Manifestations of nonlocal exchange, correlation, and dynamic effects in x-ray scattering

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Introduction

X-ray scattering is a widely used diagnostic tool, and a quantitative understanding of this basic process has fundamental importance. In practice, elastic and inelastic scattering cross sections are obtained from form factor tables, which are calculated using wavefunctions of varying accuracy. These form factors and incoherent scattering factors are derived from the A^2 term of the nonrelativistic interaction Hamiltonian [$H_{int} = (1/2mc^2)e^2A^2 - (e/mc)\mathbf{p}\bullet\mathbf{A}$]. Simple, well-known corrections to the form factor approximation (i.e., anomalous scattering factors for elastic scattering and generalized oscillator strength corrections for inelastic scattering) can then be added perturbatively. This, however, still leaves an incomplete picture in that these do not account for the inelastic second-order **p•A** terms, etc. Recent theoretical advances using relativistic S-matrix techniques allow one to treat the interaction fully at the expense of a simplified model of the atom for both elastic and inelastic scattering [1, 2]. The two approaches yield cross sections for scattering from the neon atom that differ by up to 16% in the 11–22 keV range.

Very few experiments have been reported for scattering from free atoms [3], the case for which cross sections are usually calculated. In view of the basic need for accurate and absolute measurements in free atoms to assess the importance of various approximations, we have explored scattering from neon and helium at 90º from 11–22 keV. This combined experimental and theoretical work accomplishes the following: 1) it reports precise measurements of differential x-ray scattering cross sections in neon and helium, 2) it uses these to identify the more accurate approach for this energy and momentum transfer range and Z, 3) it estimates the magnitude of corrections to the simplest theory, and 4) it outlines a method for obtaining predictions of 1% accuracy.

Methods

We measured polarization-independent differential cross sections in Ne at energies far above the K-edge (870 eV) where Compton and Rayleigh scattering cross sections are comparable. By measuring ratios, $d\sigma_{tot}(Ne)/d\sigma_{tot}(He)$ and $d\sigma_C\,(Ne)/\,d\sigma_R\,(Ne)$, we avoided difficult absolute measurements of x-ray flux and detector efficiency and were able to obtain high accuracy. Here, $d\sigma_{tot}$, $d\sigma_C$, and $d\sigma_R$ denote differential cross sections at 90º for total, Compton, and Rayleigh scattering. The ratio of total scattering in Ne to He is a good test of theory because the He total-scattering cross sections are well described by nonrelativistic, nonlocal incoherent scattering factors over this energy and momentum transfer range.

The experiment was performed on the bending magnet beamline (12-BM) at the Basic Energy Sciences Synchrotron Radiation Center (BESSRC) of the Advanced Photon Source. Monochromatic x-rays ($\Delta E/E \approx 1.4 \times 10^{-4}$) created a line source in the gas target, which was viewed simultaneously by two well-characterized Si(Li) detectors placed orthogonal to each other and to the photon beam propagation axis (Figure 1). Polarization-independent cross sections are obtained for any polarization state of the incident beam by averaging the yield in the described geometry. The detector acceptances were defined by two apertures and determined through simulation to be 90º ± 3º. Scattered x-rays passed through ≈ 25.4 mm of gas and a 125 µm Kapton window to the detectors. Gas pressure was measured to 0.25%. Higher harmonics of the incident beam entered only indirectly through run-to-run normalization, where the use of a low-Z gas in the ion chambers reduced their contributions to < 0.7%.

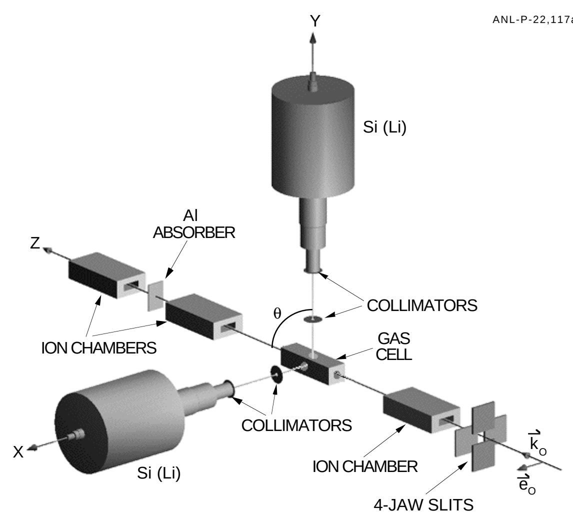

Figure 1: Experimental configuration for gaseous x-ray scattering.

Measurements were made at 11, 15, 18, and 22 keV x-ray energies and two pressures for Ne and He. Background measurements taken with an evacuated cell show no scattered light at the incident energy (22 keV in Figure 2a).

The two features at ≈ 24 and 25 keV correspond to K_α emission from In and Sb impurities in the detector. Background subtracted spectra are shown in Figures 2b and 2c.

