

Recent Advances in Compton Scattering*

L. Dobrzyński

Institute of Physics, Warsaw University Branch, Lipowa 41, 15-424 Białystok, Poland

Z. Naturforsch. **48a**, 266–272 (1993); received January 9, 1992

A review of achievements in the field of Compton spectrometry in the years that elapsed from the last Sagamore meeting (1988) is presented. Some physical problems that have either appeared as new or became clarified are described. Special emphasis is put to the results obtained by means of magnetic Compton scattering.

Key words: Compton scattering; Electron momentum distribution; Synchrotron radiation; Magnetic materials.

1. Introduction

There are so many excellent review papers describing the principles of Compton spectrometry that they may be considered almost as a bible for anyone wishing to work in this field. Here I want to quote the book edited by Williams [1], papers by Cooper [2–4] and more recent papers of Manninen [5] and Sharma [6]. The use of circularly polarized synchrotron radiation to so-called magnetic Compton scattering studies was reviewed this year by Sakai et al. [7].

In this review, recent achievements of the experimental technique are summarized and a few interesting problems in physics that have been studied recently by the Compton scattering technique are pointed out. We shall be mainly interested in studies carried out in the last three years that elapsed since the Sagamore IX meeting. Obviously, a choice of material is very subjective and does not pretend to exhaust all the problems studied. For example, we shall not deal at all with the applied research that can be carried out with the use of Compton scattering, in spite of the fact that such problems occupy people's minds (see, e.g. [8, 9]) and that their importance cannot be questioned.

2. Experimental Technique

Using essentially the idea of the high-resolution X-ray-type spectrometer as published earlier by Lou-

pias and Petiau [10], the Japanese team [11, 12] managed to install and develop a spectrometer that exhibits a momentum resolution of $0.083 p_0$ ($p_0 = 1$ a.u. of momentum $= \hbar/a_0 = 1.99289 \cdot 10^{-24}$ kg m/sec). Such a resolution is comparable with the one known from positron annihilation experiments and enables one to study details of the Fermi surface. The beautiful results obtained for vanadium single crystals [13, 14] serve as the best example. Moreover, as it will be described later, such a resolution allows for a detailed comparison of the electron momentum distributions seen by photons and by positrons. One should also point out that the spectrometer used by the Japanese group can simultaneously measure the Compton profiles along four directions.

The highly intense beam (with a flux at 60 keV of $2.5 \cdot 10^{12}$ photons/cm²/s and an energy resolution $\Delta E/E$ of 10^{-3}) with a circular polarization of about 60% was obtained at KEK, Japan, from the multipole elliptical wiggler [15], so very efficient studies of magnetic Compton scattering became possible as well. The time required for a measurement has been further reduced by using either an imaging plate or a segmented Ge detector with 13 elements. Last but not least, the photon flux has been raised a few times with the aid of a highly sophisticated construction of a quasi-doubly-bent Si-crystal monochromator [16] that is able to focus the beam both in the vertical and horizontal directions.

As we mentioned, the achievement of high resolution in Compton spectrometry allows one to solve the old problem of the difference between the momentum distribution seen by photons and the one seen by positrons. Obviously, inherent differences are expected owing to the different interactions of the probes with matter. These differences have been listed and

* Presented at the Sagamore X Conference on Charge, Spin, and Momentum Densities, Konstanz, Fed. Rep. of Germany, September 1–7, 1991.

Reprint requests to Prof. Dr. Ludwik Dobrzyński, Institute of Physics, Warsaw University Branch, Lipowa 41, 15-424 Białystok, Poland.

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discussed, e.g., in a paper by Das [17]. Such a comparison was made for a metal (Al) as well as for a semiconductor (Si) (see [18]), and found understandable in the light of general expectations.

