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1994 J. Phys.: Condens. Matter 6 9469

(http://iopscience.iop.org/0953-8984/6/44/025)

J. Phys.: Condens. Matter 6 (1994) 9469–9475. Printed in the UK

The high-resolution magnetic Compton profile of Fe–5.8 at.% Si

Y Sakurai†, Y Tanaka†, T Ohata‡, Y Watanabe‡, S Nanao‡, Y Ushigami§, T Iwazumi‖, H Kawata‖ and N Shiotani¶

† The Institute of Physical and Chemical Research, Wako, Saitama 351-01, Japan
‡ Institute of Industrial Science, The University of Tokyo, Roppongi, Minato, Tokyo 106, Japan
§ Electrical Steel Laboratory, Nippon Steel Co., Futtsu, Chiba 299-12, Japan
‖ Photon Factory, National Laboratory for High-Energy Physics, Tsukuba, Ibaraki 305, Japan
¶ Tokyo University of Fisheries, Kounan, Minato, Tokyo 108, Japan

Received 14 June 1994

Abstract. A total and a magnetic Compton profile of an Fe–5.8 at.% Si single crystal are measured with a momentum resolution of 0.12 atomic units, a much higher resolution than that of any previous works. From these the majority- and minority-spin electron momentum distributions are deduced. The results are compared with those obtained by FLAPW calculations for pure Fe.

1. Introduction

Compton scattering yields information about the electron momentum distribution, $n(\boldsymbol{p})$. The momentum distribution of electrons involved in ferromagnetism is of particular importance. In terms of band theory it provides information about spin-dependent occupation in momentum space and spin-dependent wavefunctions in the ground state and thus serves as a sensitive test of various band models. The Compton profile, $J(p_z)$ is defined as a one-dimensional projection of the electron momentum distribution,

$$J(p_z) = \iint (n_{\mathrm{up}}(\boldsymbol{p}) + n_{\mathrm{down}}(\boldsymbol{p}))\,\mathrm{d}p_x\,\mathrm{d}p_y \qquad (1)$$

where p_x, p_y and p_z are the Cartesian momentum components, the z axis being parallel to the scattering vector and $n_{\mathrm{up}}(\boldsymbol{p})$ and $n_{\mathrm{down}}(\boldsymbol{p})$ are the electron momentum densities of majority-spin and minority-spin electrons, respectively. It is related to the measured double-differential scattering cross-section by

$$\mathrm{d}^2\sigma/(\mathrm{d}\Omega\,\mathrm{d}E_2) = (\mathrm{d}\sigma/\mathrm{d}\Omega)(E_2/E_1)J(p_z) = r_0^2 A^2(E_2/E_1)J(p_z) \qquad (2)$$

where $\mathrm{d}\sigma/\mathrm{d}\Omega$ is the differential cross-section, $E_{1\,(2)}$ is the initial (final) photon energy, r_0 is the classical electron radius and A is proportional to the scattering amplitude, which is written [1]

$$A = a + \mathrm{i}\boldsymbol{b}\cdot\boldsymbol{S} \qquad (3)$$

0953-8984/94/449469+07$19.50

where the first term is for charge scattering, and the second term corresponds to magnetic scattering from the electron spin S and is smaller than the charge scattering amplitude by a factor of E_1/mc^2. The vector b is determined by the photon energy and the scattering angle. It is clear from (3) that if b is real there is no interference between charge and magnetic scattering. However, if b is complex and this corresponds to the incident photon beam having a non-zero degree of circular polarization, there is an interference term. Since the interference term is linear in $b \cdot S$, it is smaller than the charge scattering by the factor of E_1/mc^2. However, it is sensitive to the spin direction and thus carries important information. It is obvious that magnetic Compton scattering experiments require a beam of circularly polarized x-rays to make the interference term non-zero, of high energy and of large flux to overcome the factor E_1/mc^2. It is convenient to rewrite the double-differential cross-section in terms of total Compton profile, $J(p_z)$, and magnetic Compton profile, $J_{\mathrm{mag}}(p_z)$ [2],

$$\mathrm{d}^2\sigma/(\mathrm{d}\Omega\,\mathrm{d}E_2) = (\mathrm{d}\sigma/\mathrm{d}\Omega)_{\mathrm{charge}}(E_2/E_1)J(p_z) + (\mathrm{d}\sigma/\mathrm{d}\Omega)_{\mathrm{mag}}(E_2/E_1)J_{\mathrm{mag}}(p_z). \tag{4}$$

