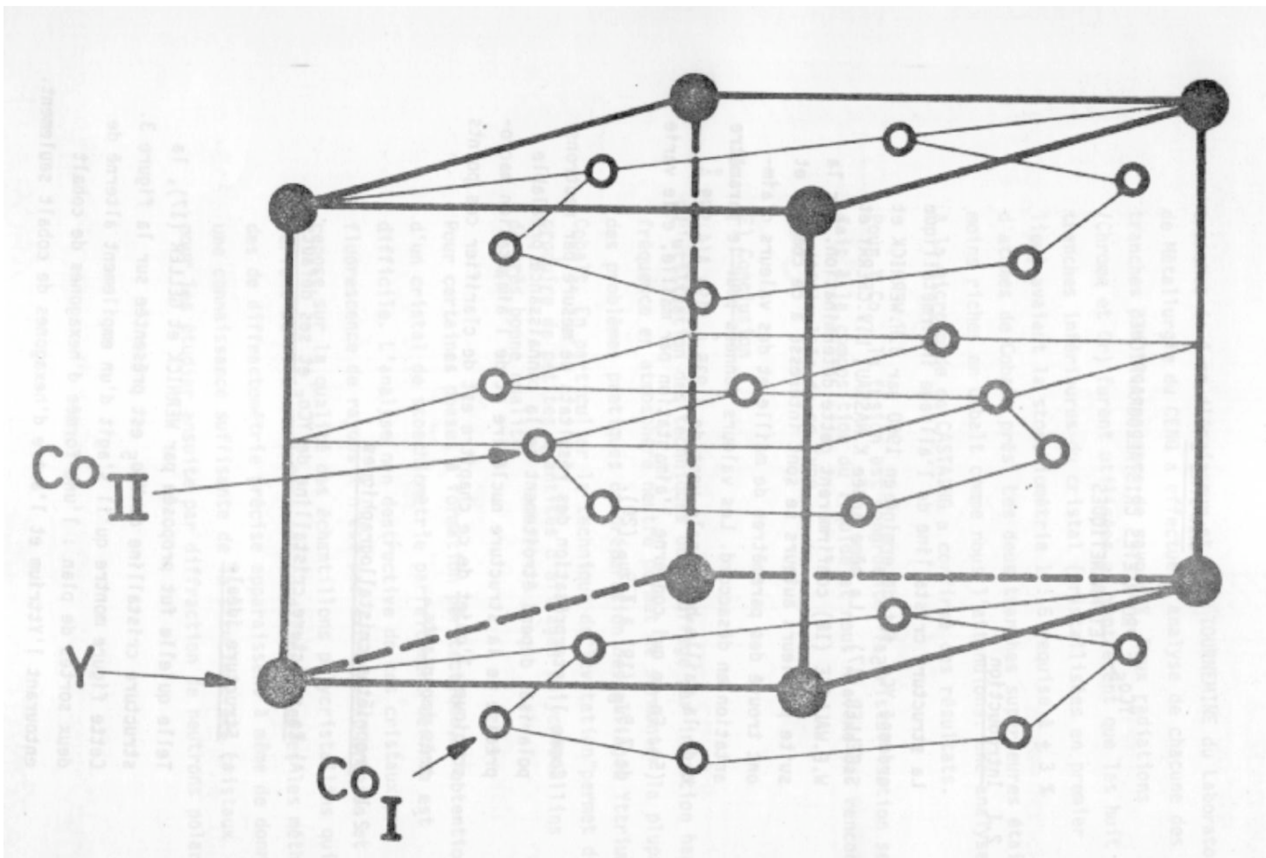
# Magnetic anisotropy

YCo5

Y2Co17

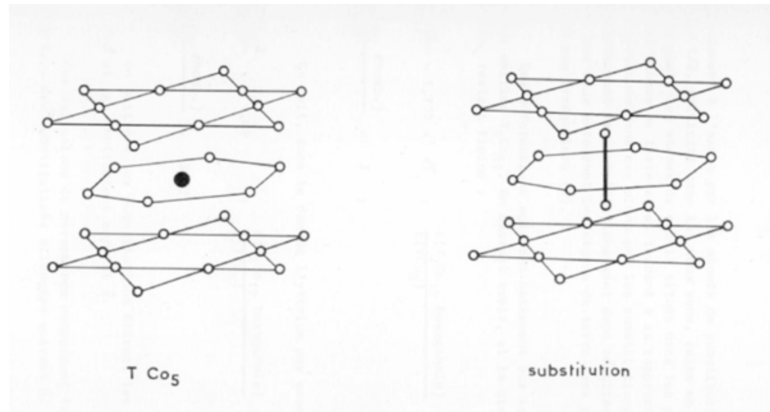


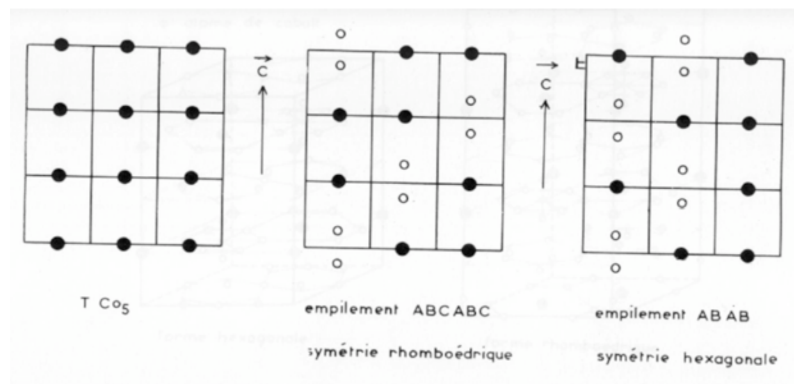
# Structure of $YCo_5$ ($CaCu_5$)



