# Hard X-ray Magnetic Circular Dichroism: application to spintronics materials

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# X-rays and magnetism



**Nobel Prize in Physics 1994: B. N. Brockhouse and C. G. Shull**

**Press release by the Royal Swedish Academy of Sciences:**

**"Neutrons are small magnets…… (that) can be used to study the relative orientations of the small atomic magnets. ….. the X-ray method has been powerless and in this field of application neutron diffraction has since assumed an entirely dominant position. It is hard to imagine modern research into magnetism without this aid."**

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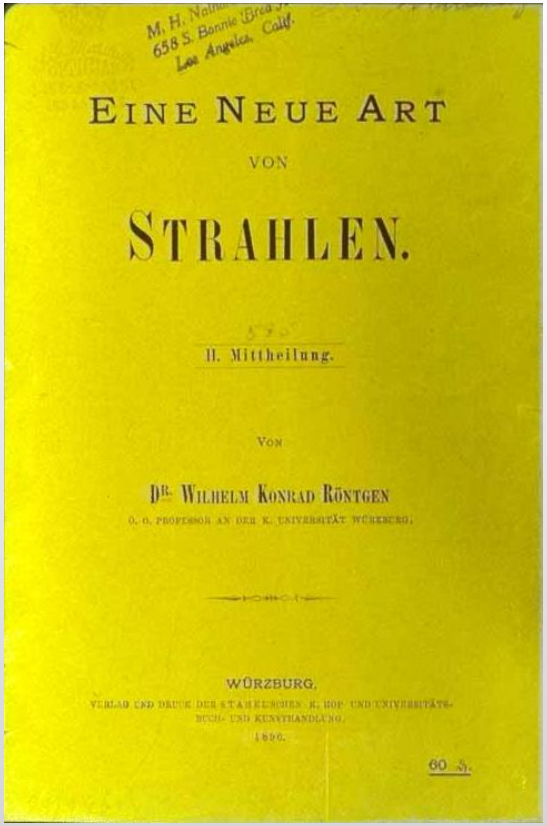
**Today:**
**X-ray magnetic circular dichroism (XMCD) is considered to be one of the most important discoveries in the field of magnetism research in the last two decades. It is hard to imagine modern research into magnetism without the aid of X-ray spectroscopy.**

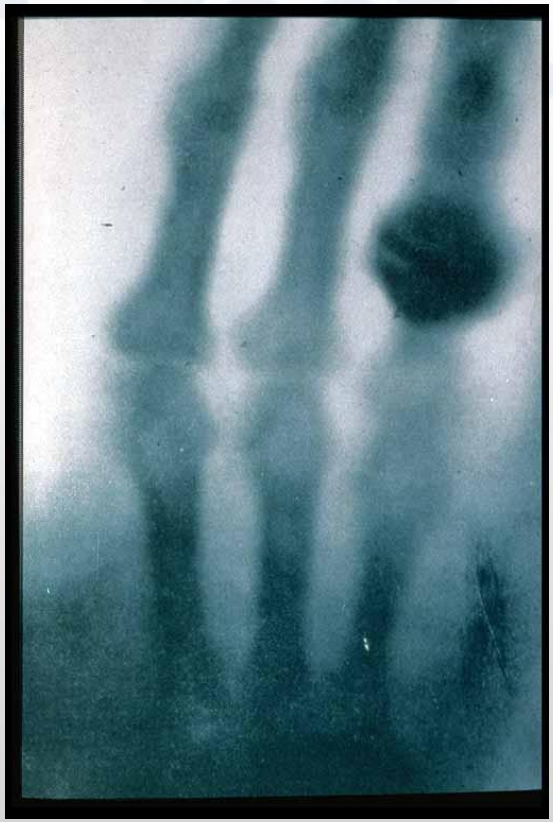
# Outline

- Introduction to X-ray Magnetic Circular Dichroism
- Experimental aspects
- Selected examples
  - Magnetism in diluted magnetic semiconductors
  - Induced magnetism in magnetic multilayers
  - X-ray detection of magnetic resonance
- Conclusions

# X-rays and Magnetism

## In 1895 Dr. W.K.Röntgen has discovered "Eine Neue Art von Strahlen"



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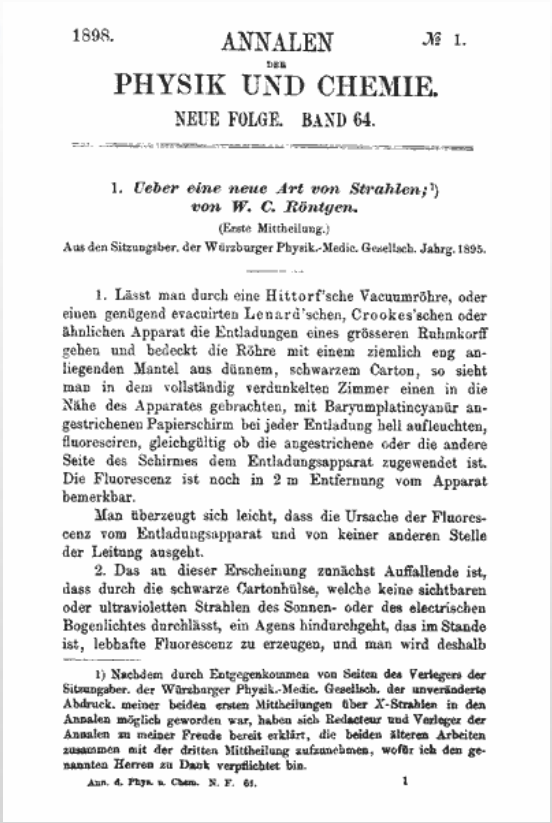


1898. ANNALEN DER PHYSIK UND CHEMIE. № 1.
NEUE FOLGE. BAND 64.

1. *Ueber eine neue Art von Strahlen;*[1]
*von W. C. Röntgen.*
(Erste Mittheilung.)
Aus den Sitzungsber. der Würzburger Physik.-Medic. Gesellsch. Jahrg. 1895.

1. Lässt man durch eine Hittorf'sche Vacuumröhre, oder einen genügend evacuirten Lenard'schen, Crookes'schen oder ähnlichen Apparat die Entladungen eines grösseren Ruhmkorff gehen und bedeckt die Röhre mit einem ziemlich eng anliegenden Mantel aus dünnem, schwarzem Carton, so sieht man in dem vollständig verdunkelten Zimmer einen in die Nähe des Apparates gebrachten, mit Baryumplatincyanür angestrichenen Papierschirm bei jeder Entladung hell aufleuchten, fluoresciren, gleichgültig ob die angestrichene oder die andere Seite des Schirmes dem Entladungsapparat zugewendet ist. Die Fluorescenz ist noch in 2 m Entfernung vom Apparat bemerkbar.

Man überzeugt sich leicht, dass die Ursache der Fluorescenz vom Entladungsapparat und von keiner anderen Stelle der Leitung ausgeht.

2. Das an dieser Erscheinung zunächst Auffallende ist, dass durch die schwarze Cartonhülse, welche keine sichtbaren oder ultravioletten Strahlen des Sonnen- oder des electrischen Bogenlichtes durchlässt, ein Agens hindurchgeht, das im Stande ist, lebhafte Fluorescenz zu erzeugen, und man wird deshalb

1) Nachdem durch Entgegenkommen von Seiten des Verlegers der Sitzungsber. der Würzburger Physik.-Medic. Gesellsch. der unveränderte Abdruck meiner beiden ersten Mittheilungen über X-Strahlen in den Annalen möglich geworden war, haben sich Redacteur und Verleger der Annalen zu meiner Freude bereit erklärt, die beiden älteren Arbeiten zusammen mit der dritten Mittheilung aufzunehmen, wofür ich den genannten Herren zu Dank verpflichtet bin.

Ann. d. Phys. u. Chem. N. F. 64. 1

*On December 28, 1895 W.C. Röntgen had submitted his manuscript*
*"On a New Kind of Ray, A Preliminary Communication" to the Würzburg Physical Medical Society.*
*On the 1st of January 1896 he sent copies of his manuscript to several renowned physicists.*

## search for polarisation of X-rays

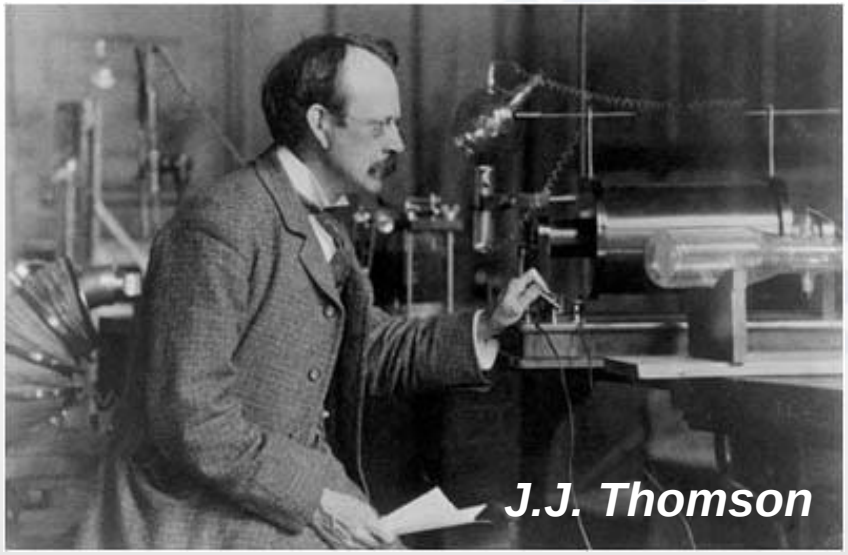
**Comptes rendus hebdomadaires des séances de l'Académie des sciences**
**Séance du lundi 27 Janvier 1896**


PHYSIQUE. — *Quelques propriétés des rayons de Röntgen.* Note de M. **Jean Perrin**, présentée par M. Mascart.

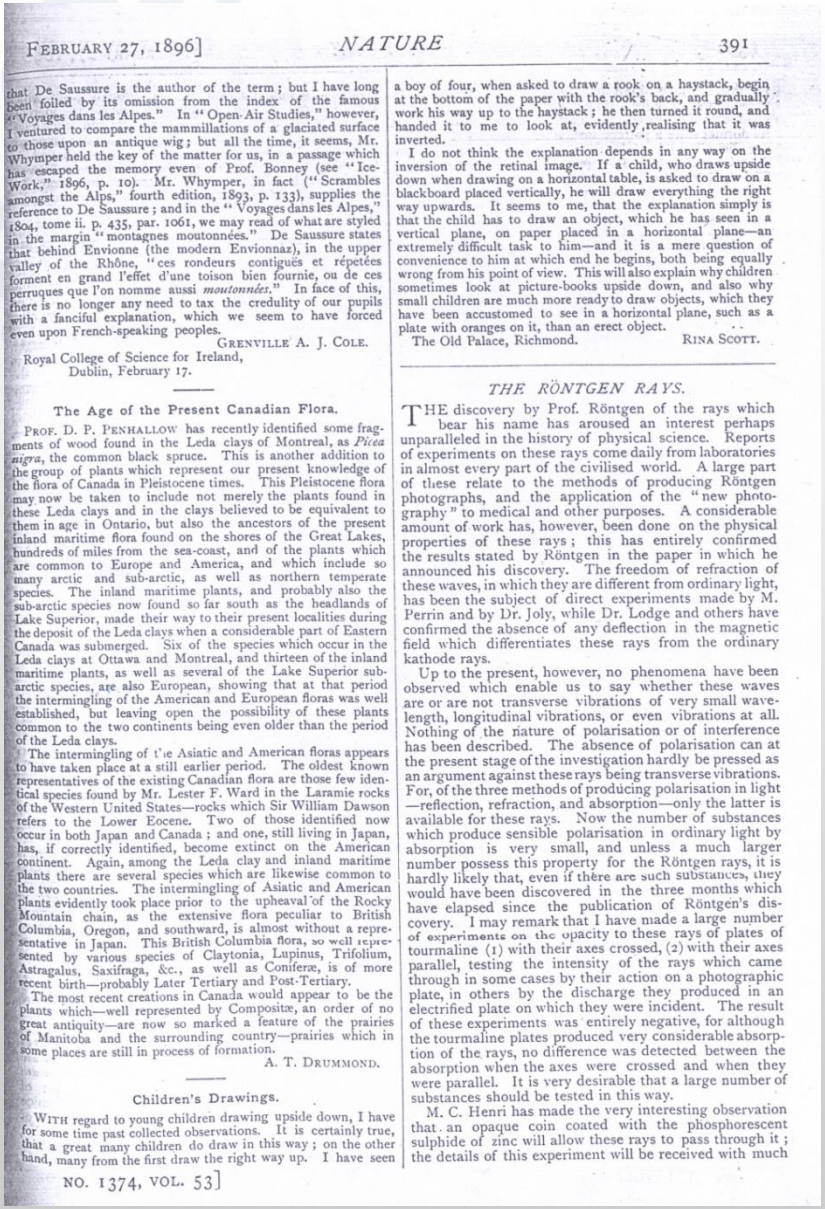
» Le bois, le papier, la cire, la paraffine, l'eau se montrèrent très transparents, l'influence de l'épaisseur restant cependant nette. Viendraient ensuite, à peu près rangés par ordre d'opacité croissante, le charbon, l'os, l'ivoire, le spath, le verre, le quartz (parallèle ou perpendiculaire à l'axe), le sel gemme, le soufre, le fer, l'acier, le cuivre, le laiton, le mercure, le plomb. Ces résultats sont encore peu nombreux, et je ne peux songer à les relier par une loi générale; toutefois, on peut remarquer, dès maintenant, que les métaux sont en général moins transparents que les autres corps, mais n'ont pas l'opacité absolue qu'ils présentent pour la lumière. Si, par exemple, on superpose trois lames de fer, d'environ $0^{mm},2$ chacune, l'opacité ne paraît atteinte que dans la région commune aux trois lames.

*C.R. Acad.Sci. Paris 122 (1896) 186-188*

# X-rays and Magnetism




"I may remark that I have made a large number of experiments on the opacity to these rays of plates of tourmaline (1) with their axes crossed, (2) with their axes parallel, testing the intensity of the rays which came through in some cases by their action on a photographic plate in others by the discharge they produced in an electrified plate on which they were incident. The result of these experiments were entirely negative, for although the tourmaline produced very considerable absorption of the rays, no difference was detected between the absorption when the axes were crossed and when they were parallel. It is very desirable that a large number of substances should be tested in this way."



FEBRUARY 27, 1896] NATURE 391

that De Saussure is the author of the term; but I have long been foiled by its omission from the index of the famous "Voyages dans les Alpes." In "Open-Air Studies," however, I ventured to compare the mammillations of a glaciated surface to those upon an antique wig; but all the time, it seems, Mr. Whymper held the key of the matter for us, in a passage which has escaped the memory even of Prof. Bonney (see "Ice-Work," 1896, p. 10). Mr. Whymper, in fact ("Scrambles amongst the Alps," fourth edition, 1893, p. 133), supplies the reference to De Saussure; and in the "Voyages dans les Alpes," 1804, tome ii. p. 435, par. 1061, we may read of what are styled in the margin "montagnes moutonnées." De Saussure states that behind Envionne (the modern Envionnaz), in the upper valley of the Rhône, "ces rondeurs contiguës et répetées forment en grand l'effet d'une toison bien fournie, ou de ces perruques que l'on nomme aussi *moutonnée*." In face of this, there is no longer any need to tax the credulity of our pupils with a fanciful explanation, which we seem to have forced even upon French-speaking peoples.

GRENVILLE A. J. COLE.

Royal College of Science for Ireland,
Dublin, February 17.

### The Age of the Present Canadian Flora.

PROF. D. P. PENHALLOW has recently identified some fragments of wood found in the Leda clays of Montreal, as *Picea nigra*, the common black spruce. This is another addition to the group of plants which represent our present knowledge of the flora of Canada in Pleistocene times. This Pleistocene flora may now be taken to include not merely the plants found in these Leda clays and in the clays believed to be equivalent to them in age in Ontario, but also the ancestors of the present inland maritime flora found on the shores of the Great Lakes, hundreds of miles from the sea-coast, and of the plants which are common to Europe and America, and which include so many arctic and sub-arctic, as well as northern temperate species. The inland maritime plants, and probably also the sub-arctic species now found so far south as the headlands of Lake Superior, made their way to their present localities during the deposit of the Leda clays when a considerable part of Eastern Canada was submerged. Six of the species which occur in the Leda clays at Ottawa and Montreal, and thirteen of the inland maritime plants, as well as several of the Lake Superior sub-arctic species, are also European, showing that at that period the intermingling of the American and European floras was well established, but leaving open the possibility of these plants common to the two continents being even older than the period of the Leda clays.

The intermingling of the Asiatic and American floras appears to have taken place at a still earlier period. The oldest known representatives of the existing Canadian flora are those few identical species found by Mr. Lester F. Ward in the Laramie rocks of the Western United States—rocks which Sir William Dawson refers to the Lower Eocene. Two of those identified now occur in both Japan and Canada; and one, still living in Japan, has, if correctly identified, become extinct on the American continent. Again, among the Leda clay and inland maritime plants there are several species which are likewise common to the two countries. The intermingling of Asiatic and American plants evidently took place prior to the upheaval of the Rocky Mountain chain, as the extensive flora peculiar to British Columbia, Oregon, and southward, is almost without a representative in Japan. This British Columbia flora, so well represented by various species of Claytonia, Lupinus, Trifolium, Astragalus, Saxifraga, &c., as well as Coniferæ, is of more recent birth—probably Later Tertiary and Post-Tertiary.

The most recent creations in Canada would appear to be the plants which—well represented by Compositæ, an order of no great antiquity—are now so marked a feature of the prairies of Manitoba and the surrounding country—prairies which in some places are still in process of formation.

A. T. DRUMMOND.

### Children's Drawings.

WITH regard to young children drawing upside down, I have for some time past collected observations. It is certainly true, that a great many children do draw in this way; on the other hand, many from the first draw the right way up. I have seen a boy of four, when asked to draw a rook on a haystack, begin at the bottom of the paper with the rook's back, and gradually work his way up to the haystack; he then turned it round, and handed it to me to look at, evidently realising that it was inverted.

I do not think the explanation depends in any way on the inversion of the retinal image. If a child, who draws upside down when drawing on a horizontal table, is asked to draw on a blackboard placed vertically, he will draw everything the right way upwards. It seems to me, that the explanation simply is that the child has to draw an object, which he has seen in a vertical plane, on paper placed in a horizontal plane—an extremely difficult task to him—and it is a mere question of convenience to him at which end he begins, both being equally wrong from his point of view. This will also explain why children sometimes look at picture-books upside down, and also why small children are much more ready to draw objects, which they have been accustomed to see in a horizontal plane, such as a plate with oranges on it, than an erect object.

The Old Palace, Richmond. RINA SCOTT.

### THE RÖNTGEN RAYS.

THE discovery by Prof. Röntgen of the rays which bear his name has aroused an interest perhaps unparalleled in the history of physical science. Reports of experiments on these rays come daily from laboratories in almost every part of the civilised world. A large part of these relate to the methods of producing Röntgen photographs, and the application of the "new photography" to medical and other purposes. A considerable amount of work has, however, been done on the physical properties of these rays; this has entirely confirmed the results stated by Röntgen in the paper in which he announced his discovery. The freedom of refraction of these waves, in which they are different from ordinary light, has been the subject of direct experiments made by M. Perrin and by Dr. Joly, while Dr. Lodge and others have confirmed the absence of any deflection in the magnetic field which differentiates these rays from the ordinary kathode rays.

Up to the present, however, no phenomena have been observed which enable us to say whether these waves are or are not transverse vibrations of very small wave-length, longitudinal vibrations, or even vibrations at all. Nothing of the nature of polarisation or of interference has been described. The absence of polarisation can at the present stage of the investigation hardly be pressed as an argument against these rays being transverse vibrations. For, of the three methods of producing polarisation in light—reflection, refraction, and absorption—only the latter is available for these rays. Now the number of substances which produce sensible polarisation in ordinary light by absorption is very small, and unless a much larger number possess this property for the Röntgen rays, it is hardly likely that, even if there are such substances, they would have been discovered in the three months which have elapsed since the publication of Röntgen's discovery. I may remark that I have made a large number of experiments on the opacity to these rays of plates of tourmaline (1) with their axes crossed, (2) with their axes parallel, testing the intensity of the rays which came through in some cases by their action on a photographic plate, in others by the discharge they produced in an electrified plate on which they were incident. The result of these experiments was entirely negative, for although the tourmaline plates produced very considerable absorption of the rays, no difference was detected between the absorption when the axes were crossed and when they were parallel. It is very desirable that a large number of substances should be tested in this way.

M. C. Henri has made the very interesting observation that an opaque coin coated with the phosphorescent sulphide of zinc will allow these rays to pass through it; the details of this experiment will be received with much

NO. 1374, VOL. 53]

## search for polarised X-rays

### Comptes rendus hebdomadaires des séances de l'Académie des sciences
### Séance du lundi 23 Mars 1896



» J'ai employé le dispositif de la chambre noire.

» Un trou de petit diamètre (1mm), percé dans une plaque de laiton, située à quelques centimètres du tube, permettait d'obtenir l'image des régions actives sur une plaque photographique placée au delà.

» Je vérifiai d'abord ainsi que les parties utiles de la plaque sont bien celles que frappent les rayons cathodiques.

» Puis je disposai sur le trajet de ces rayons un obstacle en aluminium de forme simple (étoile ou croix); l'image de cet obstacle apparut très intense. Elle n'apparut pas lorsque, au moyen d'un aimant, on empêcha les rayons cathodiques de tomber sur l'obstacle.

» Cet obstacle pouvait d'ailleurs être pris comme anode : cela ne changea rien à aucun de ces deux résultats.

» L'expérience réussit également bien avec des corps moins perméables aux rayons X que l'aluminium et le verre, tels que le cristal et le platine. Pour le montrer, j'ai construit un tube en verre transparent aux rayons X, et, à l'intérieur de ce tube, je plaçai un obstacle mi-partie en platine, mi-partie en cristal. Naturellement la plaque photographique fut disposée de manière à recevoir les rayons X émis par celle des faces de l'obstacle que frappaient les rayons cathodiques. Les images obtenues furent encore nettes et très intenses.

» Dans aucune de ces expériences, la cathode ne donna son image.

» En résumé, *aux points où une matière quelconque arrête les rayons cathodiques, se développent des rayons de Röntgen,* et il ne paraît pas s'en développer en d'autres points.

» Ces rayons divergent dans toutes les directions; seulement certaines substances, telles que le cristal, les absorbent rapidement; on comprend ainsi pourquoi les tubes en cristal ont un rendement faible, quoique la production y soit intense. Toute l'importance pratique des tubes à fluorescence verdâtre résulte de la transparence du verre dont ils sont formés (1). »

PHYSIQUE. — *Recherches concernant les propriétés des rayons* X. Note de MM. le Prince **B. Galitzine** et **A. de Karnojitzky**, présentée par M. L. Cailletet. (Extrait.)

« En poursuivant, avec de nouveaux tubes de Crookes, de formes diverses, les recherches que nous avons déjà soumises à l'Académie, nous avons obtenu de nouvelles photographies d'après lesquelles les centres d'émission des rayons X sont nettement caractérisés.

(1) Travail fait au Laboratoire de Physique de l'École Normale.



» D'après l'ensemble de nos recherches, l'existence, dans quelques cas, d'un centre d'émission d'origine anodique semble être mise désormais hors de doute. On observe en outre quelquefois que, si l'on intervertit l'anode et la cathode, le nouveau centre anodique se produit là où se trouvait auparavant le centre cathodique, mais ce dernier est toujours plus intense.

» Pour expliquer l'ensemble des faits observés, nous avons, dans notre Note précédente, proposé l'hypothèse des foyers, qui semble bien correspondre à tous les détails de nos expériences et qui, d'ailleurs, est d'accord avec ce fait, que les centres d'émission se trouvent d'ordinaire dans le voisinage de l'endroit où le verre du tube de Crookes devient fortement fluorescent, vu que c'est précisément là que l'action des rayons cathodiques se concentre. C'est cette hypothèse qui nous paraît offrir le plus de vraisemblance....

» Il se présente une autre question. Les rayons X correspondent-ils à des vibrations longitudinales ou à des vibrations transversales? Cette question pourrait être tranchée si l'on pouvait démontrer qu'ils se polarisent.

» Les premières recherches que nous avons entreprises à ce sujet ont échoué. Nous avons alors fait préparer trois petites plaques de tourmaline très minces (environ 0mm,5 d'épaisseur). Sur la plus grande se posaient les deux autres, une parallèlement et l'autre perpendiculairement à la première. S'il y a polarisation là où les plaques sont croisées, on doit s'attendre à voir l'action des rayons X affaiblie. Il va sans dire que l'action de la lumière ordinaire a été exclue et qu'on a changé plusieurs fois la position relative des petites plaques, afin d'éliminer toute influence d'inégale épaisseur ou de manque d'homogénéité. Dans les huit épreuves obtenues, on peut distinguer que là où les plaques ont été croisées l'action photochimique des rayons X a été moindre (1).

» On peut en conclure que les rayons X se polarisent et, par suite, qu'ils correspondent à des vibrations transversales. »

(1) Pour renforcer ces épreuves, nous avons eu recours à M. Bourinsky, qui a récemment indiqué une méthode ingénieuse pour renforcer les négatifs faibles, méthode basée sur le principe de la superposition des pellicules (voir *Bulletin de l'Académie impériale des Sciences de Saint-Pétersbourg*, n° 4, avril 1895). La différence des teintes a été mise ainsi hors de doute, comme on peut le voir sur les épreuves que nous avons l'honneur de soumettre à l'Académie.

*C.R. Acad.Sci. Paris 122 (1896) 717-718*

## search for dichroism with X-rays

**Comptes rendus hebdomadaires des séances de l'Académie des sciences**
**Séance du lundi 30 Mars 1896**



PHYSIQUE. — *Sur la diffraction et la polarisation des rayons de M. Röntgen.* Note de M. G. Sagnac, présentée par M. Lippmann.

« I. Pour obtenir, avec un réseau par transmission à intervalles égaux, des images réelles d'une fente lumineuse (¹), on placerait ce réseau en avant d'une image réelle fournie par un faisceau convergent. On ne peut le faire avec les rayons de M. Röntgen, qui divergent à l'extérieur du tube de Crookes et pour lesquels on ne possède pas de lentilles. Pour obtenir des images réelles d'une fente, j'ai diaphragmé par une seconde fente, derrière laquelle est placé le réseau, l'entrée d'une grande chambre noire.

» J'ai employé un réseau de $\frac{1}{10}$ de millimètre, construit par M. Gaiffe avec des fils de platine de près de $\frac{1}{20}$ de millimètre de diamètre. La partie inférieure d'un faisceau *lumineux* défini par deux fentes métalliques distantes de $7^{cm},5$ passe au-dessous de la monture métallique du réseau et forme sur le verre dépoli de la chambre noire, à $35^{cm}$ au delà du réseau, une image réelle de la première fente. La partie supérieure du même faisceau traverse à $2^{cm},5$ derrière la seconde fente le réseau placé sous l'incidence de 45°, ce qui augmente ici les déviations et les intensités des faisceaux diffractés. Ces derniers ajoutent à l'image directe autant d'images diffractées dont quatre surtout sont bien nettes. Ces cinq images de la première fente supposée peu étroite ($1^{mm}$) se renforcent en se superposant en partie et forment ainsi une image d'aspect cannelé, plus large de $7^{mm}$ environ que l'image inférieure sans diffraction.

» Cela suffit pour conclure : *Les rayons de Röntgen qui ont impressionné la plaque sensible à travers le volet de bois du châssis ne possèdent pas de longueurs d'onde supérieures à 4 centièmes de micron* (¹).

» II. Les rayons de M. Röntgen sont-ils liés à un vecteur soit longitudinal, soit transversal et, dans ce dernier cas, comment les polariser?

» On ne peut songer actuellement à les polariser par réflexion, ni par réfraction, ni sans doute par diffraction. L'émission et la diffusion seraient peut-être à essayer. Il est, en tout cas, plus simple de tenter de les polariser *par absorption*.



chroïsme. La plage des sections principales parallèles laisse passer une intensité dont l'excès sur celle de l'autre plage est mesuré par $(o^2 - e^2)^2$.

» D'ailleurs pour les différences $(o - e)$, de signes peut-être différents, relatives à différentes longueurs d'onde, les différences d'intensité correspondantes s'ajoutent toujours en faveur de la plage où les sections principales sont parallèles.

» J'ai recherché si l'effet se produit avec les rayons de M. Röntgen en disposant de tels systèmes de trois lames sur le double de papier noir qui recouvre la plaque sensible. Le temps de pose s'est élevé jusqu'à plusieurs heures. Les clichés obtenus *avec l'intensité désirable* n'ont pas révélé de dichroïsme sensible :

| Corps employés. | Quartz. | Spath. | Tourmalines brunes (¹). | Mica. | Ferrocyanure de potassium. |
|---|---|---|---|---|---|
| Épaisseurs de chaque lame. | $0^{mm},03$ | $0^{mm},4$ | $0^{mm},5$ | $0^{mm},2$ | $0^{mm},4$ à $2^{mm}$ |

» Une précaution est indispensable pour éliminer l'influence d'une petite différence de nature comme il est arrivé pour les tourmalines : on fait tourner de 90° sur place chacune des deux demi-lames supérieures de manière que les plages des sections principales parallèles et des sections croisées s'échangent mutuellement. Une différence d'intensité due au dichroïsme doit suivre, dans son déplacement, le parallélisme des sections principales. Or, la très petite différence d'intensité, aperçue avec les tourmalines, persistait toujours sur la même demi-lame.