Here

$$J_{\mathrm{mag}}(p_z) = J_{\mathrm{maj}}(p_z) - J_{\mathrm{min}}(p_z) \tag{5}$$

where

$$J_{\mathrm{maj}}(p_z) = \iint n_{\mathrm{up}}(p)\,\mathrm{d}p_x\,\mathrm{d}p_y$$

and

$$J_{\mathrm{min}}(p_z) = \iint n_{\mathrm{down}}(p)\,\mathrm{d}p_x\,\mathrm{d}p_y.$$

The magnetic Compton profile can then be obtained experimentally by subtracting the data set taken with the magnetizing field direction reversed. The total profile can be obtained by adding the data set. With an experimentally determined magnetic profile $J_{\mathrm{mag}}(p_z)$ and a total profile $J(p_z)$ it is possible to separate the Compton profiles of the majority-spin and minority-spin electrons, $J_{\mathrm{maj}}(p_z)$ and $J_{\mathrm{min}}(p_z)$.

The first magnetic Compton-profile measurements were performed by Sakai and Ono [3], who obtained circularly polarized gamma rays from the cryogenically oriented radioactive source of ^{57}Co. Although this pioneering experiment suffered from a low count rate, the result clearly demonstrated the existence of the magnetic effect predicted theoretically by Platzman and Tzoar [1]. Subsequent magnetic Compton experiments have been mostly devoted to searching for more intense sources of circularly polarized photons and thus differ significantly only in this respect. The three techniques used to date take advantage of the polarization properties of synchrotron radiation. The first of these, called the inclined-view method, makes use of the fact that synchrotron radiation emitted at a small angle above or below the electron orbital plane is elliptically polarized and therefore has some degree of circular polarization, although rather reduced in intensity compared with radiation emitted in the plane. The feasibility of this method was first proposed by Holt and Cooper [4]. Subsequently, some experiments [2, 5, 6] have made use of this simple but effective technique for the extraction of circularly polarized synchrotron radiation. In the second method, developed by Mills [7] from the method of Golovchenko and co-workers [8], an x-ray quarter-wave plate converts the linearly polarized radiation emitted in the orbital plane into elliptically polarized radiation.

The third method, proposed by Yamamoto and Kitamura [9] and successfully realized by Yamamoto and co-workers [10], is to use an insertion device, called an elliptic multipole wiggler, which produces elliptically polarized radiation on the axis of the magnet array. In the wiggler the electron orbit is elliptical and thus the normal of the orbital plane is rotating around the axis of the magnets. Consequently, when viewed on the axis at the end of the magnet array the radiation is elliptically polarized as in the inclined method and the intensity of radiation is multiplied by the number of poles of the wiggler. Therefore, the photon flux from this insertion device is the highest among the existing methods to derive elliptically polarized radiation over a very wide range of energy.

The wiggler is installed in the 6.5 GeV accumulation ring of the National Laboratory for High-Energy Physics and has been used as a source of circularly polarized photons to measure magnetic Compton profiles of Fe–Si [11, 12], Ni [13], Gd [14], amorphous Fe–Gd [15] and various other magnetic materials. The results have produced interesting information about the magnetic and electronic structure of these materials. However, all of these profiles are measured with a solid-state detector, mainly because the data acquisition time is greatly reduced and statistical accuracy is much higher than the case in which a crystal analyser and a position-sensitive detector are used for energy analysis of the scattered x-rays. Consequently, the momentum resolution is 0.7–0.8 atomic units (au), which is not high enough to make full use of the unique properties of the Compton profile that equations (1) and (5) suggest. To detect fine structures due to the existence of the Fermi surface and other band-structure effects, a momentum resolution of about 0.1 au is needed. In addition to the rather low resolution only the magnetic profiles were data processed and presented because rather lengthy processing of raw data is required to obtain a reliable total profile. In parallel with the development of these measurements with a low resolution but very high statistical accuracy, an attempt to establish a high-resolution Compton spectrometer that can provide a momentum resolution of 0.1 au for 60 keV incident x-rays was successfully made by Sakurai *et al* [16].

