A theory of magneto-X-ray effects in FCC cobalt

1990 J. Phys.: Condens. Matter 2 9451

(http://iopscience.iop.org/0953-8984/2/47/019)

J. Phys.: Condens. Matter 2 (1990) 9451–9456. Printed in the UK

LETTER TO THE EDITOR

A theory of magneto-x-ray effects in FCC cobalt

P Strange† and B L Gyorffy‡

† Physics Department, Keele University, Keele, Staffs ST5 5BG, UK and Rutherford-Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, UK

‡ Physics Department, Bristol University, Tyndall Avenue, Bristol BS8 1TL, UK

Abstract. We recall briefly the general theory of magneto-optical effects and calculate the K-edge absorption of circularly polarized radiation by FCC cobalt. In particular we study the difference in absorption rate for right- and left-handed incident radiation, the Faraday rotation and the induced ellipticity of linearly polarized radiation in order to compare the results with recent experiments.

Recently, the high intensity x-rays available from synchrotron sources have been exploited with great success to probe the electronic structure of materials to a level not previously possible [1]. Not only has this involved the refinement of conventional methods, but completely new techniques have been developed. For example, magnetic scattering has been used successfully to probe of the magnetic structure of rare earth materials [1–3]. Furthermore new spectroscopies have become feasible. An example of this is the work of Schutz *et al*, ([4, 5] for example), who have performed x-ray absorption rate measurements on a large number of materials and studied the difference in absorption rate for left- and right-circularly polarized incident radiation. The theoretical understanding of their results was provided by Ebert *et al* [6, 7] on the basis of the relativistic spin-polarized multiple scattering theory of Strange *et al* [8, 9]. Recently new experiments have been reported by Siddons *et al* [10]. They have measured the Faraday rotation of x-rays incident upon a thin layer of cobalt as a function of energy above the K-absorption edge. Also in this region they have measured the induced ellipticity as a function of energy. All these effects are a result of the interaction of the incident photon with the magnetic moment of the material. However, in a non-relativistic framework they would be identically zero. In other words they all require relativistic quantum theory to describe them.

In this letter, following Ebert *et al* [6, 7] we discuss all three effects on the basis of a relativistic spin-polarized multiple scattering theory. Firstly, we present a brief description of the formal theory. Secondly, we present a discussion of the theory and give a more qualitative description of its application to magnetic metals. Finally we illustate the theory with an explicit calculation of magneto-optical effects in cobalt.

A very concise theory for studying spectroscopies, including x-ray absorption, was developed by Durham [1]. He showed that the absorption rate for a core state i with incident radiation of wavevector $\boldsymbol{q}$ and polarization state λ could be written

$$W_i^{\boldsymbol{q}\lambda} = -\frac{1}{\Gamma}\int \mathrm{d}^3r \int \mathrm{d}^3r'\, \psi_i^*(\boldsymbol{r}) X_{\boldsymbol{q}\lambda}(\boldsymbol{r}) \operatorname{Im} G(\boldsymbol{r}, \boldsymbol{r}'; E + \hbar\omega) \times X_{\boldsymbol{q}\lambda}^*(\boldsymbol{r}')\psi(\boldsymbol{r}')\Theta(E_i + \hbar\omega - E_\mathrm{F}) \tag{1}$$

0953-8984/90/479451+06$03.50

For the relativistic version of the formalism E_i is the energy of the core state ψ_i, which is a Dirac 4-vector. E_F is the Fermi energy, $G(\boldsymbol{r}, \boldsymbol{r}'; E)$ is the 4×4 Dirac bispinor Green function which describes the electronic structure of the material, and in particular the final states available for the excited electron. It can be evaluated from scattering theory. The $X_{\boldsymbol{q}\lambda}$ are interaction operators. Within a fully relativistic and spin-polarized formalism this is given by

$$X_{\boldsymbol{q}\lambda}(\boldsymbol{r}) = -e\boldsymbol{\alpha} \cdot \boldsymbol{A}(\boldsymbol{r}) \tag{2}$$

where $\boldsymbol{A}(\boldsymbol{r})$ is the vector potential associated with the incident radiation and $\boldsymbol{\alpha}$ is the usual Dirac matrices vector. According to Strange *et al* [9] the multiple scattering form of the Green function is given by

$$G(\boldsymbol{r}, \boldsymbol{r}'; E) = \sum_{\Lambda\Lambda'} Z_\Lambda(\boldsymbol{r}, E)\tau_{\Lambda\Lambda'}(E)Z^+_{\Lambda'}(\boldsymbol{r}', E) - \sum_{\Lambda} Z_\Lambda(\boldsymbol{r}, E)J_\Lambda(\boldsymbol{r}', E). \tag{3}$$