» Si la méthode se prête à un contrôle aussi facile et se trouve indépendante de la complexité des radiations employées, elle n'est pas, malheureusement, bien sensible. On voit aisément, *dans le cas de la lumière*, que, pour déceler une différence de $\frac{1}{10}$ par exemple, entre l'unité et le rapport $\frac{o}{e}$, il faudrait que la photographie pût révéler une différence relative de $\frac{1}{10}$ entre les intensités lumineuses qui impressionnent les deux plages contiguës, ce qui dépasse déjà beaucoup ce qu'on peut espérer.

» On ne saurait donc tirer de ces expériences négatives de dichroïsme un argument de grande valeur en faveur de l'hypothèse d'un vecteur longitudinal. Elles ajoutent seulement une distinction particulière de plus entre les rayons X et les rayons lumineux que nous connaissons (²). »

(¹) Ces lames de quartz, de spath et de tourmalines, fournies par M. Verlain, étaient parallèles à l'axe. Les lames de mica et de ferrocyanure étaient obtenues par clivage.

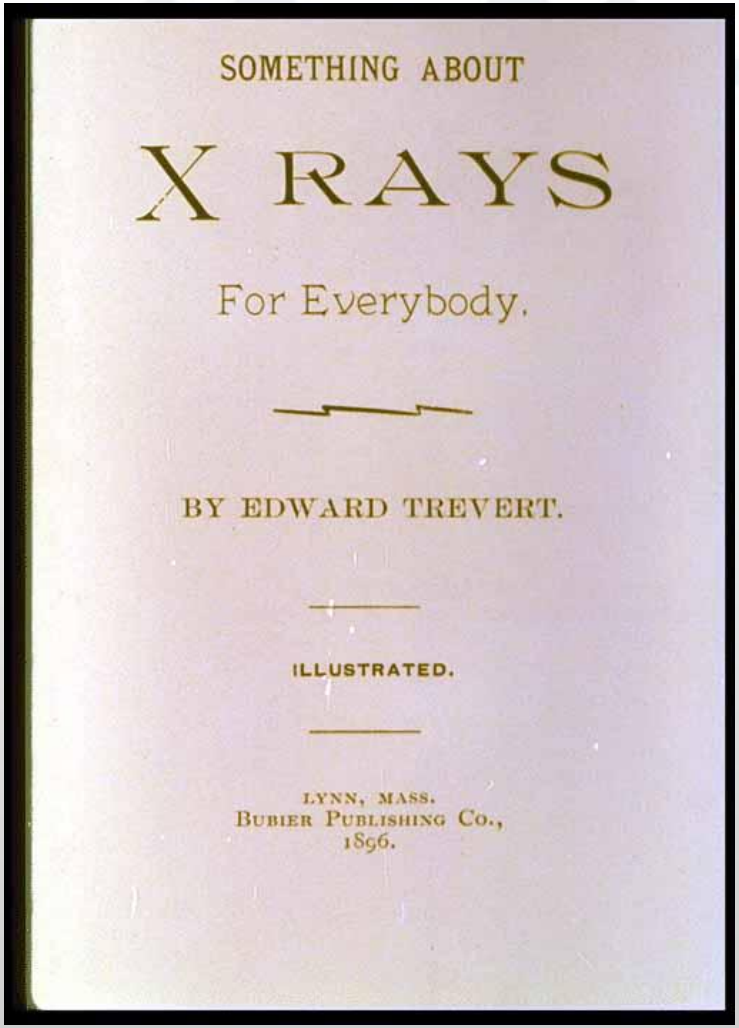
(²) Travail fait au laboratoire de M. Bouty, à la Sorbonne, en février et mars.

J'avais déjà réalisé quelques-unes de ces expériences, quand M. J.-J. Thomson a publié, dans le numéro du 27 février du journal *The Nature*, dont je viens d'avoir connaissance, la même expérience négative dans le cas de la tourmaline.

*C.R. Acad.Sci. Paris 122 (1896) 783-785*

# X-rays and Magnetism

## X-rays were everywhere and for everybody


SOMETHING ABOUT

X RAYS

For Everybody.

BY EDWARD TREVERT.

ILLUSTRATED.

LYNN, MASS.
BUBIER PUBLISHING CO.,
1896.

*How-to book gave step-by-step instructions on making radiographs, and included forms for ordering the equipment described.*

## X-rays are polarised

**Phil.Trans. A204 (1905), pp 467-479; Proc.Roy.Soc. London A, 77 (1906), pp247-255**



XIII. *Polarised Röntgen Radiation.*

*By* Charles G. Barkla, *D.Sc. (Liverpool), M.Sc. (Vict.), B.A. (Cantab.), King's College, Cambridge; Oliver Lodge Fellow, University of Liverpool.*

*Communicated by* Professor J. J. Thomson, *F.R.S.*

Received January 21,—Read February 16, 1905.

Though many attempts have been made to produce a beam of polarised X-radiation and to detect the polarisation by such methods as are applicable to ordinary light, the experiments have proved unsuccessful, and no evidence of polarity has been obtained. An arrangement of molecules such as occurs in crystals does not appear to affect a beam of this radiation transmitted through the crystalline substance.

The experiments here described were suggested by the results of an investigation of secondary radiation proceeding from gases and certain solids subject to X-rays.* for it was found that the gases experimented upon were the source of a radiation differing little in character from the primary radiation which produced it. In some respects the difference was inappreciable, as, for instance, in the absorbability of the radiations by aluminium. The primary and secondary radiations differed slightly, however, in their ionizing powers in air.† The energy of this secondary radiation was found to be proportional to the mass of gas through which the primary beam of definite intensity passed, and to be independent of the nature of the gas.

This led to the conclusion that this radiation is due to what might be called a scattering of the primary X-rays by the electrons constituting the molecules of the gas.

More recent experiments have shown that from light solids which emit a secondary radiation differing little from the primary, the energy of this radiation obeys the same law.

The phenomenon of secondary radiation from metals, however, is *apparently* much more complex, for in addition to secondary X-rays differing enormously in character from the primary, the metal radiator emits negative corpuscles. The total energy of these secondary radiations and the energy of the secondary X-rays alone are subjects

* C. G. Barkla, 'Phil. Mag.,' pp. 685–698, June, 1903, and pp. 543–560, May, 1904.

† See note, 'Phil. Mag.,' p. 549, May, 1904.





In conclusion, I have to thank Professor Schuster for lending me the radium used in these experiments. To Dr. Hutton I am indebted for the kind way in which he placed his electric furnaces at my disposal, and for his advice as to the best methods of obtaining the temperatures required.

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*Polarisation in Secondary Röntgen Radiation.*

By Charles G. Barkla, D.Sc. (Liverpool), M.Sc. (Victoria), B.A. (King's College, Cambridge), Demonstrator and Assistant Lecturer, University of Liverpool.

(Communicated by Professor J. J. Thomson, F.R.S. Received January 17,—Read February 8, 1906.)

In a paper on "Polarised Röntgen Radiation," * the writer gave an account of experiments which demonstrated the partial polarisation of a beam of X-rays proceeding from the antikathode of an X-ray focus-tube, and verified the theory previously given† of the production of secondary X-rays in light substances.‡

In that paper it was shown that the secondary radiation proceeding in a direction perpendicular to that of propagation of the primary radiation from certain substances placed in that primary beam should, according to the theory put forward, be plane polarised. From gases, however, the secondary radiation was not sufficiently intense to produce a tertiary of measurable intensity, and thus the polarisation of the secondary from them was not verifiable. On the other hand, though heavy metals were found to emit secondary radiation of sufficient intensity and ionising power to produce appreciable tertiary effects, in these metals the production of secondary radiation is a more complex phenomenon, and evidence of polarisation of the secondary beam is not to be expected from experiments upon them.

For the secondary radiator a substance had to be chosen which emitted a radiation of considerable intensity, yet differing very little in character from the primary. It had been shown that from such substances the intensity of radiation is proportional merely to the quantity of matter passed through by the primary of given intensity. A substance permitting the passage of the

* 'Phil. Trans.,' A, vol. 204, 1905, pp. 467—479.

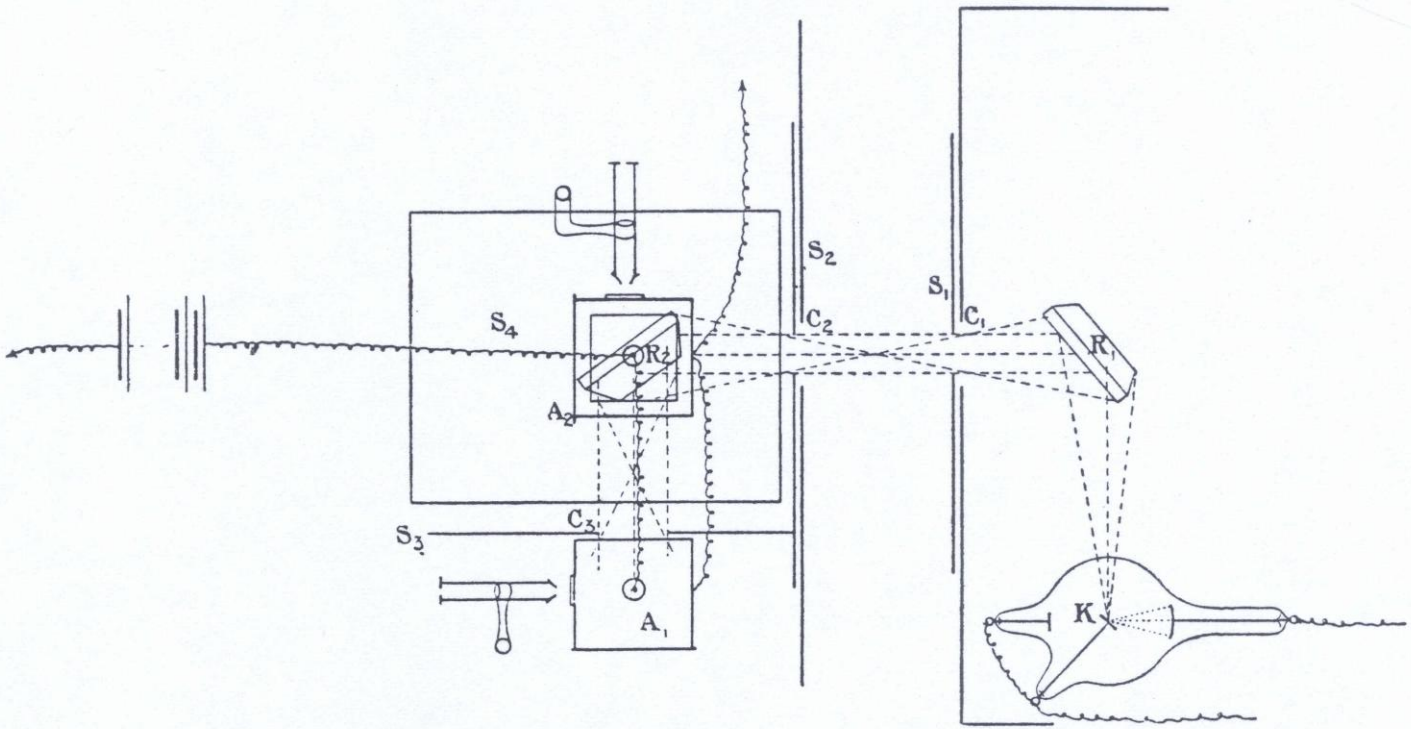
† J. J. Thomson, 'Conduction of Electricity through Gases,' p. 268; C. G. Barkla, 'Phil. Mag.,' June, 1903, and May, 1904.

‡ More precisely, substances of low atomic weight.

# X-rays and Magnetism

## X-rays are polarised

Shortly after I arrived at the conclusion as to the origin of secondary radiation from gases, Professor WILBERFORCE suggested to me the idea of producing a plane-polarised beam by means of a secondary radiator and of testing the polarisation by a tertiary radiator. The secondary radiation from gases under ordinary conditions is,




Plan of apparatus, showing position of bulb giving maximum deflection of electroscope $A_1$ and minimum of electroscope $A_2$.

***Experimental set-up of Prof. Charles G. Barkla***

# X-RAY ABSORPTION SPECTROSCOPY AND MAGNETISM



LXXXIII. *Some Experiments on Polarized Röntgen Radiation.* *By* J. Crosby Chapman, *B.Sc., Tutor in Mathematics, King's College, London; late Research Student of Gonville and Caius College, Cambridge* *.

*Introduction.*

ALL the experimental evidence obtained within the last few years indicates the fundamental identity of X-rays and light: this evidence includes experiments on the properties of the scattered, fluorescent, and corpuscular radiations, as well as on the polarization, interference, reflexion, and the velocity of X-rays. These seem to establish the theory that X-rays are merely light-waves of exceptionally short wave-length. Previous to the recent experiments on interference, one of the most significant properties which Röntgen rays had been shown to possess was that of polarization. Barkla †, as early as 1906, showed that it was possible to produce a secondary beam of X-rays which shows fairly complete polarization.

Up to the present all experiments on the rotation of the plane of polarization have been made using light of a wave-length of the order of that found in the visible spectrum. The results indicate that the magnitude of the rotation in quartz and other active substances varies *inversely* as some power of the wave-length. This power over a considerable portion of the spectrum examined is the square, but it does not hold over a wider range.

On the assumption that Röntgen radiation is merely light of exceptionally short wave-length, it seemed possible that a great rotation of the plane of polarization of X-rays might be obtained under suitable conditions.

The object of this present research, suggested by Professor Barkla, was to test directly whether anything corresponding to a rotation of the plane of polarization could be obtained with X-rays.

*Apparatus.*

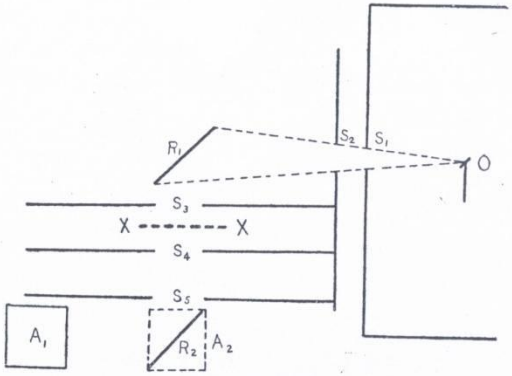
The apparatus was essentially similar to that used in previous experiments on X-ray polarization. X-rays from the anticathode O of an X-ray tube passed through the slits $S_1$ and $S_2$ in lead screens on to the first radiator $R_1$; a portion of the rays scattered from this radiator passed through the slits $S_3$, $S_4$, and $S_5$ in further lead screens, on to the radiator $R_2$. At equal horizontal and vertical distances of

* Communicated by Prof. C. G. Barkla, F.R.S. The expenses of this research have been partly covered by a grant from the Royal Society.

† Barkla, Proc. Roy. Soc. Series A, vol. lxxvii. pp. 247–255 (1906).



about 12 cm. from the radiator $R_2$, two similar electroscopes, $A_1$ horizontal, and $A_2$ vertical, were placed so as to subtend the same solid angle at the plate $R_2$, the face of which was equally inclined to vertical and horizontal lines drawn from the centre of the radiator to the electroscopes $A_1$ and $A_2$.




$A_2$ (Dotted lines) vertically above $R_2$

The substance which was being tested as regards its power of rotating the plane of polarization was placed at XX, so that the secondary partially polarized beam of rays from $R_1$ passed directly through it before striking $R_2$. A full account of this apparatus and the conditions which it must satisfy is given in the original paper *, where it is demonstrated that while there is but little polarization in the primary beam, yet in the secondary beam there is a considerable amount of polarization. This follows from elementary considerations of the usual theory of scattering.

In the present experiment, the primary and secondary beams were horizontal. When this was the case, and both the radiators at $R_1$ and $R_2$ consisted merely of a scattering agent such as carbon, it was found that the horizontal tertiary beam passing into $A_1$ was of much greater intensity than the vertical tertiary beam passing into $A_2$. When, however, the carbon plate at $R_2$ was replaced by a plate capable of giving a preponderating characteristic radiation, which has been shown to be distributed equally in all directions †, the intensities of the radiation reaching the

* Barkla, Proc. Roy. Soc. Series A, vol. lxxvii. pp. 247–255.

† Barkla and Ayres, Phil. Mag. Feb. 1911, pp. 271–278.

**Phil. Mag. 25, 792-802 (1913)**

# X-RAY ABSORPTION SPECTROSCOPY AND MAGNETISM



Combining this result with others obtained :—

TABLE IX.

| Polarization of direct secondary beam. | Polarization of secondary beam after transmission through sugar solution. |
|---|---|
| 3·5 | 3·5 |
| 3·6 | 3·7 |
| 3·4 | 3·3 |
| 3·4 | 3·4 |
| 3·4 | 3·6 |
| Mean value . 3·5 | 3·5 |

These results clearly prove that no measurable rotation of the plane of polarization of X-rays is produced by a sugar solution which is active for light in the visible spectrum.

The interpretation of these results is not that a fundamental difference exists between X-rays and light, but the experiments rather indicate that the usual formulæ which determine the relation between rotation and wave-length for light in the visible spectrum are quite inadequate when light of exceptionally short wave-length is concerned.

*Absorption of polarized X-rays by Iron in Magnetized State.*

It is legitimate to assume that the absorption of X-rays by an atom of any given substance must depend to a certain extent on two factors :—(1) the period of the X-radiation absorbed ; (2) the natural period of vibration of the electrons within the atom.

In order to explain double refraction and other phenomena peculiar to crystalline substances, it is necessary to suppose that the period of vibration of the electrons is dependent on the direction of vibration with respect to the atomic system.

Now in a polarized beam of X-rays, the electrical vibrations are largely in one direction. Consider now such a beam being transmitted through iron in which, since it is unmagnetized, there is no regular orientation of the orbits of the electron in the system. If, now, the iron is magnetized, the orientation becomes more regular, and if absorption of X-radiation is dependent on the direction of its own electrical vibration relatively to that of rotation of the electron in the



absorbing substance, it might be expected that there would be a considerable alteration in the absorption of the material. With such an idea in mind, the following experiment was tried.

A polarized beam of X-rays was passed through iron when unmagnetized, and later when magnetized. The absorption of the polarized beam was determined in each case. In the first experiment, the field produced was perpendicular to the sheet of iron absorbing and parallel to the direction of the X-rays absorbed. In the second part the field was perpendicular to the direction of the X-rays absorbed and parallel to the sheet of iron absorbing. The field was produced in the first instance by a coil of wire, and in the second case by a powerful horseshoe electromagnet. In each case a standardizing electroscope was employed to allow for changes in the primary beam.

The results are given in tabular form.

Magnetic field perpendicular to X-rays and parallel to iron sheet.

H = 30,000 gauss.

| Absorption by iron unmagnetized. | Absorption by iron magnetized. |
|---|---|
| 68·0 per cent. | 68·0 per cent. |
| 67·2 ,, | 67·0 ,, |
| 20·1 ,, | 20·3 ,, |
| 20·5 ,, | 20·1 ,, |

Magnetic field parallel to X-rays and perpendicular to iron sheet.

H = 320 gauss.

| Absorption by iron unmagnetized. | Absorption by iro magnetized. |
|---|---|
| 53·0 per cent. | 53·1 per cent. |
| 31·7 ,, | 31·0 ,, |
| 31·7 ,, | 32·4 ,, |
| 31·8 ,, | 31·9 ,, |

Thus the absorption of a polarized beam of X-rays by iron is not appreciably dependent upon whether it is magnetized or unmagnetized ; that is, absorption of X-rays is independent of the orientation of the electronic orbits inside the atom, so far as they are affected by the magnetic state.

# X-RAY ABSORPTION SPECTROSCOPY AND MAGNETISM

*Phys. Rev. 3 (1914) 306-313*



THE EFFECT OF MAGNETIZATION ON THE OPACITY OF IRON TO RÖNTGEN RAYS.

BY A. H. FORMAN.

IN a general way it would seem natural to expect that a change in the molecular arrangement of iron by magnetization, which produces the phenomenon of magneto-striction, should to some extent also affect the opacity of the iron to Röntgen rays. The investigations along this line have all given negative results. Experiments[1] in which the polarized secondary rays were used instead of the ordinary Röntgen rays have been tried with negative results.

*Phys. Rev. 7 (1916) 119-124*



THE EFFECT OF MAGNETIZATION ON THE OPACITY OF IRON TO RÖNTGEN RAYS.

BY A. H. FORMAN.

IN a previous paper[1] the results covering the investigation of the effect of magnetizing the iron in a plane perpendicular to the path of the Röntgen rays were published. The results were negative with a set up of apparatus sensitive enough to detect a change of one part in ten thousand under the most favorable conditions.

# X-RAY ABSORPTION SPECTROSCOPY AND MAGNETISM



Phys. Rev. 22, 320–323 (1923)

## THE EFFECT OF A MAGNETIC FIELD ON THE ABSORPTION OF X-RAYS

By Joseph A. Becker[1]

Abstract

**Effect of a magnetic field on the absorption of x-rays.**—A continuation of previous work (Phys. Rev. **20**, p. 134, 1922) with improved specimen holders and technique. *Iron, aluminum, and carbon* showed no changes in the absorption coefficient as great as one part in 10,000 (due to a magnetic field of 17,000 gauss) when the tube was operated at 90 kv peak. *Carbon, wood, and lithium* also showed effects smaller than the limit of accuracy of the apparatus—2 parts in 10,000 for 61 kv, 4 parts in 10,000 for 46 kv, and 10 parts in 10,000 for 30 kv. If the effect exists, then, it is very small.

# X-rays and Magnetism

## *On question about the magnetic rotation of plane polarised primary X-rays*

**Zeitschrift fur Physik, 39, 886-900 (1926)**

### Zur Frage nach der magnetischen Drehung der Polarisationsebene primärer Röntgenstrahlen.

Von W. Kartschagin und E. Tschetwerikowa in Moskau.

Mit 3 Abbildungen. (Eingegangen am 16. September 1926.)

In der vorliegenden Arbeit wurde eine Wirkung des magnetischen Feldes auf die Polarisationsebene der Röntgenstrahlen bei ihrem Durchgang durch Paraffin und Eisen untersucht. Auf Grund der Resultate der Arbeit und der Versuche bei einigen Annahmen, die Folgerungen der Elektronentheorie der Dispersion auf die zu behandelnde Frage anzuwenden, ziehen wir folgenden Schluß: Beim Durchgang primärer Röntgenstrahlen durch Paraffin kann die magnetische Drehung der Polarisationsebene wegen der Kleinheit des Drehungswinkels nicht beobachtet werden. Beim Durchgang der primären Strahlen durch Eisen kann man eine Drehung der Polarisationsebene erwarten, aber eine genaue Messung des Drehungswinkels ist sehr schwierig.

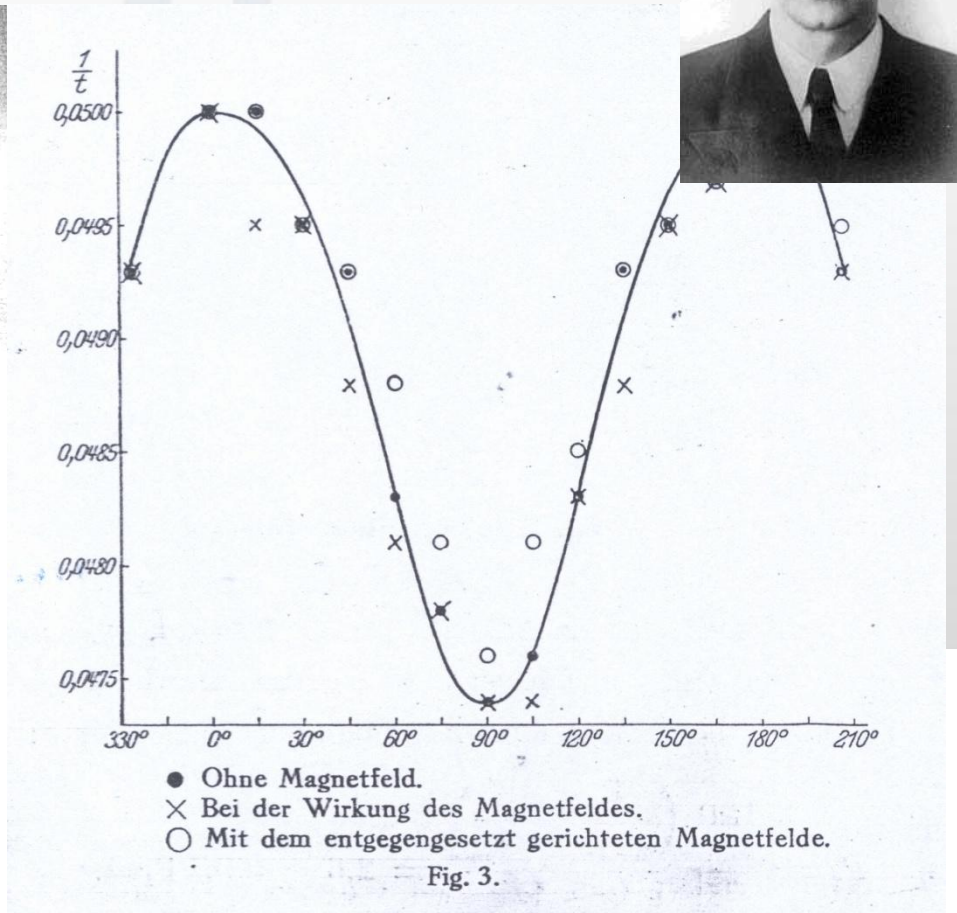
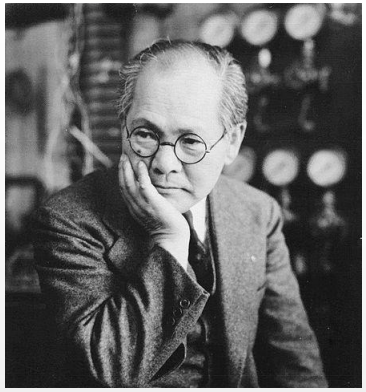



Fig. 3.

**In this work, an effect of a magnetic field on plane polarised X-rays scattered by Paraffin and Iron is studied. Far from the absorption edge the rotation can not be observed due the weakness of the rotation angle. Whereas at an absorption edge of ferromagnetic materials one can expect to measure the magnetic rotation eventhough precise measurements are very difficult.**

# X-rays and Magnetism



**Zeitschrift fur Physik, 52, 853-868 (1929)**

**On the scattering of radiation by free electrons according to the new relativistic quantum dynamics of Dirac**

**Über die Streuung von Strahlung durch freie Elektronen nach der neuen relativistischen Quantendynamik von Dirac.**

Von **O. Klein** und **Y. Nishina** in Kopenhagen.

(Eingegangen am 30. Oktober 1928.)

Auf Grund der neuen, von Dirac entwickelten relativistischen Quantendynamik wird die Intensität der Comptonstreustrahlung berechnet. Das Resultat zeigt Abweichungen von den entsprechenden Dirac-Gordonschen Formeln, die von der zweiten Größenordnung hinsichtlich des Verhältnisses der Energie des primären Lichtquants zu der Ruheenergie des Elektrons sind.

**interaction between photons and magnetic electrons is considered for the first time**

$$\frac{d\sigma}{d\Omega} = \alpha^2 r_c^2 P(E_\gamma,\theta)^2 [P(E_\gamma,\theta) + P(E_\gamma,\theta)^{-1} - 1 + \cos^2(\theta)]/2$$

$$P(E_\gamma,\theta) = \frac{1}{1 + (E_\gamma/m_e c^2)(1-\cos\theta)}$$

PHYSICAL REVIEW VOLUME 92, NUMBER 4 NOVEMBER 15, 1953

## Compton Scattering of 2.62-Mev Gamma Rays by Polarized Electrons*,†

S. B. GUNST‡ AND L. A. PAGE
*University of Pittsburgh, Pittsburgh, Pennsylvania*
(Received July 20, 1953)

The differential cross section for Compton scattering of a circularly polarized photon by an electron with given initial spin orientation can be written as a sum of the common Klein-Nishina formula for no polarization and a term sensitive to polarization. The total cross section is $\sigma=\sigma_0\pm\sigma_1$. A measurement of the transmission of 2.62-Mev gamma rays through iron magnetized along the transmission direction relative to that through unmagnetized iron gives the absolute value of $\sigma_1$ for this energy, if the number $\nu_s$ of polarized electrons per iron atom at saturation is known. For $\nu_s=2.06$, $\sigma_1/\pi r_0^2=0.089\pm0.007$. This agrees with the theoretical value 0.093. Alternatively, the theoretical $\sigma_1$ and the measurements would yield $\nu_s=1.97\pm0.15$.

The application of the method of this experiment to measurement of gyromagnetic ratios for ferromagnets is suggested, as is its application to the analysis of circularly polarized radiation.