The main objective of this paper is to demonstrate that with the high-resolution spectrometer and ample circularly polarized photons delivered from the elliptic multipole wiggler, it is actually possible to measure a total profile and a magnetic Compton profile, and thus the majority-spin and minority-spin electron momentum distributions with a momentum resolution of 0.12 au, a resolution long sought after in the field of spin-dependent Compton-profile measurements.

2. Experimental procedures and data processing

A single crystal of Fe–5.8 at.% Si plate with a thickness of 0.2 mm, the normal of which is [110], was grown by the secondary recrystallization method [17]. The [100] direction of the specimen is set along the x-ray scattering vector. The specimen is clipped across the pole pieces of a C-type electromagnet. The angle between the direction of the magnetization and the scattering vector is 35°. The experimental set-ups are schematically drawn in figure 1. The photon source is the elliptic multipole wiggler. A quasidoubly-bent monochromator delivers 2×10^{11} photons of 60 keV and of degree of circular polarization about 0.6 to a specimen per second with an energy resolution of 1.5×10^{-3}. As described in detail in [16] the high-resolution spectrometer consists of a Cauchois-type crystal analyser and a sheet of imaging plate as a position-sensitive detector. To obtain the magnetic profile J_{mag} two raw profiles N_{up} and N_{down} must be recorded independently but in the same conditions. Therefore the spectrometer is modified to accommodate two sheets of imaging plate, one,

IP_{up}, for N_{up} and the other, IP_{down}, for N_{down}. After N_{up} has been recorded on IP_{up} for 10 s the x-ray beam is shut and the direction of magnetization of the specimen is reversed. Then IP_{up} is mechanically withdrawn into a radiation-shielded chamber and IP_{down} comes out from the shielded chamber and takes the position to be exposed. Then N_{down} is recorded on IP_{down} for the next 10 s. This cycle is repeated continuously for 12 h. Then the imaging plate is read out. To accumulate sufficient numbers of events the measurements lasted for 4 d.

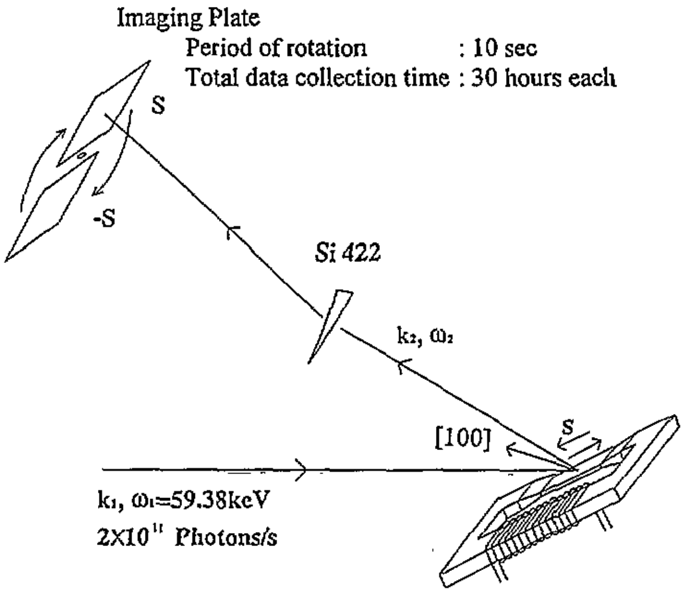

Figure 1. A schematic diagram of a high-resolution magnetic Compton-scattering experiment. The single crystal of Fe–5.8 at.% Si is clamped across the poles of an electromagnet. The scattered x-rays are energy analysed by a Cauchois-type crystal analyser and recorded on a set of imaging plates, which is rotated synchronously with the periodic reverse of the magnetic field.

