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Need for further inelastic scattering measurements at x-ray energies

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Abstract

With recent improvements in both theory and experiment for scattering of x-rays from atoms, it is possible now to make a more quantitative comparison, and see whether agreement is being obtained within the much more stringent limits set by the present calculations and measurements. Comparing with present theory, measured whole atom Compton scattering cross sections in the photon energy range 11 to 40 keV using synchrotron x-ray sources demonstrate that a dramatic improvement in the precision of scattering measurements has been achieved. However, circumstances are also identified in which further experimental data is needed in order to test the adequacy of present theoretical approaches.

Keywords: Compton scattering; incoherent scattering factor; x-rays

1. Introduction

Inelastic x-ray scattering, discovered by A.H. Compton (1923), commonly known as Compton scattering, has been a subject of continued investigations since its discovery, both in its own right as one of the fundamental processes of photon-atom interaction and also due to its application in studying electronic properties in condensed matter. Compton scattering was originally derived for free electrons, although the scattering we usually observe is from electrons bound to the atom. Compton scattering from bound electrons is the inelastic scattering of photons by atoms in which an electron is ejected from the atom. In the case of scattering from free electrons, the outgoing photon energy is determined uniquely by its scattering angle, while scattering from bound electrons is characterized by outgoing photons having a spectral distribution at any given scattering angle.

Recent high-precision scattering experiments (accurate to the level of 1 to 2%) performed using synchrotron sources, coupled with advances in theoretical calculations, give us the opportunity to make a more quantitative comparison between theory and experiment. All theoretical calculations involve various assumptions and approximations,

while experimental results are subject to experimental uncertainties. The experimental accuracy in recent years has reached a level such that the accuracy of the underlying assumptions used in theoretical calculations can be examined. With this in mind, we here compare the experimental results with theoretical predictions, to see whether agreement is being obtained within the stringent limits in accuracy of the calculations and measurements.

Recent scattering experiments performed (Jung et al., 1998; Young et al., 2001) on the Advanced Photon Source (APS) at Argonne National Laboratory for helium and neon at x-ray energies in the range 4.39-22 keV opened up a new discussion on the adequacy of different theoretical calculations. Carney and others (Carney et al., 2000, Jung et al., 1998; Young et al., 2001) considered the various theoretical approaches and estimated the magnitudes of necessary corrections to each of them. Any particular correction is to be applied to an approximate theory in which that correction has not been included, in order to obtain a superior result. This composite approach was characterized as yielding "best" predictions.

In this paper, we will be only concerned with whole atom Compton scattering. Such cross sections were measured in one of the APS experiments. More precisely, Jung et al. (1998) measured the ratio of whole atom inelastic scattering cross sections to that of elastic scattering, for neon at a 90 degree scattering angle for four different energies in the range 11 to 22 keV. Synchrotron measurements of these cross sections have also been reported by Namito et al. (1995) for six elements at 90 degrees for the photon energy range 20-40 keV. We present a comparison of experimental results with the commonly used incoherent scattering factor (ISF) predictions as well as with "best" predictions, to understand what needs further attention. A comparison of measured whole atom cross sections in the photon energy range 11 to 40 keV, using conventional and synchrotron sources, also demonstrates the dramatic improvement in the precision of scattering measurements when synchrotron sources have been used.

A brief description of various theoretical calculations, their inherent deficiencies and the corrections needed to improve predictions, is presented in section 2. The various types of experimental measurement of Compton scattering are discussed in section 3. Comparisons of experiments with ISF and "best" predicted values are discussed in section 4, identifying some circumstances which require further experimental data to confirm the adequacy of theoretical treatments. These results are presented in Figures 1-4.