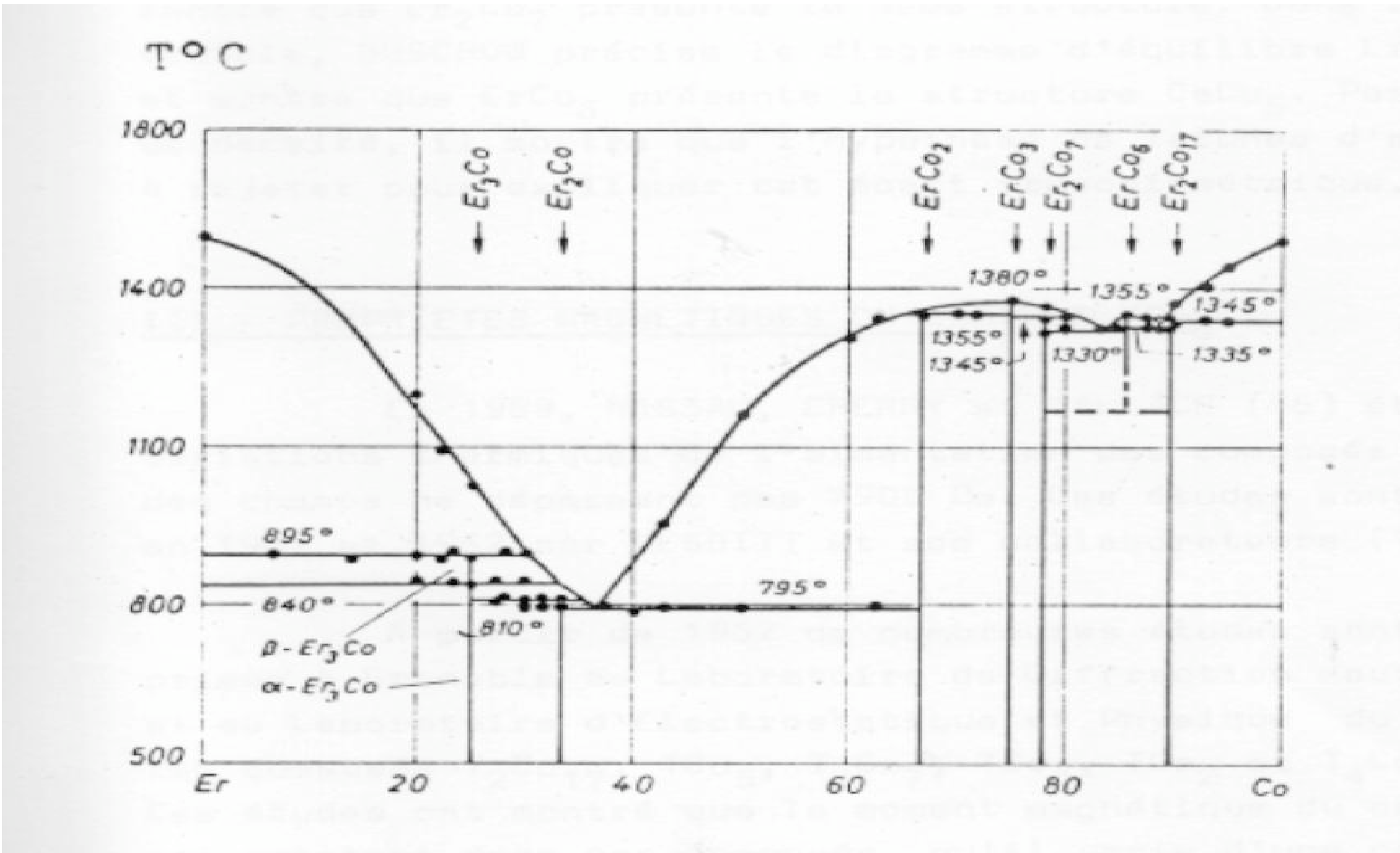
# Structure of $Y_2Co_{17}$

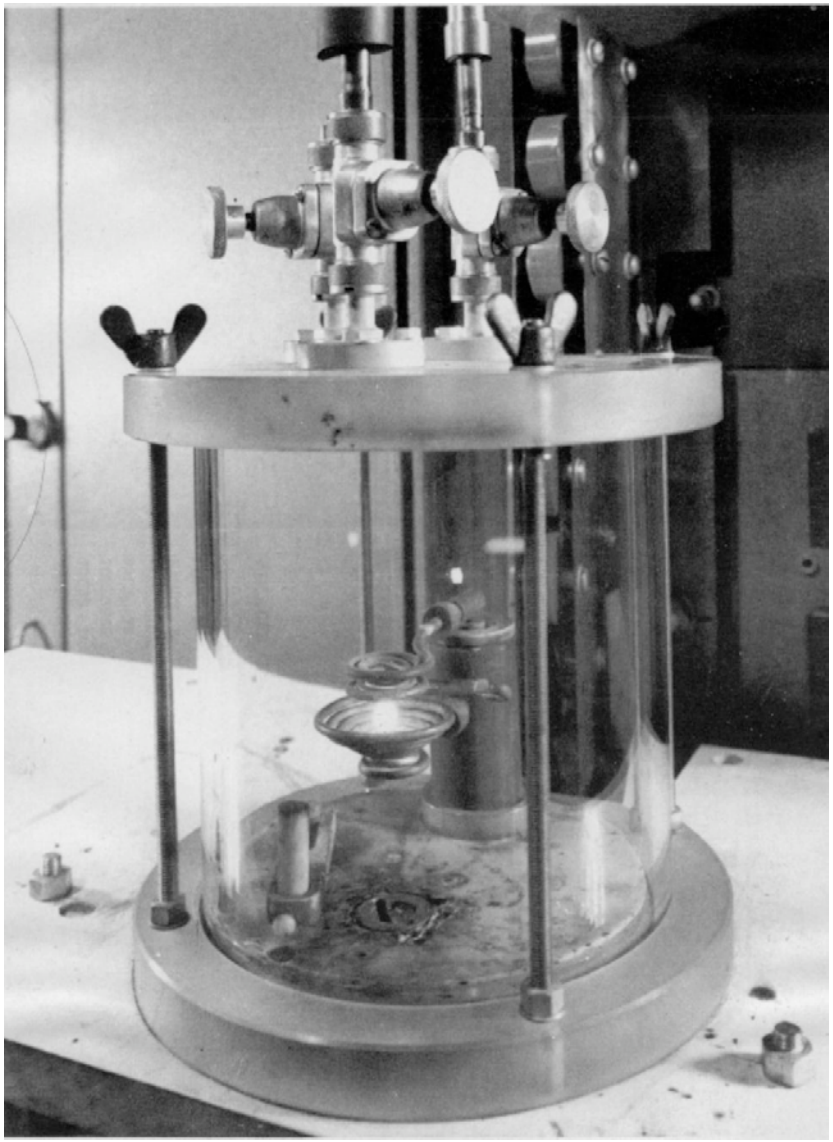
- Substitution of one third of the Y atoms by a pair of Co (Co dumbbell)
- These substitutions are ordered
- According to the order of the substitutions one get either the rhomboedral form (ABCABC..) or the hexagonal form (ABAB…)



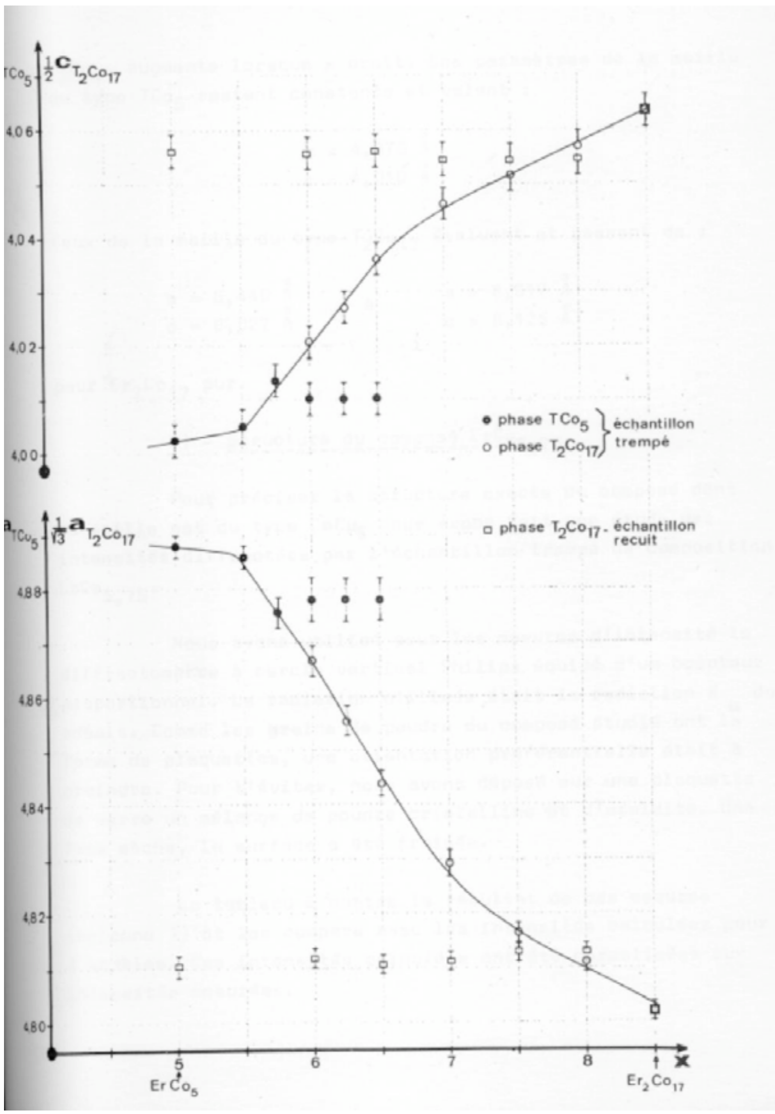
In 1966 Bushow published the phase diagramme Er-Co :
$ErCo_5$ does not exist; $ErCo_6$ exists (with the $CaCu_5$ structure)





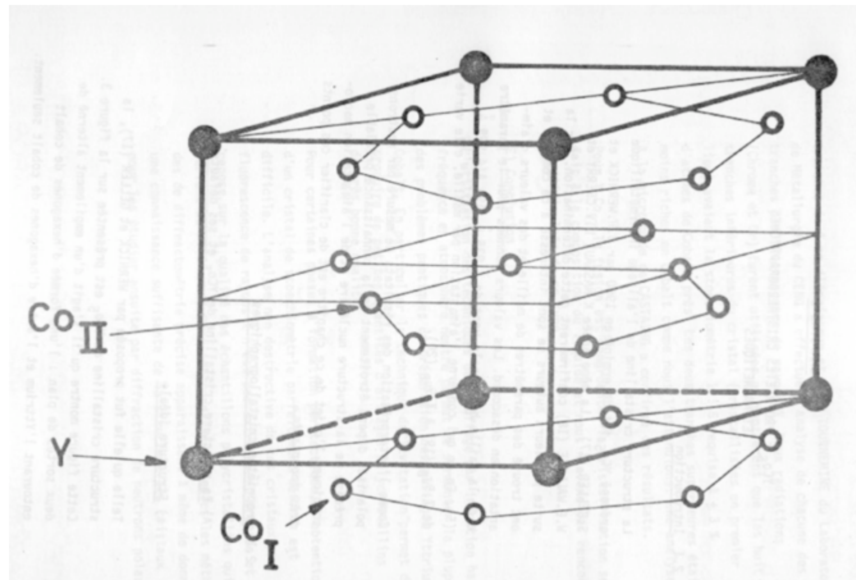
- First job of Francis:
- **Prepare samples (polycristalline) with stoichiometry beween $ErCo_5$ and $Er_2Co_{17}$**
- Use the levitation furnace to prepare quenched samples
- Anneal the samples
- Study the samples with an X rays powder camera in order to understand what does mean « $ErCo_6$ »

