INELASTIC SCATTERING AT HIGH PHOTON ENERGIES

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Abstract

Determination of electron momentum densities by Compton scattering where the scattered photon and the recoil electron are detected in coincidence, and determination of spin densities from the magnetic contribution to Compton profiles are reviewed. The present day limitations of the methods arise from insufficient photon flux at high energies, which necessitates the use of Ge detectors for the energy analysis of the scattered radiation. A helical small-gap undulator for the TESLA facility is described. The energy of the fundamental line is 200 keV, and with bent crystal and multilayer optics 10^{13} circularly polarized photons can be focused on a spot of a few microns in diameter. This makes possible the use of an efficient crystal spectrometer, and count rates of 10^4 to 10^5 and momentum resolution of 0.05 a.u. can be achieved. The 3-level time structure – 80 fsec bunches separated by 93 nsec in 1 msec train with 5 Hz repetition rate – provides unique possibilities for gating and use of pulsed lasers and high-field magnets.

1. Introduction

The present discussion concerns only of a sub-set of inelastic scattering, namely scattering of high-energy photons from electrons. This is called deep inelastic or Compton scattering, where a bound electron is scattered to continuum states, and the energy of the scattered photon is less than the initial photon energy. The conservation laws are

$$\begin{aligned} \varepsilon_2 &= \varepsilon_1 + \omega_1 - \omega_2 = \varepsilon_1 - \omega \\ \mathbf{p}_2 &= \mathbf{p}_1 + \mathbf{q}_1 - \mathbf{q}_2 = \mathbf{p}_1 - \mathbf{q} \end{aligned} \tag{1}$$

where ε_1 (<0) is the electron binding energy, ω_1 the initial photon energy, ω_2 that after scattering, and ε_2 the energy of the recoil electron. The corresponding momenta are indicated by $\mathbf{p}_1$ and $\mathbf{p}_2$ for the electron, and by $\mathbf{q}_1$ and $\mathbf{q}_2$ for the photon. In a "complete" scattering experiment all these quantities are observed for a given event, i.e. by a coincidence measurement. Such a measurement would yield the spectral electron momentum density (SEMD),

$$\rho_j(\mathbf{p}_1,\varepsilon_1) = \rho_j(\mathbf{p}_1)\, \delta[\varepsilon_1 - \varepsilon_j(\mathbf{p}_1)] \,, \qquad (2)$$

where $\rho_j(\mathbf{p}_1)$ is the EMD of the jth band, and $\varepsilon = \varepsilon_j(\mathbf{p}_1)$ is the dispersion relation. In the extended zone scheme $\mathbf{p}_1 = \mathbf{k}_1 + \mathbf{G}$, where $\mathbf{k}_1$ is the crystal momentum, and $\mathbf{G}$ indicates the zone.

The present day (γ, eγ) or (X,eX) experiments are incomplete in many respects. The energy of the scattered photon is observed using a Ge detector, which has an energy resolution of 0.3% at best, and the energy resolution of the electron detector is not sufficient for separation of the different bands. Still there have been substantial advances in the last few years, and the results of those experiments are reviewed in the following [1- 4].

2. Electron momentum density and Compton scattering

In the following we consider only experiments where the EMD is summed over all bands and binding energies,

$$\rho(\mathbf{p}_1) = \sum \rho_j(\mathbf{p}_1) \,. \qquad (3)$$

The triple-differential scattering cross section is proportional to the EMD,

$$(d^3\sigma/d\omega_2 d\Omega_\gamma d\Omega_e) = p_2(\omega_1/\omega_2)(d\sigma/d\Omega_\gamma)_{KN}\, \rho(\mathbf{p}_1) \,. \qquad (4)$$

The subscript γ indicates the scattered photon, subscript e the recoil electron, and KN the Klein-Nishina scattering cross section. The simple relationship between the cross section and the EMD is based on the so-called "impulse approximation", which assumes that the potential where the electron is moving does not change during the scattering event. Calculations for hydrogen-like atoms indicate that the momentum transfer should be an order of magnitude larger that the initial electron momentum [5]. This condition is fulfilled in all the cases that are discussed in the following.