2. Theoretical methods

The theory of inelastic x-ray scattering is, in general, based on the relativistic treatment of the interaction between the electromagnetic field and bound electrons. Different theoretical schemes, namely incoherent scattering factor (ISF) approximation, impulse approximation (IA), S-matrix formalism, are available to calculate the inelastic or Compton scattering cross sections. An overview of the theories used in Compton

scattering calculations is available (Bergstrom et al., 1997). Both ISF and IA are based on the A^2 term of the non-relativistic interaction Hamiltonian, and they neglect the dynamic effects associated with the ***p.A*** term. ISF uses a closure approximation in its derivation, involving a sum over all inelastic procedures (including excitations). As a result, it calculates inelastic scattering for the total atom, including Raman scattering. Kinematic restrictions on allowed transitions (due to binding) are ignored in ISF. ISF has been calculated using non-relativistic Hartree-Fock wave functions. Correlation effects on the atomic structure can be included in the ISF calculation. Details of the ISF calculation, and frequently used tables of scattering factors S (q,Z) at different momentum transfers q and for different elements, are given by Hubbell et al. (1975). IA assumes that the atomic electrons are free, and therefore it is expected to be valid for photon energies much greater than the binding energy of the interacting electron at large scattering angles. Binding effects in IA enter only through the momentum distribution of the free electrons. IA does not include Raman scattering. ISF modifies the free electron scattering by the incoherent scattering factor, while IA modifies free electron scattering result by the Compton profile of the bound electrons. Tables of Compton profiles for all shells of all elements have been given by Biggs et al. (1975). ISF calculates the singly differential cross sections, while IA obtains the doubly differential cross section for Compton scattering ($d^2\sigma/d_'d_$) in energy and direction of the scattered photon, through the Compton profile for electrons of different shells. Singly differential cross sections can also be obtained in IA integrating the doubly differential cross section over scattered photon energies. ISF and IA are generally considered to be 'simplest' calculations.

Dynamic effects in scattering have been obtained more recently, using a second order S-matrix calculation which includes the **p.A** term or the full relativistic interaction. The approach more fully treats the dynamics of the scattering process, including resonances and the infra-red rise in the soft photon region (Suric et al., 1991; Bergstrom et al., 1993a). However, it is an independent particle approximation (IPA) calculation, with electron states described in a common self-consistent Dirac-Slater type central potential, neglecting correlations among electrons. A computer code (Bergstrom *et al.,* 1993b) calculates the doubly differential cross section for Compton scattering based on exact numerical evaluation of the relativistic second order external field S-matrix element within the IPA model. In these calculations no further approximations are made and all intermediate states in the field are taken into account. This has sometimes been referred to as an 'exact' calculation. However, the approximations made in the S-matrix calculation i.e. neglect of non-local exchange effects and electron-correlation effects, are not adequate in the regions close to thresholds. In order to obtain energy integrated cross sections, one integrates over the spectrum of final energies, applying a low-energy cutoff (reflecting experimental detector sensitivity) so as to exclude the infrared divergent region which is present in Compton scattering with soft final photons. To obtain whole atom cross sections one must sum all sub-shell cross sections and also add a separate calculation of the Raman scattering cross sections.

It is to be understood that all theoretical approaches have limitations and no one of them is fully adequate. Carney and Pratt (2000) estimated the magnitude of the various corrections to simplest theories (dynamic, non-local exchange, electron correlation and

relativistic effects) and developed a general scheme to obtain «best» predictions (except close to thresholds), considering them as linearly independent perturbative corrections. Symbolically, this may be expressed as

$$(d\sigma/d\Omega)_{best} = (d\sigma/d\Omega)_{theory}\ (1+\Sigma_j\ \delta_j).$$

Here $(d\sigma/d\Omega)_{theory}$ represents any of the theoretical predictions (S-matrix, ISF, IA) and the summation is over the corrections δ_j which should be applied to the theoretical method used. The S-matrix calculations require correction for non-local exchange and for correlation. In case of ISF, the correction essentially stems from the dynamic effects. This correction is large for low momentum transfers. A comparison of S-matrix and ISF predicted values with the "best" predictions has been presented for neon at five photon energies at four scattering angles (Carney and Pratt, 2000). ISF is found to be adequate at higher energies (accurate within about 1% or better for energies above 25 keV, for all angles), while it differs from "best" predictions by as much as 50% for 1.5 keV at smaller scattering angles. ISF and "best" predictions are shown in Figs. 3-4.