In order to deduce J_{mag} care must be taken that both raw profiles, N_{up} and N_{down}, are measured in the same experimental conditions, such as the total number of incident photons and the scattering geometry. One exception is the direction of the magnetization of the specimen, which is reversed periodically. Although during the measurement it is impossible to measure the exact total number of incoming photons on the specimens, after the measurement the total number in each direction can be made the same by normalization based on the fact that the integrated intensity of the elastic peak is proportional to the total number of incident photons and is independent of the direction of magnetization. Therefore, first the measured N_{up} and N_{down} are normalized using the area under the elastic peak and then subtraction is carried out. The resultant $(N_{up} - N_{down})$, which contains only the spin-dependent term, is corrected for the spin- and energy-dependent scattering cross-section, the formula for which is given by Grotch *et al* [18], the energy-dependent efficiency of the analyser and imaging plate and absorption in the specimen. In this process we have assumed that the background noise is independent of the direction of the magnetization and the effect of multiple scattering is spin independent, and thus both disappear after the subtraction. In principle the area under the corrected $(N_{up} - N_{down})$ profile should correspond to the number of magnetic electrons in the specimen. So, instead of obtaining the absolute magnetic profile from the experiment using somewhat ambiguous geometrical factor and degree of polarization, the corrected $(N_{up} - N_{down})$ is area normalized to the number of the magnetic electrons per atom in the alloy. The number is deduced to be 2.0 from the bulk magnetization data of the alloy by Bozorth [19]. The magnetic Compton profile thus obtained, $J_{mag}(p_z)$, is shown in figure 2.

The number of events accumulated is about 8×10^5 at the Compton peak and 3×10^7 under the profile for N_{up} and N_{down}. The statistical accuracy of the data is determined by the number of photons absorbed by the imaging plate, the uniformity of the imaging-plate response and the electronic noises generated in the read-out process. Ito and Amemiya [20] studied this complex problem. The relative standard deviation as a function of photon numbers absorbed by the imaging plate is summarized in figure 4 of [20]. To determine the error bars in the present data, the number of photons in some representative channels are converted to the units of the abscissa of figure 4 of [20] and the standard deviations are read off.

To deduce the total profile $J(p_z)$ the procedure described by Shiotani *et al* [21] is closely followed. But, here, instead of using the step at the K edge, the experimental tail part, -10 au $< p_z < -8$ au and $+8$ au $< p_z < +10$ au, is fitted with a linear background, a theoretical core profile and a normalization constant. The above-mentioned normalized N_{up} and N_{down} are added to eliminate the spin-dependent contributions. Then the sum $(N_{up} + N_{down})$ is corrected for the background noise, the energy-dependent scattering cross-section, the formula of which is given by Ribberfors [22], the energy-dependent efficiency of the analyser and imaging plate, and absorption in the specimen. To determine the background noise the core-electron profile for Fe–5.8 at.% Si is calculated from the Hartree–Fock atomic core-electron profiles of Fe and Si [23]. The effect of double scattering is estimated by the Monte Carlo method programmed by Sakai [24]. The area under the corrected valence part of the $(N_{up} + N_{down})$ is normalized to 7.77, which is the effective number of valence electrons per atom in the specimen. The obtained valence electron profile, $J_{val}(p_z)$, is shown in figure 3.

Addition of $J_{mag}(p_z)$ to $J_{val}(p_z)$ gives the profile of the majority electrons, $J_{maj}(p_z)$, and subtraction of $J_{mag}(p_z)$ from $J_{val}(p_z)$ yields the profile of minority electrons, $J_{min}(p_z)$. In figure 3 $J_{maj}(p_z)$ and $J_{min}(p_z)$ are also shown.

3. Discussion

It is well known that the value of the magnetic moment per Fe atom in the Fe–Si alloy system remains the same as that of pure Fe when the Si concentration is small. Terakura [25] explained this experimental fact by calculating the electronic structure of an Si atom in ferromagnetic Fe. The calculation showed that the change of the number of majority-spin electrons within the Wigner–Seitz sphere at an Si atom was -3.08 and that of the minority-spin electrons was -0.93.