Here $Z(\boldsymbol{r}, E)$ and $J(\boldsymbol{r}, E)$ are the regular and irregular solutions of the Kohn–Sham–Dirac equation, respectively. $\tau_{\Lambda\Lambda'}(E)$ is the scattering path operator given by the integral of the inverse of the KKR matrix over the first Brillouin zone. To calculate the absorption rates we only need the imaginary part of the Green function and hence in principle it is only necessary to retain the first term in equation (3). In practice this is not the case as it turns out to be computationally efficient to evaluate the Green function at complex energies. At complex energies the second term is non-zero. Substituting equation (3) into equation (1) we find that the absorption rate is given by the scattering path operator multiplied by some interaction operator matrix elements.

We obtain the vector potential by noting that the x-rays have an electric field vector $E = E_0 e^{i(\boldsymbol{q}\cdot\boldsymbol{r}-\omega t)}$. This can be related to the vector potential via Maxwell's equations. To calculate the matrix elements we expand the exponential part of the interaction operator as a series. Keeping only the first term (which is 1) leads to the conventional relativistic dipole selection rules. In the results which follow we have kept the first two terms in the series.

This theory is sufficient to show a difference in the absorption rate for left- and right-circularly polarized incident x-rays. The reason for this is easy to see. Right circularly polarized x-rays are described by the electric field vector $\boldsymbol{E}_- = (E_x, -iE_y, 0)$ and left-circularly polarized x-rays by $\boldsymbol{E}_+ = (E_x, iE_y, 0)$. Obviously when the scalar product of these two with the alpha matrices is taken, different matrix elements will be non-zero in the resulting matrices. This would occur in non-relativistic quantum theory as well. However in the non relativistic case the radial parts of the matrix elements would be the same as they are independent of the m-quantum number. This leads to a cancellation of the difference in the absorption rates. In the relativistic case the radial parts of the matrix elements are dependent on the m-quantum number and hence the same cancellation does not occur.

In addition to describing the behaviour of circularly polarized light as it passes through a magnetic material, the above theory works well for linearly polarized radiation. From our point of view the most useful formulation of the Faraday effect was given by Bennett and Stern [11] and by Smit [12]. They have shown that the real part of the diagonal components of the optical conductivity tensor σ^1_{xx} are proportional to the sum of the power absorbed from left-circularly polarized and right-circularly polarized incident rays. Furthermore they show that the difference in absorption between right- and left-circularly polarized radiation is proportional to the imaginary part of

the off-diagonal component of the optical conductivity tensor σ^2_{xy}. Their power absorbed is a sum over all processes which absorb radiation of frequency ω. In the experiments we wish to describe the incident radiation is tuned to some core level energy, and we assume excitation of this core electron to the conduction band is the only absorption process which contributes to the Faraday effect. The imaginary part of the diagonal component and the real part of the off-diagonal component of the optical conductivity tensor can then be found using the Kramers–Kronig relations. As discussed by Smit [12] and Ebert *et al* [7] the optical effects we wish to calculate are then given by

$$\Phi = \phi_F + i\epsilon_F = \sigma_{xy}/\sigma_{xx}n \tag{4}$$

where ϕ_F is the Faraday rotation and ϵ_F is the ellipticity.

We now turn to a relatively simple qualitative discussion of how the Faraday effect and ellipticity arise. When linearly polarized light enters a metal it is split into left- and right-circularly polarized components of equal amplitude. These then travel through the metal without interference. On exiting from the material the beam recombines into a single polarization state. The material involved will have a different complex refractive index for each polarization state. In general both the real and imaginary parts may differ and so the emerging two beams will have different amplitudes and phases. This will lead to a small amount of elliptical polarization being introduced, and an apparent rotation of the major axis of polarization.

Of course, the degree of absorption, rotation and induced ellipticity depends on the thickness of material the x-ray passes through. However, by doing the experiments for several thicknesses of material the thickness can be divided out and curves characteristic of the material can be found.

We have carried out the calculations indicated above from first principles using a spin-polarized potential derived from a self-consistent, spin-polarized LMTO calculation. The material chosen for this was FCC cobalt, on whichn some high quality experimental data became available recently.