PHYSICAL REVIEW B VOLUME 2, NUMBER 9 1 NOVEMBER 1970

## Magnetic Scattering of X Rays from Electrons in Molecules and Solids

P. M. Platzman
*Bell Telephone Laboratories, Murray Hill, New Jersey*

and

N. Tzoar*
*City College of the City University, New York City, New York*
(Received 2 June 1970)

The scattering of moderately high-energy x rays from electrons in magnetic solids is analyzed. We show that (a) the incoherent Compton scattering of polarized x rays can be used to determine the spin-dependent momentum distribution function of electrons in ferromagnetic materials, and (b) the coherent Bragg scattering of unpolarized x rays can be used to determine the magnetic structure of antiferromagnetic solids below their transition temperature.

Volume 39A, number 2 PHYSICS LETTERS 24 April 1972

## OBSERVATION OF MAGNETIC SUPERLATTICE PEAKS BY X-RAY DIFFRACTION ON AN ANTIFERROMAGNETIC NiO CRYSTAL

F. De BERGEVIN and M. BRUNEL

*Laboratoire de rayons-X, Cédex 166, 38–Grenoble-Gare, France*

Received 14 February 1972

We observe on a NiO single crystal two superlattice X ray diffraction peaks, which disappear above the Néel point. Their intensities, $4 \times 10^{-8}$ smaller than normal ones, agree with those evaluated from photon-electron spin interaction.

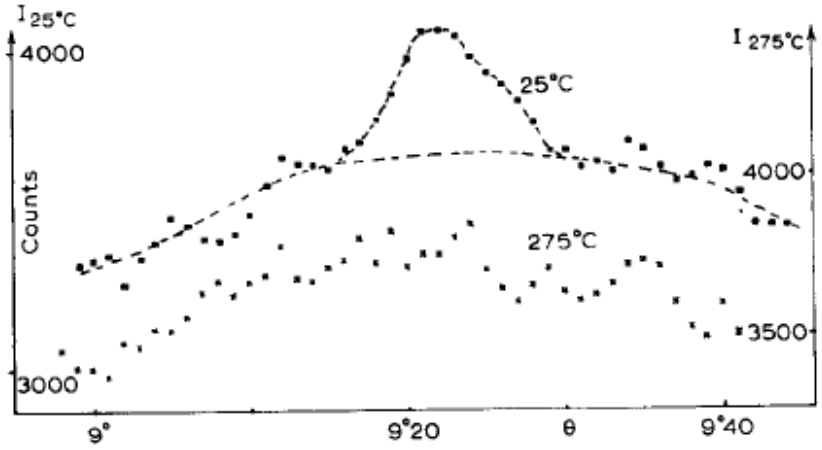



Fig. 1. Intensity $I_t(\theta)$ near the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ position at $t = 25°$ C and 275° C in counts/225 min. The hump which cover the interval could be due to some impurity.

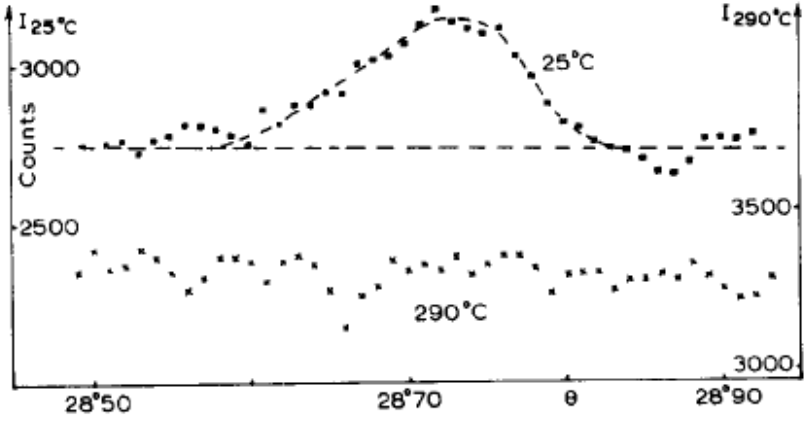



Fig. 2. Intensity $I_t(\theta)$ near the $(\frac{3}{2}\frac{3}{2}\frac{3}{2})$ position at $t = 25°$C and 290°C in counts/225 min.

*Acta Cryst.* (1983). A**39**, 84–88

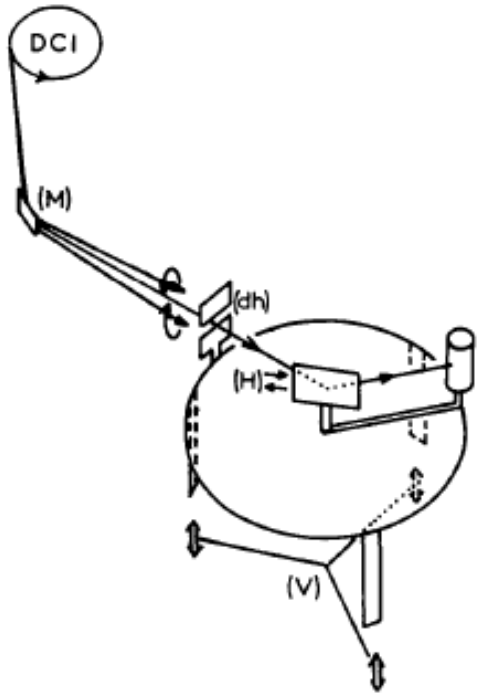
## Etude de la Polarisation Circulaire du Rayonnement Synchrotron dans la Gamme des Rayons X par Diffraction sur un Composé Ferrimagnétique

PAR M. BRUNEL, G. PATRAT ET F. DE BERGEVIN

*Laboratoire de Cristallographie, Centre National de la Recherche Scientifique, Laboratoire associé à l'USMG, 166 X, 38042 Grenoble CEDEX, France*

ET F. ROUSSEAUX* ET M. LEMONNIER

*LURE, Université de Paris-Sud,* 91405 *Orsay CEDEX, France*




### Conclusions

Malgré l'imprécision des résultats due, d'une par à la durée limitée des expériences, d'autre part aux instabilités du faisceau, nous avons pu montrer l'intérêt de la diffraction magnétique pour l'étude de la polarisation circulaire d'un faisceau de rayons X de longueur d'onde voisine de 1 Å. Les résultats sont cohérents, à la fois avec les calculs de polarisation circulaire du rayonnement synchrotron, et avec les calculs de diffraction magnétique des rayons X. L'utilisation du rayonnement synchrotron pour des études de structure magnétique pourrait être complémentaire de la diffraction neutronique, en particulier dans le cas des corps absorbant trop les neutrons (Gd par exemple) ou dans le cas où l'effet du moment orbital est difficile à séparer de celui du moment de spin (ces deux effets ne se combinent pas de la même façon pour les rayons X et les neutrons); toutefois elle semble limitée par la faible précision que l'on peut atteindre actuellement.

# Basics of XMCD

***The first serious approach to the problem of absorption of circularly polarized X-rays***

PHYSICAL REVIEW B VOLUME 12, NUMBER 11 1 DECEMBER 1975

## Calculation of the $M_{23}$ magneto-optical absorption spectrum of ferromagnetic nickel

J. L. Erskine*
*Department of Physics, University of Illinois, Urbana, Illinois 61801*

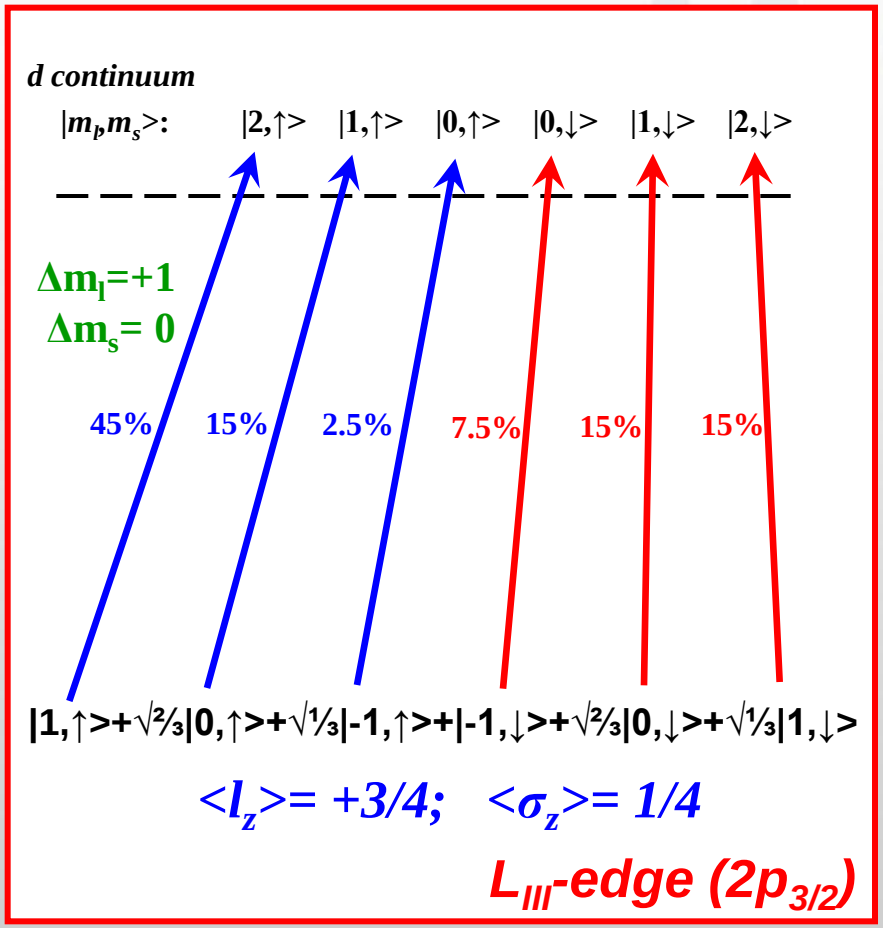
E. A. Stern†
*Department of Physics, University of Washington, Seattle, Washington 98195*
(Received 28 April 1975)

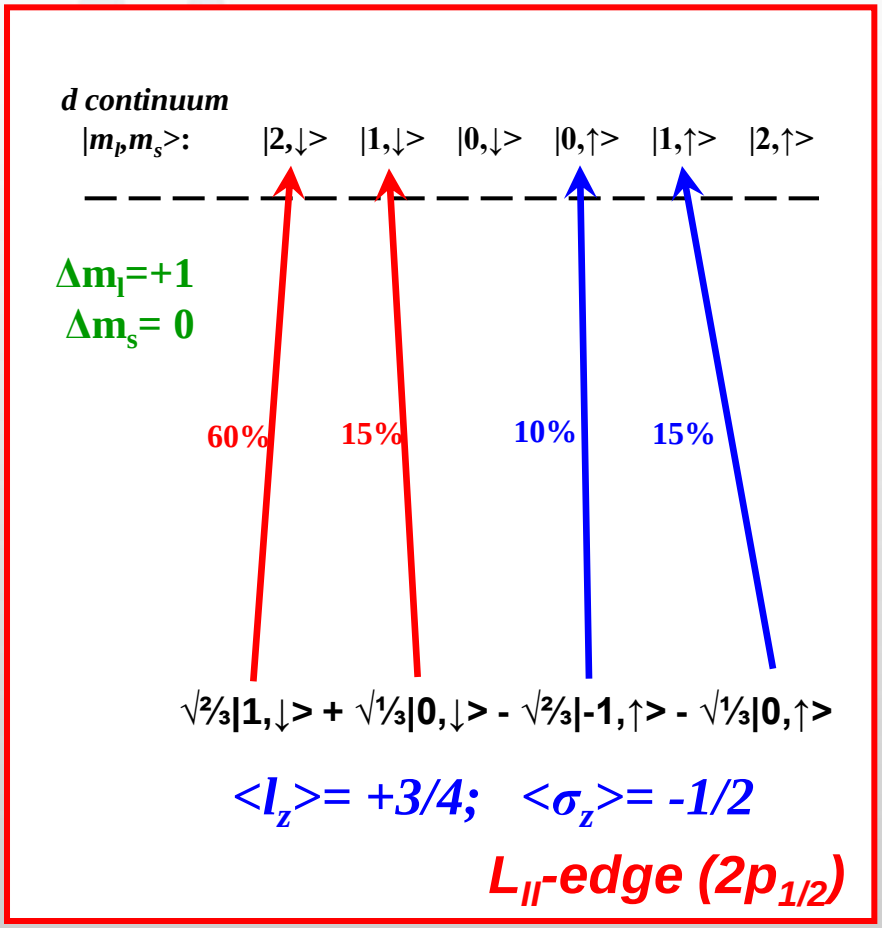
The $M_{23}$ magneto-optical absorption spectrum of ferromagnetic nickel is calculated using an approach similar to the component state-density method that has been successfully used in obtaining valence-band emission and absorption x-ray spectra of metals. The $M_{23}$ magneto-optical effects result predominantly from spin-orbit splitting of the $3p$ core state in conjunction with the final $d$-state spin polarization. The calculated spectrum exhibits features that are directly related to electronic structure parameters including the $3p$ core spin-orbit splitting, and the unfilled $d$-band spin polarization. Temperature variations in the magneto-optical structure can be used to determine separately the exchange-splitting variation and spin-wave excitation contributions to the decrease in the magnetization. Experimental verification of these predictions should provide insight into the applicability of the Stoner model to ferromagnetic nickel and may be helpful in resolving some of the apparently conflicting results of other experimental probes of the spin polarization near the Fermi level in nickel.

***Two-step model***

# Two Step Model of XMCD

## Absorption of a right circularly polarized photon



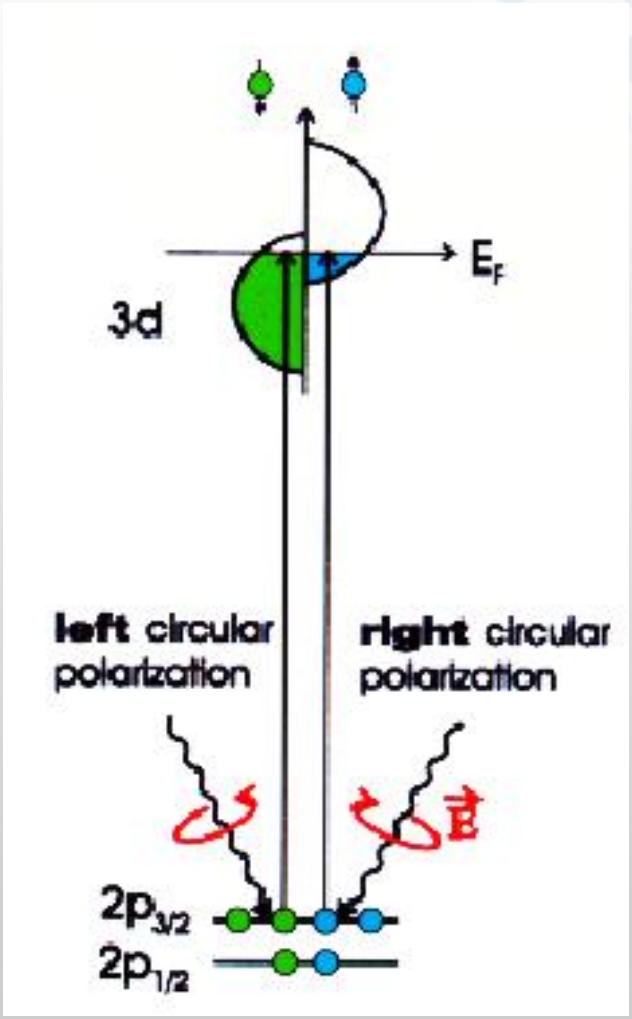



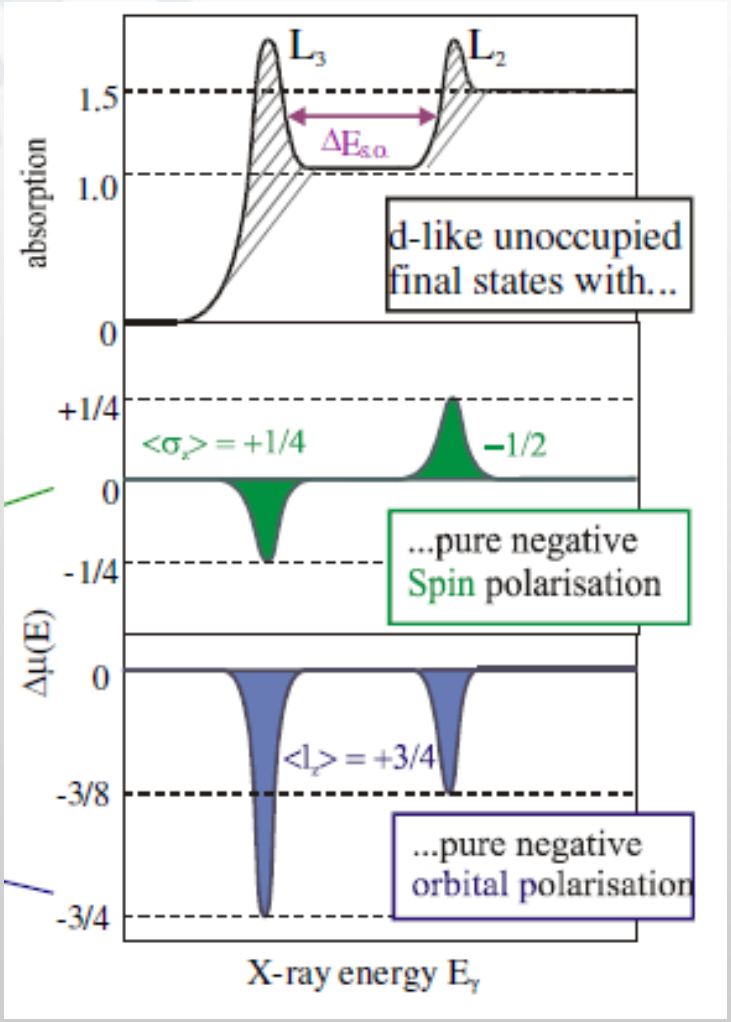

**Excited photoelectrons are spin polarized**

# Basics of XMCD

## Two Step Model of XMCD

**Exchange splitting of the valence band is driving the second step**



## First experimental evidence of influence of magnetism on X-ray absorption

VOLUME 55, NUMBER 19 PHYSICAL REVIEW LETTERS 4 NOVEMBER 1985

**Strong Magnetic Dichroism Predicted in the $M_{4,5}$ X-Ray Absorption Spectra of Magnetic Rare-Earth Materials**

B. T. Thole, G. van der Laan, and G. A. Sawatzky
*Physical Chemistry Department of the Materials Science Center, University of Groningen, 9747 AG Groningen, The Netherlands*
(Received 21 August 1985)

A theory is presented which predicts an anomalously large magnetic dichroism in the $M_{4,5}$ x-ray absorption-edge structure of rare earths in magnetically ordered materials. Polarized synchrotron radiation can therefore be used to determine accurately the magnitude, the orientation, and the temperature and magnetic field dependence of the local rare-earth magnetic moment in a large variety of magnetically ordered materials and thin films.

PACS numbers: 78.20.Ls, 78.70.Dm

PHYSICAL REVIEW B VOLUME 34, NUMBER 9 1 NOVEMBER 1986

**Experimental proof of magnetic x-ray dichroism**

Gerrit van der Laan, Bernard T. Thole, and George A. Sawatzky
*Physical Chemistry Department of the Material Science Center, University of Groningen, NL-9747 AG Groningen, The Netherlands*

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*Research Institute for Materials, University of Nijmegen, Toernooiveld 10, NL-6525 ED Nijmegen, The Netherlands*

Jean-Marc Esteva and Ramesh Karnatak
*Laboratoire pour l'Utilisation du Rayonnement Electromagnétique, F-91405 Orsay Cédex, France*

J. P. Remeika
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Hanna A. Dabkowska
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(Received 24 April 1986)

What we believe to be the first experimental results have been obtained on strong magnetic x-ray dichroism in the $M_{4,5}$ absorption spectra of magnetically ordered rare-earth materials, in accordance with recent predictions.

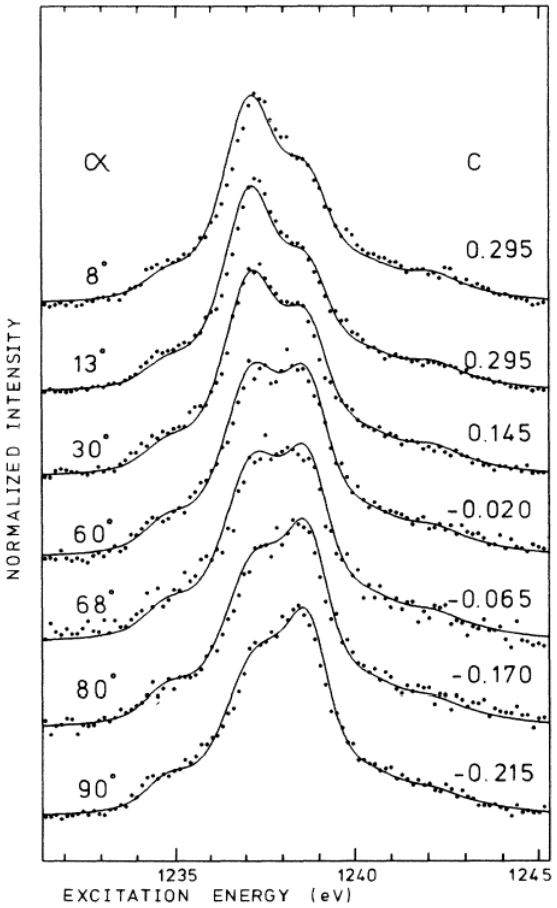


FIG. 1. Experimental $M_5$ absorption spectra of TbIG at various values of $\alpha$, which is the angle between the polarization vector of the x rays and the [111] magnetization direction. The solid lines are fits using Eq. (5). The optimum values of $C$ are indicated.

## First experimental evidence of XMCD

VOLUME 58, NUMBER 7 PHYSICAL REVIEW LETTERS 16 FEBRUARY 1987

### Absorption of Circularly Polarized X Rays in Iron

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and

R. Frahm and G. Materlik

*Hamburger Synchrotronstrahlungslabor am Deutsches Elektronen-Synchrotron DESY, D-2000 Hamburg 52, West Germany*

(Received 22 September 1986)

The transmission of synchrotron radiation through magnetized iron at energies above the $K$-absorption edge shows relative differences for right and left circular polarization of several times $10^{-4}$. The observed spin dependence of the near-edge photoabsorption is proportional to the difference of the spin densities of the unoccupied bands. In the extended absorption region up to 200 eV above the Fermi level a small spin-dependent absorption is observed and thus is expected to give information on the magnetic neighborhood of the absorbing atom.

PACS numbers: 75.50.Bb, 75.10.Lp, 75.25.+z, 78.70.Dm

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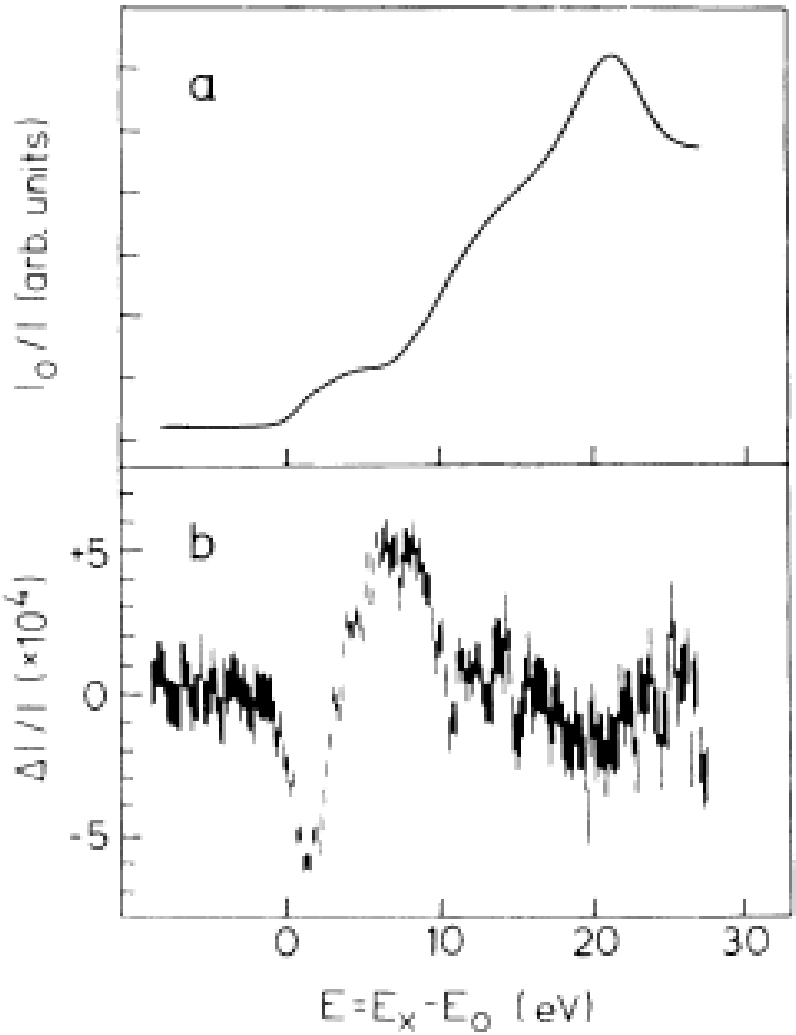
PACS numbers: 75.50.Bb, 75.10.Lp, 75.25.+z, 78.70.Dm

# Basics of XMCD

## The first Fe K-edge XMCD spectrum



RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 42, NUMBER 11 15 OCTOBER 1990-I

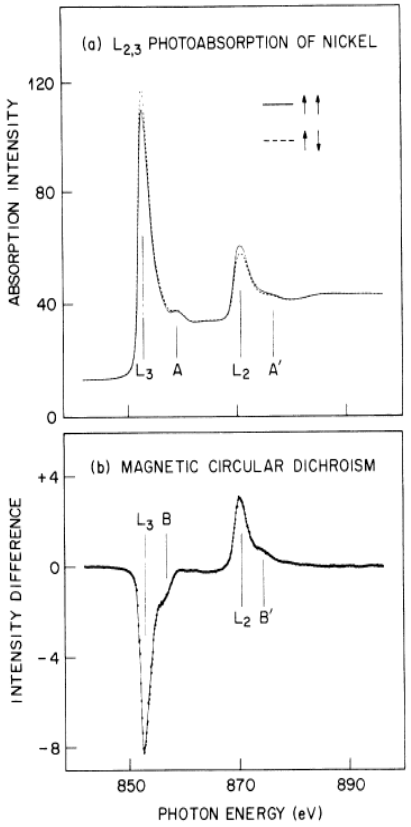
## Rapid Communications

*Rapid Communications are intended for the accelerated publication of important new results and are therefore given priority treatment both in the editorial office and in production. A Rapid Communication in* **Physical Review B** *should be no longer than 4 printed pages and must be accompanied by an abstract. Page proofs are sent to authors.*

### Soft-x-ray magnetic circular dichroism at the $L_{2,3}$ edges of nickel

C. T. Chen, F. Sette, Y. Ma, and S. Modesti
*AT&T Bell Laboratories, Murray Hill, New Jersey 07974*
(Received 2 March 1990)

Magnetic circular dichroism (MCD) has been observed at the $L_{2,3}$ absorption edges of ferromagnetic nickel by use of circular-polarized soft-x-ray synchrotron radiation. The MCD intensity ratio between the $L_2$ and the $L_3$ edges is found to differ appreciably from that predicted by a simple exchange-split-valence-band model. Fine MCD features, imperceptible in the absorption spectra, are also observed and a tentative interpretation is given. This work, demonstrating the feasibility of MCD measurements in the soft-x-ray region, provides a new approach to study $3d$ and $4f$ ferromagnetic systems with their respective dipole-permitted $2p \rightarrow 3d$ and $3d \rightarrow 4f$ transitions.



# XMCD Sum Rules

**Sum rules relate experimental XMCD spectra to the spin and orbital moments**

VOLUME 68, NUMBER 12 PHYSICAL REVIEW LETTERS 23 MARCH 1992

**X-Ray Circular Dichroism as a Probe of Orbital Magnetization**

B. T. Thole,[1] Paolo Carra,[2] F. Sette,[2] and G. van der Laan[3]

[1]*Department of Chemical Physics, Materials Science Centre, University of Groningen, Nijenborgh 16, 9747 AG Groningen, The Netherlands*

[2]*European Synchrotron Radiation Facility, BP 220, F-38043 Grenoble CEDEX, France*

[3]*Daresbury Laboratory, Science and Engineering Research Council, Warrington, WA4 4AD, United Kingdom*

(Received 2 December 1991)

A new magneto-optical sum rule is derived for circular magnetic dichroism in the x-ray region (CMXD). The integral of the CMXD signal over a given edge allows one to determine the ground-state expectation value of the orbital angular momentum. Applications are discussed to transition-metal and rare-earth magnetic systems.