Although the band structure of pure Fe is disturbed by addition of Si, it is of interest to note that the previous low-resolution magnetic profile of Fe–5.8 at.% Si measured by Sakai *et al* [11] and Tanaka *et al* [12] has shown a very good agreement with the theoretical profile of pure Fe given by Kubo and Asano [26] using the FLAPW method. A comparison of the present high-resolution profiles and the latter theory is given in figures 2 and 3. The theory is convoluted with the experimental resolution of 0.12 au. It should be noted that in the theory the area under the total profile is 8.0, and that for the magnetic profile is 2.07. Thus

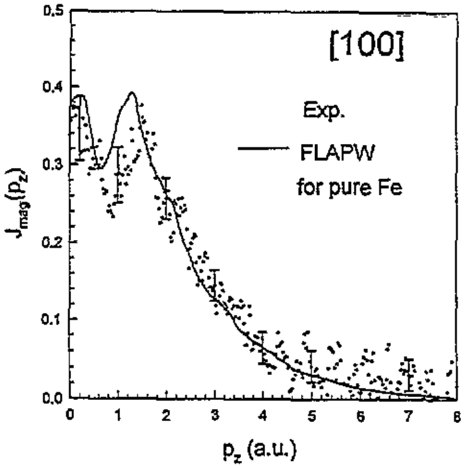

Figure 2. The magnetic Compton profiles along the [100] direction. The dots represent the experiment on Fe–5.8 at.% Si and the solid curve represents the theory for pure Fe calculated by Kubo and Asano [26]. (See the text for the normalization of the profiles.)

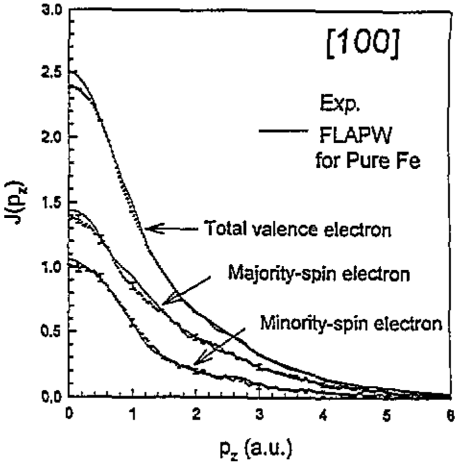

Figure 3. The total valence electron, majority-spin- and minority-spin-electron Compton profiles. The dots represents the experiments on Fe–5.8 at.% Si and the solid curves represent the theory for pure Fe. (See the text for the normalization of the profiles.)

the area under the profile of the majority-spin electrons is 5.04 and that for the minority-spin electrons is 2.97. On the other hand in the experiment the area under the total profile is 7.77 and that under the magnetic profile is 2.0. By considering the above-mentioned difference in the area under the profiles the overall shape of the total profiles shows a fairly good agreement between the theory for pure Fe and the experiment on Fe–5.8 at.% Si except that the theory has a shoulder around 1.0 au. There is little difference in the profiles of the minority-spin electrons. On the other hand in the profiles of the majority-spin electrons a difference between the theory and experiment is obvious around $p_z = 1.0$ au. According to Kubo and Asano the shoulder around $p_z = 1.0$ au in the theoretical profile originates mainly from the sixth-band contribution. There is no fifth or sixth band in the minority-spin band. The magnetic profiles show this difference in an enhanced way. According to the band-wise analysis of the magnetic profile shown in figure 2(a) of [26] the dip around $p_z = 0.7$ au is formed mainly by the first-, the third- and the sixth-band contributions. The shoulder around $p_z = 1.4$ au is mainly due to the third- and the sixth-band contributions. So, it can be said that the difference between the theory for pure Fe and the experiment on Fe–5.8 at.% Si shown in the profile of the majority-spin electrons and the magnetic profile indicates that the majority-spin band of pure Fe is more perturbed by addition of Si than the minority-spin band. This conclusion is qualitatively consistent with Terakura's theory.

Acknowledgments

The authors wish to express their thanks to Professor Kubo and Professor Asano for sending their results of FLAPW calculations for pure Fe and for their valuable discussion on the experimental results. They also thank Professor Sakai for his helpful discussion. This work has been performed with the approval of the Photon Factory Programme Advisory Committee (proposal No 90-228).

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