# An Improved Apparatus for Nondipolar Photoelectron Angular Distribution Measurements

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## Introduction

New aspects of the photoionization of atoms, molecules, and materials have been investigated in recent years with studies of nondipole photoelectron asymmetries using tunable synchrotron radiation. The physics of photoionization processes is largely understood using the dipole (E1) approximation. In the multipole expansion of the exact form of the photon-electron interaction, the E1 term is the dominant contribution. A first-order correction comes through the interference between E1 and the quadrupole term (E2).[1] This first-order correction is observable as a forward-backward asymmetry in the angular distribution of the emitted photoelectrons. The asymmetry depends on the dynamics of the photoionization process, i.e., the E1 and E2 transition matrix elements and their respective complex phases. The observable asymmetries generally increase with photon energy. Using hard x-ray beamlines at Brookhaven's National Synchrotron Light Source and Argonne's Advanced Photon Source (APS), we have measured nondipolar asymmetry parameters for atomic inner shells.[2] In this report we describe a new experimental apparatus for gas-phase measurements of nondipolar photoelectron asymmetries. The apparatus has been successfully used to determine the nondipolar asymmetries in Kr-1s and Br-1s photoionization.

## Method

The apparatus consists of a vacuum chamber with four parallel-plate electron analyzers (PPAs) mounted 90° from each other on a computer-controlled turntable. The sample gas is let into the chamber by an effusive jet. The x-ray beam intersects the sample gas close to the nozzle of the jet. The four PPAs are aligned such that they view the interaction region at the "magic" angle of 54.7° with respect to the vertical. A detail of the spectrometer nozzles and the effusive jet is shown in Fig. 1. For the measurement of the nondipolar asymmetry, the analyzers are positioned along the diagonals in space: two in the forward hemisphere, and two in the backward hemisphere. Using these particular directions and averaging between the two analyzers in each of the two hemispheres, eliminate any influence of the polarization properties of the x-rays on the result of the nondipolar asymmetry measurement. The difference between the signal in the forward and backward hemispheres, divided by their sum, is directly proportional to the nondipolar asymmetry parameter. The use of four analyzers (rather than one as in the earlier apparatus), which measure the same part of the electron spectrum of the sample at the same time at different angles, greatly increases the efficiency of the data-taking process and diminishes the need for critical normalizations.
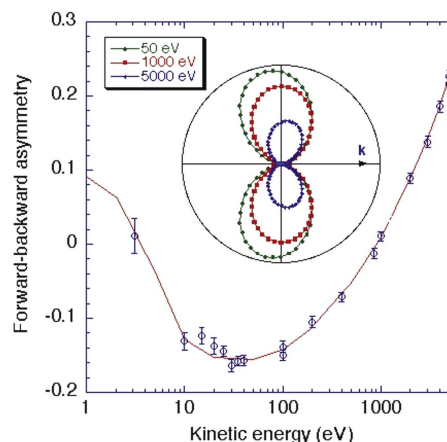


*FIG. 2. Forward-backward asymmetry in the described geometry for Kr-1s photoionization. A positive (negative) value of the forward-backward asymmetry denotes predominantly forward (backward)-directed electron emission. The circular insert shows representative emission patterns as polar plots.*

## Results and Discussion

We have measured the nondipolar forward-backward asymmetry of Kr 1s photoionization from threshold to 8 keV above the Kr-*K* edge. The results of this measurement are shown in Fig. 2, together with a theoretical prediction based on the inclusion of higher-order corrections to the photon-atom interaction.[3] It can be seen from Fig. 2 that theory predicts that the emission will be forward-directed close to threshold but will change direction within a few eV from threshold. The lowest data point at 3-eV excess energy gives a vanishing asymmetry, and at 10 eV, a pronounced backward-directed emission pattern is observed. In perfect agreement with the prediction, the data points show the energy dependence of the nondipolar asymmetry going through another sign change near 1000 eV to increasingly positive values with increasing excess energy. The circular insert in Fig. 2 shows the emission patterns of three representative excess energies as a polar plot. The relative sizes of the patterns reflect the decrease in the cross section with energy. It can be seen that all three cases exhibit the typical two-lobed pattern of dipolar photoemission.



*FIG. 1. Closeup view of the entrance nozzles of the four PPAs and the effusive gas jet.*

In comparison to the forward-backward symmetric pattern at 1000 eV, however, the ones at 50 eV and 5000 eV are clearly skewed into the backward and forward directions by the nondipolar interaction. Similar results were obtained for Br 1s photoionization in $Br_2$ and $CF_3Br$.

## References
[1] See, e.g., A. Bechler and R. H. Pratt, Phys. Rev. A **39**, 1774 (1989) and **42**, 6400 (1990).

[2] B. Krässig et al., Phys. Rev. Lett. **75**, 4736 (1995); M. Jung et al., Phys. Rev. A **54**, 2127 (1996); S. H. Southworth et al., APS User Activity Report, ANL Report 00/5 (2001).

[3] L. LaJohn, private communication. For details on the calculation, see L. LaJohn and R. H. Pratt, Phys. Rev. A **58**, 4989 (1998