3. Experiment

Historically Compton scattering was experimentally observed using an x-ray tube as a source and an ionization chamber as a detector (Compton et al., 1923a). With the availability of better sources and improved detectors Compton scattering measurements were reinitiated from time to time. Precise experimental results within an uncertainty of 1% are now possible, using synchrotron sources and high resolution detectors.

A complete measurement of Compton scattering for a given incident photon energy which detects scattered photon and ejected electron (but not their spin and polarizations) involves four observables, namely, the direction and energy of the scattered photon, and the direction and energy of the scattered electron, described by the cross section $d^4\sigma/d_'dE'd__\ d_e$. Here $_'$ is the scattered photon energy, E' the scattered electron energy, $__$ the solid angle of the emitted photon and $_e$ the solid angle of the ejected electron.

Measurements of the singly differential cross section $(d\sigma/d__)$ i.e. the scattering cross section integrated over scattered photon energies, electron energies and electron directions, are the simplest and are the most extensive. Numerous measurements of the doubly differential cross sections $(d^2\sigma/d_'d__)$ have also been made. A few measurements of triply differential cross sections $(d^3\sigma/d_'d__d_e)$ integrated over electron energies have been reported (Bell et al., 1990; Bell et al., 1991; Kurp et al., 1996). Measurement of the triply differential cross sections is particularly difficult, both because the direction of the recoiling electron is altered by multiple scattering within the target and because a coincidence measurement is required.

Measurements can be made of whole atom scattering, i.e. measuring the scattering intensity without looking at which bound electrons contribute to the scattering. Or

measurements can be made shell-wise, where the energy and angle of the scattered photon are observed along with the sub-shell from which an electron is ejected in scattering. This sub-shell may be determined if the scattered photon is detected in coincidence with an energy measurement of ejected electron or in coincidence with the fluorescence x-rays characteristic of a particular shell. Scattered photons detected in coincidence, for example, with characteristic K x-rays led to selective study of K-shell Compton scattering. A few measurements on L-shell scattering have also been reported (Kane 1992). When the direction of the recoil electron is measured in coincidence with the scattered photon, we get the whole atom triply differential cross section. A review of the measurements of singly and doubly differential scattering cross sections from sub-shells and the whole atom is available (Kane 1992).

Here we are concerned with the whole atom singly differential cross section. Measurements have been reported using conventional x-ray sources (x-ray tubes and radioactive sources) and more recently using synchrotron sources. The easy availability of conventional sources over a wide photon energy range had allowed measurements covering a wide range of photon energies and target elements. In contrast measurements with synchrotron sources have so far been limited to a few photon energies (especially lower energies) and a few elements (Jung et al., 1998; Young et al., 2001; Namito et al., 1995) as described earlier. Other recent measurements (Rao et al., 1994, 1996; Garg et al., 1993; Kumar et al., 2001; Shah et al., 2001; Simsek 2000; Erzeno_lu et al., 1998) have been reported using conventional sources, for elements in the range $13 \leq Z \leq 82$ and for various photon energies in the range 14 to 88 keV. Both kinds of measurements have been performed using solid state detectors. The reported maximum experimental uncertainties are 10% for measurements done with conventional sources, 2.5% for measurements done with synchrotron sources. For the purpose of the present paper we restrict our discussion to measurements in the energy range 11 to 40 keV, in which deviations from ISF predictions were reported using conventional sources. These measurements of Compton scattering were performed at 90 degree scattering angle. Results of the two classes of experiments are compared in Figures 1 and 2.

4. Comparison and Discussion

In the range of the existing experimental data there is little difference between ISF and "best" predictions. We compare the experimental results with ISF predicted values in figures 1 and 2. A sharp contrast between the two types of measurements can be noted. Measurements using conventional sources are scattered, differing in magnitude from ISF by 5% to more than 50% (much above the stated uncertainties of 10%), while all measurements using synchrotron sources fall within a narrow band of few percent differences. However, use of synchrotron sources may not always be favored. In coincidence experiments, due to the high random coincidence rate in synchrotron sources, it has been argued that the use of conventional sources is favored (Laukkanen *et al.* 1996).