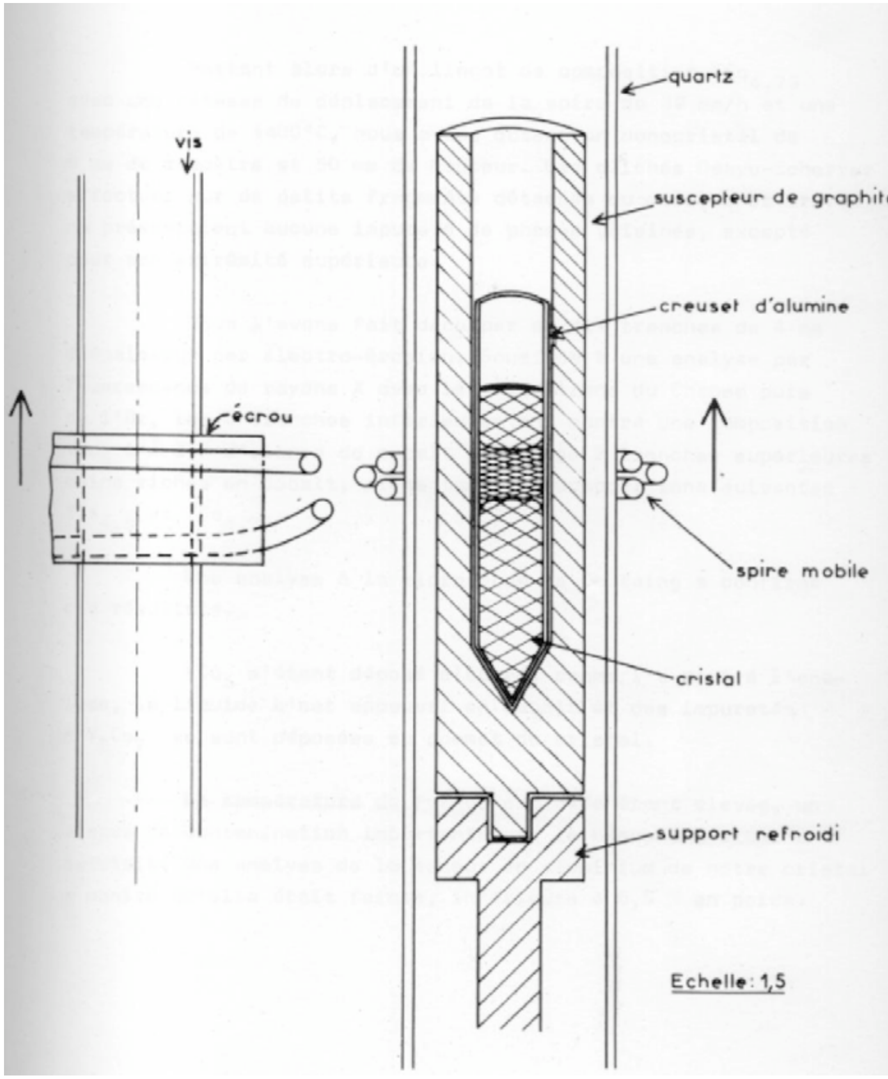
# Lattice constants of the samples $ErCo_x$




- When the number of substitutions increases a shrinks and c swallows
- For annealed samples the $CaCu_5$ phase does not exist: only the $Er_2Co_{17}$ phase
- For quenched samples, the $CaCu_5$ phase exists for $ErCo_x$ samples with $5.50<x<5.75$
- The substitutions stabilize the $CaCu_5$ phase, **but they are not ordered** and **do not give rise to the superlattice reflections**
- The lattice constant measurement is a way to estimate the number of substitutions

- In a $RECo_5$ or in a $YCo_5$ crystal a possible defect is the presence of random substitutions of RE or Y atoms by dumbbells of Co





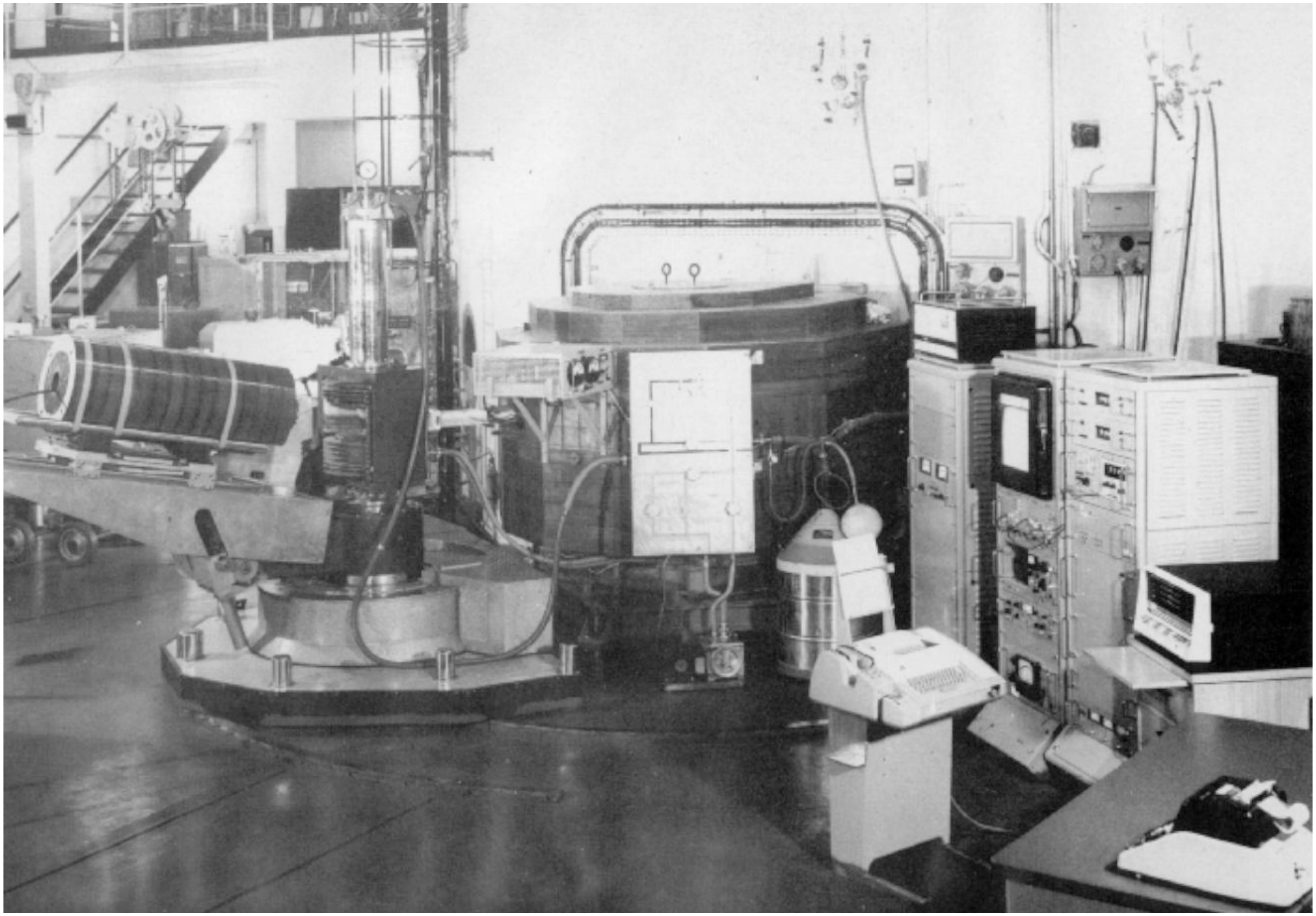

- Second job of Francis :
- **Prepare a single crystal of $YCo_5$**

- Use an induction furnace with a moving spire
- Use a cone shaped alumina crucible and a graphite susceptor

- As $YCo_5$ is not congruent choose an intermediate composition,
- Cut the final rod in slices,
- Make analyses to choose the right slice

