

Atomic Form Factor Measurement on Beryllium and Aluminum by Using Inelastic X-ray Scattering

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Introduction

The atomic form factor is one of the fundamental physical quantities in solid-state physics. It is a Fourier transform of an electron distribution around a nucleus and carries information on the electron wave function. Since a complicated many-body problem is involved in this calculation, it is generally difficult to obtain an exact solution for the atomic form factor. Some calculations do use approximate methods that are believed to be accurate enough for practical use for materials. The tabulated values of the atomic form factors for isolated atoms based on these calculations can be found in the literature [1]. However, the accuracy of these calculations should be verified experimentally. Furthermore, the atomic form factor in aggregate could be slightly different from the atomic form factor of an isolated atom because of the reconfiguration of the electron distribution. The atomic form factors of several systems have been studied by using well-established experimental methods [2-5] and theoretical methods [6-8], and fairly good results have been obtained. Since the atomic form factors were determined from Bragg reflections in these methods, measurements were limited to the lowest-order Bragg reflections. Typically the lowest-order reflection for a given crystal structure is around 30 nm^{-1} in the reciprocal momentum space. On the other hand, the strongest contributions to the atomic form factor from valence electrons can be found in the region up to 35 nm^{-1} . Therefore, direct measurements in this region where the valence electron contribution dominates can hardly be achieved by the methods stated above. Access to this region can be achieved by phonon intensities measured by inelastic x-ray scattering. Furthermore, this method allows us to study charge densities without any limitation in the momentum space.

Methods and Materials

A method for determining atomic form factors has been developed by means of inelastic x-ray scattering. This method employs precise measurement of phonon intensities as a function of momentum and energy transfers in the millielectronvolt regime produced by an incident x-ray energy of 21.657 keV. These experiments were performed at APS beamline 3-ID. The general setup is shown in Fig. 1.

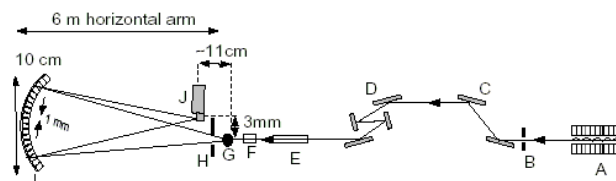


FIG. 1. Schematic view of the spectrometer developed at beamline 3-ID. The beam comes from an undulator (A) and premonochromator (C), then passes through the high-resolution monochromator (D) and focusing mirror (E) before it illuminates the sample (G). The scattered intensity is focused by the analyzer (I) into the detector (J). The ionization chamber (F) monitors the incident flux on the sample. The slit systems (B and H) determine the source size.

The resolution function of the spectrometer was experimentally determined from a plexiglass sample, and a full width at half-maximum (FWHM) of 2.2 meV energy resolution was measured. The requirements for very high energy resolution and the basic principles of such instrumentation are discussed in References 9 and 10. In this work, the x-ray atomic form factors of beryllium and aluminum single crystals along the $[0\ 0\ \xi]$ principal symmetry direction are reported in order to obtain reliable information on the outer electron charge densities in these elements. In the experiment, 13-mm-diameter beryllium and 2.05-mm-thick aluminum single crystals with a 99.9999% high purity level were used. The rocking curves for beryllium and aluminum $(0\ 0\ 2)$ reflection appeared as double or single peaks. The peak widths for beryllium and aluminum are about 0.02° and 0.05° , respectively.

Results and Discussion

Figures 2 and 3 show the spectra corresponding to measurements at different momentum transfers along the $[0\ 0\ \xi]$ direction for beryllium and aluminum, respectively. The momentum transfer axis is given in units of $2\pi/c$, where c is the lattice constant of the given sample along the c axis direction. For hcp-structure beryllium, $\xi = 1/2$ corresponds to the first Brillouin zone. Meanwhile, for fcc-structure aluminum, the first Brillouin zone is at $\xi = 1$.