## Orbital sum rule

$$\int_{j^+ + j^-} (\mu^+ - \mu^-) = \frac{2l(l+1)}{l(l+1) + 2 - c(c+1)} \times C \times \langle L_z \rangle$$

## Spin sum rule

$$\int_{j^+} (\mu^+ - \mu^-) - \frac{c+1}{c} \int_{j^-} (\mu^+ - \mu^-) = C \times [A\langle S_z \rangle + B\langle T_z \rangle]$$

VOLUME 70, NUMBER 5 PHYSICAL REVIEW LETTERS 1 FEBRUARY 1993

**X-Ray Circular Dichroism and Local Magnetic Fields**

Paolo Carra,[1] B. T. Thole,[1,2] Massimo Altarelli,[1] and Xindong Wang[3]

[1]*European Synchrotron Radiation Facility, BP 220, F-38043 Grenoble CEDEX, France*

[2]*Department of Chemical Physics, Materials Science Center, University of Groningen, Nijenborgh 16, 9747 AG Groningen, The Netherlands*

[3]*Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011*

(Received 13 July 1992)

Sum rules are derived for the circular dichroic response of a core line (CMXD). They relate the intensity of the CMXD signal to the ground-state expectation value of the magnetic field operators (orbital, spin, and magnetic dipole) of the valence electrons. The results obtained are discussed and tested for transition metals and rare earths.

$$T = \sum_i (s_i - 3r_i(r_i \cdot s_i)/r_i^2) \qquad C = \frac{1}{n_h} \int_{j^+ + j^-} (\mu^+ + \mu^- + \mu^0)$$ - *X-ray absorption cross section per hole;*

$$A = \frac{l(l+1) - 2 - c(c+1)}{3c} \qquad B = \frac{l(l+1)[l(l+1) + 2c(c+1) + 4] - 3c(c-1)^2(c+2)^2}{6c \cdot l(l+1)}$$

# XMCD Sum Rules

$$\langle L_z \rangle = -\frac{4}{3} \cdot C \cdot (A + B)$$

$$\langle S_z \rangle - 7\langle T_z \rangle = -2C \cdot (A - 2B)$$

$$\frac{\langle L_z \rangle}{\langle S_z \rangle - 7\langle T_z \rangle} = \frac{2}{3} \cdot \frac{(A + B)}{(A - 2B)}$$

**in the case of $L_{3,2}$ absorption edges**

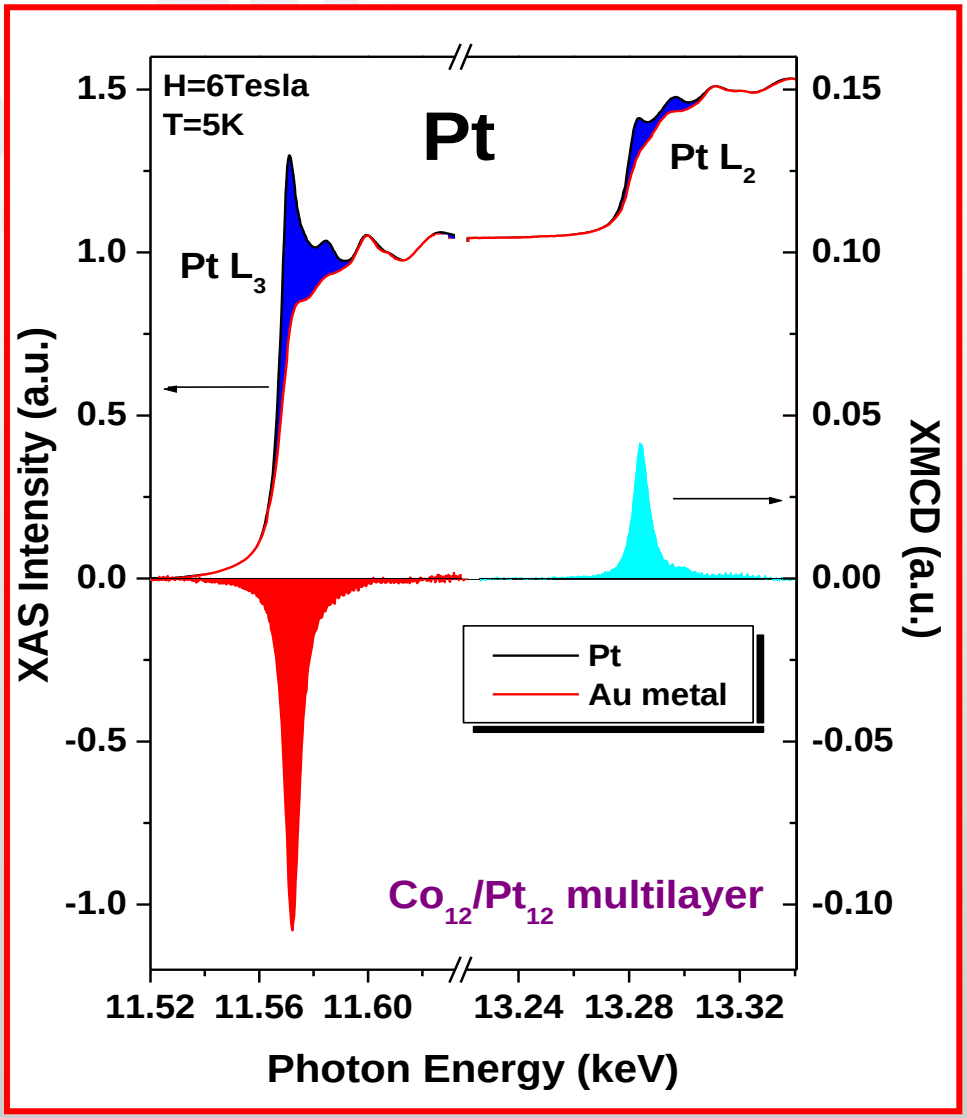
$$C = \frac{(n_h^{Pt} - n_h^{Au})}{A}$$

**A ~ integrated intensity of transitions into unoccupied *d* band**

$n_h^d$ = number of holes in *d* band

$$\langle T_z \rangle \ll \langle S_z \rangle$$

**(if S.-O. is small)**

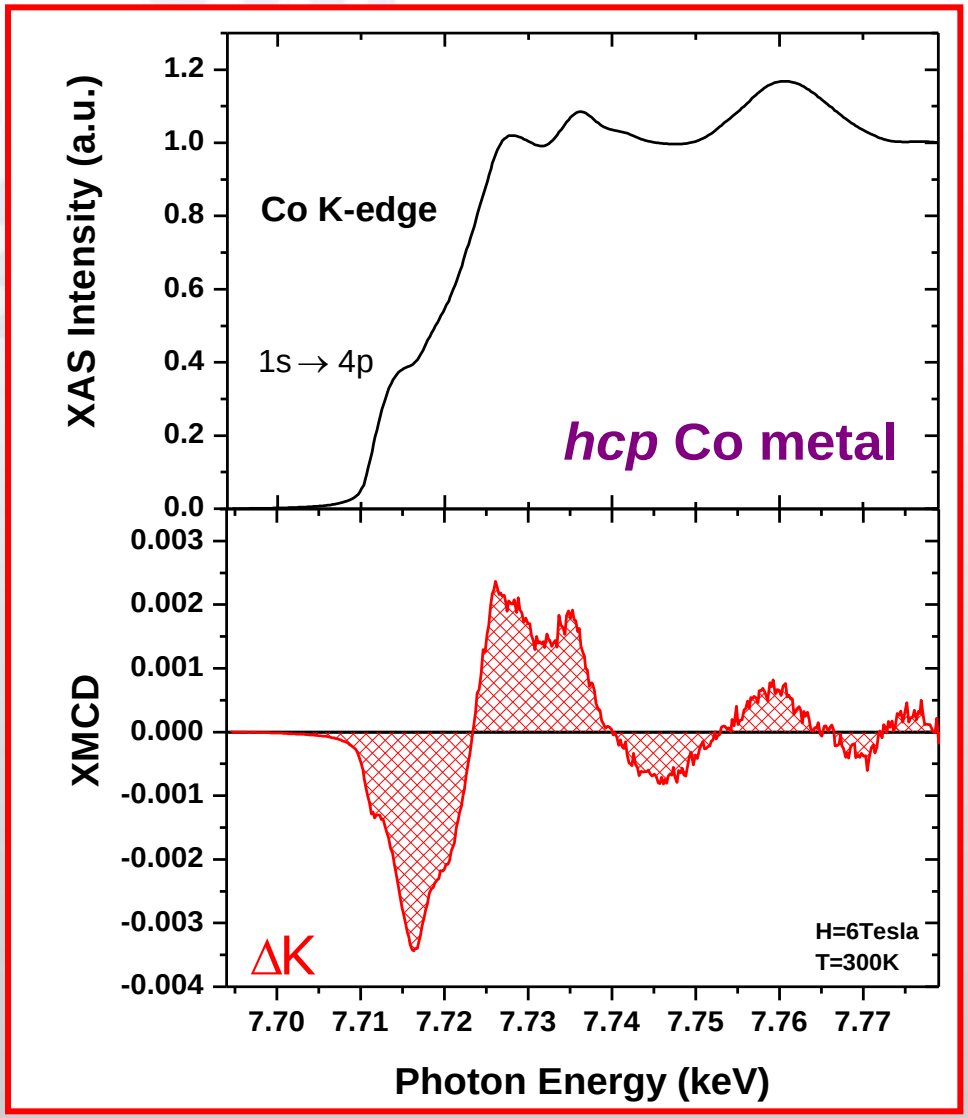


# XMCD Sum Rules

$$\underset{(E1\text{-}E1)}{\langle L_z^p \rangle} + \varepsilon \underset{(E2\text{-}E2)}{\langle L_z^d \rangle} = - \; C \, \Delta K$$

$$C = \frac{n_{4p} + n_{3d}}{A_{4p} + A_{3d}}$$

- A ~ integrated intensity of transition into unoccupied states
- n ~ number of holes



# XMCD Sum Rules

J. Phys.: Condens. Matter 6 (1994) L491–L495. Printed in the UK

LETTER TO THE EDITOR

## Magnetic absorption dichroism and sum rules in itinerant magnets

P Strange

Physics Department, Keele University, Staffs ST5 5BG, UK

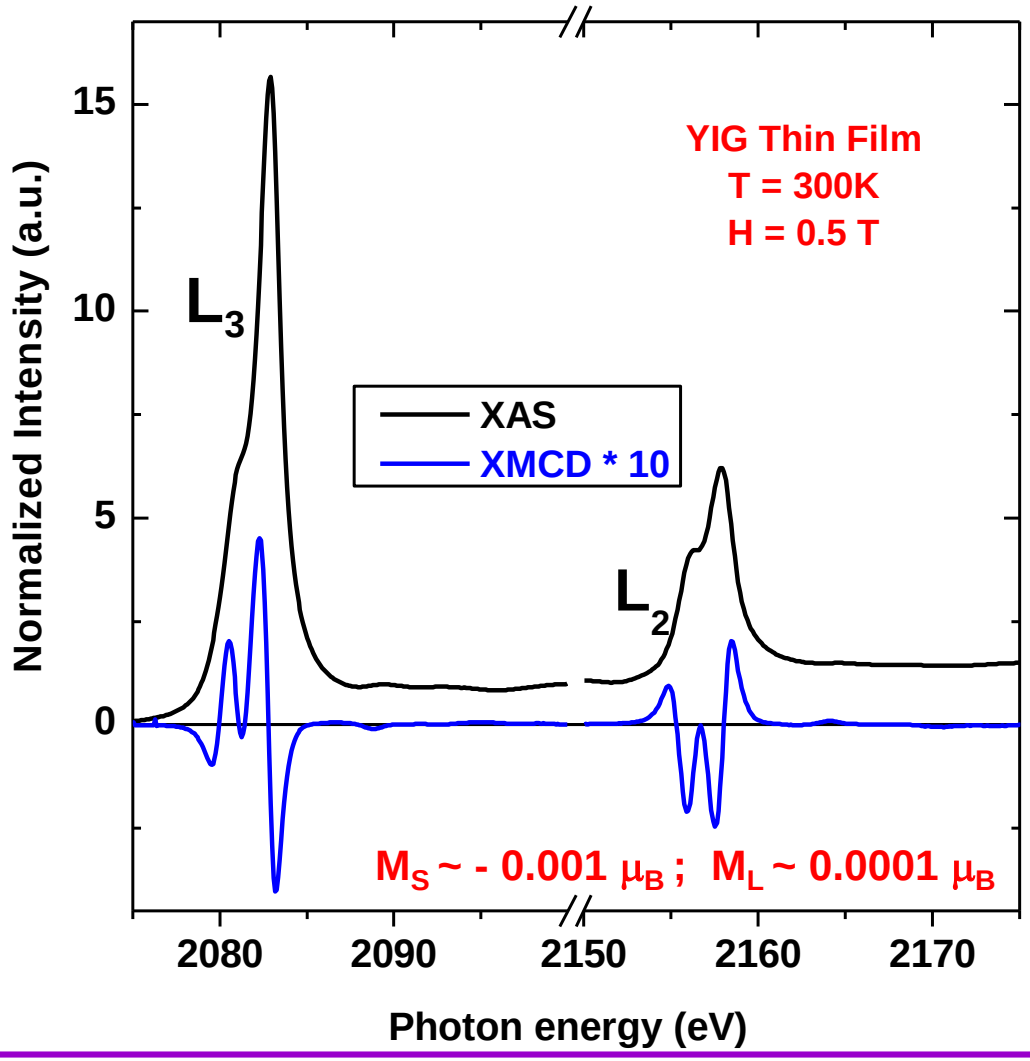
of the electrons in the dipole allowed states. This has proved to be a very useful and enlightening step forward in our understanding. However, there are still several questions to be answered. Firstly, the orbital and spin magnetic moments are energy integrated quantities, but the dichroism curves are often highly structured [5–7] and so the sum rules do not extract all the information from a dichroism experiment. Secondly, the models used

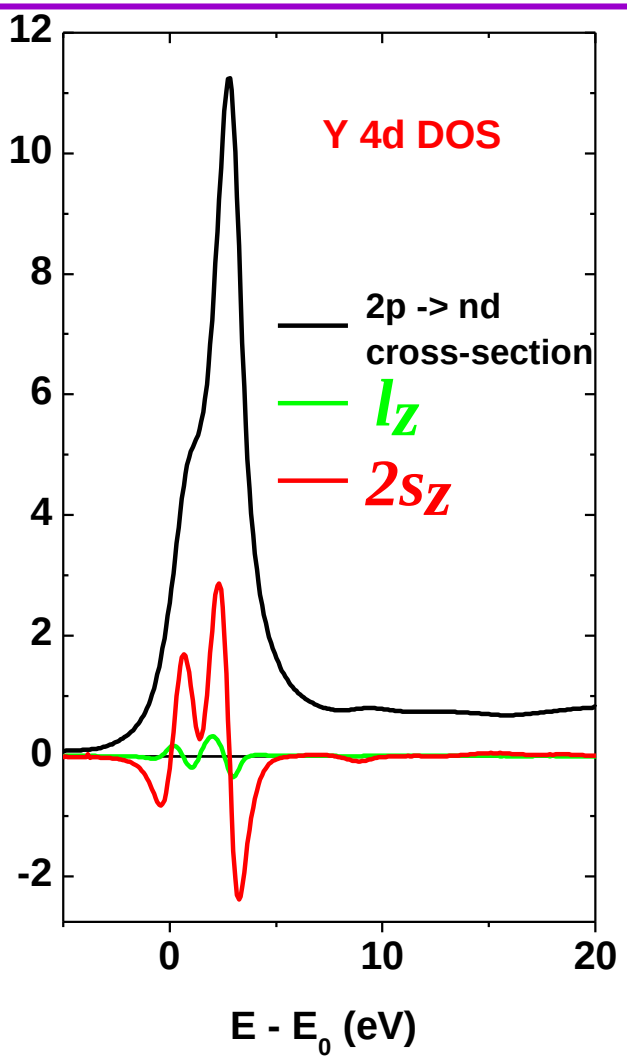
*C. Guo, Phys. Rev. B57 (1998), 10295* *H. Ebert et al. Phys. Rev. B60 (1999), 7156*

$$\Delta\mu_{L3}(E-E_0)+\Delta\mu_{L2}(E-E_0)\approx\frac{\partial}{\partial E}\langle L_z\rangle_d$$

$$\Delta\mu_{L3}(E-E_0)-2\Delta\mu_{L2}(E-E_0)\approx\left[\frac{\partial}{\partial E}\langle S_z\rangle_d+7\frac{\partial}{\partial E}\langle T_z\rangle_d\right]$$

# XMCD Sum Rules

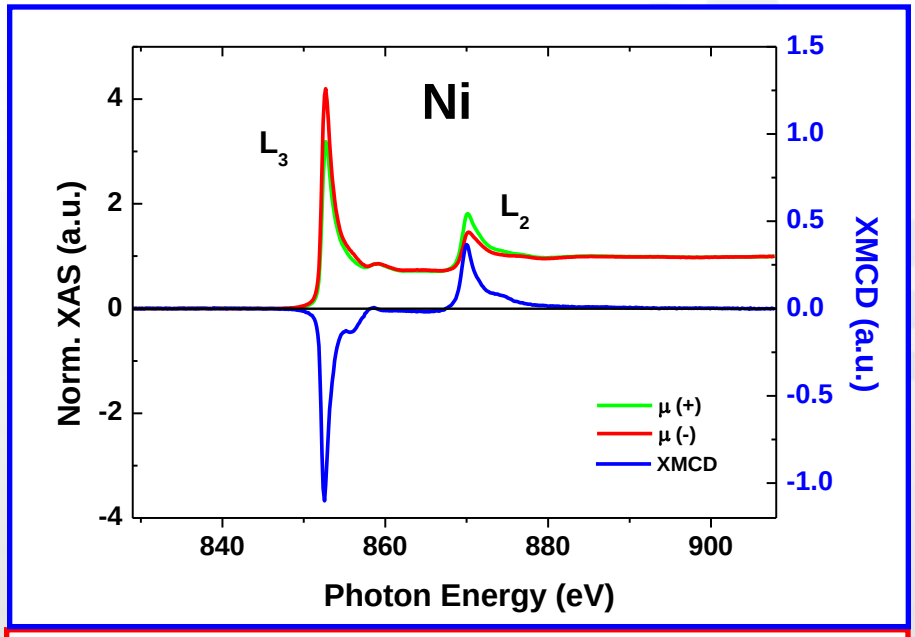


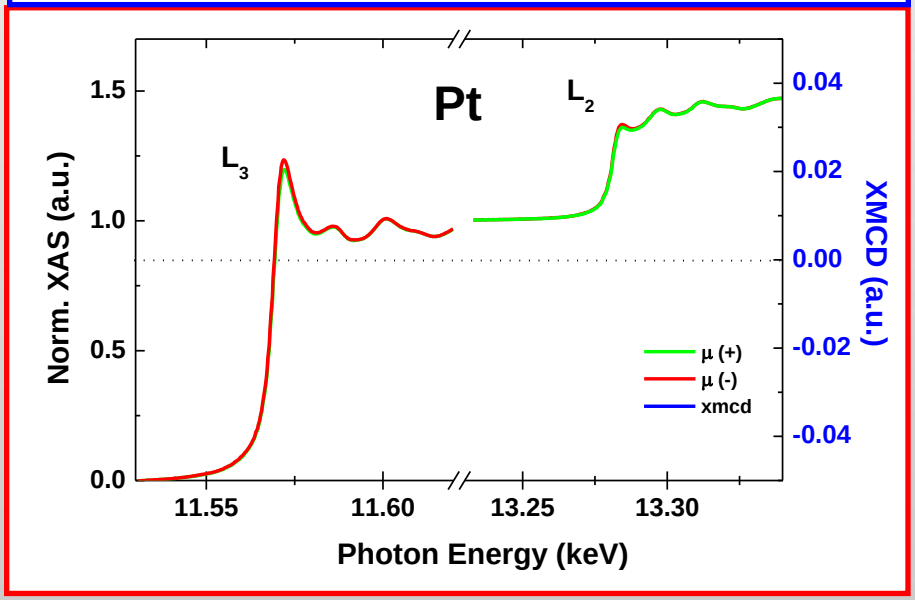




**Crystal field split 4d states of Y are strongly spin polarized with zero moment**

*A. Rogalev et al, J. Magn. Magn. Mat. 321, 3945 (2009)*

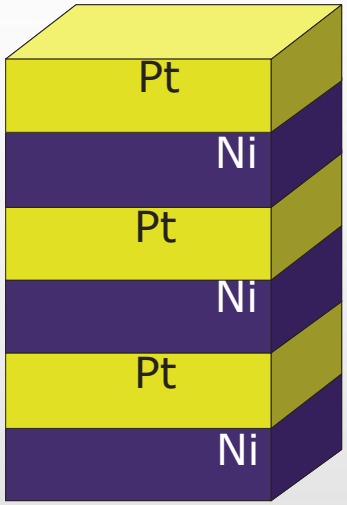
# Element Selectivity of XMCD




## $Ni_2 / Pt_2$ multilayer

T ~ 10K
H = ± 5 T

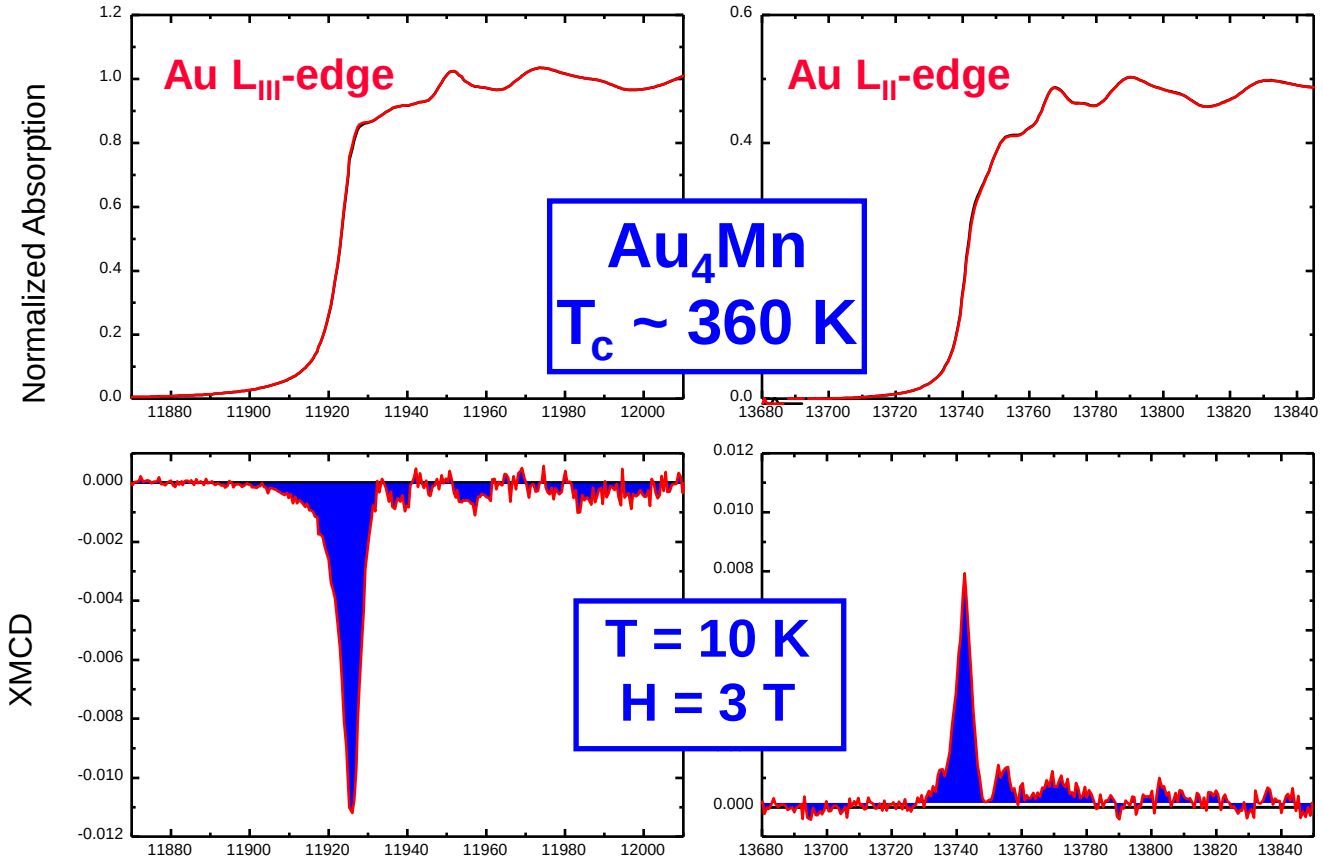



## RESULTS

- **Ni magnetic moments:**
  $\mu_S^{3d}=0.35\ \mu_B$/atom
  $\mu_L^{3d}=0.038\ \mu_B$/atom
- **Pt induced magnetic moments:**
  $\mu_S^{5d}=0.14\ \mu_B$/atom
  $\mu_L^{5d}=0.03\mu_B$/atom

*F. Wilhelm et al., Phys. Rev. Lett., 85, 413 (2000)*

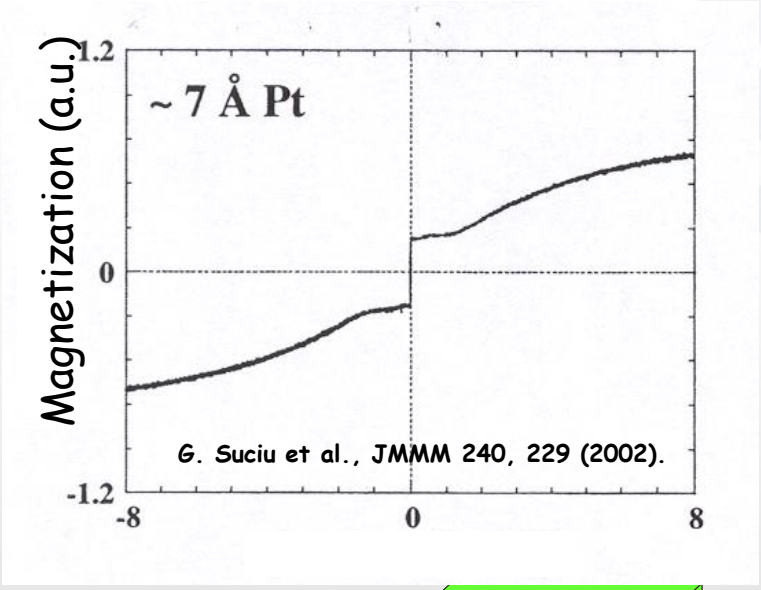
# Sensitivity of XMCD



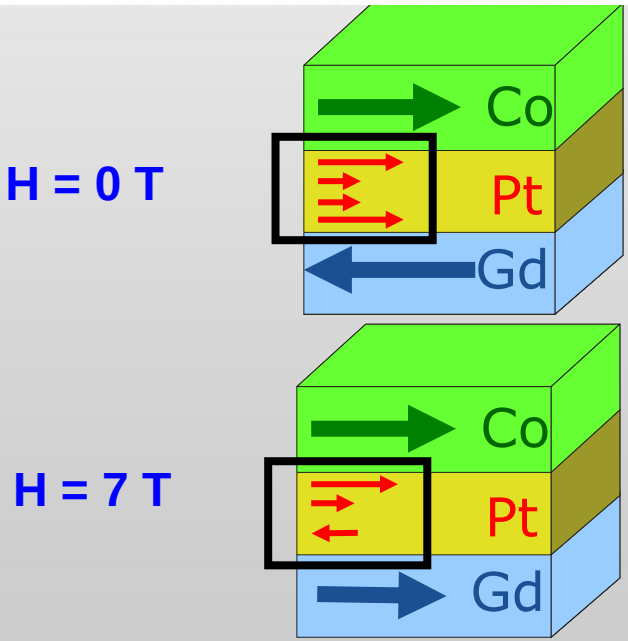

$\langle S_z \rangle = 0.0353(5)\mu_B$ $\langle L_z \rangle = 0.0054(5)\mu_B$ (per Au atom)