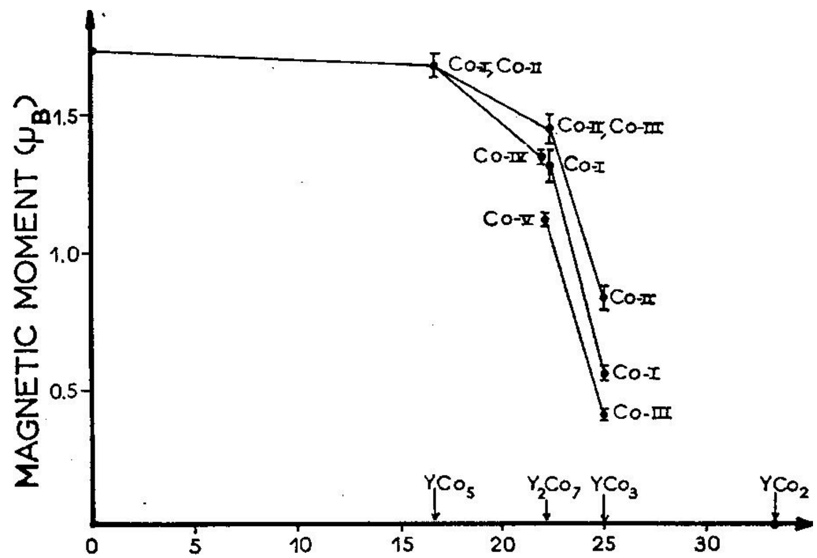
- **Go to the polarized neutron spectrometer DN2 in Mélusine**

# DN2 in Mélusine



# 1967-1968 Thèse de 3eme cycle

- Precise measurement of the Co magnetic moments in $YCo_5$
- Compare with those already measured in $Y_2Co_7$, $YCo_3$ and $YCo_2$



# In the mean time….

- Francis got a fellowship from the ILL (Jacrot)
  - -to make a thesis in Bertaut's laboratory
  - -to build a polarized neutron diffractometer for the ILL
- Conference of Koehler in CENG to explain the polarization analysis (uniaxial polarization analysis)
- Propose to build a multipurpose instrument D5
  - - for spin density and form factors measurements
  - - for polarization analysis

# Francis' double life

- In CENG

  Measuring the Co form factors and the spin density in $YCo_5$

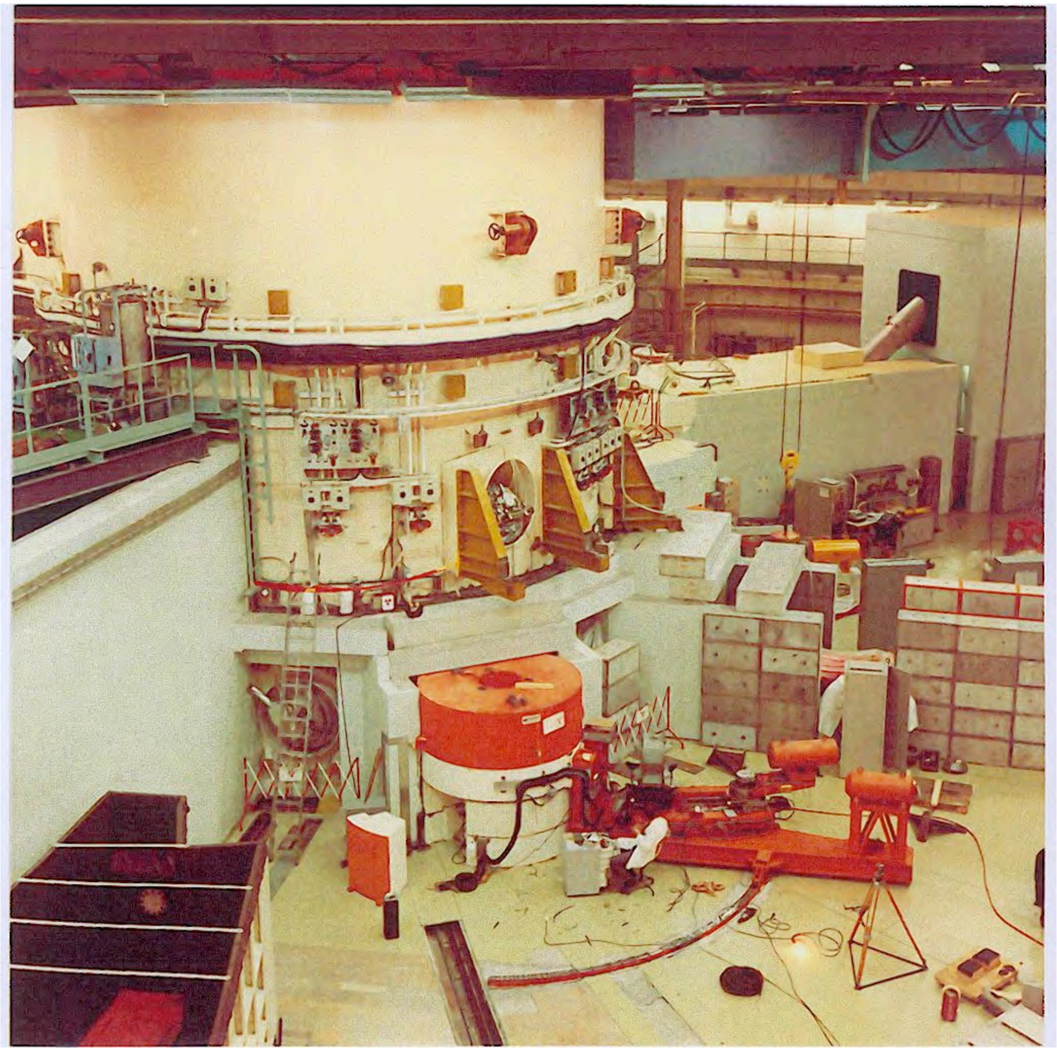
  -many reflections
  -several wavelengths
  -several crystals

- In ILL

  Supervising the design and the construction of D5

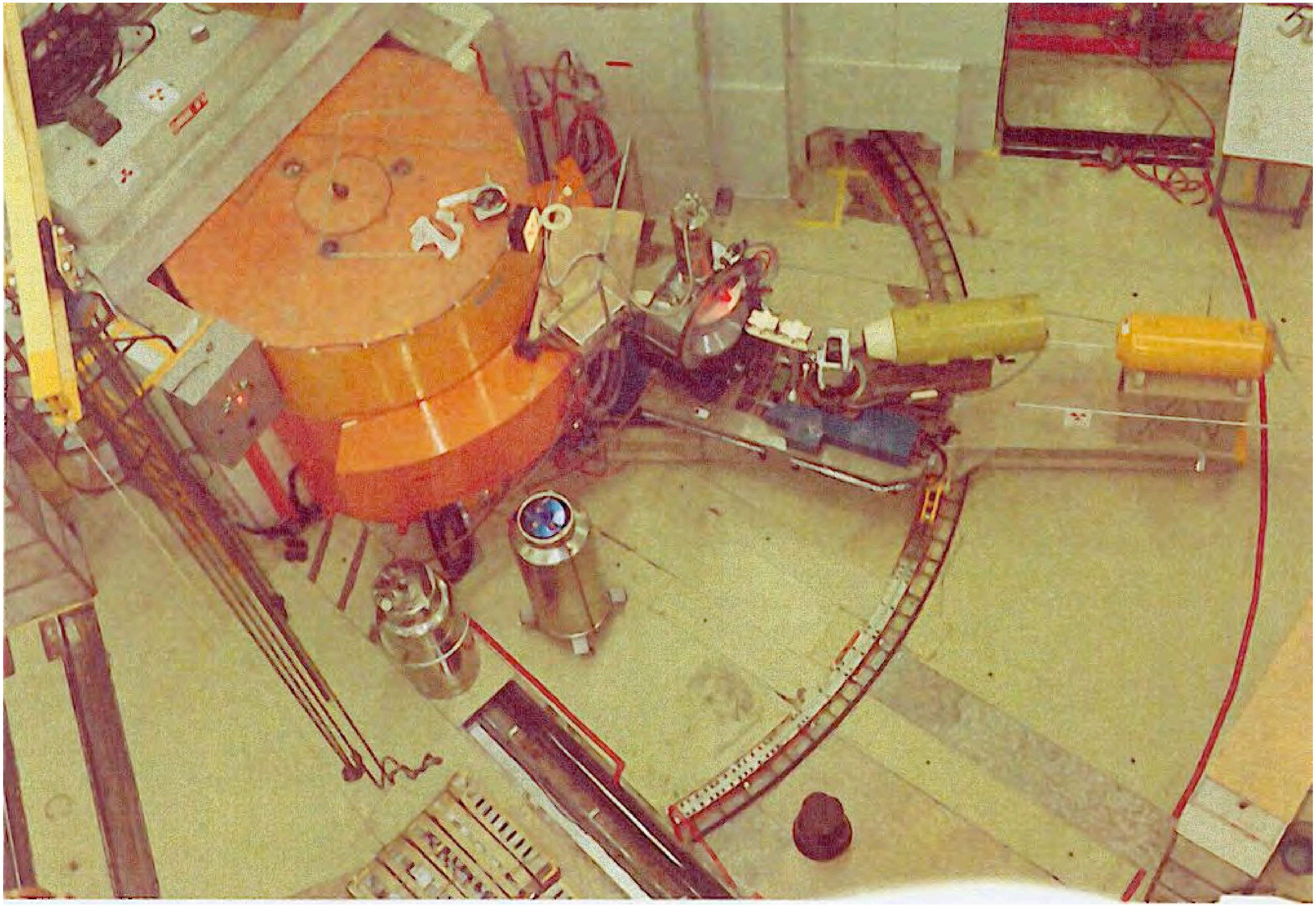
  -Guy Gobert
  -Bernard Mathieu
  -George Messoumian