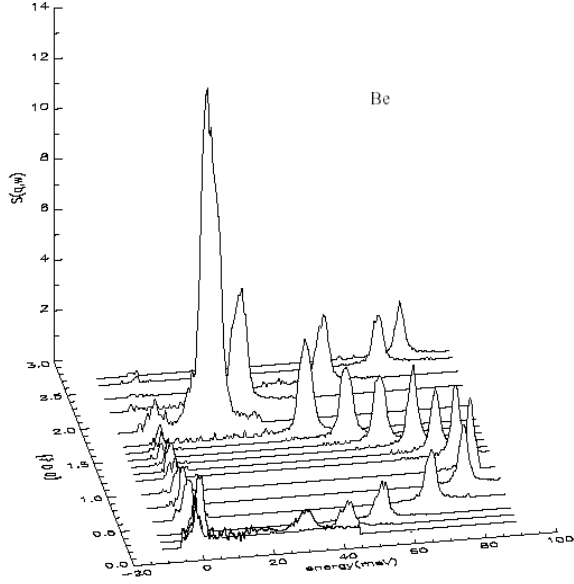


FIG. 2. Energy scans for beryllium along the $[0\ 0\ \mathbf{x}]$ direction for longitudinal modes.

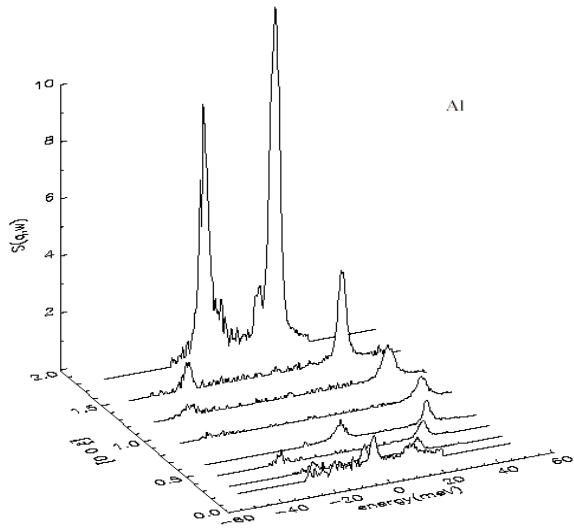


FIG. 3. Energy scans for aluminum along the $[0\ 0\ \mathbf{x}]$ direction for longitudinal modes.

Since there was elastic scattering due to either impurities in the sample or some other reason, only the energy-loss side of the excitations was recorded for the beryllium case (Fig. 2). For the case of aluminum, however, both the energy-loss and energy-gain sides of the excitations were recorded to determine the energy scale, except for the $\xi = 0.6$ scan.

The phonon dispersion relations determined from the energy positions of the measured data are presented in

Fig. 4 for beryllium and aluminum. Longitudinal and transverse modes for beryllium and only the longitudinal mode for aluminum are compared to the previous results from inelastic neutron scattering [11, 12]. The comparison of the present work with the neutron data shows an agreement of 2% or better.

Atomic form factors calculated from measured integrated intensities of the phonon excitations are presented as circles in Fig. 5 for beryllium and in Fig. 6 for aluminum. In these figures, measured atomic form factors are compared to Hartree-Fock calculations of the free-atom form factor (solid line) by using the χ^2 minimization method. In addition, in Fig. 5, measured form factors are also compared to the pseudopotential calculation [13] of liquid beryllium. In Figs. 5 and 6, the

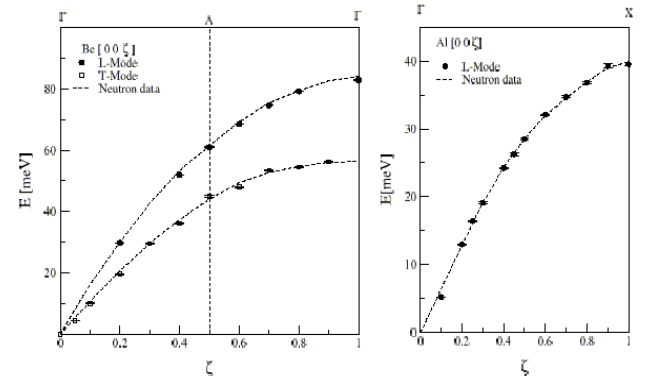


FIG. 4. Dispersion relations for beryllium and aluminum along the $[0\ 0\ \mathbf{x}]$ direction.