In figure 1 we show the total x-ray absorption spectrum as a function of energy above the Fermi energy. We have included in this and all diagrams an instrumental broadening. The curve is essentially in perfect agreement with the experimental work of Siddons *et al* [10] and Schutz *et al* [5]. In figure 2 we show the difference in absorption rate for left- and right-circularly polarized x-rays. Note that the magnitude of this curve is three orders of magnitude smaller than figure 1, yet we still observe good qualitative agreement with the experimental results of Schutz *et al*. Figure 3 displays the Faraday rotation up to approximately 40 eV above the Fermi energy. Here the agreement with experiment is less good, although we still reproduce the main features of the spectrum seen by Siddons *et al* [10]. Certainly the theoretical results are of the correct order of magnitude and, on the whole, the correct sign. Finally in figure 4 we show the ellipticity as a function of energy, Again we are able to reproduce the observed behaviour on a qualitative level.

Figures 1 and 2 have been Lorentzian broadened with a width 1.8 eV and figures 3 and 4 with a width 1.25 eV. The degree of agreement between the theory and experiment can be improved in various parts of the energy range by changing the broadening factor. It should be noted that in the experiment measurements were taken with the field parallel and antiparallel to the incident x-ray wavevector. The results of these two experiments should be of the same magnitude and opposite sign. This is not the

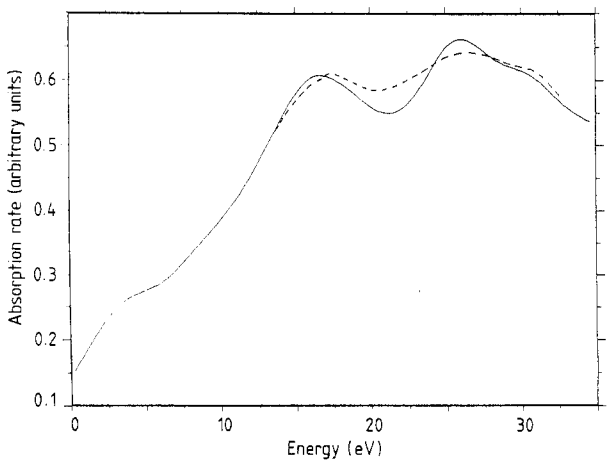

Figure 1. The total K-Edge x-ray absorption spectrum for FCC cobalt as a function of energy above E_F: full curve, theory; broken curve, experiment.

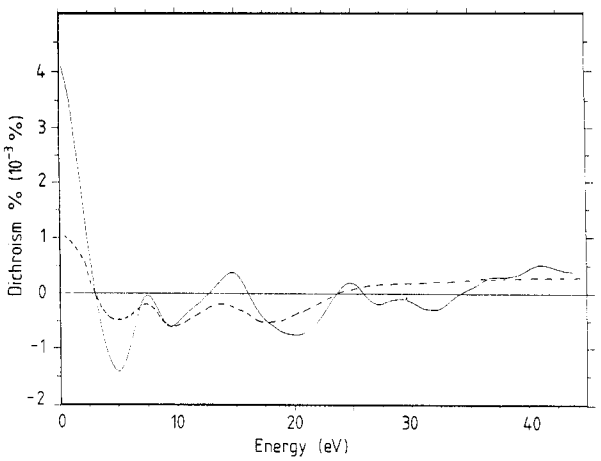

Figure 2. The difference in absorption rate for between left- and right-circularly polarized incident x-rays close to the K-edge: full curve, theory; broken curve, experiment. The experimental results are taken from Schutz *et al* (1988).

case because the experiment used a polycrystalline sample. The discrepancy between the two experimental curves is a measure of the uncertainty in the results. Finally the experiment was performed on a material containing 10% iron to stablise the FCC phase. The effect of this on the Faraday rotation measured is not known. Hence a direct comparison between experiment and theory is fraught with uncertainty.

In conclusion we have showed that the conventional theory of magneto-optical behaviour in solids at optical frequencies gives a good account of the corresponding phenomena for x-rays at the K-absorption edge of cobalt. As occurs at optical frequencies, the simple explanation of the effects is in terms of the difference in absorption rate for left- and right-circularly polarized x-rays. We have presented the first ever quantitative first-principles calculation of the Faraday rotation and elliptic-