A particularly interesting achievement, which followed from very early dreams of physicists doing Compton-profile studies, was gained in Hamburg, where a (γ , $e\gamma$)-spectrometer was installed and tested successfully [19, 20]. In addition to a conventional measurement of the scattered γ -ray, the recoil electrons were detected in coincidence with the γ -particles, thus leading to a complete knowledge of the initial momentum of the target electron. The authors proved that for a sufficiently tight collimation and appropriate angular resolution one can identify an electron-photon signal undistorted by multiple scattering of the electron. The multiple scattering of electrons, which always presented a problem in Compton profile studies of solids, is diminished only when the sample is sufficiently thin. In fact, using computer simulations (the authors developed a special code that takes account of this multiple scattering) it was shown that the contribution of unscattered ejected electrons to the total scattering is saturated already in a layer of about 0.1 μm thickness. Initially the authors used a ^{54}Cr source ($E_\gamma = 320.08$ keV), so the overall coincidence signal was rather weak [19]. Nevertheless, they showed that this technique should be efficient in combination with a strong synchrotron radiation source. Indeed, results obtained at the DORIS storage ring [20] proved that one can await many novel results, especially in studies of thin films.

The formidable problem of the multiple scattering of electrons in the well-known (e , $2e$)-scattering technique may be largely diminished when using highly energetic protons instead of electrons. This fact became a basis of the experiment performed by Spies and Bell [21] with the use of 21 MeV protons impinging on thin (less than 0.1 μm thick) foils of silver and gold. In an experiment like that, the spectral line shape of the ejected electrons (i.e. number of recoiled electrons vs. their energies) instead of the “classical” Compton profile, i.e. the line shape of the scattered photons, is observed. The use of so extremely thin foils intends to minimize the multiple scattering of electrons in the solid under study. The reduction of the sample volume, which would be felt severely by a low flux of detected particles in any more conventional experiment, is compensated here by a much larger scattering cross-section. Indeed, the leading term in

the scattering cross-section in the case of the (p , ep) experiment describes the Rutherford scattering, which is several orders of magnitude larger than the Klein–Nishina cross-section. In addition, the available fluxes of protons are another 1000 times higher than the photon fluxes from γ -sources. In fact, the problems with using such a technique may arise from a radiation damage and local heating of the sample, so the low sample volume presents the least important problem. Therefore, this technique should be particularly useful in studies of very thin, yet self-supporting films. Besides, it should be advantageous in studies of heavy elements, for which the photoabsorption would present a problem difficult to overcome when using high-energy γ -sources.

Ending this section it is worth mentioning that the reasoning used in the description of Compton scattering may be equally successfully used also in epithermal neutron scattering. Indeed, neutrons with energies of a few electron volts can experience inelastic scattering in a similar way as photons do, but this time the scattering unit is not an electron, but a whole atom or, more precisely, the nucleus. By observing the line shape of the scattered neutrons one can get information on the momentum distribution of an atom in a solid. Such a type of scattering was called deep inelastic neutron scattering, and the interested reader may find its description, e.g., in a paper by Mayers and Holt [22 a] as well as in the papers by Evans et al. [22 b] and Simmons [22 c].

3. Some Selected Results

3.1 Validity of the Impulse Approximation

The very basis of interpretation of Compton profiles relies on the so-called impulse approximation. It is assumed that the energy transferred to the electron is much larger than its binding energy E_B . Having obtained high recoil energy, E_r , the ejected electron forgets its initial state and may be considered as a plane wave. This can work well when $E_r \gg E_B$. However, if both energies are comparable, then – according to Platzman and Tzoar [23] – the error made in assigning the observed intensity to the Compton profile expected within the impulse approximation must go roughly as $(E_B/E_r)^2$. The situation close to the electron binding energy was studied carefully by Issolah et al. [24] for the 1s electrons in carbon. They considered the result of applying the so-called “hydrogenic approxi-