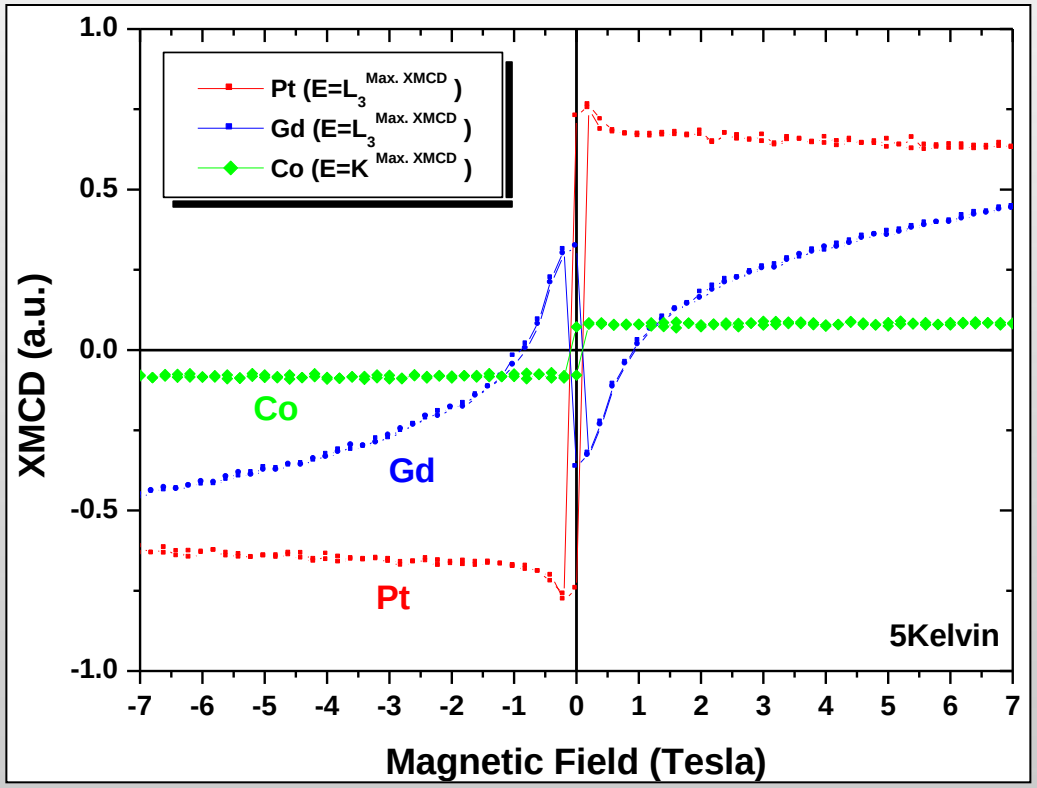
To compare with $4.15\mu_B$ per Mn atom

# Element Selective Magnetometry




## $Co_{17}/Pt_7/Gd_{16}/Pt_7$ multilayer



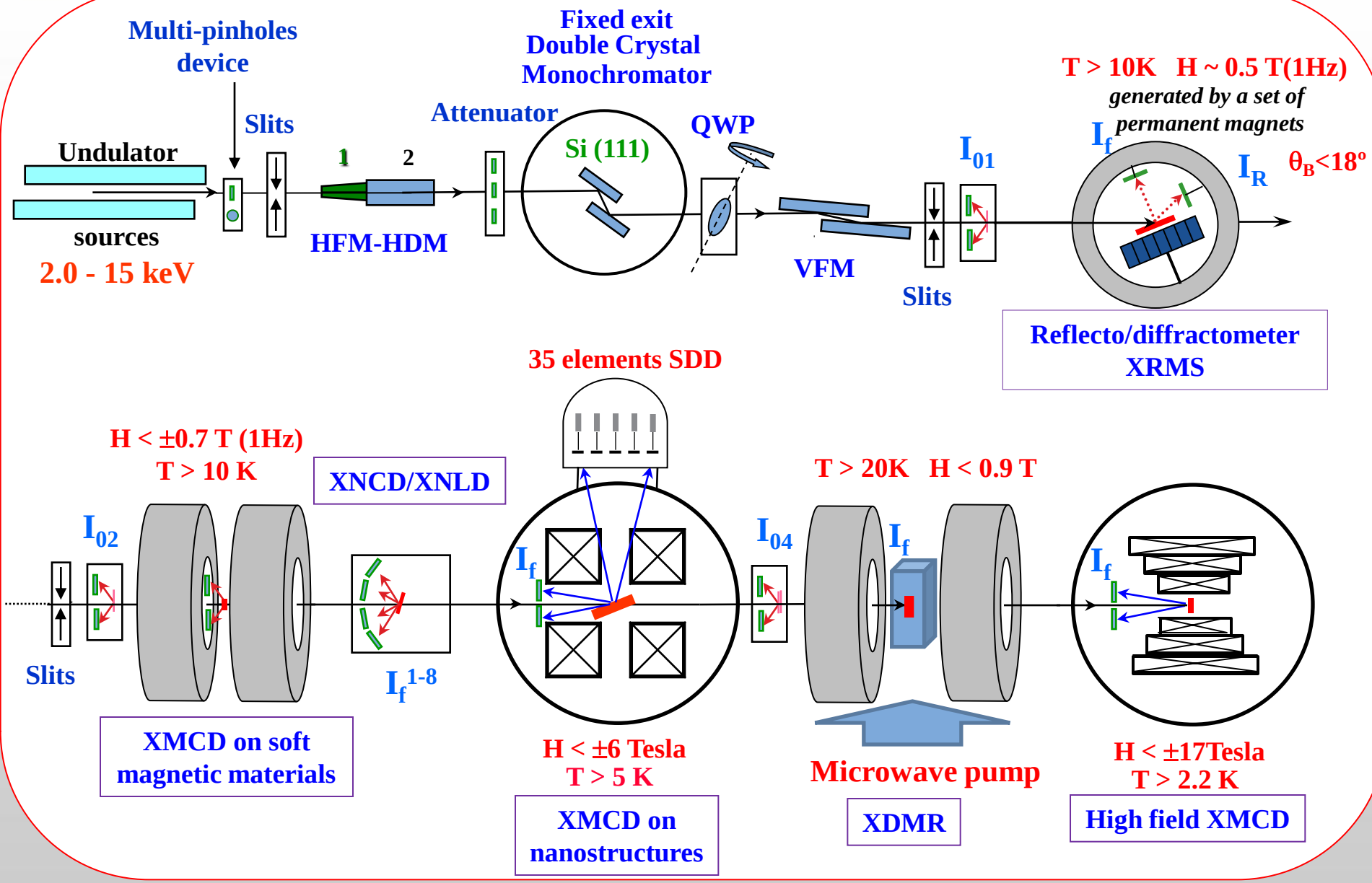
# XMCD Experiment

**Quantity to measure: $\Delta\mu = \mu^+ - \mu^-$**

$\mu^+$, $\mu^-$ => **Absorption cross-sections for CP X-rays with**
**( + ) helicity *parallel* to the sample magnetization**
**( - ) helicity *antiparallel* to the sample magnetization**

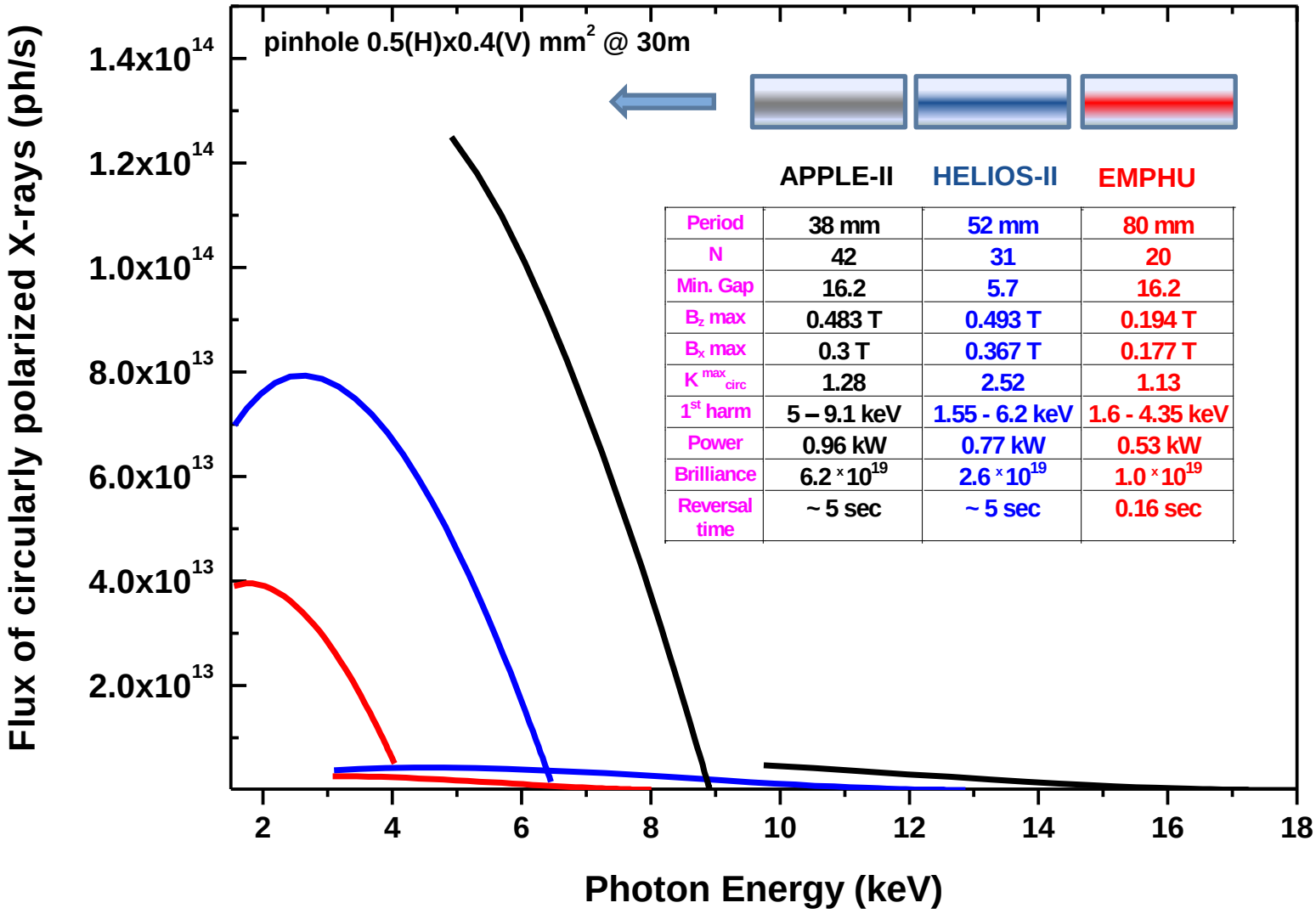
- **Highly performing X-ray detectors**
- **Magnetic field to magnetize a sample**
- **Source of monochromatic circularly polarized X-rays**

**The best possible at the 3rd generation synchrotron radiation facilities**


Multi-pinholes device
Slits
Undulator
sources
2.0 - 15 keV
1
2
HFM-HDM
Fixed exit Double Crystal Monochromator
Attenuator
Si (111)
QWP
VFM
Slits
$I_{01}$
T > 10K H ~ 0.5 T(1Hz)
generated by a set of permanent magnets
$I_f$
$I_R$
$\theta_B$<18°
Reflecto/diffractometer XRMS
35 elements SDD
H < ±0.7 T (1Hz)
T > 10 K
XNCD/XNLD
$I_{02}$
Slits
$I_f^{1-8}$
XMCD on soft magnetic materials
$I_f$
H < ±6 Tesla
T > 5 K
XMCD on nanostructures
T > 20K H < 0.9 T
$I_{04}$
$I_f$
Microwave pump
XDMR
$I_f$
H < ±17Tesla
T > 2.2 K
High field XMCD

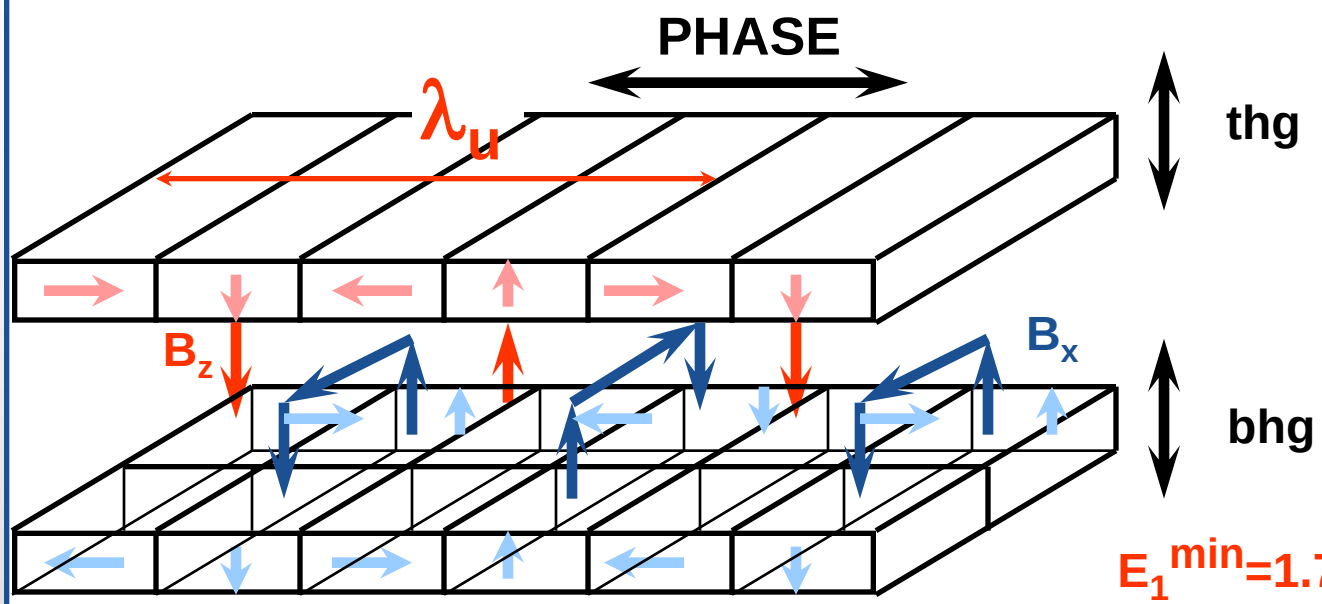
# ESRF Beamline ID12






| | APPLE-II | HELIOS-II | EMPHU |
|---|---|---|---|
| Period | 38 mm | 52 mm | 80 mm |
| N | 42 | 31 | 20 |
| Min. Gap | 16.2 | 5.7 | 16.2 |
| $B_z$ max | 0.483 T | 0.493 T | 0.194 T |
| $B_x$ max | 0.3 T | 0.367 T | 0.177 T |
| $K^{max}_{circ}$ | 1.28 | 2.52 | 1.13 |
| 1st harm | 5 – 9.1 keV | 1.55 - 6.2 keV | 1.6 - 4.35 keV |
| Power | 0.96 kW | 0.77 kW | 0.53 kW |
| Brilliance | $6.2 \times 10^{19}$ | $2.6 \times 10^{19}$ | $1.0 \times 10^{19}$ |
| Reversal time | ~ 5 sec | ~ 5 sec | 0.16 sec |

# Helios-II Undulator source




**31 periods**

**$\lambda_u$ = 52 mm**

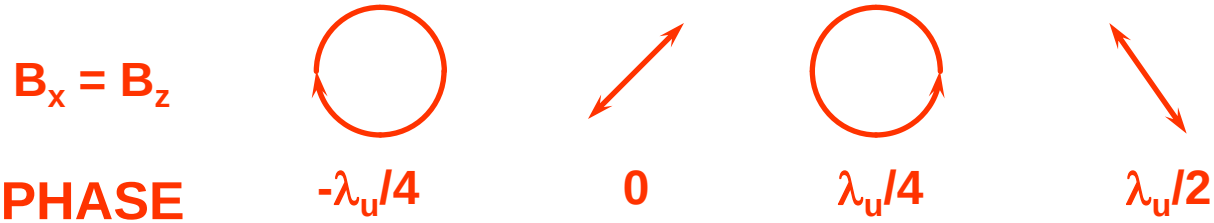
**thg = 6.0 mm**
**($B_z$=0.493 T)**
**bhg = 6.0 mm**
**($B_x$=0.349 T)**

**$E_1^{min}$=1.7 keV, $E_1^{max}$=6.2 keV**

*P. Elleaume, J. of Syn. Rad., **1**, 19-26 (1994)*

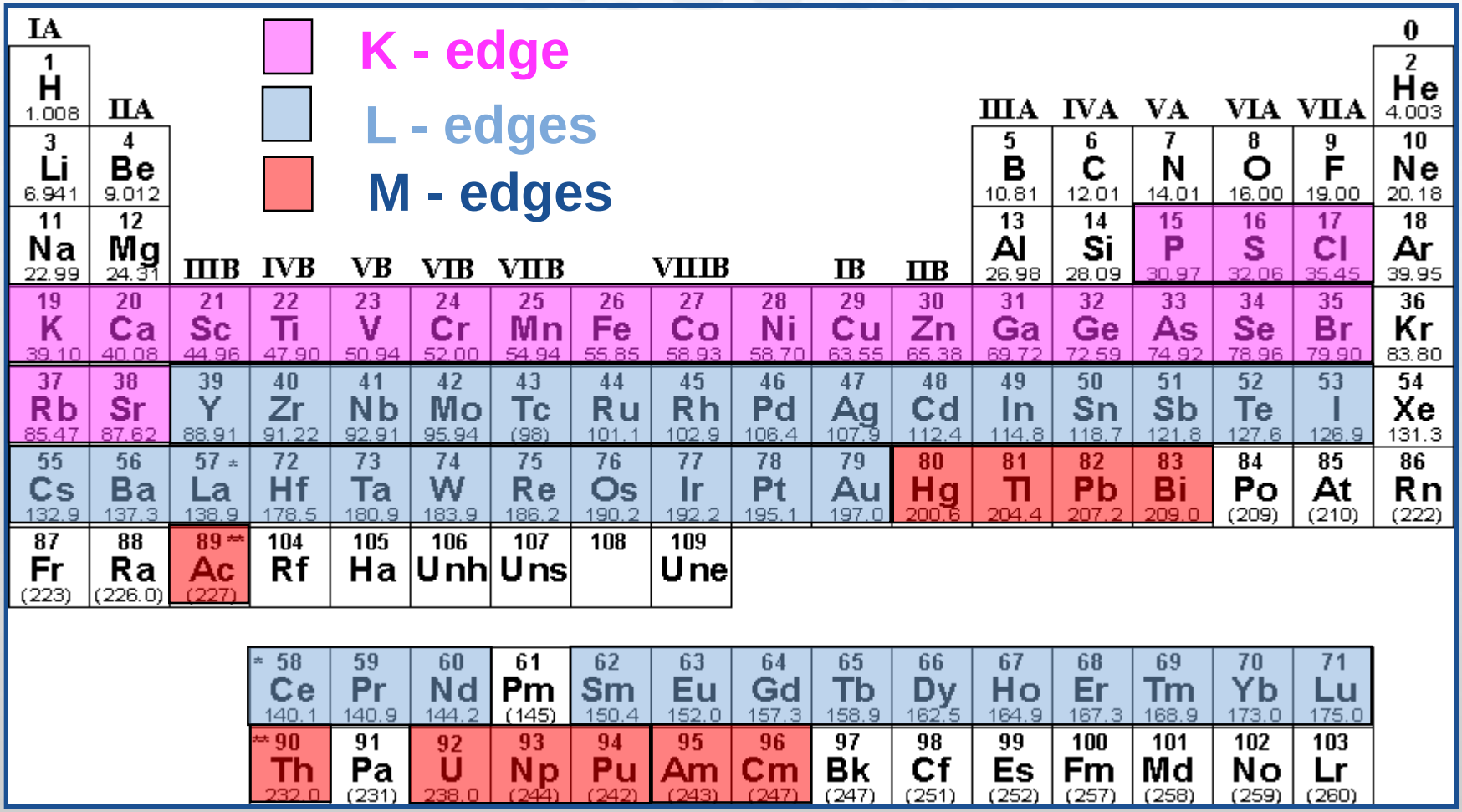
**Flux: $10^{14}$ ph/s/0.1%bw**

***Full control of polarization: flipping time ~ 5 seconds***

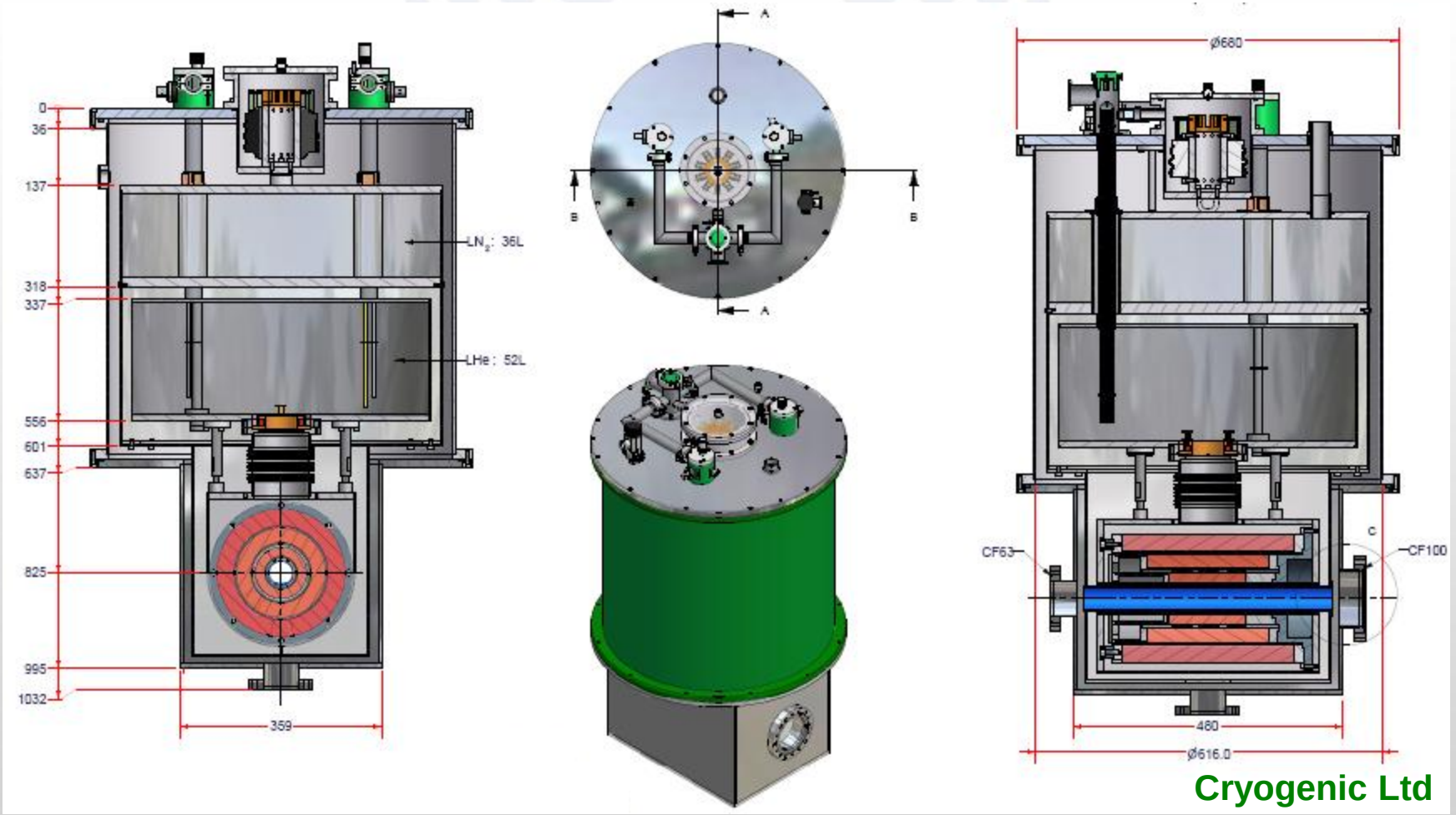


# XMCD experiment at ID12

**2.05 keV - 15 keV**



# High Field Magnet




**Magnet is a solenoid consisting of 3 coils ( 2 of Nb3Sn and outer of NbTi ) .**
**The maximum field is 17T with homogeneity over 10mm DSV of 0.1 %.**
**Maximum ramp rate is 2Tesla/minute. Cryogens autonomy is about 48 h.**

# High field XMCD set-up

# Magnetism in DMS

## Semiconductor materials that are claimed to exhibit hysteresis and spontaneous magnetisation at 300 K

wz-c-(Ga,Mn)N, (In,Mn)N, (Al,Mn)N, (Ga,Cr)N, (Al,Cr)N
(Ga,Fe)N, (Ga,Cu)N, (Ga,Gd)N, (Ga,Eu)N

(Ga,Mn)As, (In,Mn)As, (Ga,Mn)Sb, (Ga,Mn)P:C

(Zn,Mn)O, (Zn,Ni)O, (Zn,Co)O, (Zn,V)O, (Zn,Fe,Cu)O, (Zn,Cu)O

$(Ti,Co)O_2$, $(Ti,V)O_2$, $(Ti,Cr)O_2$, $(Sn,Co)O_2$, $(Sn,Fe)O_2$, $(Hf,Co)O_2$
$(Cd,Ge,Mn)P_2$, $(Zn,Ge,Mn)P_2$, $(Cd,Ge,Mn)As_2$, $(Zn,Sn,Mn)As_2$

(Ge,Mn), (Ge,Cr), (Ge,Mn,Fe)

$(La,Ca)B_6$, C, $C_{60}$, $HfO_2$, $TiO_2$-K

XMCD has demonstrated that non of them is intrinsically ferromagnetic at RT

*A. Ney et al, New Journal of Physics 12, 013020 (2010)*

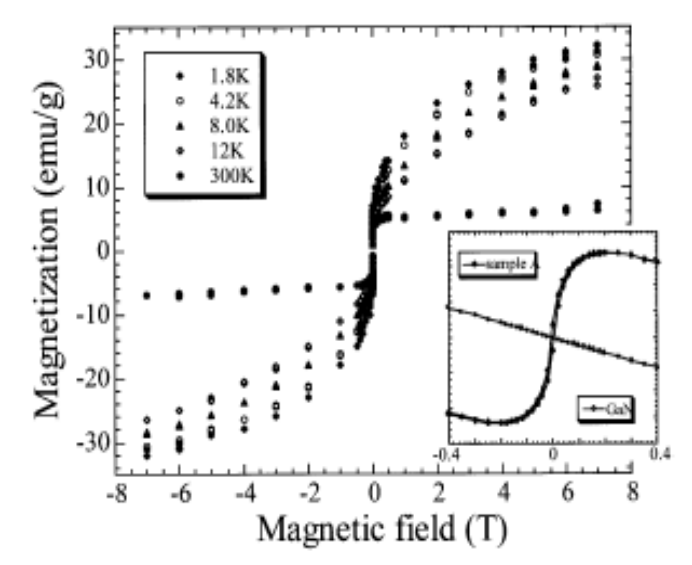
# Magnetism in DMS

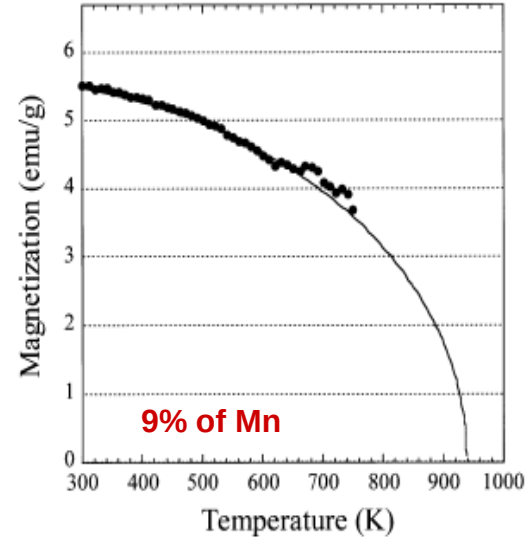
## GaN : Mn

**The most controversial diluted magnetic semiconductor**

**SQUID magnetometry measurements:**

- **Ferromagnetism** $2K < T_c < 940K$



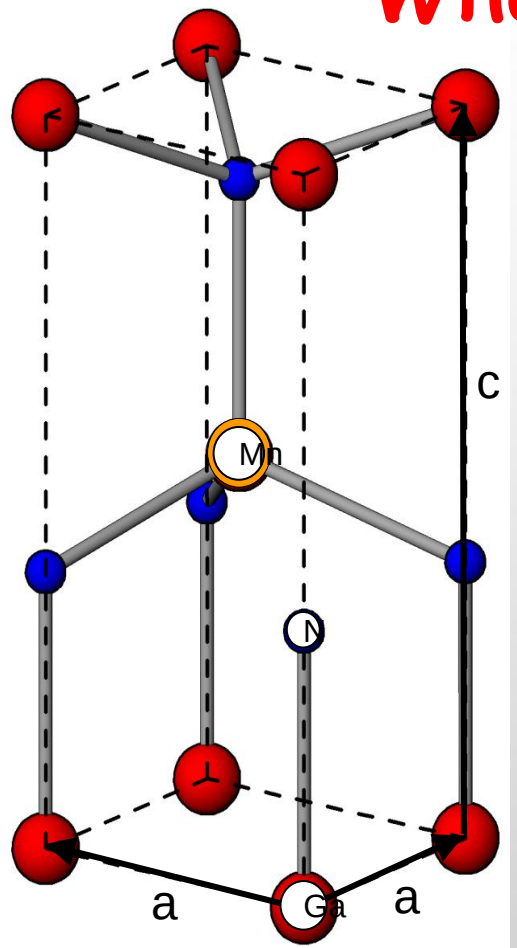




*S. Sonoda et al., J. Crys. Growth 237-239, 1358 (2002)*

- **Paramagnetism**

*A. Wolos et al., PRB 69, 115210 (2004)*

# What do we need to know more ?




Space group 186
Structure : $P6_3mc$
Non-centrosymmetric

**1. How are 3d atoms incorporated in the lattice?**

- **Single phase**
- **Presence of (magnetic) secondary phases**
- **Clusters formation**