Experimental Compton scattering cross sections for neon are compared with ISF and "best" predictions in Figure 3. It is to be noted that in obtaining the experimental Compton scattering cross sections from the measured Compton to Raleigh ratio (Jung et al. 1998), we have used the "best" predicted Raleigh scattering cross sections (Carney and Pratt 2000). As already noted, there is hardly any difference between ISF and "best" predicted values at higher photon energies. This is expected, as ISF is believed to be valid at high photon energies. ISF is higher than the "best" predicted values at lower photon energies. The difference between ISF and "best" predicted values differ as much as 19% for 5.415 keV and by about 38% at 1.486 keV. The ISF result of Hubbell et al. includes excitation channels as well as ionization and therefore includes Raman scattering, while the "best" prediction presented here is only for the Compton scattering. However it has been found by Young et al. (2001) that the Raman contribution to total scattering is less than 1% for neon in the energy range 5 to 15 keV. Therefore the large difference between ISF and "best" predictions at lower energies reflects the importance of dynamic corrections.

It is evident that further inelastic scattering measurements around 10 keV and lower are warranted to confirm the adequacy of the theoretical treatment, especially because we know that the perturbative treatment in the "best" calculations will fail at lower energies. But of course these are also situations in which cross sections are smaller, making measurements more difficult. In addition, separation of the elastic and inelastic peaks will be difficult and may be beyond the capability of present day detectors. Further, with the lack of precision measurements at other angles than 90 degree, we are not in a position to test the adequacy of the different theoretical approaches at other angles. Therefore, measurements at a range of angles are also needed to confirm the adequacy of the theories in this range. These studies, and corresponding theoretical predictions, are also needed over a range in Z.

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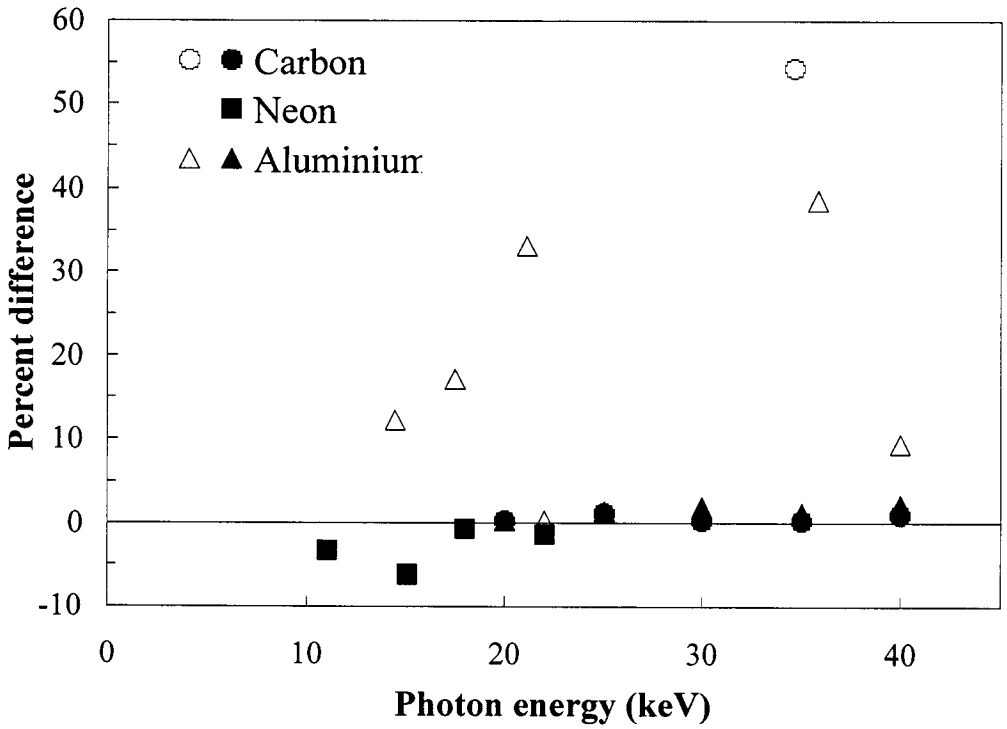

Fig. 1. Comparison of experimental Compton scattering cross sections (Garg et al. 1993; Rao et al., 1994, 1996; Namito et al., 1995; and Jung et al., 1998) for Carbon (Z=6), Neon (Z=10) and Aluminium (Z=13), measured using conventional and synchrotron sources, with ISF predicted values. Solid symbols represent measurements using synchrotron sources and hollow symbols represent measurements with conventional sources.