mation" in which the recoiled electron is considered as moving in a Coulombic potential of Z^*/r , where Z^* is an effective nuclear charge. They tried also to improve calculations by going to a more self-consistent approach with improved potential for the final state. The result of this analysis was twofold. Firstly, they state that close to the binding energy (300 eV in this case) a shift of the Compton peak should take place. In addition, the peak value of the profile, $J(0)$, should change as well. From the quantitative point of view both changes are dependent on the approximation used. In fact, Namikawa and Hosoya [25] claimed to observe a shift of the Compton profile as well as a Raman-type peak close to the K-edge of iron and copper. Their findings, however, were put in doubt by Manninen [26], who suspected that this result could be due to false coincidences overlooked in [25]. Much earlier, in 1979, Pattison and Schneider [27] have found that the impulse approximation works well for Au and Pb. At the K-edge only a step in the profile was observed, and the size of this step totally corresponded to a simple subtraction of the contribution of two 1s electrons from the ensemble of electrons taking part in the scattering. In principle, what should matter is the value of ka , where k is the wavevector of the recoil electron and a is a typical dimension of the corresponding electron orbit. Close to $ka = 1$ we can expect distortions of the Compton profile. In the experiments carried out by Manninen et al. on silver [28] and on Cu and Zr [29], the energies of the radiation used were 141 keV and 59.54 keV, respectively. In the experiment of [29] the scattered photon was additionally detected in coincidence with the X-ray fluorescence radiation in order to separate the contribution of the K-shell itself to the scattering. These measurements have shown, however, that the Compton profile is undistorted in spite of the fact that, e.g., in the case of Zr the value of ka was as low as 0.67. Similarly to the result of Pattison and Schneider [27], the size of the step seen at the K-edge was fully explainable by the removal of two 1s-electrons from the scattering. This rather unexpected situation is difficult to explain, the more as it seems that such phenomena like Compton-profile shift and a change of its maximum value is observed when the energy transfer takes place close to the L-edge [30].

3.2 Correlation Effects in Transition Metals

Since the extensive studies of the Compton profiles of copper by Bauer and Schneider [31, 32] it is known

that band theories based on a single-electron approximation overestimate the scale of anisotropies, i.e. the differences between the Compton profiles observed along different crystallographic directions. A similar effect was observed, e.g., in Ni [33]. The origin of this effect is sought in the electron–electron correlations, which are neglected in single-electron approximations. Indeed, the so-called Lam-Platzman correction [34], which can correct the results obtained within the local-density approximation, turned out to be rather efficient although its main disadvantage consisted in the fact that it was isotropic in momentum space. The first successful attempt to calculate an anisotropic correction for electron–electron correlations was done for chromium and vanadium [35, 36] and showed that in principle we should now understand full directional dependences of the Compton profiles of transition metals. Obviously, an improvement of the momentum resolution in the experiment can reveal new features; so this last statement should be taken with some reservation.

3.3 Bond Properties in Semiconductors

Valence-electron Compton profiles of silicon have been a subject of detailed studies of Hansen, Pattison and Schneider [37], who did a 3-dimensional reconstruction of the electron momentum distribution. Their results have been obtained with a gold source, which is known to give a resolution in momentum space of the order of $0.4 p_0$. Recently other high-quality results obtained with synchrotron radiation of 29.5 keV and a momentum resolution $0.084 p_0$ have been published by Sakai et al. [38]. It was clearly shown that pseudopotential calculations, which are lacking a proper treatment of the core-orthogonalization problem, are inferior with respect to the tight-binding model. At the same time the directional properties of the electron momentum distribution turned out to require still some revision.

The deficiencies of pseudopotential calculations were also seen in studies of the Compton profiles of one of the most popular III–V semiconductors, GaAs. Two experiments were recently performed [39, 40]. In both experiments the directional properties of the profiles have been studied, and in particular the reciprocal form factor $B(s)$ was of interest. In order to convince themselves that the results obtained do not suffer from any appreciable systematic errors owing to the source, in the experiment carried out by the

mixed English-Japanese team [39] two sources, viz. ^{241}Am and ^{198}Au , were used. The experiments show that the pseudopotential theory predicts both the scale and oscillation period of the directional dependence of the Compton profile. Nevertheless, the theoretical values of $J(0)$ are by about 5% higher than found in the experiment. This is just due to the neglect of the core-orthogonalization terms, which give rise to high-momentum components in the profile. Because this effect is isotropic, it does not influence the directional properties, and neither is of importance for $B(s)$ at large distances s .

In comparison with Ge, the scale of the anisotropy is decreased. This is marking an increasing ionicity of the bonds [41]. In this respect, the studies of the anisotropies of Compton profiles are supplementing very efficiently the studies of charge density distributions.