# D5 : one of the first instruments ready at the ILL start

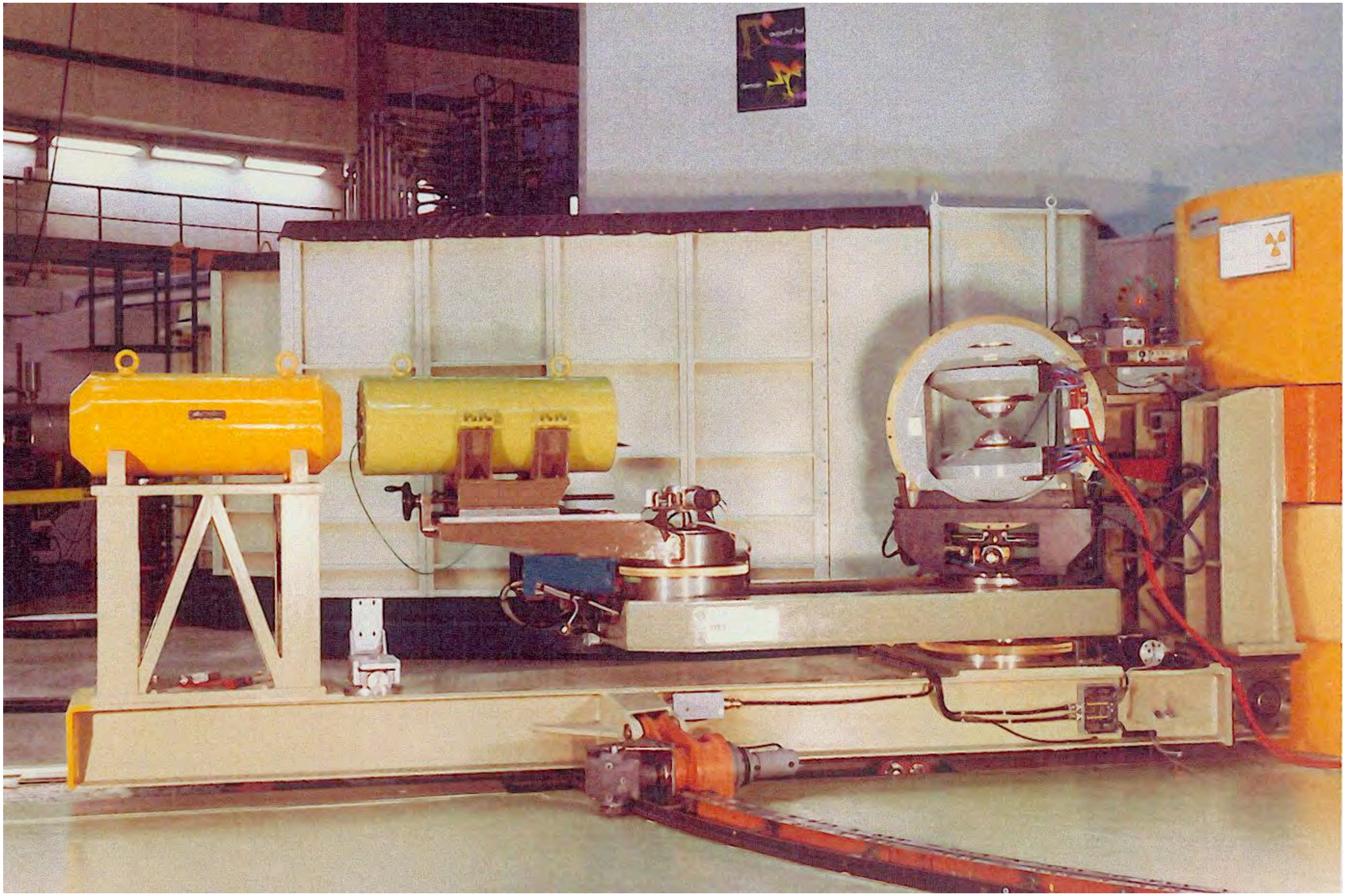


- Triple axis
- Built in Darmstadt
- Variable wavelength
- Hot source

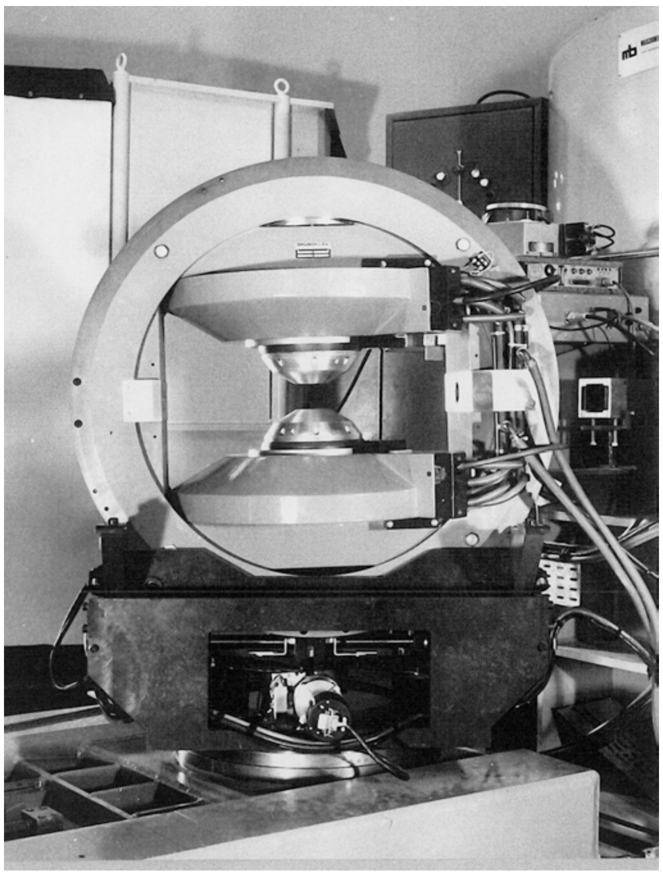
# Polarization analysis



Instrument mounted on tracks (DN2)
Each motion automatic



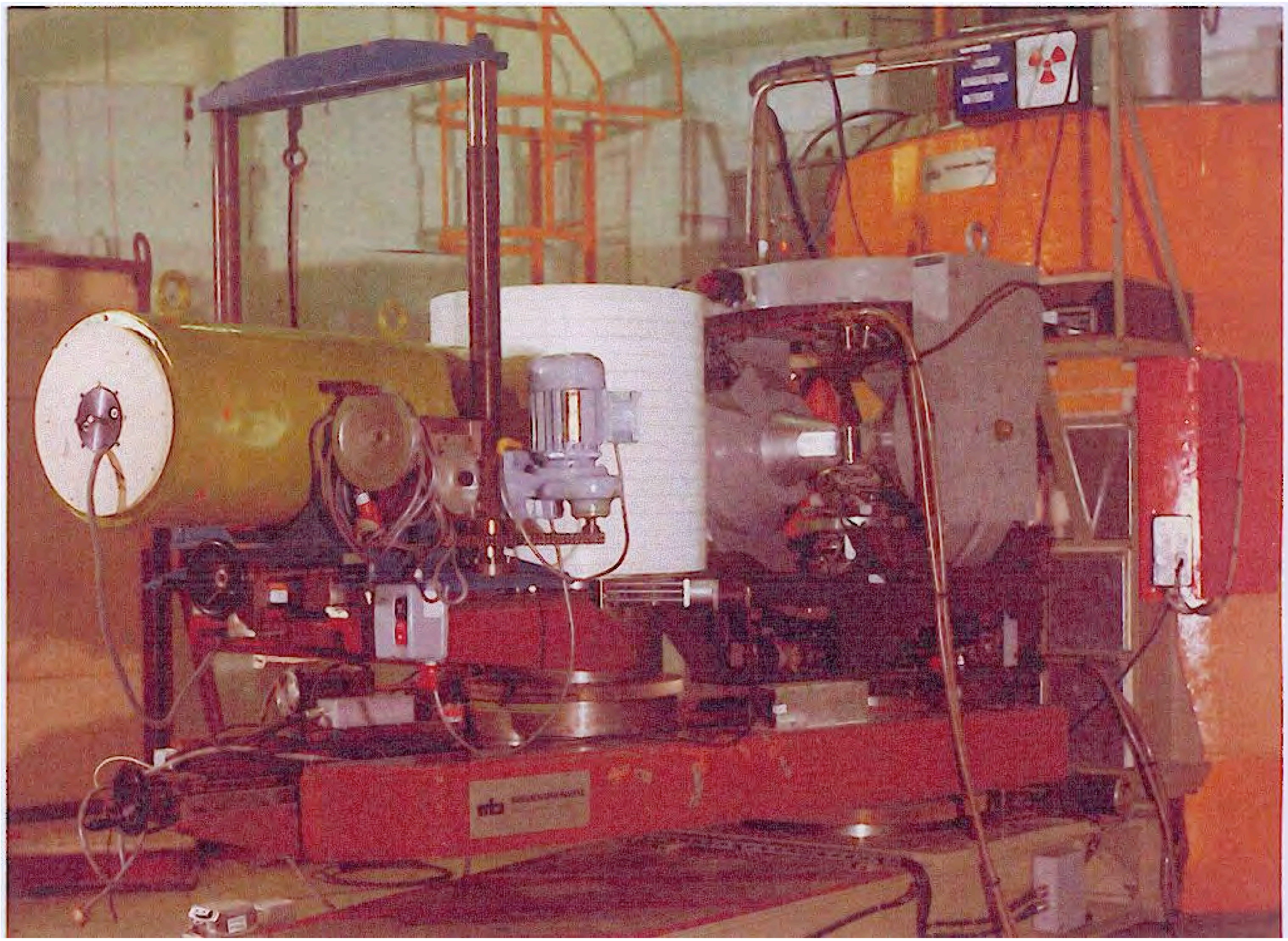
# Magnet with a circular frame

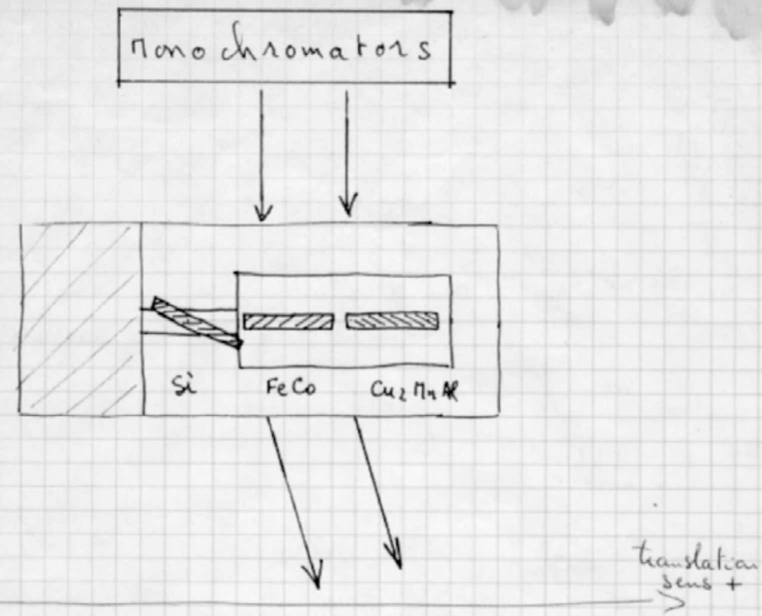


- Field either vertical or horizontal

- Mounted on a kind of eulerian craddle

also an horizontal field magnet
also the  possibility to lift the counter






| cristal | translation | inclination |
|---|---|---|
| Silicium | butée (+) -300<br>Fe Co +5700 | |
| Fe Co | silicium -5700<br>Heussler +4700 | 0 |
| Heussler | butée (−)<br>Fe Co -4700 | -200 |