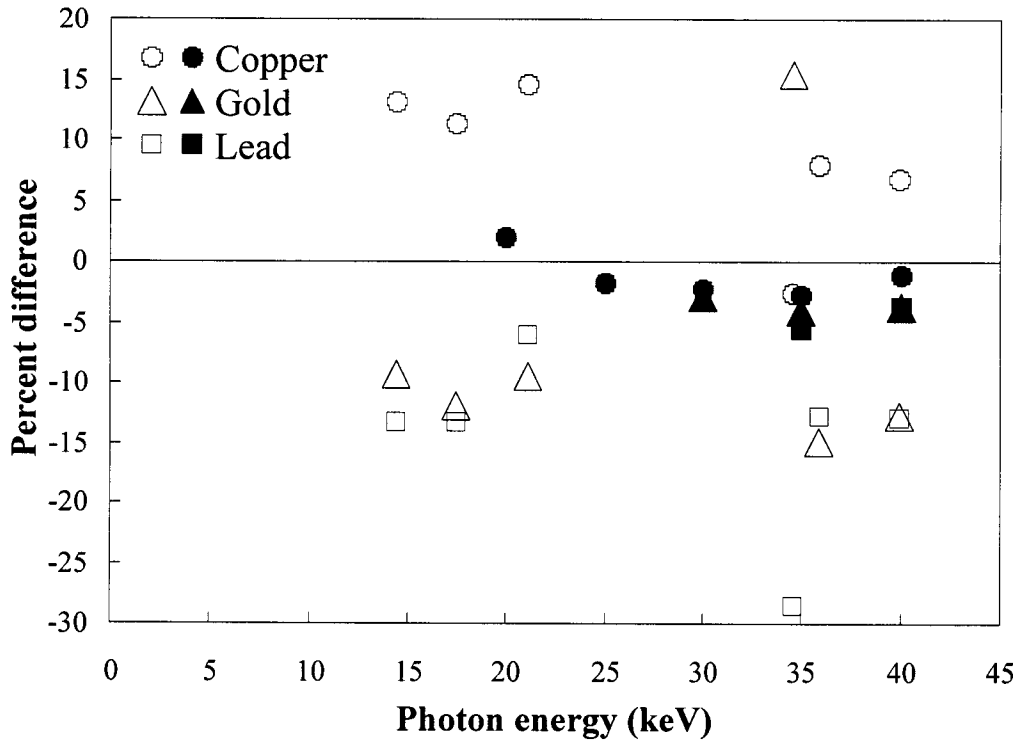

Fig. 2. Comparison of experimental Compton scattering cross sections (Garg et al., 1993; Namito et al., 1995 and Rao et al., 1994, 1996) for Copper (Z=29), gold (Z=79) and lead (Z=82), measured using conventional and synchrotron sources, with ISF predicted values. Solid symbols represent measurements using synchrotron sources and hollow symbols represent measurements with conventional sources.