The energy of the photon scattered from an electron at rest ($\mathbf{p}_1$= 0) is

$$\omega_{2,0} = \omega_1/[1 + (\omega_1/mc^2)(1 - \cos \Theta)] \,, \qquad (5)$$

where Θ is the scattering angle. The angle α between $\mathbf{q}_1$ and the direction of the recoil electron ($\mathbf{p}_2$= - $\mathbf{q}_0$) is given by

$$\cot \alpha = (1 + \omega_1/mc^2) \tan (\Theta/2) . \qquad (6)$$

The initial electron momentum $\mathbf{p}_1$ causes a Doppler shift in ω_2, and the direction of the recoil electron deviates from α. The spectrum of the scattered radiation is analyzed by an energy-dispersive detector or spectrometer, and this yields the component of the electron momentum in the direction of the scattering vector $\mathbf{q}$,

$$p_{1z} = mc\,[r - 1 + (\omega_2/mc^2)(1 - \cos \Theta)]/[1 + r^2 - 2r \cos \Theta]^{1/2} , \qquad (7)$$

where $r = \omega_2/\omega_1$. In atomic units, mc = 137.

The components p_{1x} and p_{1y} of the initial electron momentum are obtained when the direction of the recoil electron is observed in coincidence with the scattered photon. However, multiple scattering distorts $\mathbf{p}_2$, and the unscattered electrons come from the top layer corresponding to the last mean free path. This is only a few tens on nanometers at recoil energies of 50 keV. Extensive Monte Carlo calculations demonstrate that the scattered electrons contribute to an almost flat background, and sharp features in the EMD, which arise from abrupt changes in the occupation number density, are clearly visible in the contribution of unscattered electrons [6]. This gives confidence that the EMD of valence electrons can be extracted from the measured triple-differential scattering cross section.

The experimental set-up for a $(\gamma,e\gamma)$ coincidence measurement has been described earlier [2]. The photon detector is placed near the backscattering direction, and the center of the electron detector is opposite to $\mathbf{q}_0$. Recently at the ESRF the single-element Ge detector was replaced by a 13-element detector, which enhances the coincidence rate by an order of magnitude, when the data from different detector elements are combined. Even then the coincidence rate is low, of the order of a few tens of events per second. Typically, the incident photon energy is 150 keV, so that $\omega_{2,0}$ is about 100 keV. The electron detector is a 16x16 element array of Si diodes, which has a granularity of 0.14 a.u. in p_{1x} and 0.28 a.u. in p_{1y}. The energy resolution of the Ge detector corresponds to a momentum resolution of 0.4 a.u. in p_{1z}, and the other uncertainties include the bandwidth of the primary beam, solid angle of the Ge detector, and the size of the beam on the sample. When the momentum errors are combined and the energy conservation condition is imposed the variance vector of $\mathbf{p}_2$ is found to be (0.18;0.43;0.20) a.u.

The advantages of Compton scattering in Fermi surface studies are evident in the cases where methods like deHaas-vanAlphen cannot be applied due to imperfections or lack of translational symmetry in the crystal, like in alloys and quasicrystals. The SEMD can be determined by an (e,2e) experiment, which is similar to a $(\gamma,e\gamma)$ experiment, except that the projectile is an electron instead of a photon. The cross section is high, which allows the use of electron spectrometers and determination of the dispersion relation for individual bands. However, multiple scattering is a serious problem in (e,2e) scattering, because it concerns both the incident and ejected electrons [7]. The EMD can be determined with good resolution by a positron annihilation experiment, where the angular correlation of the annihilation quanta is measured (ACAR). The limitation of this method is that the EMD is weighed by the positron wave function, so that certain features are emphasized and some others suppressed. The results of ACAR and (γ,eγ) experiments on graphite and fullerene have been compared recently [3]. Positron annihilation occurs preferentially with π-electrons, and because of the p-character of these electrons, the EMD has an apparent double-hump shape in the $p_{1x}p_{1y}$-plane (p_{1z} is along the c-axis). This artifact is not seen in the EMD determined from the (γ,eγ) experiment, because in Compton scattering all electrons are on the same footing. A comparison between graphite and fullerene indicates that the electrons are more delocalized in the spherical shell-like structure of the fullerene than in the layered structure of graphite.

The above results show that the (γ,eγ) method has a great potential in studies of electronic ground state in solids, and in many ways it is a unique spectroscopy. On the other hand, the present-day limitations are obvious: low count rates and insufficient momentum resolution. Radiation from a very long undulator at a high-energy linear accelerator may remove these limitations, as discussed in the last section of this paper.