**Hot source**

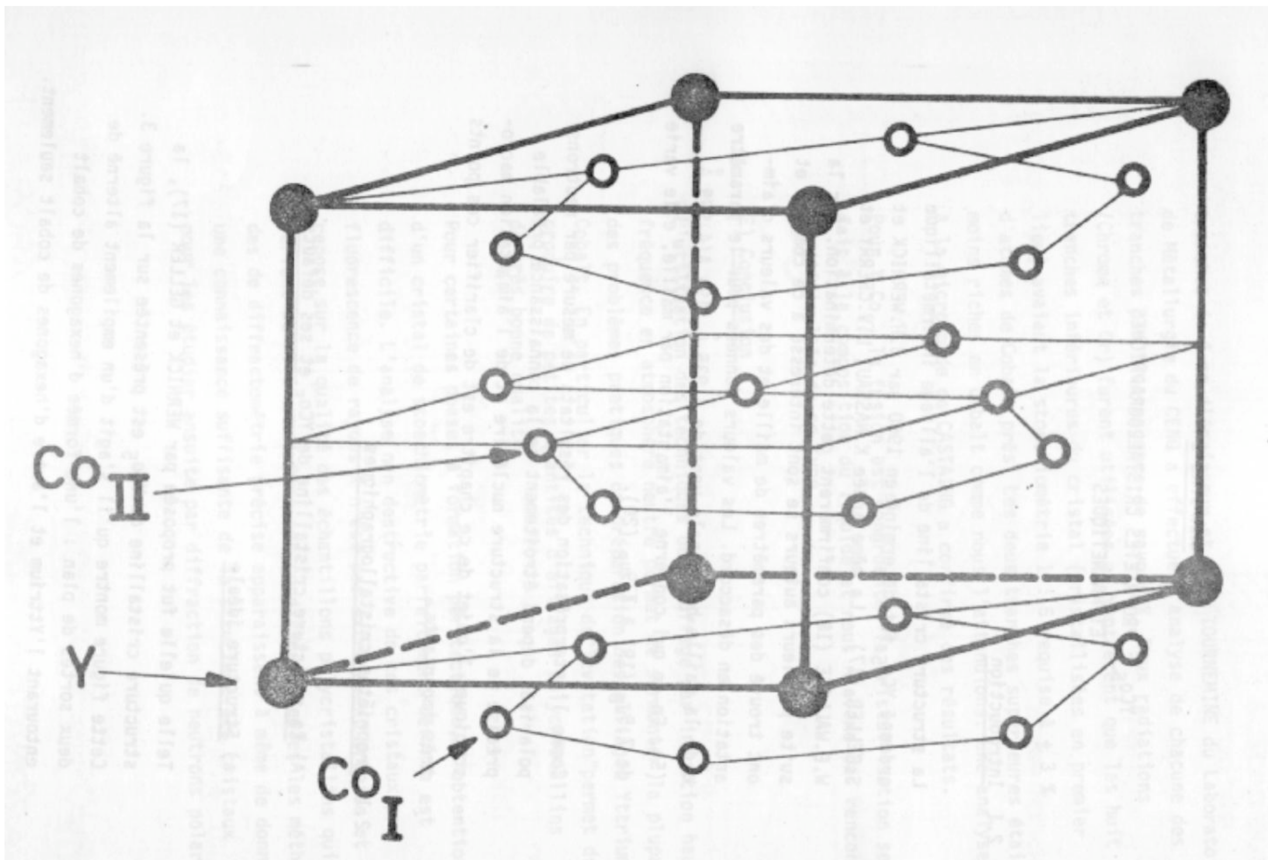
very good for extinction corrections

wavelengths and filters

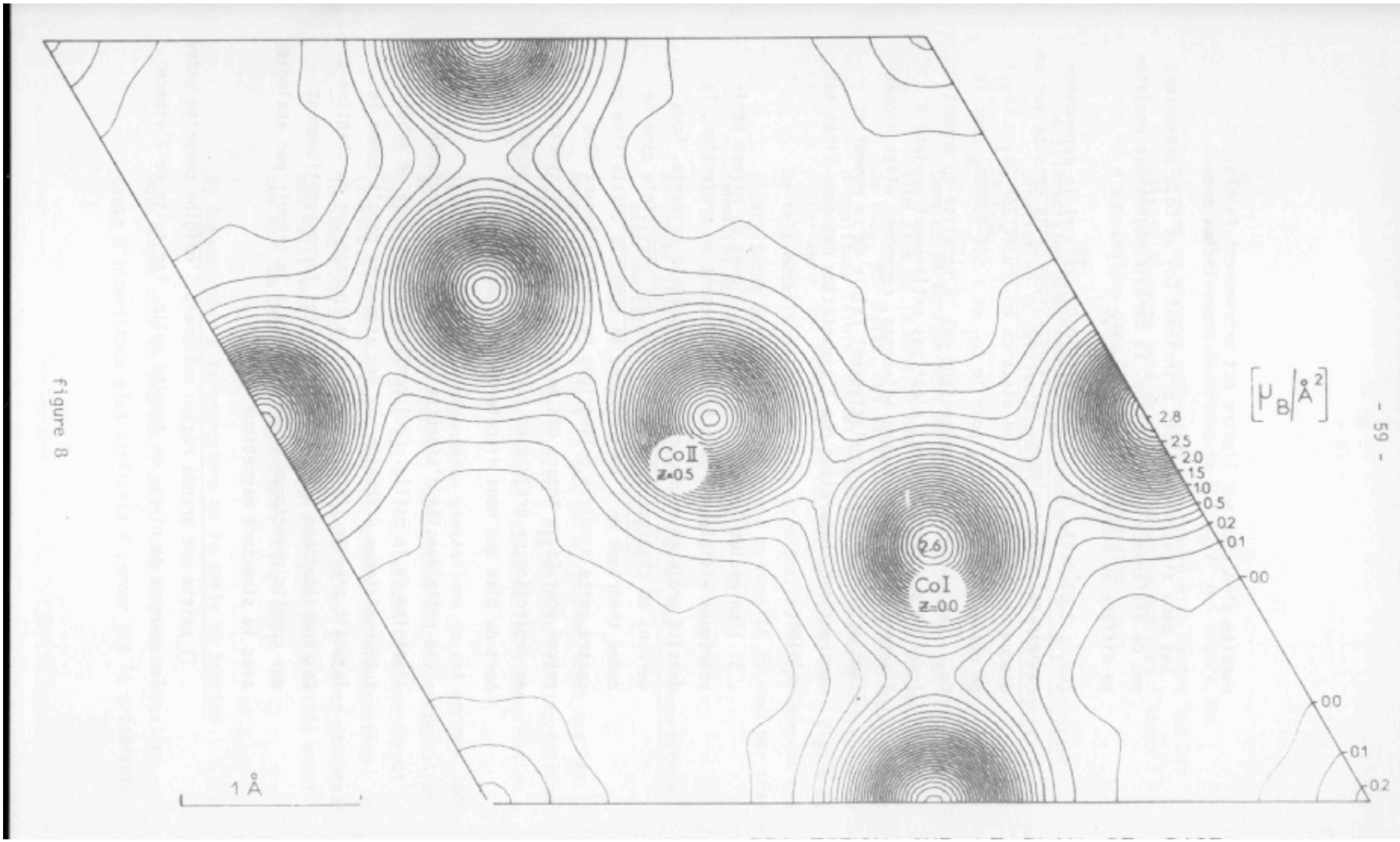
| | |
|---|---|
| $\lambda < 0.50$ A | no filter |
| $\lambda = 0.74$ A | Er filter |
| $\lambda = 0.84$ A | Er filter |
| $\lambda = 1.05$ A | Pu filter |

# First sample tested on D5:$YCo_5$

one week on D5=two years on DN2 (Mélusine)



# Projection of the spin density



# Spin and orbit

the magnetic anisotropy is carried mainly by site $Co_{I}$
(in the plane containing Y)

<table>
<tr><td></td><td></td><td colspan="7">Résultats de l'affinement</td></tr>
<tr><td>Site principal</td><td>Moment total localisé</td><td>proportion de spin</td><td colspan="5">caractère orbital</td></tr>
<tr><td>$Co_{I}$</td><td>1.80(3)</td><td>0.64(7)</td><td>$\varpi\ d_{z^2}$ 0.22(4)</td><td>$\varpi\ d_{xz}, d_{yz}$ 0.06(19)</td><td colspan="3">$\varpi\ d_{x^2-y^2}, d_{xy}$ 0.72</td></tr>