From the methodological point of view we note that the studies of [39] show a positive feature at $s \cong 0.8 a_0$ in the difference of $B(s)$ along [111] and [100] directions, whereas a negative dip was observed in the studies of [40]. This contradiction may have a source in a noise introduced by the data-handling procedures. However, because the disagreement with theoretical results is particularly severe in this region, we feel that the data should be reanalysed.

3.4 Magnetic Compton Profiles of Elemental Ferromagnets

Probably the most exciting latest achievements are due to extraction of the magnetic interaction of photons with matter and particularly its contribution to the Compton effect. We should like to quote here the efforts of the English and Japanese groups. The first was using the inclined-orbit method of obtaining the required circularly polarised beam of photons [42, 43]. The Japanese group used an elliptical multipole wiggler, a quasi-doubly bent Si-monochromator and a segmented Ge solid-state detector [7, 16, 44]. Both groups studied nickel and iron, and both presented data from which a deficiency of "magnetic" electrons with low momenta follows as a clear-cut phenomenon. This deficiency or dip in the magnetic Compton profiles is interpreted as being due to the negative polarization of conduction electrons.

A concept of the negative polarization of the conduction band was expressed in the early sixties and

was based on the magnetic form factor measurements of iron [45], cobalt [46] and nickel [47] carried out by means of neutron diffraction. The conclusion of all these measurements was that the spin densities observed consist of a 3d part peaking at the atomic positions and the uniform background of negative polarization owing to the conduction electrons. This last conclusion concerning the uniform background was put in doubt, e.g., in [48]. The recent Compton results now show that indeed the polarization of conduction electrons is almost twice as small as claimed by early neutron diffraction results, so the latter must have contained a contribution coming presumably from the d-d exchange polarization, which should also lead to the negative moments far away from the atoms.

The correctness of the interpretation of dips observed in the magnetic Compton profiles as due to the conduction-electron polarization is strengthened by the studies on gadolinium [49], in which, instead of a dip, an extra positive contribution to the magnetic profile at low momenta was observed. Moreover, if one assumed that the magnetic moment of f-electrons visible in the relative wide magnetic profile extending to about $10 p_0$ is $7 \mu_B$ (as it follows from Hund's rule) then the area under the extra profile observed below about $2 p_0$ turned out to be $(0.53 \pm 0.08) \mu_B$. Therefore, one could easily explain the well-known magnetic moment of the gadolinium atom $(7.63 \pm 0.02) \mu_B$ as composed of a localized 4f contribution of $7 \mu_B$ and the conduction-electron contribution of about $0.6 \mu_B$.

Very recent results on the magnetic Compton scattering from alloys were reviewed by Sakai [50].

3.5 What About the Orbital Magnetic Moment?

In accordance with the elementary theory of magnetic photon scattering [51], the orbital magnetic moment in Compton scattering should be clearly distinguished from the spin moment contribution as in the case of elastic scattering. The experiments along these lines have been performed by the English team [43, 52] who measured the fractional magnetic effect in the total Compton scattering as a function of the sample orientation with respect to the incoming beam of photons. Such a dependence should be more or less linear and, most importantly, show a change of sign of the magnetic effect. The crossing point of the line with the abscissa should characterize the gyromagnetic ratio.

The first experiments have been carried out on Fe and Co, and two contradicting results have been obtained [53]: after the first impression of having an agreement with the expectations, the experiment repeated by the English team in Japan showed no difference between the results obtained for the two metals. What was, however, much more surprising was that for HoFe_2 , which has been chosen as a good candidate for testing theory because the ratio of the orbital to the spin-moment was expected to be higher than 3 at room temperature, the experimentally found crossing point was not different from the one found for iron itself. These experiments seem to indicate that in the Compton process, in which a relatively large energy transfer takes place, the orbital magnetic moment does not contribute to the scattering.

Two remarks can be made here. Obviously, such a kind of experiment can be hardly recommended if the only purpose is the measurement of gyromagnetic ratios: it is too expensive and time consuming. At the moment the most important question is correctness of the theory by Lovesey [51]. Because of the finding described in [53], one should consider whether the experimental results, especially on HoFe_2 , were not subjected to a systematical experimental error owing, e.g., to the lack of magnetic saturation. Because the experiments were carried out by a technique in which a relatively low magnetizing field of about 0.5 T was switched back and forth, one can worry whether the orbital moment did follow the direction of the field. However, if such doubts could be rejected, the theory of magnetic Compton scattering would need to be severely revised. If for some reasons Compton scattering were not sensitive to the orbital magnetic moment, its comparison with the results of magnetic neutron scattering, which is sensitive to the total magnetic moment, would be of great interest.