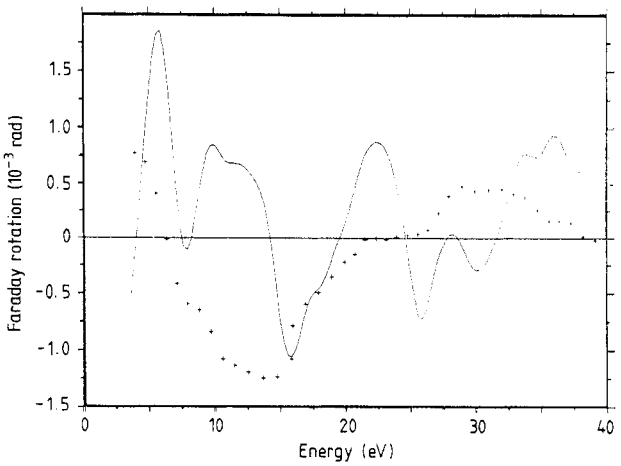

Figure 3. The Faraday rotation for FCC cobalt for incident linearly polarized x-rays close to the K-edge for FCC cobalt: full curve, theory; crosses, experiment. The experimental results are taken from Siddons *et al* (1990).

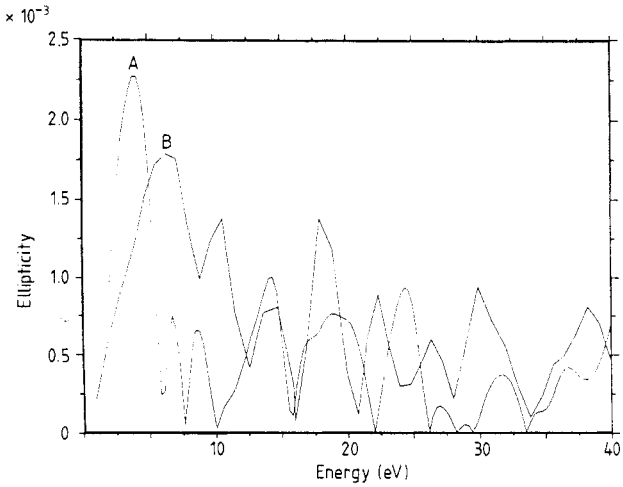

Figure 4. The induced ellipticity for FCC cobalt for incident linearly polarized x-rays close to the K-edge for FCC cobalt: curve A, theory; curve B, experiment. The experimental results are taken from Siddons *et al* (1990).

ity for a core electron. Finally, we note that all the magneto-optical data presented here contains a wealth of information about the electronic and magnetic structure of the materials. The structure observed in figures 2–4 contains a wealth of information about the details of the electronic structure of cobalt, particularly about differences in the m_j quantum number character of bands above the Fermi energy. Evidently more experiments on single and pure crystals would be eminently worthwhile. Moreover, studying the effect of changing the relative orientation of the crystal and the incident photon beam could also yield interesting results. We hope that measurements of magneto-optical behaviour will become an important tool for probing the subtle properties of matter determined by the interplay between spin–orbit coupling and

spin-polarization.

We would like to thank Professor M Hart for showing us his group's data prior to publication. We would also like to thank him and Dr H Ebert for some useful discussions.

References

[1] Durham P J 1984 *The Electronic Structure of Complex Systems* ed P Phariseau and W M Temmerman (New York: Plenum)
[2] Blume M 1985 *J. Appl. Phys.* **57** 3615
[3] Lovesey S W 1985 *J. Phys C: Solid State Phys.* **20** 5625
[4] Schutz G, Wagner W, Wilhelm W, Kienle P, Zeller R, Frahm R and Materlik C 1987 *Phys. Rev. Lett.* **58** 737.
[5] Schutz G, Frahm R H, Wiencke R, Wilhelm W, Wagner W and Kienle P 1989 *Rev. Sci. Instrum.* **60** 1661
[6] Ebert H, Strange P and Gyorffy B L 1988 *J. Physique Coll.* **12** C8 31
[7] Ebert H, Strange P and Gyorffy B L 1988 *J. Appl. Phys.* **63** 3055
[8] Strange P, Staunton J B and Gyorffy B L 1984 *J. Phys. C: Solid State Phys.* **17** 3355
[9] Strange P, Ebert H, Staunton J B and Gyorffy B L 1989 *J. Phys.: Condens. Matter* **1** 2959
[10] Siddons D P, Hart M, Amemiya Y and Hastings J B 1990 *Phys. Rev. Lett.* **64** 1967
[11] Bennett H S and Stern E A 1965 *Phys. Rev.* **137** A448
[12] Smit J 1971 *Magnetic Properties of Materials (Inter-University Electronics Series)* (New York: McGraw-Hill)