<tr><td>$Co_{II}$</td><td>1.74(2)</td><td>0.87(5)</td><td>$\varpi\ d_{z^2}$ 0.15(2)</td><td>$\varpi\ d_{xz}$ 0.23(4)</td><td>$\varpi\ d_{yz}$ 0.26(4)</td><td>$\varpi\ d_{x^2-y^2}$ 0.20(4)</td><td>$\varpi\ d_{xy}$ 0.16</td></tr>
</table>

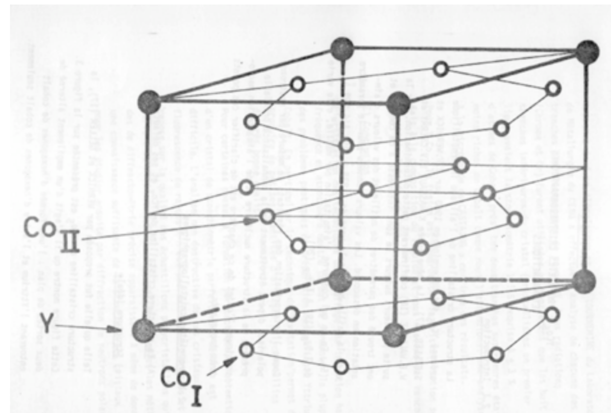
# Practical consequences to make permanent magnets

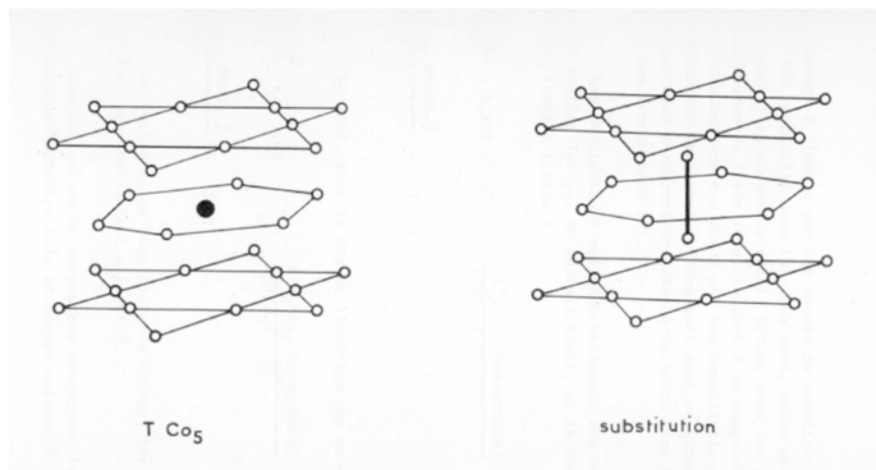
- When substituting Co by another metal, it is important to know in which sites this new metal goes
  - -Fe goes preferentially in site $Co_{II}$ ⇒
  - -Ni goes preferentially in site $Co_{I}$
- The dumbbell substitutions destroy the anisotropic site $Co_{I}$

# Dumbbell substitutions kill the magnetic anisotropy

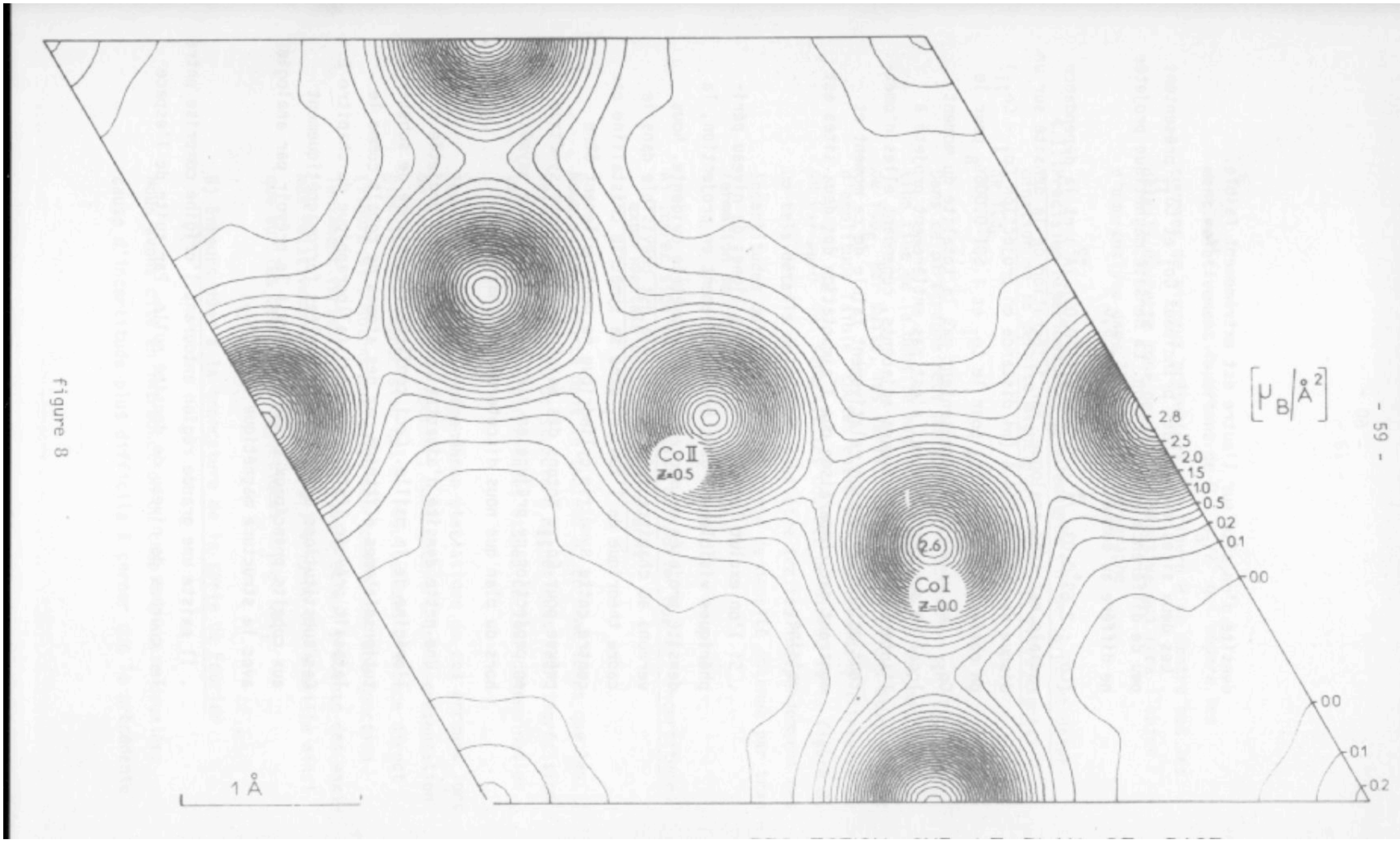
Around a substitution, the $Co_I$ hexagon around the Y vacancy shrinks