3. Magnetic Compton scattering

Magnetic Compton scattering is a new method to study magnetization distributions in ferro- and ferrimagnets. The electromagnetic field interacts with the magnetic moment of the electron, and it has been shown both theoretically and experimentally that the small change in the scattering cross section is due to the spin moment only. On the other hand, in neutron scattering the contribution of the total magnetic moment is observed, so that combination of these results with those from x-ray scattering makes it possible to separate and fully describe the magnetization distributions.

Most of the recent developments in magnetic Compton scattering have taken place at the High Energy Beamline of the ESRF, where sufficient monochromatic flux of circularly polarized radiation is available at energies of few hundred keV. Theory of inelastic magnetic scattering has been reviewed recently, and the cross section was verified up to 1 MeV [8]. In addition to the Klein-Nishina cross section there is a term of the form,

$$(d\sigma/d\Omega)_M = (r_e^2/2)(q_2/q_1)^2 \, [(\cos\Theta - 1)\, P_c\, \mathbf{S}.(\mathbf{q}_1\cos\Theta + \mathbf{q}_2)/mc], \tag{8}$$

where r_e is the classical electron radius, P_c the degree of circular polarization, and **S** the Stokes parameter for the spin polarization of the electron. It is seen that the sign of the magnetic contribution to the scattering cross section can be changed by reversing P_c or **S**. The magnetic Compton profile due to uncompensated electron spins is defined analogous to the Compton profile J_C due to charge scattering,

$$J_M(p_{1z}) = \iint [\rho^{\uparrow}(\mathbf{p}_1) - \rho^{\downarrow}(\mathbf{p}_1)]\, dp_{1x}dp_{1y} \;, \tag{9}$$

i.e. as the projection of the spin-EMD on the scattering vector. The integral over J_M is normalized to the number of unpaired electrons of the scattering unit (atom, molecule, unit cell), and this in turn is scaled against the integral over J_C, which is the total number of electrons in the scattering unit. The magnetic Compton profile is a small fraction of the total profile, a few percent at most, but nevertheless, it provides detailed information about the distribution of spin magnetization, particularly when profiles corresponding to many crystallographic directions are measured.

Almost all of the magnetic Compton profiles have been measured using an energy-dispersive Ge detector. The momentum resolution is about 0.35 a.u. under ideal conditions, and this sets an upper limit for the count rate that can be handled without degrading resolution. The total count rate is boosted by an order of magnitude with the use of the 13-element detector, so that magnetic effects can be studied on 0.1% level. The momentum resolution is sufficient for studies where magnetization is due to local moments corresponding to different atomic orbitals. The contributions of individual atoms can be separated, as demonstrated in several cases [9]. However, it is evident that the scope of magnetic Compton scattering studies can be widened enormously, if the resolution can be improved to reveal band structures, and fast detectors can be used.

4. Compton scattering with high energy undulator radiation

It is obvious that a highly collimated and intense undulator beam in the energy regime of a few hundred keV will open up many new applications for magnetic and coincidence Compton scattering. Discussion about these possibilities is based on published calculations of the brilliance of the SASE-FEL sources. It seems likely that the SASE process does not work for x-ray energies higher than a few tens of keV, so that the question becomes that of optimizing a normal undulator source for the particle energies and beam parameters available at high energy linear accelerators.

For most inelastic scattering experiments the relevant quantity is the spectral flux on the sample, so that integration over the source size and beam divergence is needed. The common feature of the proposed linear colliders is that the average beam current is much less than in the circular accelerators built for production of synchrotron radiation. For instance, at TESLA the average current is about 100 microamperes, i.e. three orders of magnitude less than at the ESRF or other 3rd generation sources. This loss in spectral brightness can be compensated at high photon energies when the fundamental undulator energy is at 200 keV, say. Even more important is the availability of intense circularly polarized radiation at high photon energies. So far, the sources of circularly polarized photons have been asymmetrical or elliptical wigglers, and phase-plates that split a linearly polarized beam in two components with 90 degree phase-shift. None of these solutions is very efficient at very high photon energies. It is also important that there are 3 very different time scales in the bunch structure of the high-energy linear colliders, which will allow different gating in the scattering experiments.

A specific undulator construction is studied to give a concrete idea of the prospects of Compton scattering studies at 4th generation synchrotron radiation laboratories. The design parameters are based on the bunch structure, emittance, and electron energy given in the design report of the TESLA facility [10]. A few undulators for production of high-energy radiation are described in that report, but here we present results for an optimized helical undulator, where the fundamental energy is 200 keV. The design is based on a small gap and undulator period, with values that are still feasible technically. The undulator parameters and calculated performance are summarized in Table 1. For comparison, brightness and flux in the central cone are given for the undulator at the 12 GeV storage ring PETRA. It is seen that the flux from the TESLA source is less than that from the PETRA undulator, but there are other features that make the present suggestion very attractive.