**Concentration limit ?**

**2. What is the site of 3d atoms ?**

- **Substitution**
- **Interstitial**
- **Substitution and interstitial**

**3. What is the valence state of 3d atoms?**

**4. Is it really a diluted magnetic semiconductor or ferromagnetic clusters in semiconducting matrix?**

# Hard X-ray Dichroisms

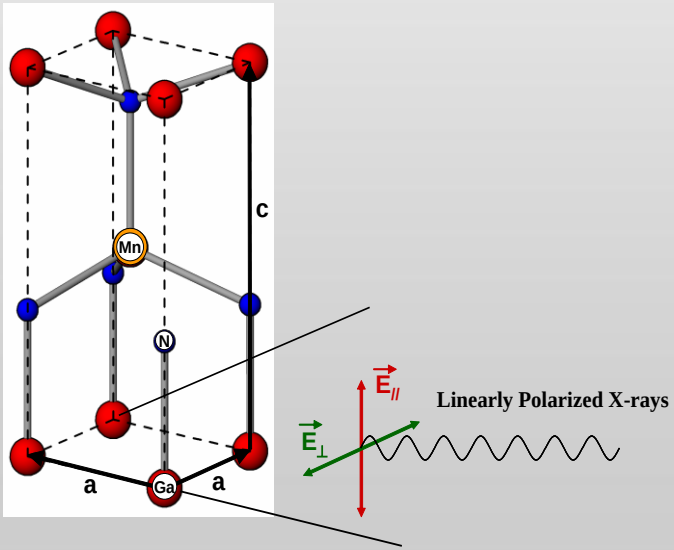
## Structural characterization

**Linearly polarized X-rays**

$$XLD = \mu^{\perp}(E) - \mu^{//}(E)$$

**XLD measures the anisotropy of the unoccupied density of states at absorbing atom**

- **high sensitivity to local environment (symmetry, bonding, electronic structure)**



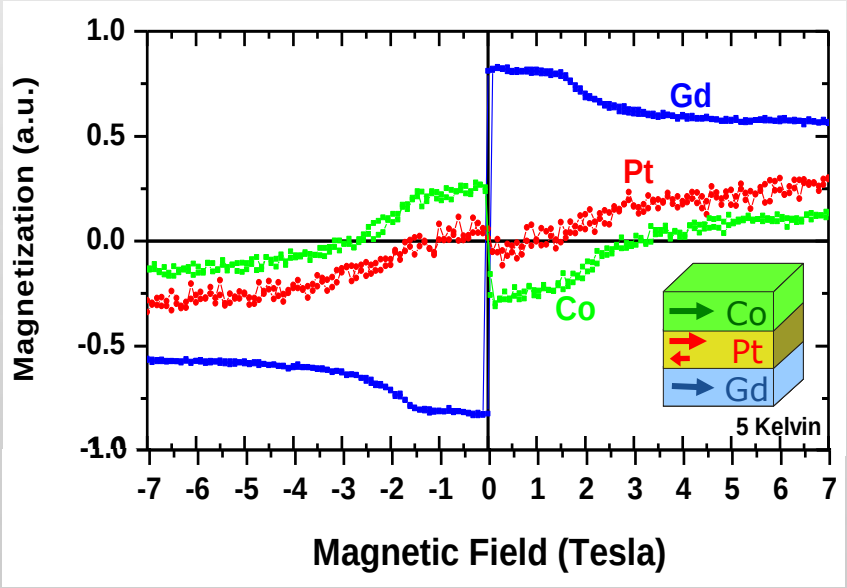

## Magnetic characterization

**Circularly polarized X-rays**

$$XMCD = \mu^{R}(E) - \mu^{L}(E)$$

**XMCD measures amplitude and directions of the spin and orbital moments carried by absorbing atom**

- **element selective magnetization measurements**

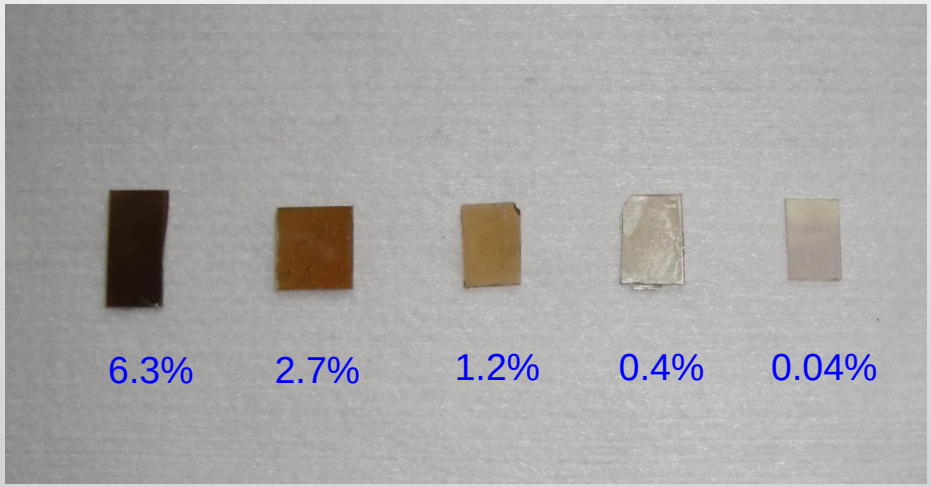


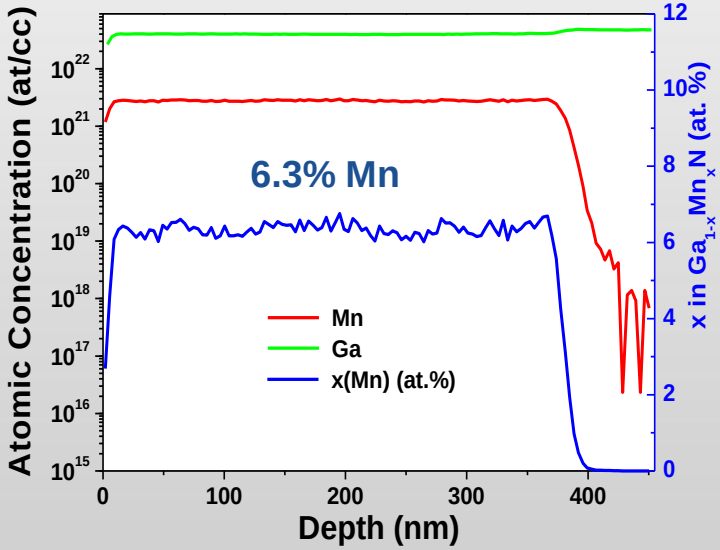
# Our GaMnN samples

Samples were grown by plasma assisted molecular beam epitaxy (PAMBE)

E. Sarigiannidou*, E. Monroy and H. Mariette
*Equipe mixte CEA-CNRS-UJF "Nanophysic and Semiconductors", DFRMC/SP2M CEA, Grenoble, France*
** LMGP/LTM, INP -Minatec, Grenoble, France*

## $Ga_{1-x}Mn_xN$ epitaxial film (400nm)



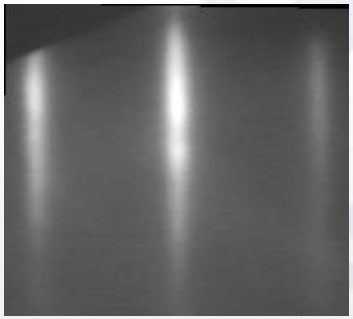




Homogeneous Incorporation of Mn from 0.04 up to max. 6.3 at.%

# Standard Macroscopic Characterization

## ➢ in-situ RHEED

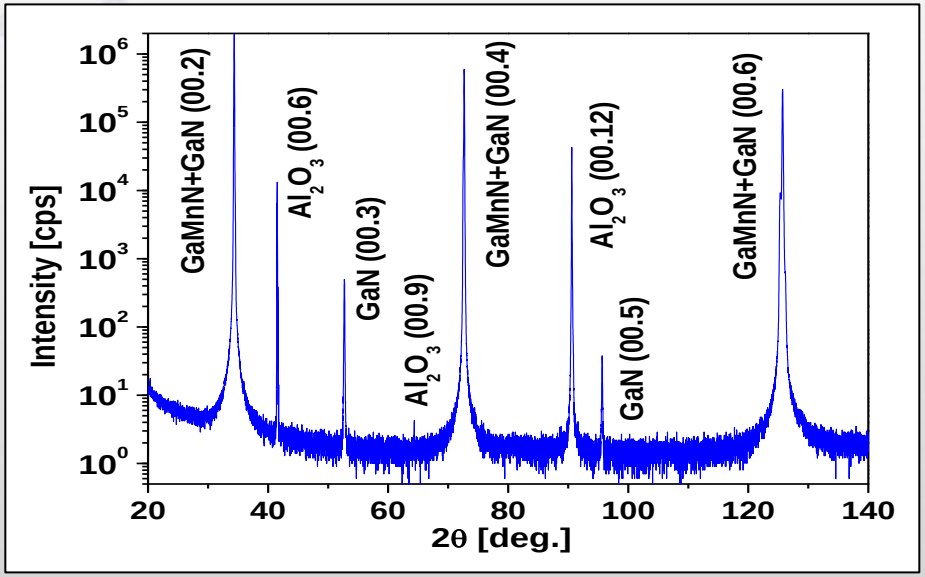
monitoring the growth



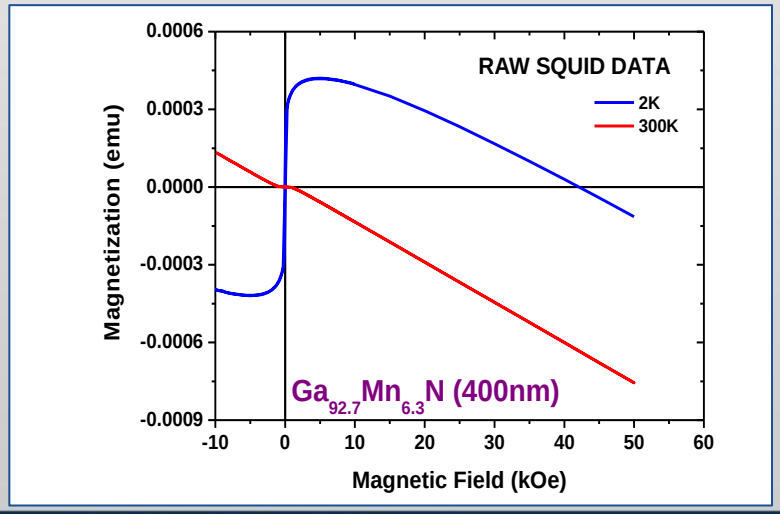
- ❖ extremely Surface sensitive
- ❖ smooth surfaces
- ❖ absence of secondary phases
- ❖ no information about the incorporation

## ➢ High Angle X-ray Diffraction

- ❖ detection limit of ~1% of sample volume
- ❖ absence of secondary phases
- ❖ difference in the lattice parameter
- ❖ not sensitive to amorphous phases

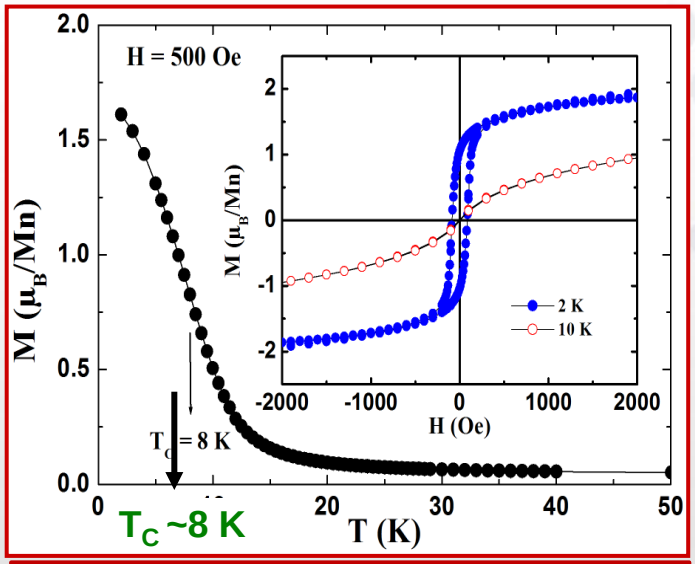


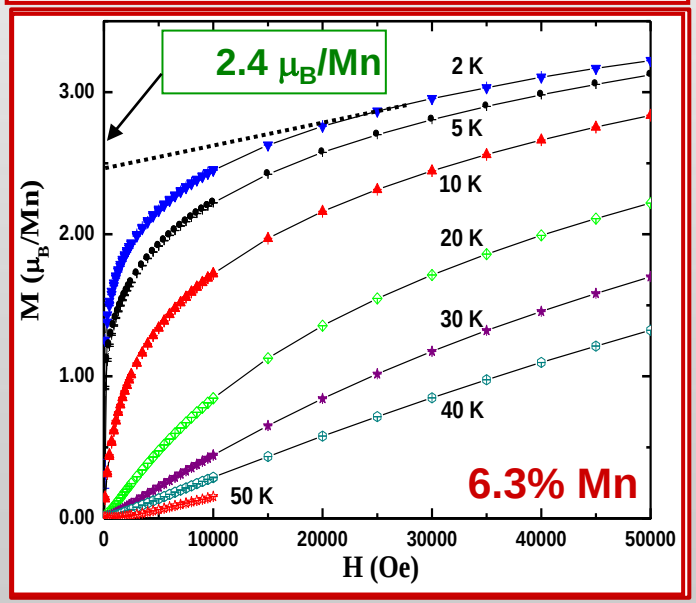

## ➢ SQUID




- ❖ measure the whole sample including substrate, cap layer, inclusions, impurities
- ❖ non trivial extraction of a true magnetic response

# Macroscopic Magnetic Characterization



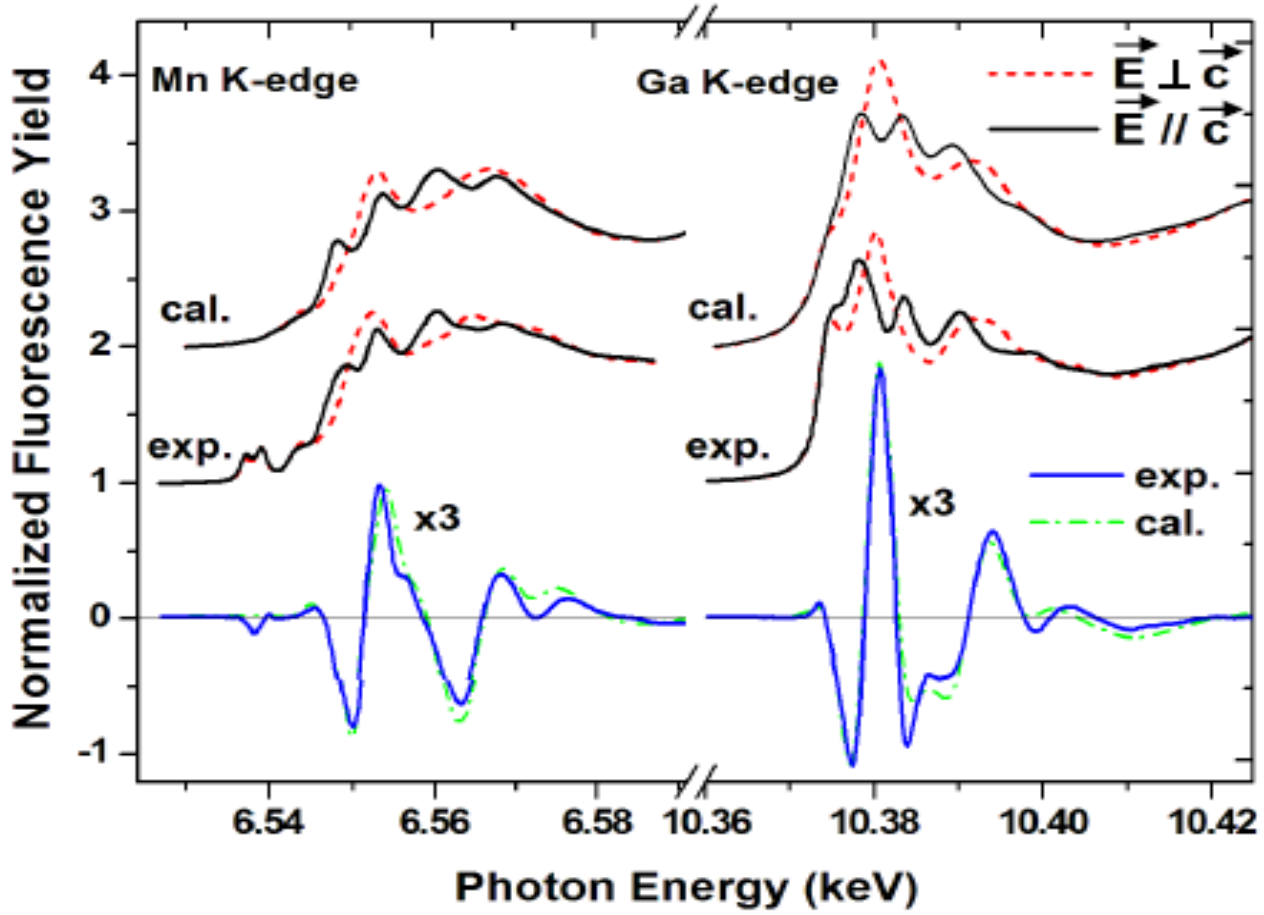

Magnetic Field ⊥ *c* crystal axis

Corrections from the diamagnetic/paramagnetic contribution: measurements on a blank substrate

Easy axis ⇒ in-plane

- Opening of the hysteresis loop at 2 K
  coercive Field ≈ 100 Gauss
- Thermal Variation ➔ $T_C$ = 8 K
- Remanent magnetization 44% of the spontaneous magnetization ➔ 2.4$\mu_B$ per Mn

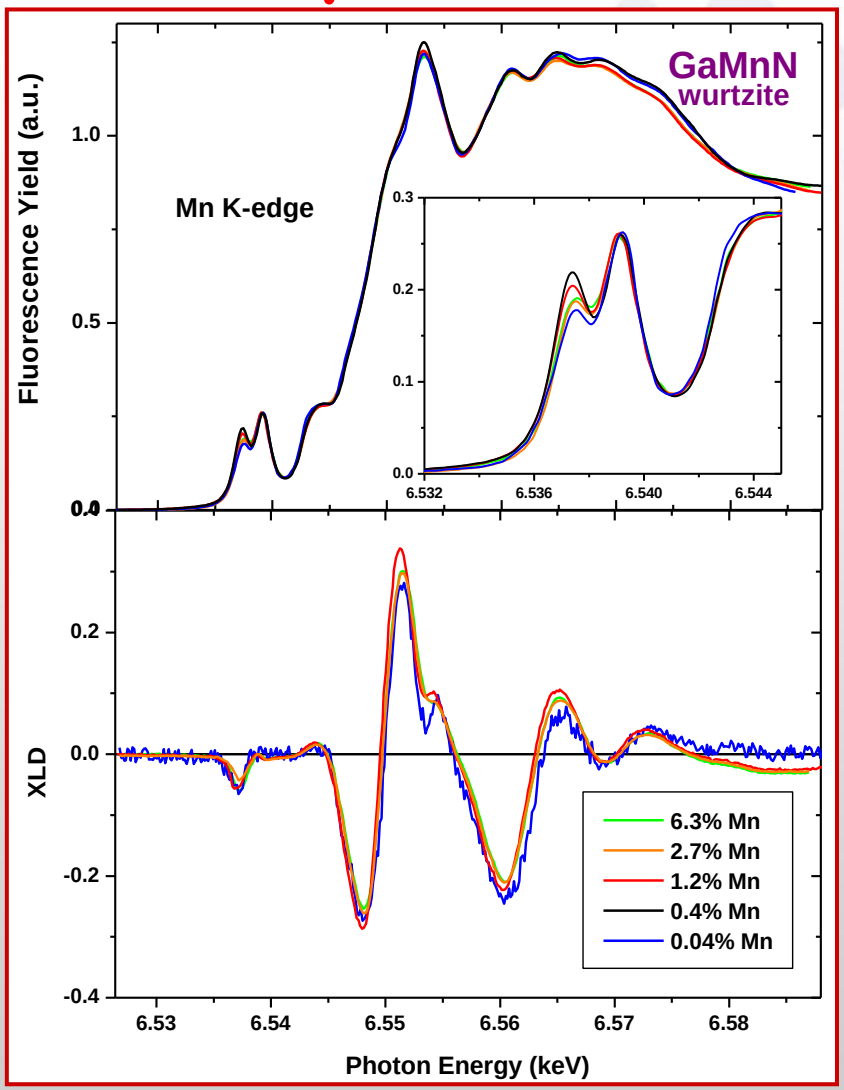
# X-ray Linear Dichroism (XLD) Results




- Mn atoms are Ga substituted
- No secondary parasitic or clusters phases

*E. Sarigiannidou et al. Phys. Rev. B74, 041306(R) (2006)*

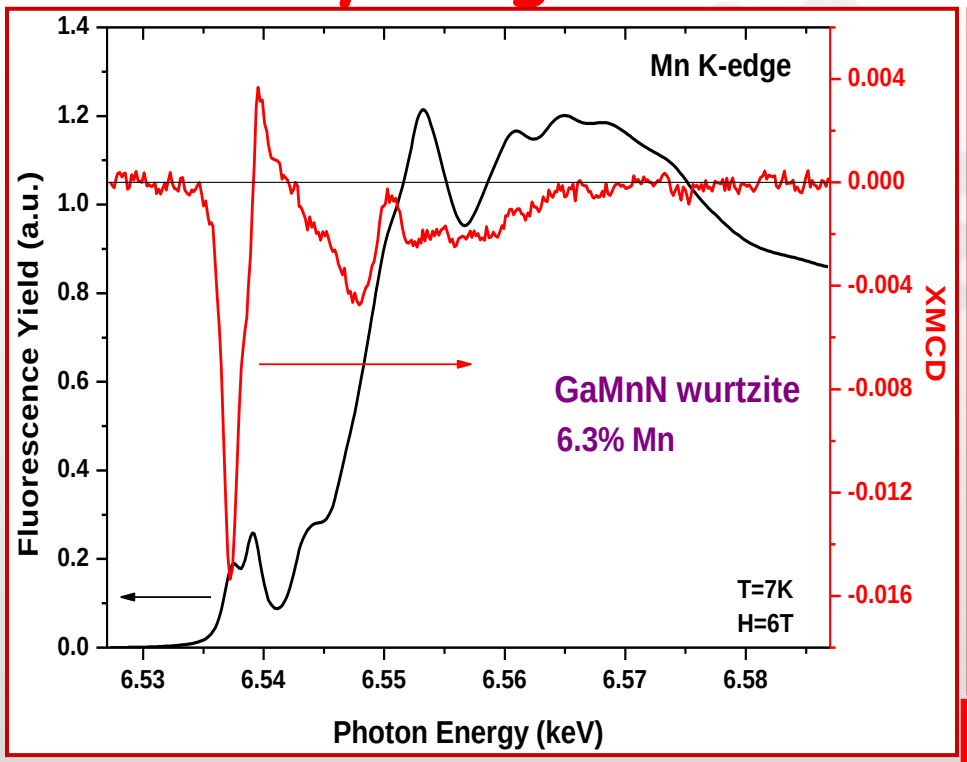
# X-ray Linear Dichroism (XLD) Results

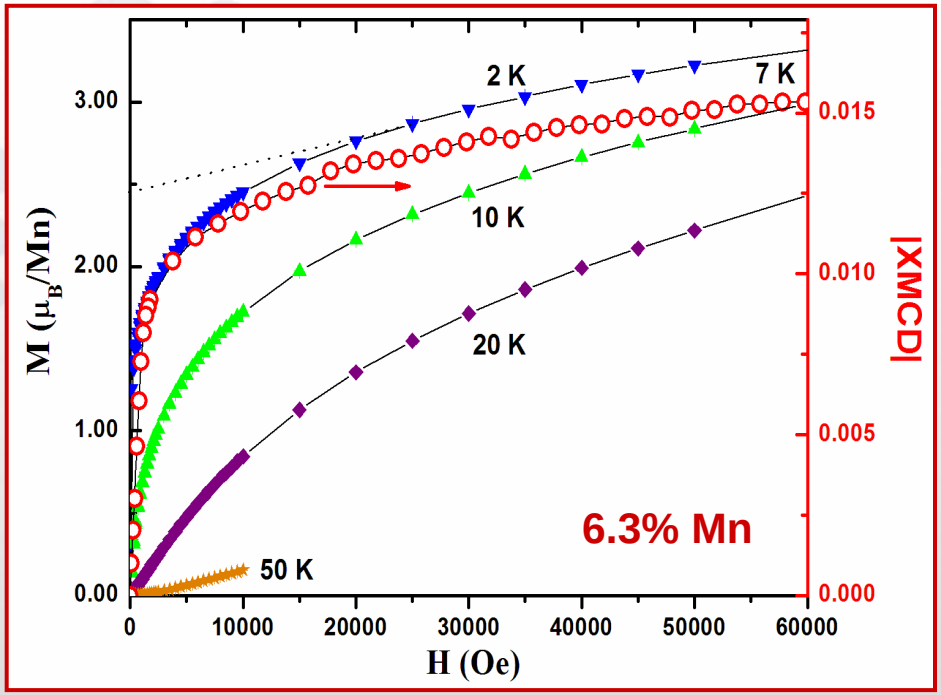



- All the Mn K-edge XANES spectra have similar spectral shape
  - small differences are observed at the first pre-edge peak (narrowing of 3d band)
- XLD spectra are identical for all samples from 0.04% up to 6.3% of Mn
  - No secondary parasitic or clusters phases 0.04% Mn is the reference spectrum (presence of secondary phases is unlikely)

**Mn is perfectly incorporated up to at. concentration of 6.3%**

# X-ray Magnetic Circular Dichroism Results




**XMCD Mn K-edge:**

**probes the orbital magnetization**

Raw XMCD Magnetization Curve
(no corrections needed)

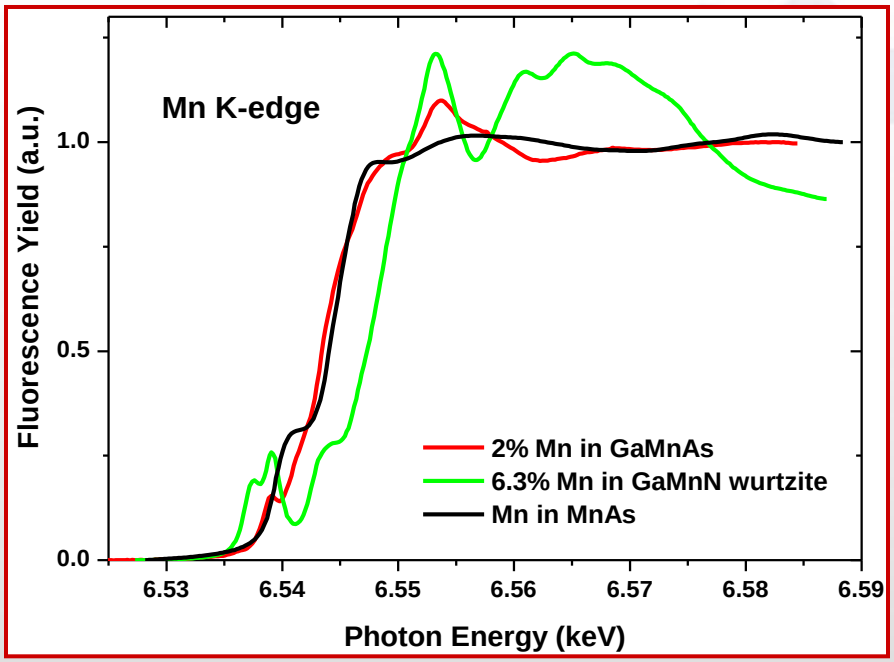
➔ Intrinsic Magnetism of (Ga,Mn)N

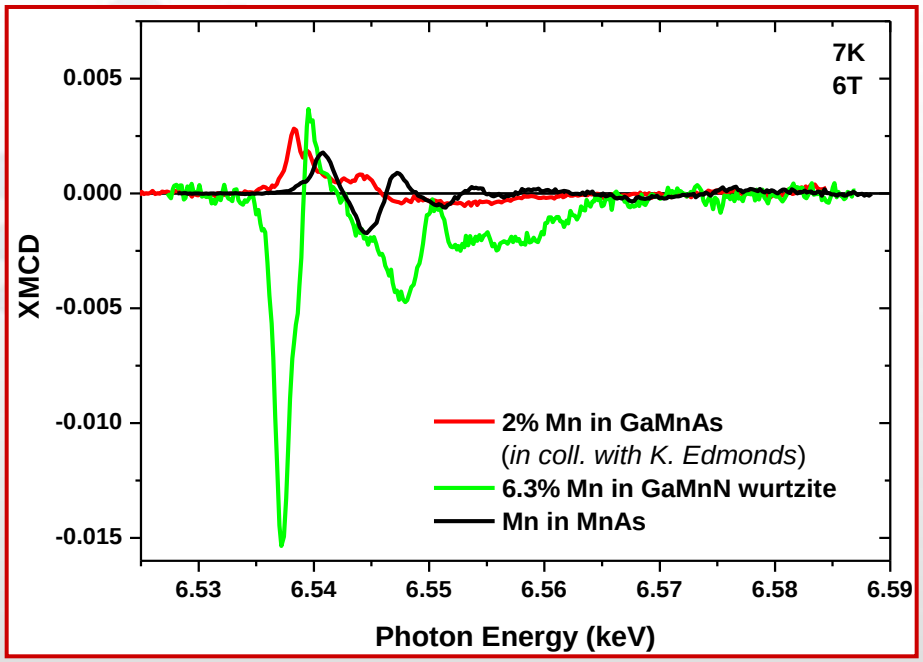
**Large Signal (~ 1.4%) ➡ Large Orbital magnetic moment ➡ mainly $Mn^{3+}$**