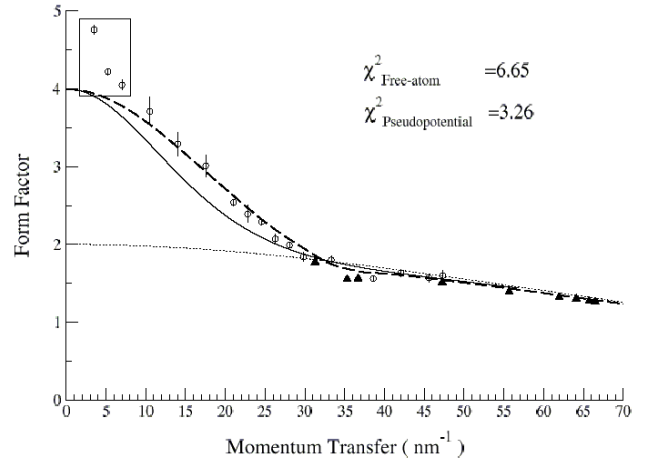


FIG. 5. Comparison of measured atomic form factor (circles) with free-atom Hartree-Fock calculation (solid line) and metallic-atom form factor calculated by pseudopotential (dashed line) in the $[0\ 0\ \mathbf{z}]$ direction for beryllium. Triangles show the previous experimental data determined from Bragg reflections [2].

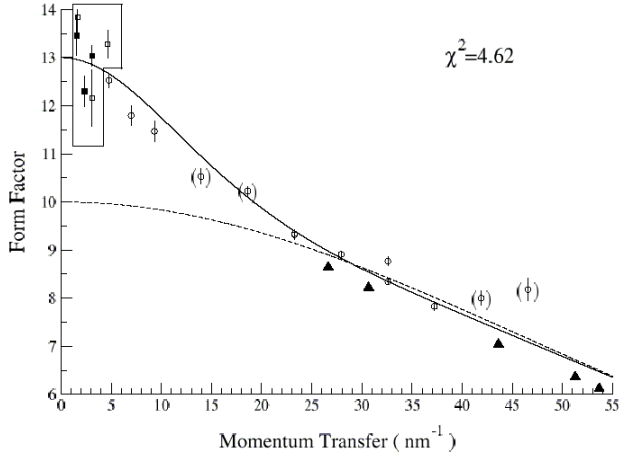


FIG. 6. Comparison of measured atomic form factor (circles) with free-atom Hartree-Fock calculation (solid line) in the $[0\ 0\ z]$ direction for aluminum. Triangles show the previous experimental data determined from Bragg reflections [14].

box shows the region where the measurements become uncertain as a result of the poor momentum resolution of the analyzer. In the analysis, measured phonon frequency widths for aluminum show broadening around the Brillouin zone as a result of the phonon-phonon interaction [11], and those points are shown with the brackets in Fig. 6. The overall χ^2 values fitted to the free-atom calculation for both beryllium and aluminum and to the pseudopotential calculation for beryllium are also included in the figures. In the case of beryllium, better agreement is obtained when the measurements are compared to the pseudopotential calculation. However, present experimental data are substantially higher than the free-atom Hartree-Fock calculation in the low momentum transfer region. In the case of aluminum, the measured experimental data are in overall agreement with the free-atom Hartree-Fock calculation.

In contrast to previous studies with elastic x-ray scattering (Bragg reflections), charge densities can be studied by phonon intensities without any limitation in the momentum space with this method. Hence, complementary to previous experiments, for the first time, atomic form factor measurements are extended to a very

low momentum transfer region (down to 7 nm^{-1}) by using the inelastic x-ray scattering method. Since the dispersion is much higher in the small momentum transfer region, the error associated in the measurements in the region $Q < 5\text{ nm}^{-1}$ can be minimized if the higher energy and momentum resolution are used in the spectrometer. In this work, we have presented the measurement in one specific direction. It is worth repeating the experiment for other major symmetry directions to see whether big changes are present in the charge distribution between the directions.

Acknowledgments

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