4. Concluding Remarks

First of all the author wishes to apologize for not citing all the papers which appeared from 1988. It does not mean that they were found less relevant: this review reflects mainly the very subjective interest of the author.

The development of synchrotrons, especially the European Synchrotron Radiation Facility in Grenoble, which relatively soon will be operational, gives hopes that the momentum resolution in Compton spectrometry will be substantially improved also at

higher photon energies of, say, few hundreds of keV. On the other hand, tremendous intensities obtained from the sources, enhanced at the sample position by focusing monochromators, enable one to study even small single crystals, so quite unique materials can be expected to be studied. The use of sophisticated insertion devices like, e.g., the multipole helical wiggler, is adding a new dimension to the problem, viz. production of highly intense circularly polarized beams, which can be used for efficient measurements of magnetic Compton scattering. Speaking about the experimental technique, we should also like to point out once again the first attempts to measure the truly 3-dimensional electron momentum distribution by means of the $(\gamma, e\gamma)$ coincidence technique [19, 20] and the use of highly energetic protons [21] as particles ejecting electrons. Both techniques are expected to deliver many interesting results, particularly concerning thin films.

The development of experimental methods is accompanied by a tremendous increase of power of computational techniques. The best example was presented during this meeting by Prof. A. Bansil, who showed that accurate band-structure calculations can be performed for substances as complicated as high- T_C superconductors. We quoted also an example of theoretical calculations of the direction-dependent electron-electron correlation effect on the Compton profile of transition metals [36]. It seems that also the influence of the core-orthogonalization terms on a Compton profile calculated by means of the pseudopotential theory is a well understood problem. In the light of these successes of theory the problem of the validity of the impulse approximation in the interpretation of the experiments seems to be anachronistic. Nevertheless, the fact that this approximation is sufficient in the description of the Compton profile at a K-edge remains still an unresolved puzzle.

The studies of magnetic Compton scattering deliver for the first time three-dimensional pictures of the momentum distributions of the "magnetic" electrons and are hoped to clarify the rôle of conduction electrons in magnetism. Indeed, this is sometimes a key point for testing any band-theoretical calculations of magnetism of metals, and Compton scattering plays here a unique rôle. The great competitor – the magnetic neutron diffraction – brings results concerned mostly with d- or f-electrons, and a contribution of conduction electrons is hardly seen by this method directly. On the other hand, neutrons are sensitive

enough to see the contribution of the orbital magnetic moment to the scattering. This is because this moment leads to a different angular dependence of the scattering of neutrons than the spin moment does. At present, the rôle of the orbital magnetic moment in the Compton scattering of photons is unclear. However, if the present theory [51] is correct, it potentially offers a clear-cut distinction between the two types of magnetic scattering. Therefore, one should impatiently await further experiments along these lines, planned by the English team in collaboration with the Japanese one*.

Acknowledgements

I should like to express my thanks to the Organising Committee for inviting me to Konstanz and offering a possibility of giving this talk. The Deutscher Akademischer Austauschdienst (DAAD) has been very helpful with its financial support. I have benefited a lot from discussions with Professors Malcolm J. Cooper and Seppo Manninen as well as Dr. Eugeniusz Żukowski, whom I owe my thanks. Thanks are also due to my colleague, Mr. Andrzej Andrejczuk, for many interesting talks. The work was partly sponsored through the grant No. 2 2350 91 02 of the Committee of Scientific Research in Poland.

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* *Note added in proof:* The results of the experiment just mentioned above were recently published [M. J. Cooper, E. Żukowski, S. P. Collins, D. N. Timms, F. Itoh, H. Sakurai, *J. Phys.: Condensed Matter* **4**, L399 (1992)]. They clearly show that the orbital magnetization is hardly seen in the magnetic Compton scattering.