*E. Sarigiannidou et al. Phys. Rev. B74, 041306(R) (2006)*

# Magnetism in DMS

## GaMnN versus GaMnAs and MnAs

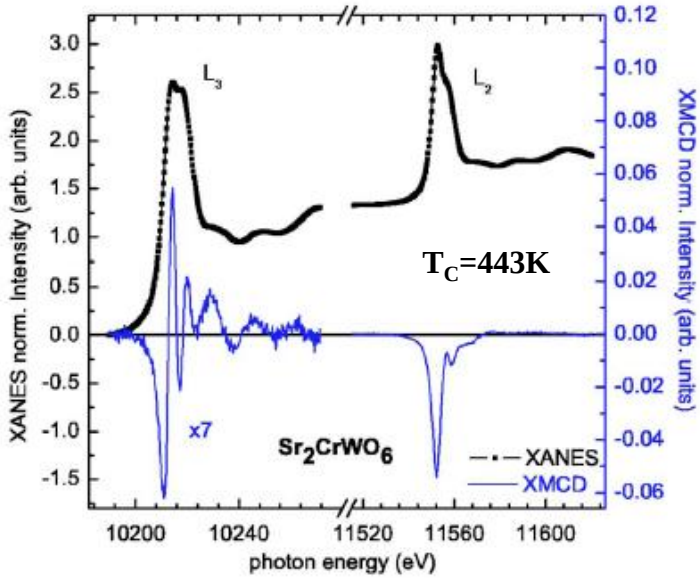



- XANES of Mn in GaMnN at higher energy : different electronic structure
- Presence of two pre-edge peaks ⇒ $Mn^{3+}$
- Difference in XMCD :
  1. Mn ($d^5$) in MnAs is ±0.2% and integral is nearly zero
  2. Mn ($d^{5+\delta}$) in GaMnAs is +0.25% and positif
  3. Mn in GaMnN is -1.4% and negatif

mainly $Mn^{3+}$ ($d^4$)

# Induced magnetism




## W induced magnetic moments:

$\mu_S^{5d} = -0.33\ \mu_B$ $\mu_L^{5d} = 0.12\mu_B$

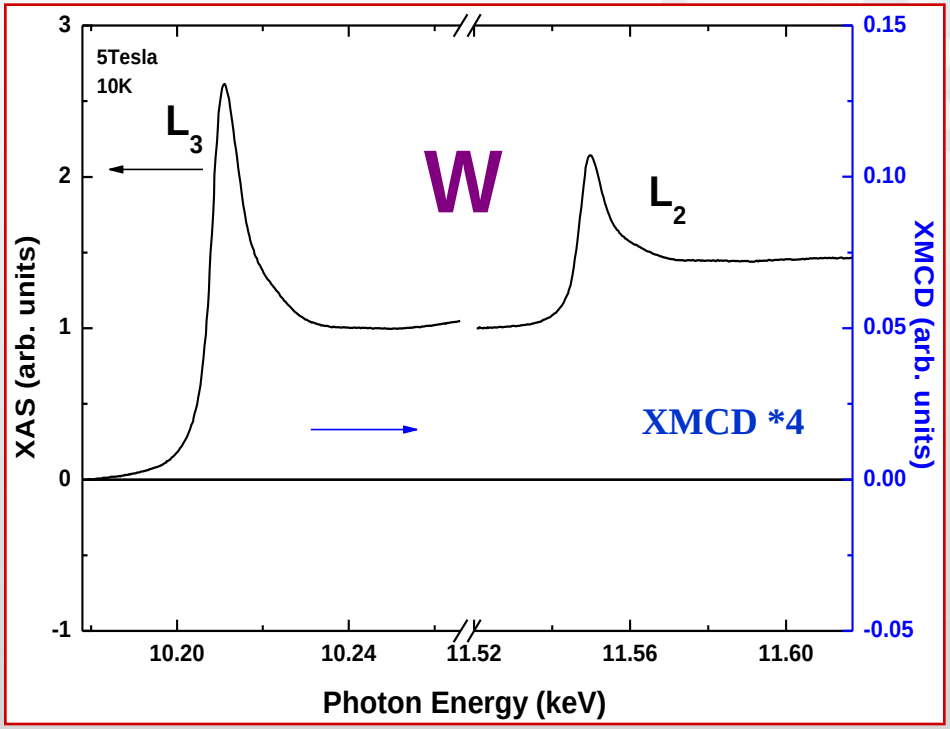
**Ferrimagnetic ordering in both compounds**

$\mu_S^{5d} = -0.22\ \mu_B$ $\mu_L^{5d} = 0.1\mu_B$

*P. Majewski et al., Phys. Rev. B, 72, 132402 (2005)*

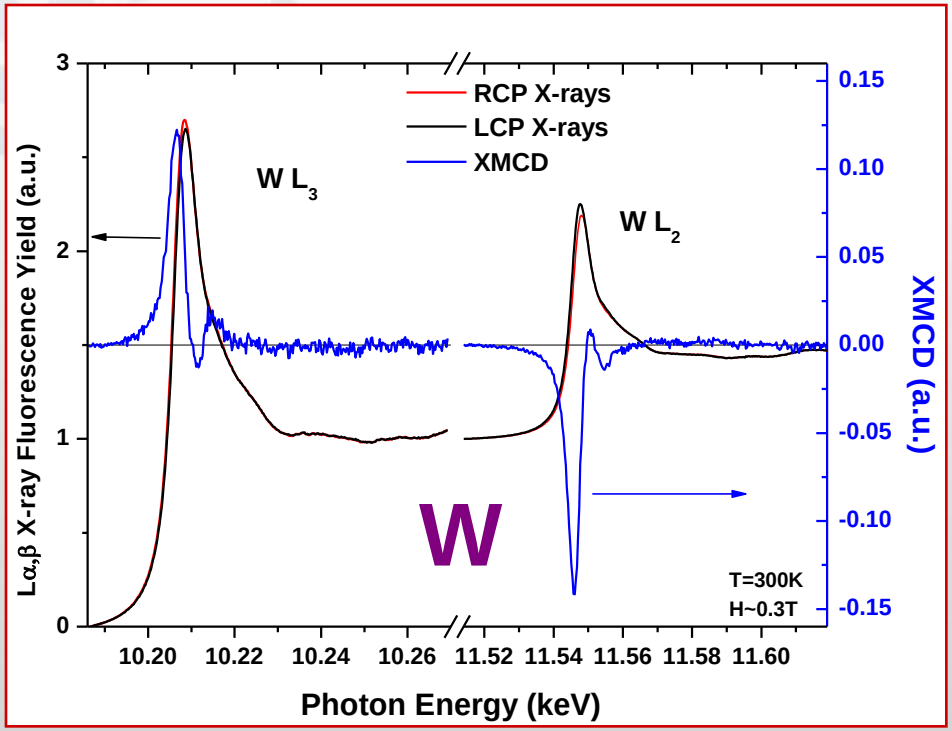
# Induced magnetism

## $[Fe_5/W_3]_{30}$ multilayer




*Wilhelm F. et al., Phys. Rev. Lett. 87 (2002), 207202*

## $Fe_{0.97}W_{0.03}$ thin film




*In collaboration with E.Majkova (Slovakia)*

**W induced magnetic moments are different in the multilayer and in the alloy**

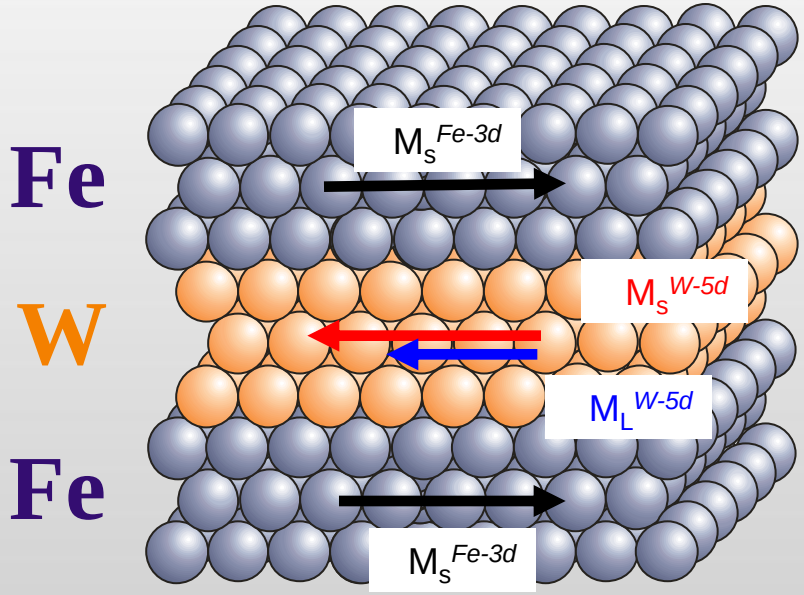
# Induced magnetism

$M_{spin}^{W\text{-}5d}$ = - 0.17 $\mu_B$/atom

$M_{orbital}^{W\text{-}5d}$ = - 0.015 $\mu_B$/atom

$M_{tot}^{W\text{-}5d}$ = - 0.185 $\mu_B$/atom

$M_{orbital}^{W\text{-}5d}/M_{spin}^{W\text{-}5d}$ = + 0.09

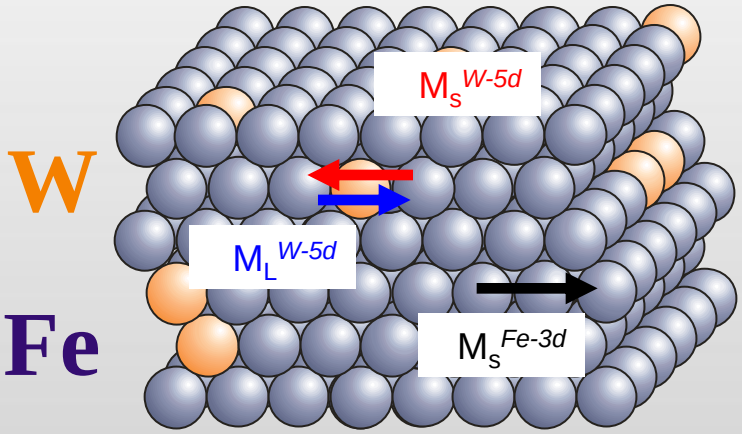



$M_{spin}^{W\text{-}5d}$ = - 0.70 $\mu_B$/atom

$M_{orbital}^{W\text{-}5d}$ = + 0.03 $\mu_B$/atom

$M_{tot}^{W\text{-}5d}$ = - 0.67 $\mu_B$/atom

$M_{orbital}^{W\text{-}5d}/M_{spin}^{W\text{-}5d}$ = - 0.043




Dimensionality may break even the well-established atomic rules (third Hund's rule)

# Who Breaks the Third Hund's Rule ?

- **Charge transfer 5d W ↔ 3d Fe**

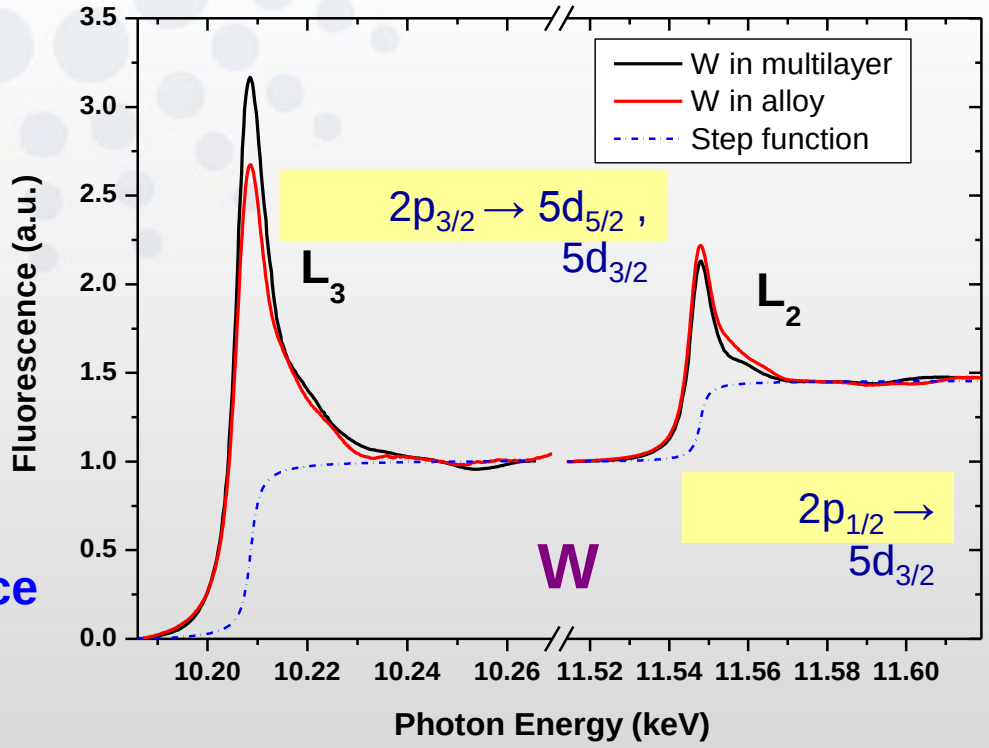
**Multilayer Fe/W :** $n_h^{W\text{-}5d}$ **= 6.11 holes**

⇒ $n_h^{5/2} / n_h^{3/2} = \underline{3.17}$

**Alloy $Fe_{97}/W_3$ :** $n_h^{W\text{-}5d}$ **= 5.72 holes**

⇒ $n_h^{5/2} / n_h^{3/2} = \underline{2.10}$

- **Local symmetry: impurity vs interface**




- **Spin-other-orbit coupling**

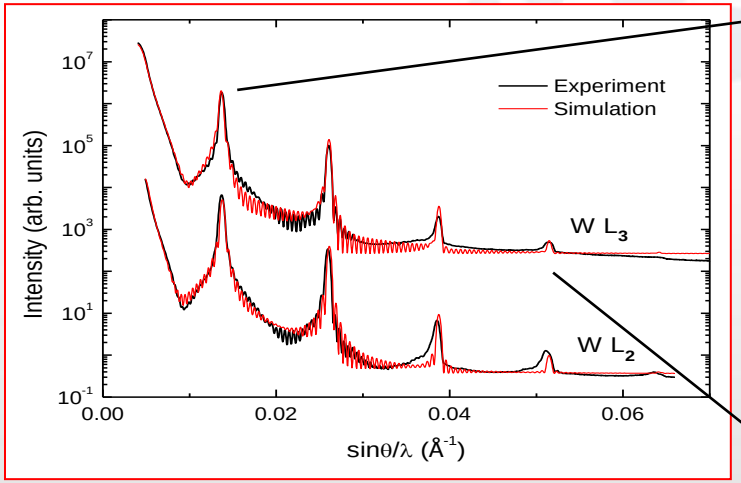
$J_{inter} . S_z^{Fe} . S_z^{W} > \lambda_{inter} . S_z^{Fe} . L_z^{W} > \lambda_{intra} . S_z^{W} . L_z^{W}$

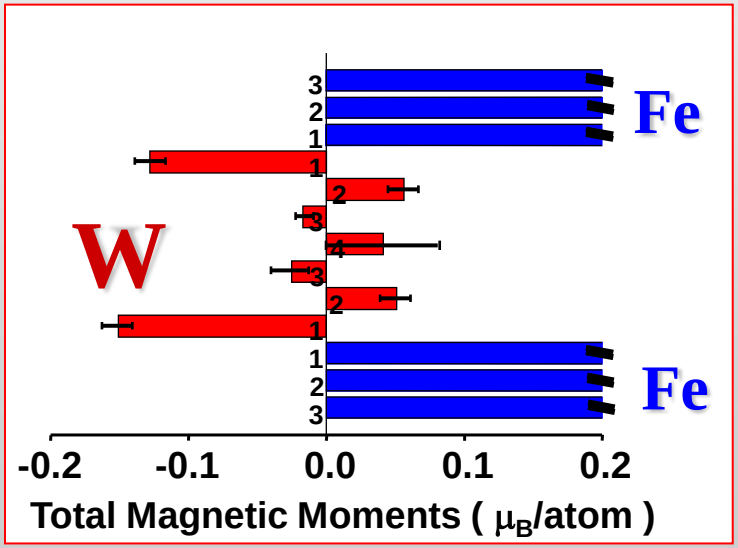
Inter-atomic spin-spin coupling (antiferromagnetic coupling)

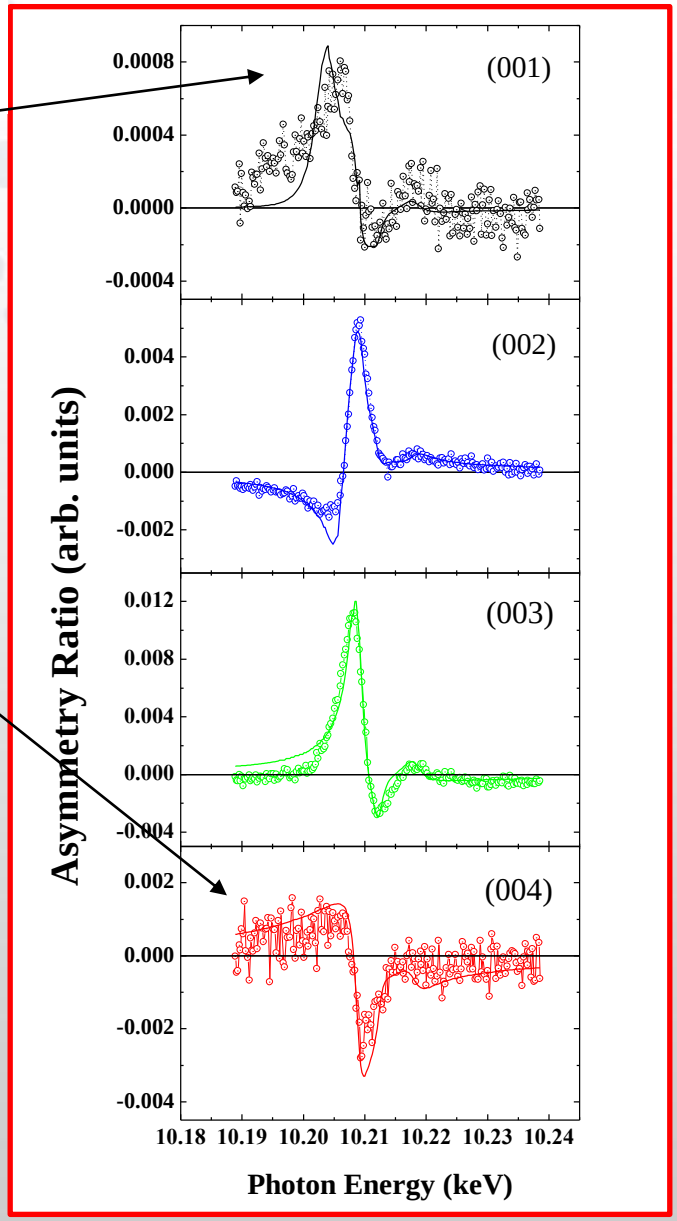
Inter-atomic antiferro Fe-W spin-other-orbit interaction

Intra-atomic W spin-orbit coupling (antiparallel) (third Hund's rule)

# Magnetization Profile in Fe/W



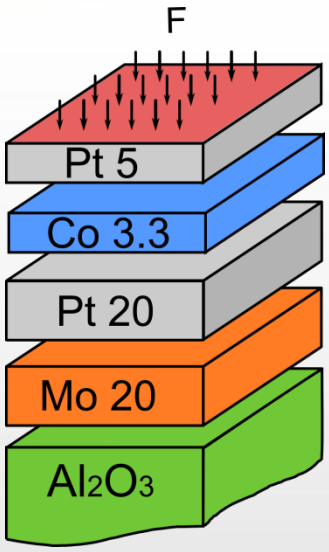



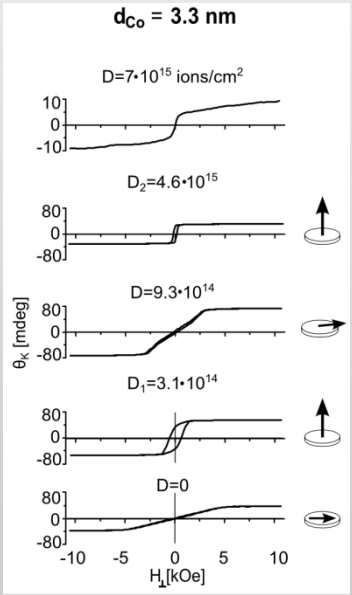




*N. Jaouen et al., Phys. Rev. B 70, 094417 (2004)*

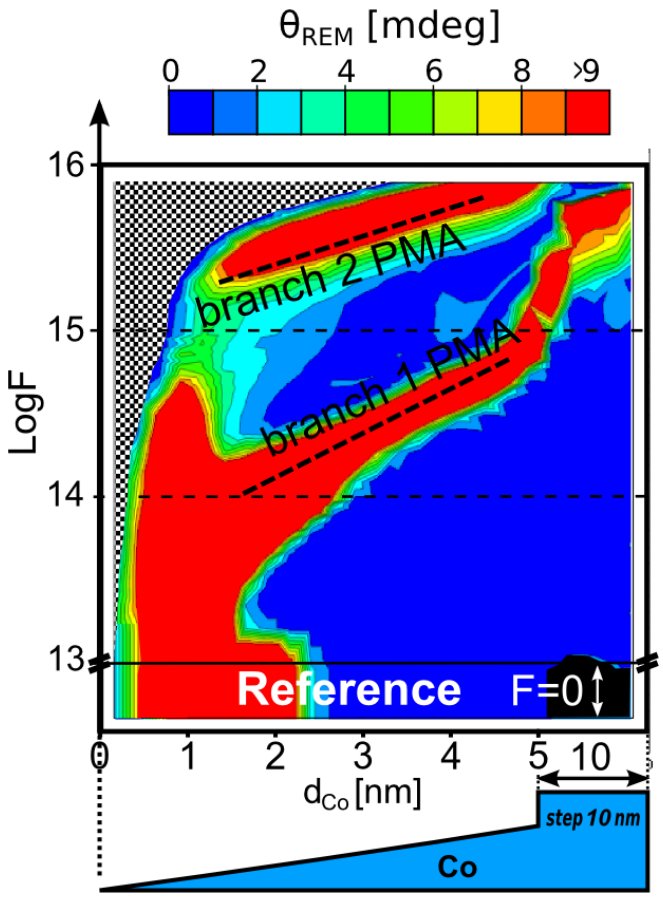
# Magnetism in Ga+ irradiated Pt/Co/Pt

## Magneto-optical Kerr studies

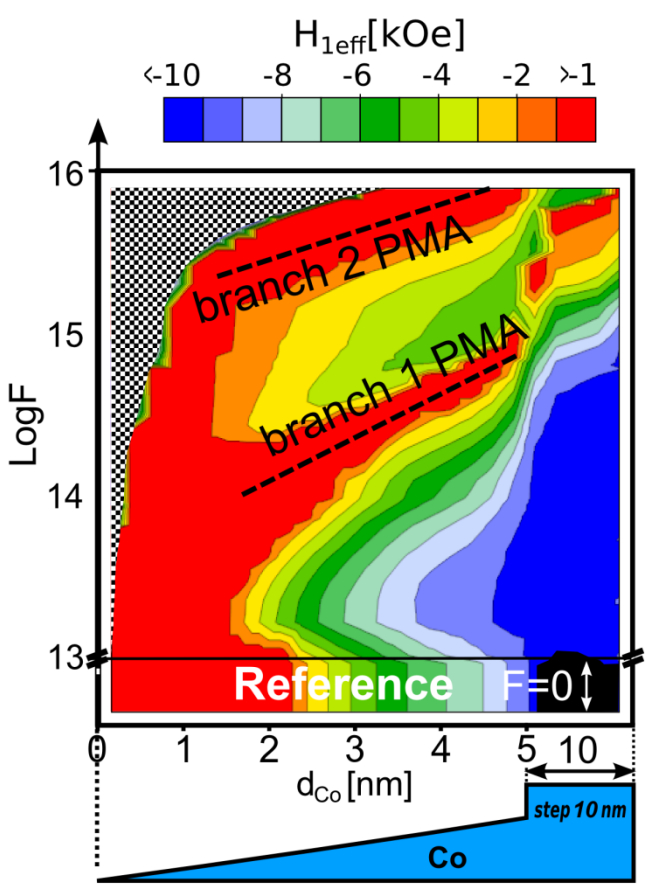


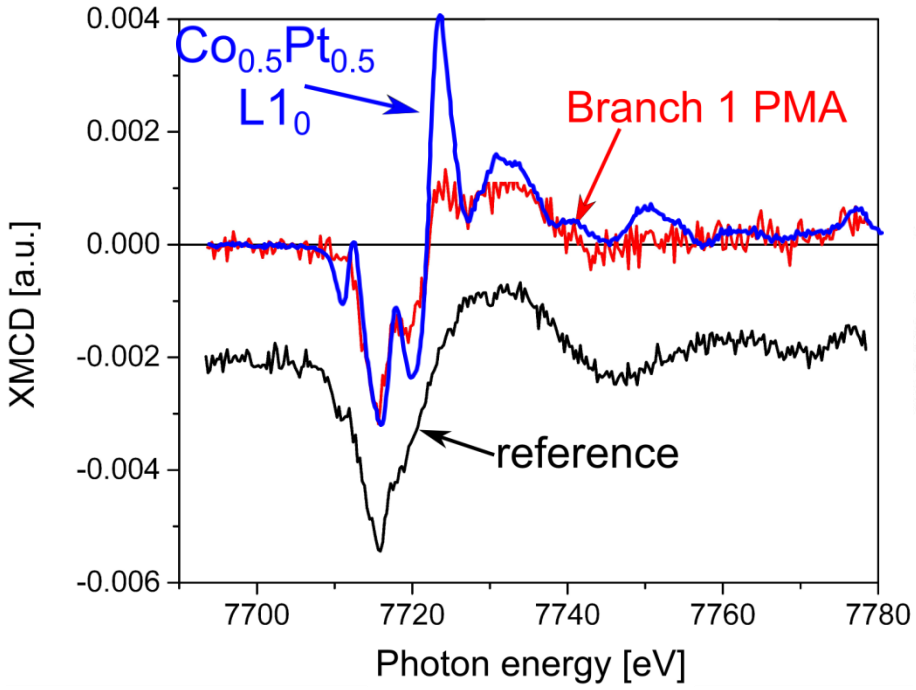

**Remanence $\theta_{REM}$**

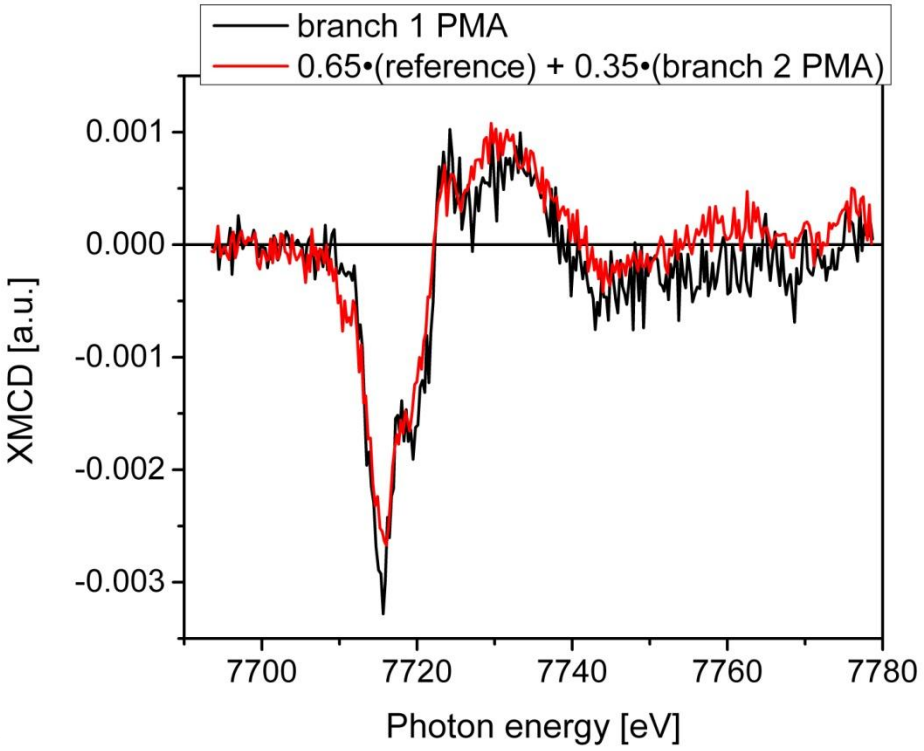



**Effective anisotropy field $H_{1eff}$**



# XMCD studies at the Cobalt K edge



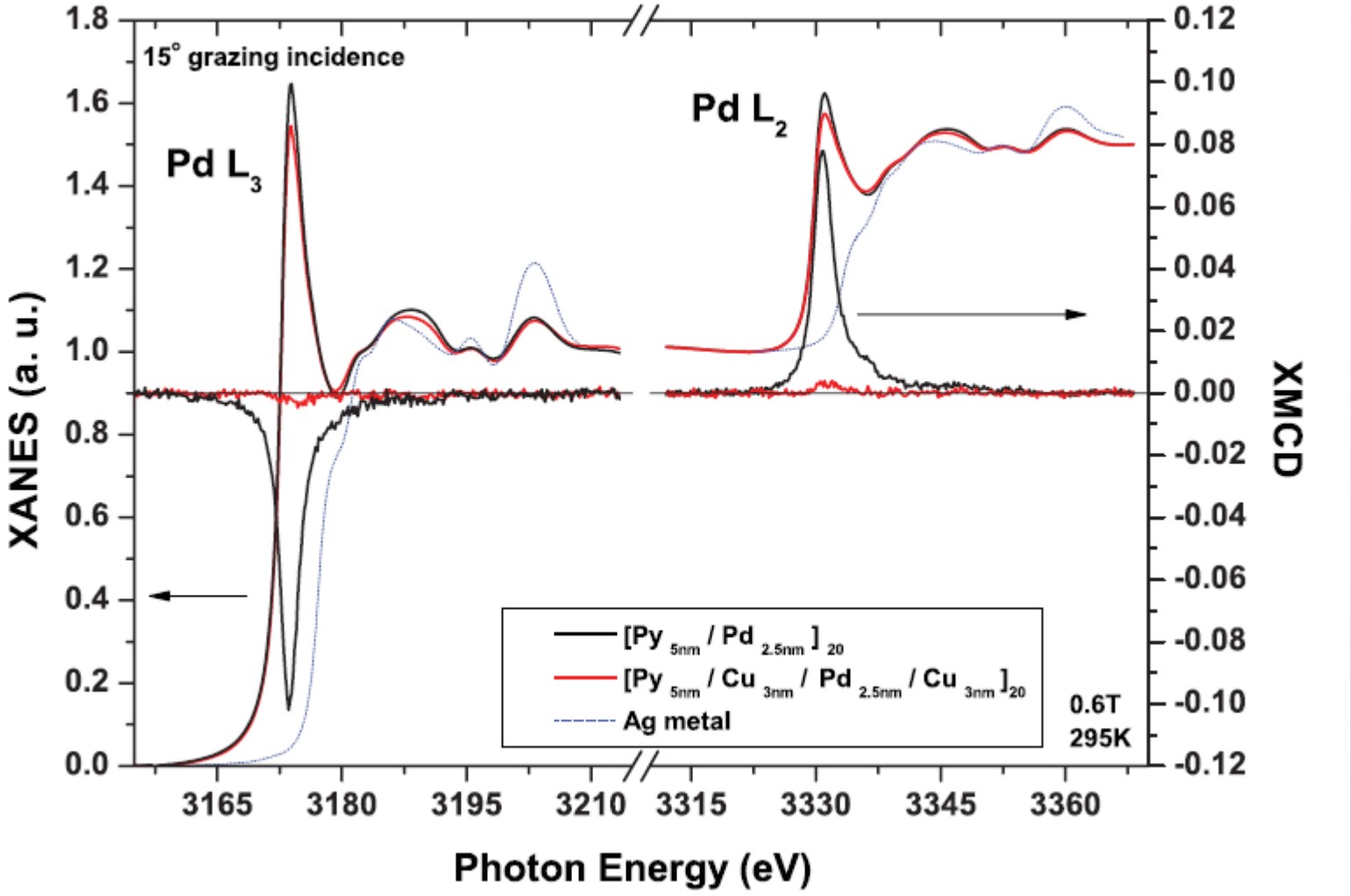




**PMA branches 1 and 2 are due to formation of a CoPt alloy at upper and lower interfaces, respectively**

A. Maziewski, P. Mazalski, Z. Kurant, M.O. Liedke, J. McCord, J. Fassbender, J. Ferré, A. Mougin, A. Wawro, L.T. Baczewski, A. Rogalev, F. Wilhelm, T. Gemming, *Phys. Rev. B* 85, 054427 (2012).