Within the central cone, which is actually a ring in the case of a helical undulator, the beam is fully circularly polarized under ideal conditions. For the undulator specified in Table 1, the diameter of the cone is about 0.4 mm at 100 m from the source for the fundamental energy of 200 keV, when the emittance effects are ignored, and the total flux is 1.87×10^{13} ph/sec/0.1% BW for the average current of 0.1 mA. The total power of the beam is 1.0 kW, but only a small fraction is absorbed in any optical element, because the dominant interaction is Compton scattering. For instance, the energy absorption coefficient for Si is 0.08/cm for energies between 100 keV and 1 MeV, so that only 8 W is absorbed in a 1 mm thick monochromator crystal. It has been demonstrated that efficient monochromators of optimized bandwidths can be realized by bent Si crystals [11], and in the present case 10^{13} photons in a 0.1% band can be focused on a line that is a few microns wide. Focusing in the other direction can be effected by a bent multilayer.

The beam described above can be used for probing local properties of materials, e.g. magnetization due to electron spins in layered structures. At the same time, the small active volume of the sample makes it possible to use high-resolution crystal spectrometers based on the Rowland circle geometry, e.g. a scanning spectrometer with a Laue-type analyzer crystal. Under ideal conditions, a momentum resolution better than 0.05 a.u. is achieved, and from a "thick" sample a typical count rate is 10^4 to 10^5 cps at the Compton peak. Fast and efficient scintillation detectors can be used, which removes the count rate limitations of Ge detectors. This opens a way to study the band structure of electron spin distributions.

The 3-level time structure of the TESLA facility offers unique possibilities for scattering experiments. First, the 93 nsec interval between the 80 fsec bunches is long enough for gating the electron and photon detectors. This makes coincidence measurements very simple. It can be extrapolated from the present-day experiments that the count rate from a layer corresponding to the free mean path of the recoil electron is about 10^2 cps. There are about 10^5 bunches/sec, so that the scattered photon and electron from a given bunch come from the same event with high probability. The coincidence rate may be too low to allow the use of a high-resolution crystal spectrometer, but the Ge detector can be replaced by a bolometer, which is well suited for measuring low count rates and has an energy resolution better than 0.1% . Second, scattering from molecules at excited states is possible at least in principle, when laser pulses comparable with the 80 fsec bunch duration are focused on the sample. Third, the slow repetition rate of 5 Hz makes it possible to use a pulsed magnet to deliver a magnetic field of several tens of Teslas during the 1 msec long bunch train. This opens the way for studies of new classes of magnetic materials by Compton scattering, particularly when small volumes can be probed.

I summary, undulator radiation with the fundamental energy in the 200 keV regime offers new possibilities for inelastic scattering studies. The flux from a TESLA undulator is comparable with that from the PETRA undulator, but the brightness is three orders of magnitude larger, and the useful spectral range begins at energies where the spectrum at PETRA is falling off rapidly. These properties make possible the use of optimal x-ray optical elements, so that very a high flux density at the sample position is achieved. Probably the most important feature of the TESLA source is the 3-level time structure of the beam, which allows probing of short-living excitations on one side and application of relatively slow pulsed fields on the other. The present article discusses only the Compton scattering experiments, but it is evident that other fields of high energy x-ray scattering, such as nuclear scattering, will become possible when undulator radiation is available at energies reaching several MeV.

Table 1: Parameters and properties of a helical undulator designed for the TESLA Facility. For comparison, flux and brightness values for the PETRA undulator are given (Hasylab Annual Report, 1995).

	TESLA	PETRA	
Undulator type	helical	planar	
Length (m) – period (mm)	30 - 30		
Gap (mm)	8	16	
Field (T) - K_x/K_y	0.51 – 1.5/1.5		
Electron energy (GeV)	45.4	12	
Current (mA)	0.1	60	
Flux at 200 keV	1.87×10^{13}	3×10^{13}	ph/sec/0.1% BW
Brightness	1×10^{19}	1×10^{16}	ph/sec/mrad^2/0.1% BW
Total power (kW)	1.0	7.0	

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