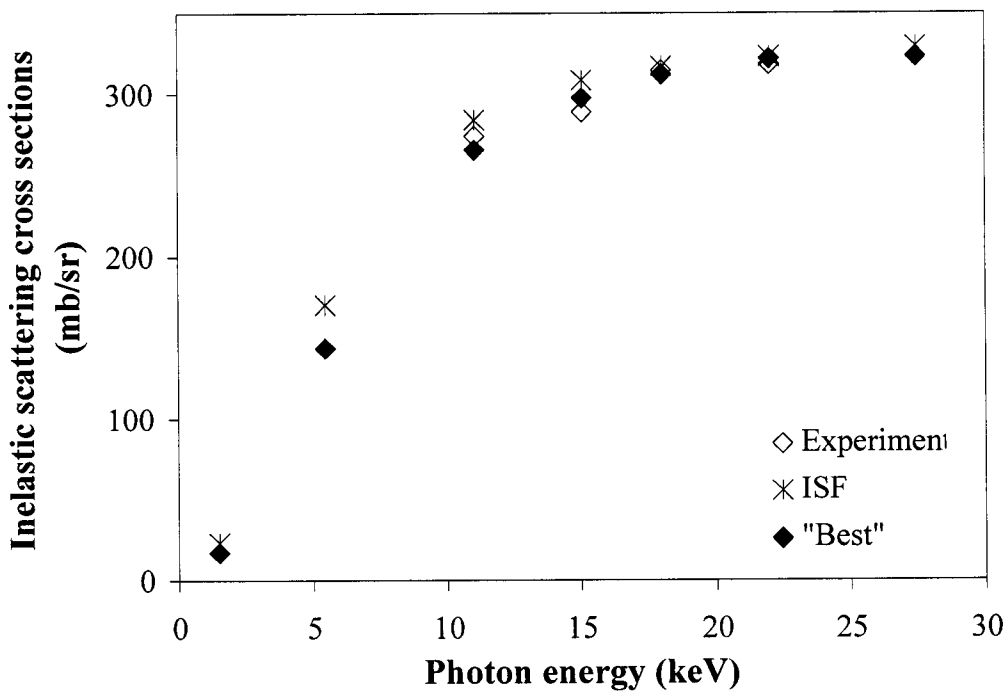

Fig. 3. Comparison of experimental Compton scattering cross sections (Jung et al., 1998) for neon at 90 degree scattering angle with ISF and "best" predicted results.

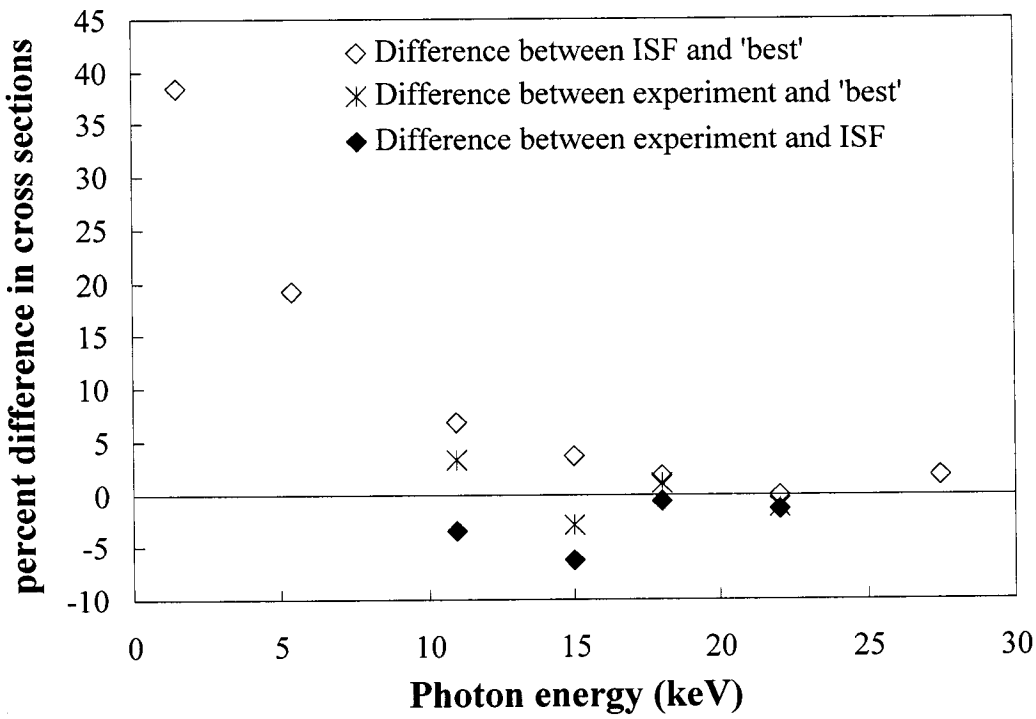

Fig. 4. Percent difference between ISF and "best", experiment (Jung et al., 1998) and ISF, and experiment (Jung et al., 1998) and "best" predicted values, for neon at 90 degree scattering angle.