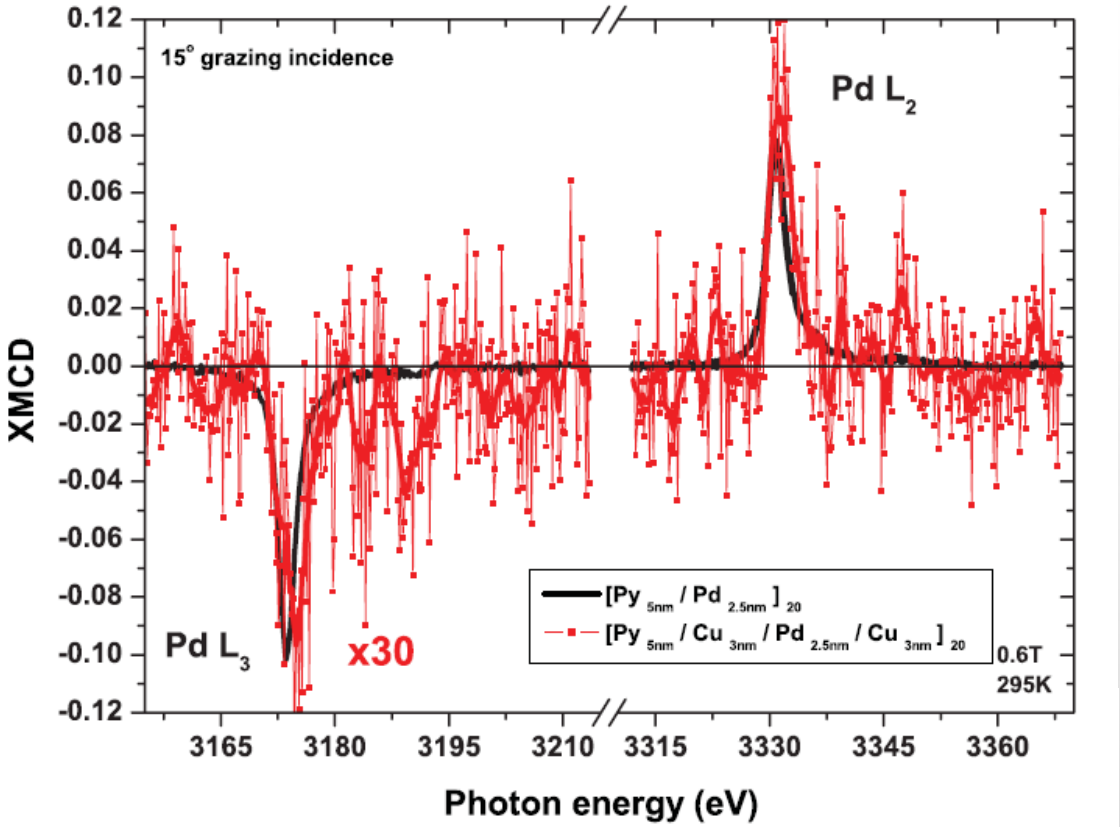
# Exchange coupling in multilayers

## Indirect exchange coupling in Py/Cu/Pd system




**Pd acquires a moment even through a 3nm thick Cu layer**

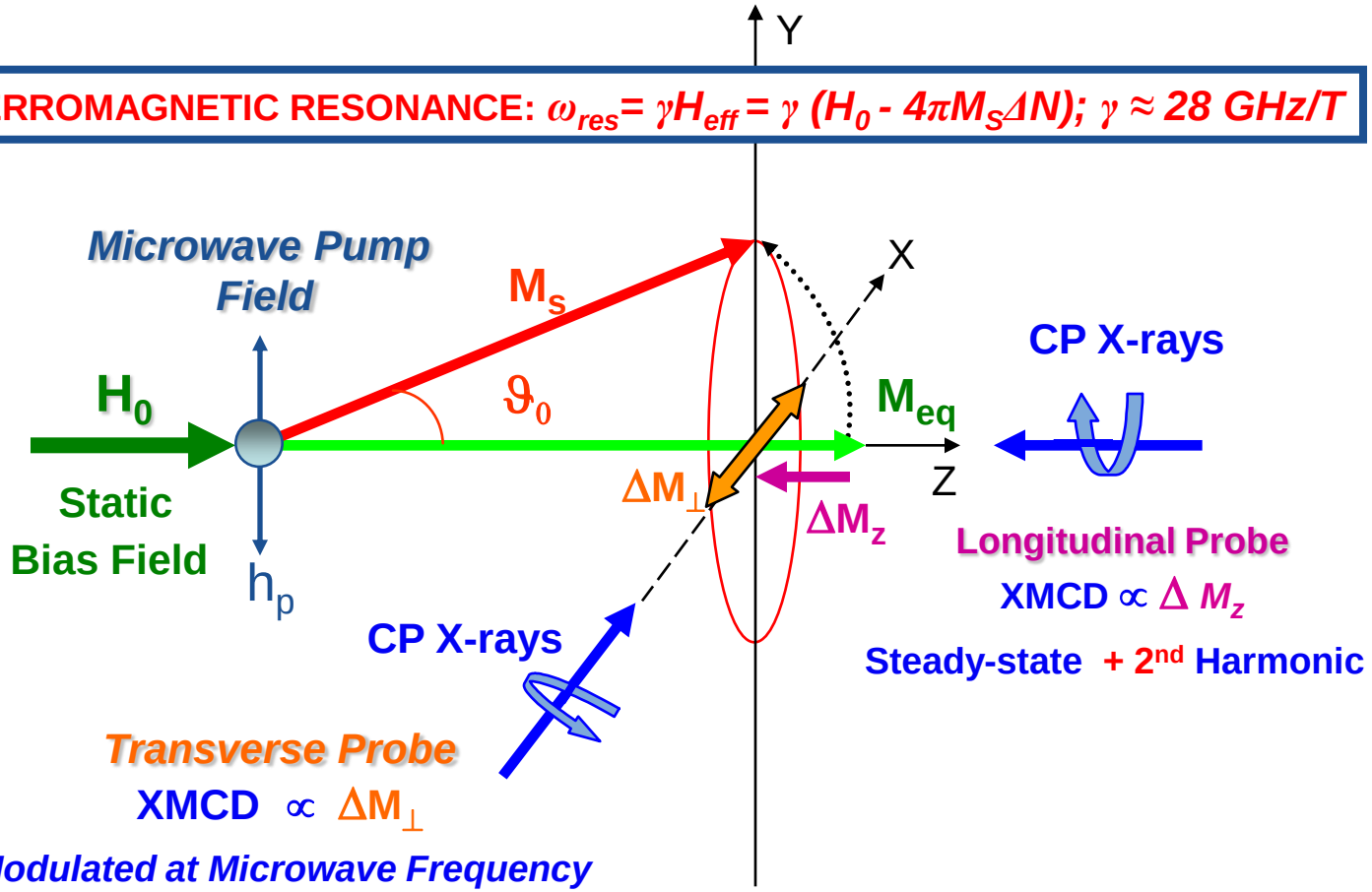
# Exchange coupling in multilayers




| Sample | $m_{Pd}^{tot}$ ($\mu_B$/at) | $m_L/m_S$ | $n_{h,4d}$/at |
|---|---|---|---|
| Py/Pd | $+0.1160 \pm 0.0007$ | $0.0485 \pm 0.002$ | 1.36 |
| Py/Cu/Pd | $+0.0036 \pm 0.0007$ | $0.028 \pm 0.08$ | 1.31 |

W.E.Bailey, A. Ghosh, S. Auffret, E. Gautier, U.Ebels, F. Wilhelm, A. Rogalev, *Phys. Rev. B* 86, 144403 (2012).

# A Probe of Magnetization Dynamics using XMCD

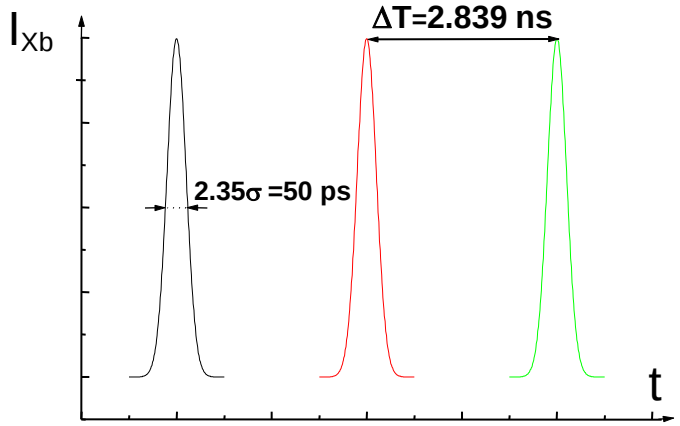



Small Precession angle $\vartheta_0$ → High Microwave Pumping Power Required !..

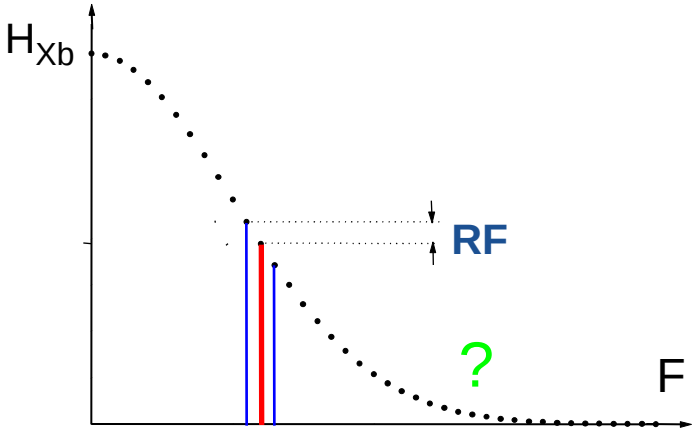
J. Goulon, A. Rogalev et al. J. Synchrotron Radiation 14 (2007) 257

# X-ray detection of magnetic resonance

## Time-domain: Bunch structure




## Frequency domain: Harmonics




**RF = 1/ΔT = 352.2023 MHz**

## GAUSSIAN MULTI-BUNCHES

$$I_{Xb}(t) = I_0 \sum_N \delta(t - N.\Delta T) \otimes \frac{1}{\sigma\sqrt{2\pi}} \exp\left\{-\frac{t^2}{2\sigma^2}\right\}$$

**ESRF:** $\delta t_{fwhm} = 2.35\ \sigma = 50$ ps $\quad \Delta T = 2.839$ ns

**FT** ⇕ *cf. E. Gratton (1992)*

$$H_{Xb}(F) = I_0 .RF \sum_N \delta(F - N.RF) \times \exp\left[-2(\pi\sigma F)^2\right]$$

$\sigma_F$ (*Freq. Distribution*) = $(2\pi\sigma)^{-1}$ = 7.48 GHz

$F_{1/2}$ (hwhm) = 8.79 GHz (N = 25)

*i.e. in the microwave X-band*

➡ **HETERODYNE DETECTION OF XDMR**

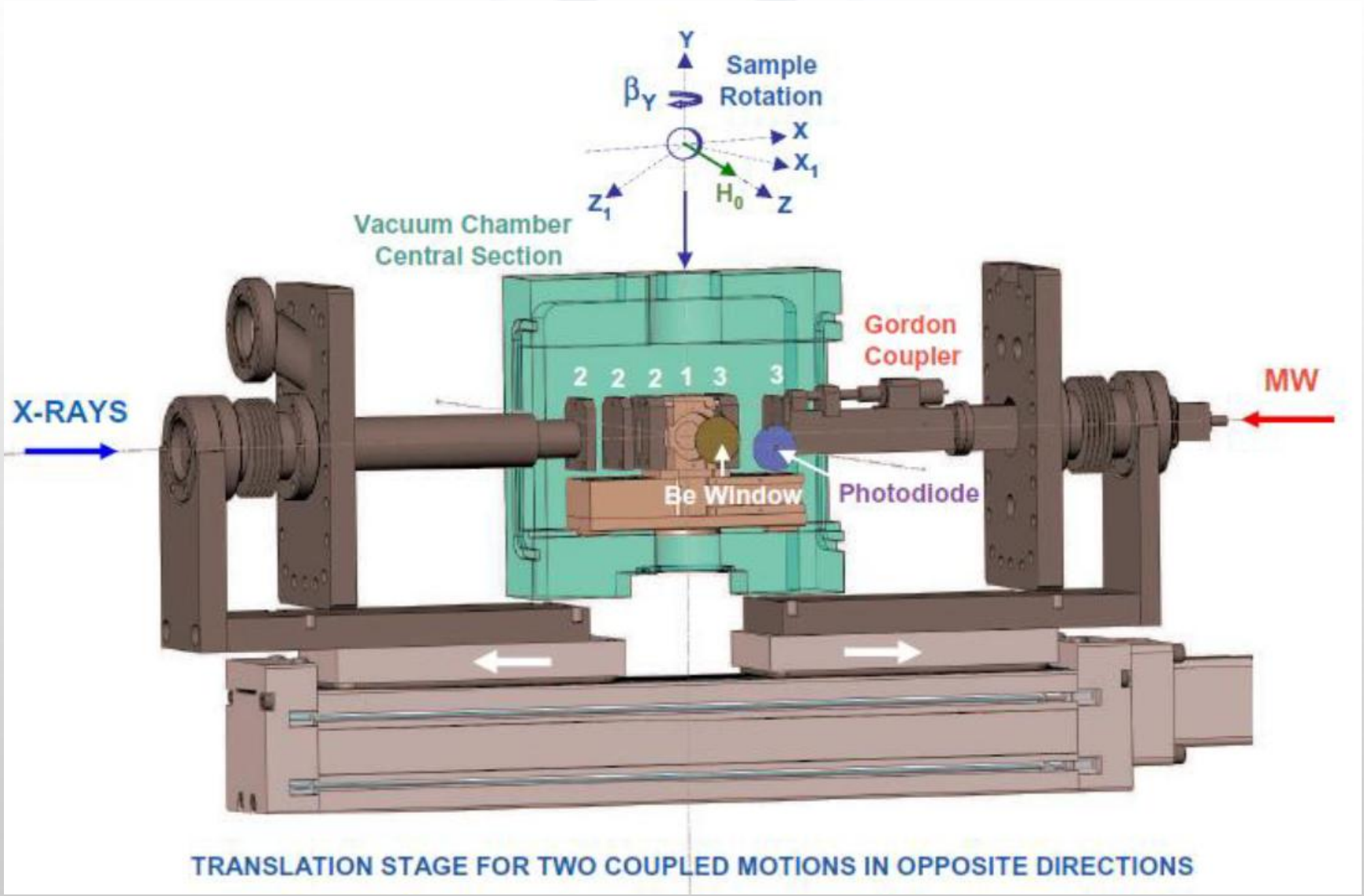
***USING HARMONICS*** **N x RF**

***… AS INTERNAL LOCAL OSCILLATOR***

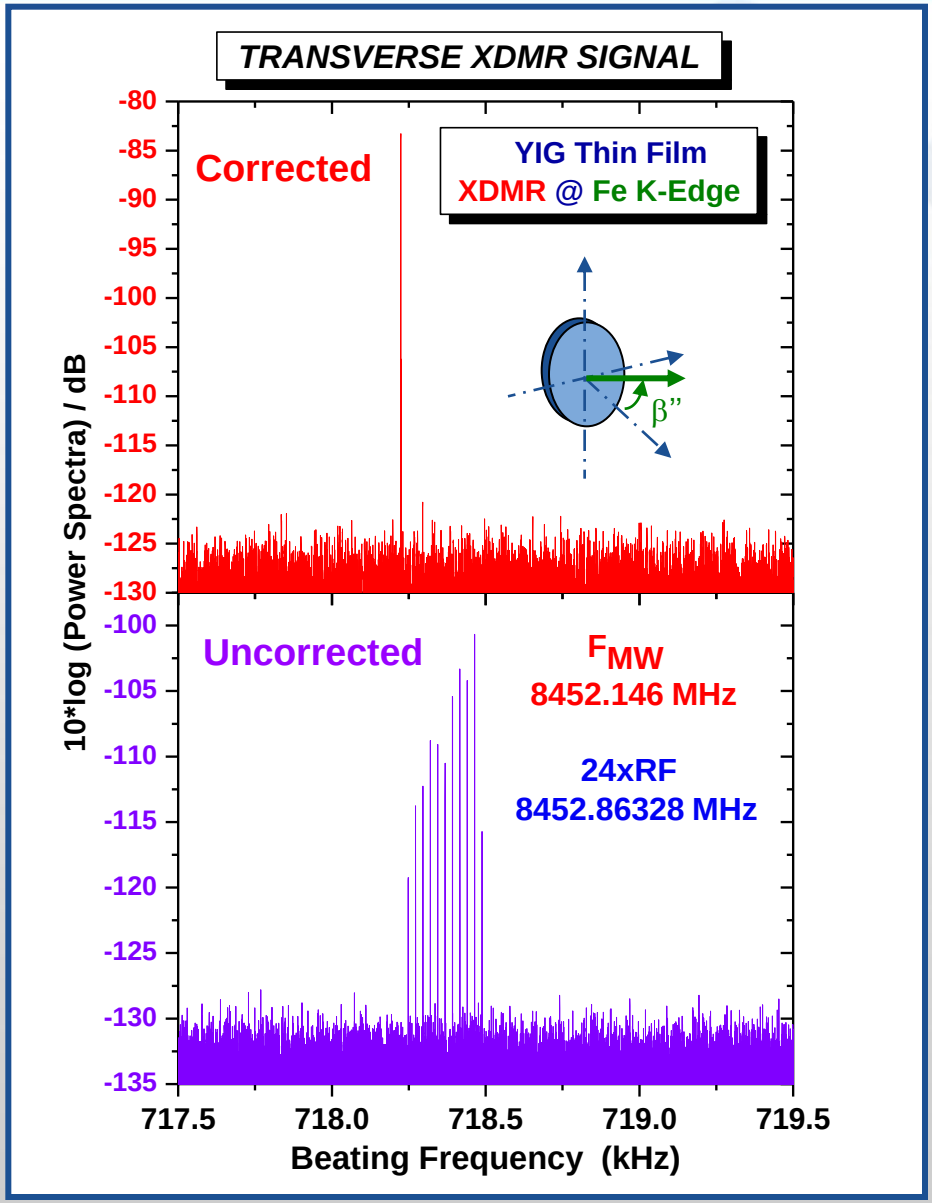
**LO = 24 x RF** = 8452.848 MHz

# X-ray detection of magnetic resonance

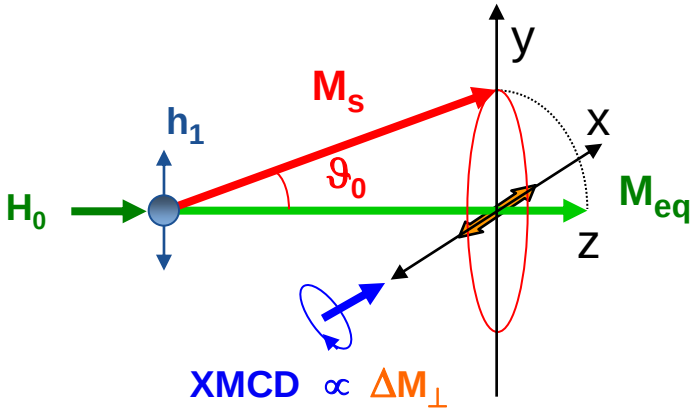
## New tunable X-band $TE_{102}$ microwave cavity for XDMR experiments



# X-ray detection of magnetic resonance




## XDMR IN TRANSVERSE GEOMETRY




*Oscillating at Microwave Frequency*

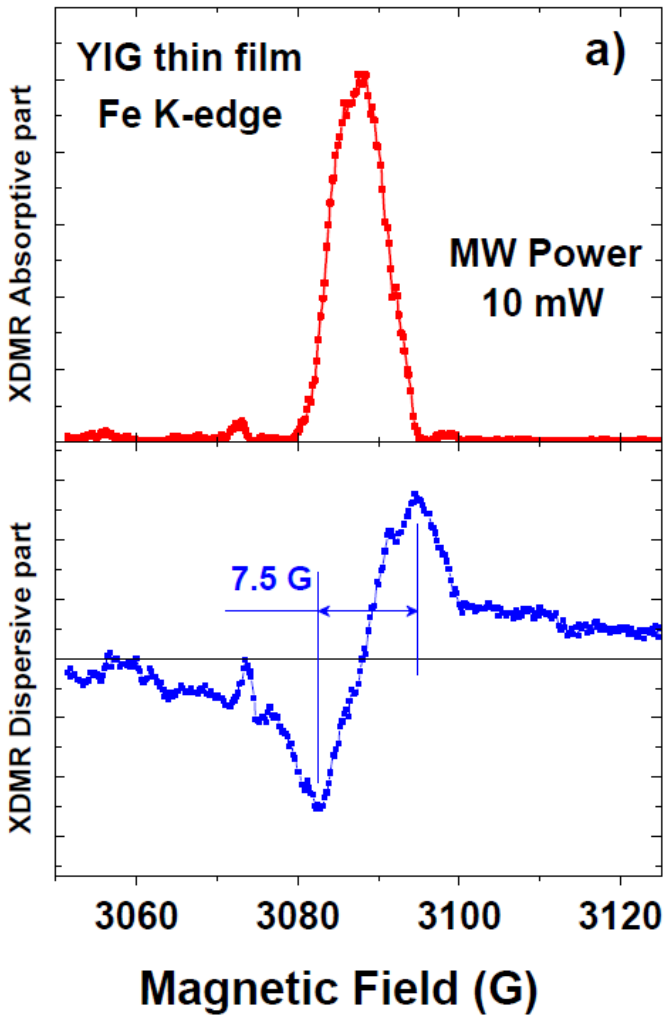
**BEATING FREQUENCY: 718. 223166 kHz**

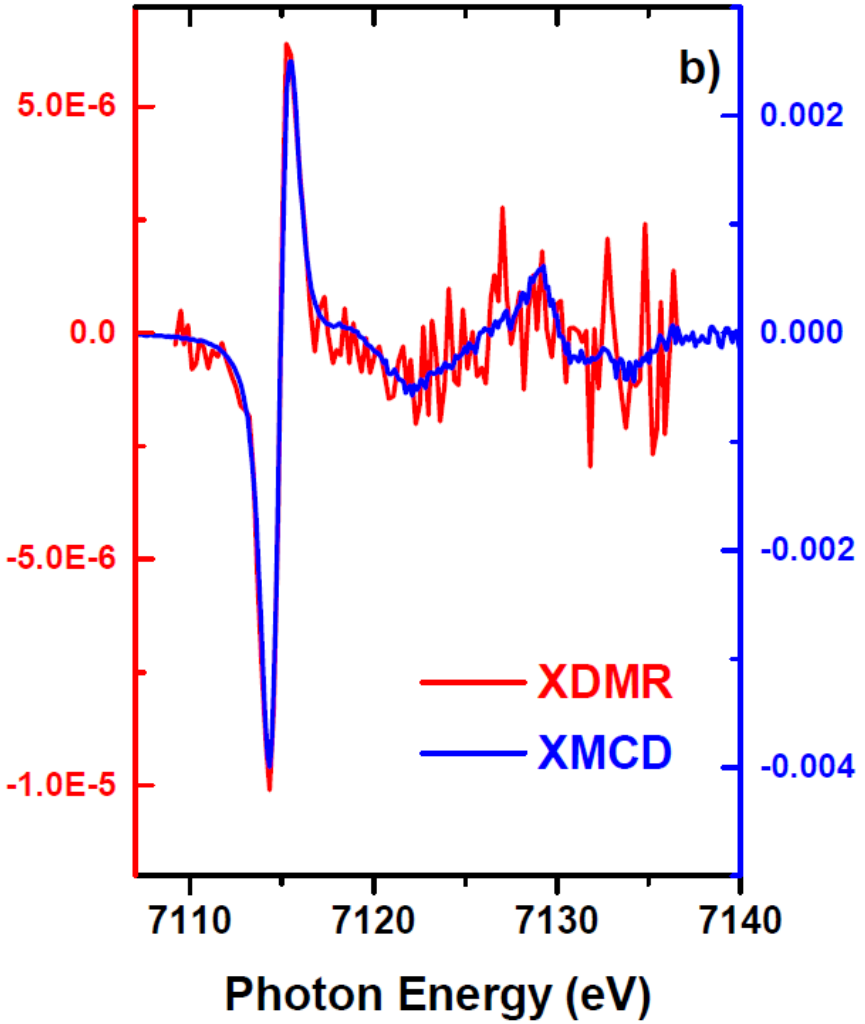
**UNEXPECTED DIFFICULTY**

*RF Frequency is an ADJUSTABLE parameter for Storage-Ring operation*

- **XDMR Signal shifts by 24 x 1 Hz at each re-adjustment**
- **Real -Time Correction of $F_{MW}$**
- ***Super –Heterodyne Detection***

# *Super-Heterodyne Detection of XDMR Spectra*



# To conclude

X-ray Magnetic Circular Dichroism
is very powerful spectroscopic tool
to unravel the microscopic origin of magnetism

BUT

XMCD is only a small part of the story …

# THANK YOU