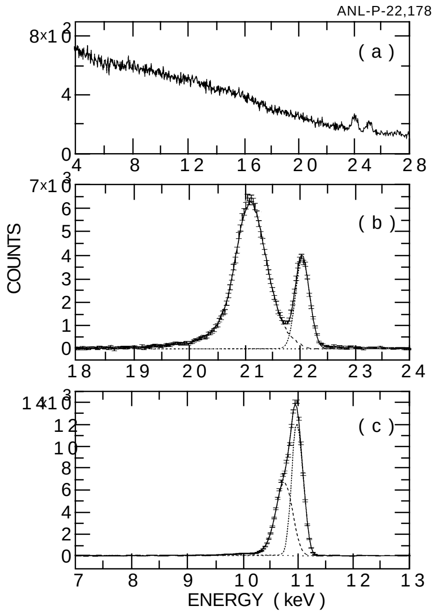

Figure 2: Energy spectra in the Si(Li) detector oriented normal to the polarization plane. (a) Background spectrum with empty cell, (b) scattering from Ne at 22 keV, and (c) scattering from Ne at 11 keV. For (b) and (c), decomposition of the Compton and Rayleigh cross sections is shown in dashed lines.

Total-scattering cross sections were corrected for detector live time and then normalized to integrated ion chamber current (including dark count and x-ray transmission corrections). Extrapolation to zero pressure provides corrections of ≈ 2% for Ne, but the He data exhibit no pressure-dependent effects. Experimental errors of ≈ 1% are obtained, which are dominated by systematic error due to x-ray beam walk during this early experiment.

Compton and Rayleigh scattering contributions were separated using the well-characterized detector response and the impulse approximation Hartree-Fock Compton profiles. The whole atom profile was computed as the sum of the 1s, 2s, and 2p subshell profiles and was then used with the detector response to generate a Compton line shape. The amplitudes and positions of the generated Compton and Rayleigh line shapes were varied in a four-parameter fit to the experimental data. Examples of these fits are shown in Figures 2b and 2c. Reliability of the separation is ≈ 8% at 11 keV where there is considerable overlap, and ≈ 2 % at 22 keV.

Results and Discussion

Table 1 shows experimental and theoretical values for the ratio of total scattering in Ne to He and the Ne Compton-to-Rayleigh ratio. The theoretical cross sections for 90° are used since the weighted average over the field of view is identical to ≈ 10^{-3}. The two columns following the experimental result show widely available theoretical results. Note that for the total-scattering cross section, the Hubbell's simple scattering factors [4] are in good agreement with experiment, whereas for the Compton to Rayleigh ratio they are not. The opposite is true for a more sophisticated theory, where modified form factors (MFF) plus anomalous scattering factors (ASF) are used for the Rayleigh cross section [5].

Table 1. Comparison of experiment and theory.

Energy (keV)	Experiment	Hubbell Hubbell	MFF+ASF Hubbell
	$d\sigma_{tot}(Ne)/d\sigma_{tot}(He)$		
11	7.31 (10)	7.25	7.83
15	5.74 (14)	5.89	6.10
18	5.61 (6)	5.60	5.70
22	5.49 (10)	5.45	5.50
	$d\sigma_C(Ne)/ d\sigma_R(Ne)$		
11	0.95 (8)	1.03	0.89
15	2.06 (9)	2.30	2.06
18	3.06 (7)	3.21	2.98
22	4.09 (8)	4.30	4.10

The contradiction is resolved by examining the approximations made in these theories. The "simplest" theory is obtained using the form factor and impulse approximations with local, nonrelativistic, independent particle (IPA) bound state wave functions (Herman-Skillman). The simplest theory can be corrected for five effects (nonlocal exchange, electron correlation, relativity, dynamics, and Raman scattering) [6]. Table 2 shows percentage corrections for our situation. Applying these corrections to existing calculations, one is able to obtain good agreement (≈ 1%–2% level) with both measured cross section ratios. Neither Hubbell's tabulation nor the MFF + ASF prediction do well in both situations at all energies.

Table 2. Percentage corrections for Compton and Rayleigh scattering cross sections for effects beyond the impulse and form factor approximations using local nonrelativistic IPA bound state wave functions. [6]

		Compton			
E(keV)	Nonlocal	Cor	Rel	Dyn	Raman K
11	1.4%	-3.3%	-0.1%	-1.2%	0.0%
15	1.1%	-1.0%	-0.1%	0.4%	0.0%
18	0.9%	-0.4%	-0.1%	0.2%	0.0%
22	0.7%	-0.2%	-0.1%	-0.6%	0.0%
		Rayleigh			
E(keV)	Nonlocal	Cor.	Rel.	Dyn.	Raman L
11	-9.5%	0.9%	-0.2%	4.8%	0.2%
15	-6.2%	0.9%	-0.4%	3.9%	0.2%
18	-3.3%	0.4%	-0.4%	3.3%	0.1%
22	-1.2%	0.1%	-0.5%	2.6%	0.1%

In conclusion, none of the current theories for which numerical values are available can alone properly describe x-ray scattering in neon (and presumably other light elements). The size of dynamic effects indicates a need to go beyond the form factor and impulse approximations to obtain accurate cross sections. While current IPA S-matrix theories for Rayleigh and Compton scattering are known to work well for high-Z elements and high energies, for low Z they are less reliable because they do not include nonlocal exchange and electron correlation effects.

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[5] Results within the MFF + ASF approximation for Rayleigh scattering are available at http://www.phys.llnl.gov/V-Div/scattering/elastic.html.

[6] Further details can be found in the full publication of this work, M. Jung *et al.*, *Phys. Rev. Lett* **81**, 1596 (1998).