The Co atoms of the shrinked hexagon have lost the magnetic anisotropy of the former $Co_I$ positions

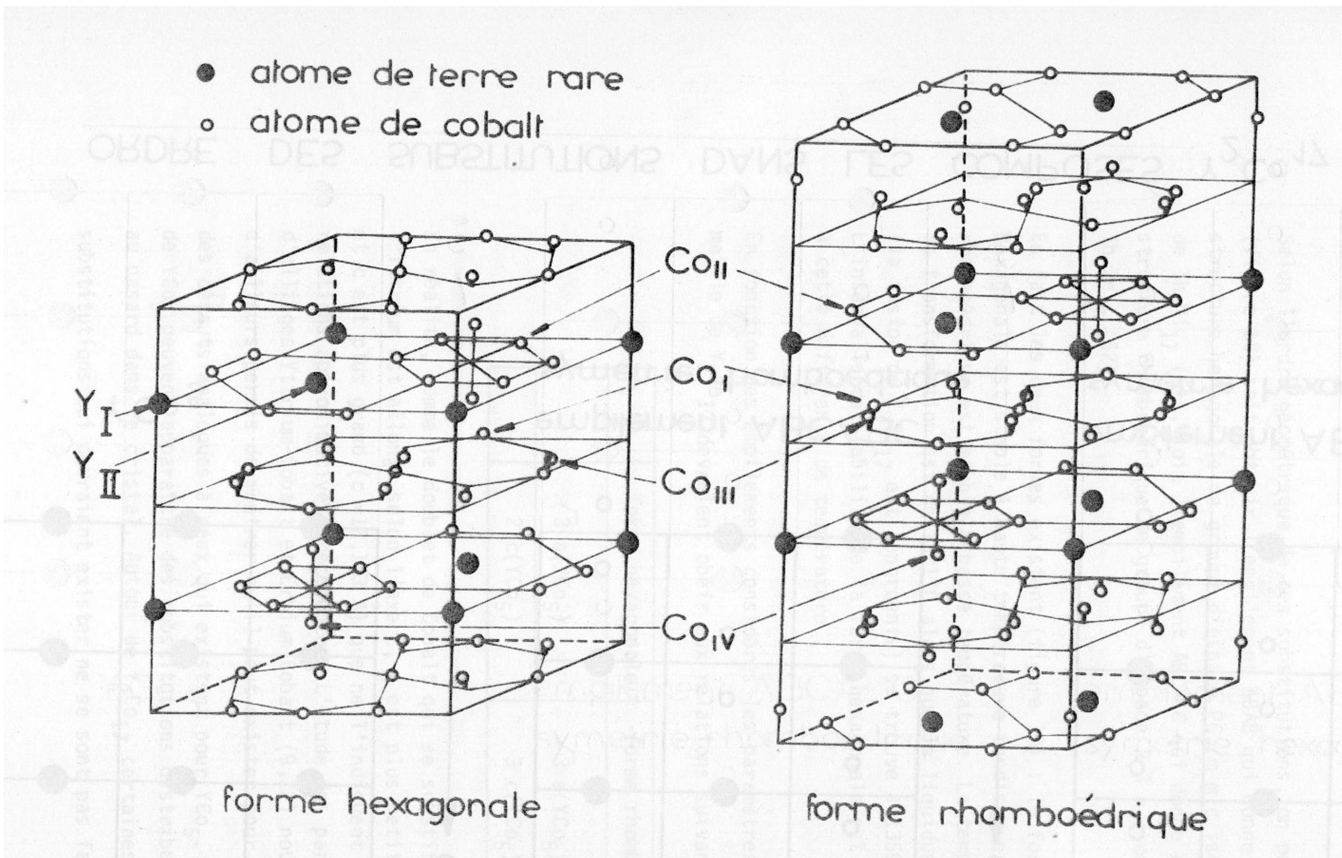


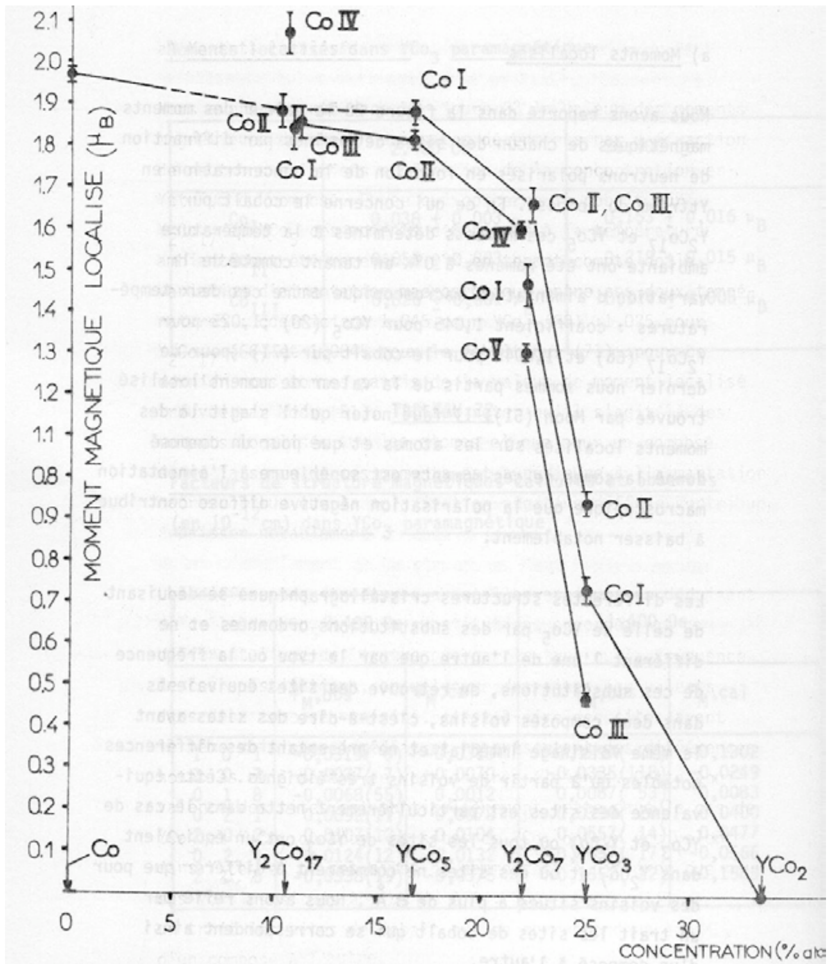
# Spin density



# This explains the low magnetic anisotropy of the compounds $Y_2Co_{17}$



# The variations of the localized magnetic moment of Co



# After Francis'thesis…

- He spent one year in Oak Ridge
  to work on superconducting flux lines

- He came back with ideas to invent a new flipper device
  based on superconductivity

- The British had joined the ILL and built a new instrument : D3
  Francis left D5 and went to D3….