

Young's Double-slit Experiment using Core-level Photoemission from N₂

The Young's double-slit experiment (YDSE) provides the simplest and most fundamental example in which the superposition of two coherent waves of light leads to interference oscillations of the light intensity. Photoionization of homonuclear diatomic molecules represents conceptually a similar phenomenon for the electron waves. The interference of the coherent electron waves emitted from two indistinguishable atoms leads to intrinsic interference oscillations. Cohen and Fano [1] were the first to derive the equation for the total photoionization cross section for the H₂ molecule including this YDSE effect. Subsequent theoretical [2] and experimental studies of ionization of H₂ and D₂ molecules with different projectiles were performed.

Core-level photoemission from molecules like N₂ provides a new opportunity to investigate coherent emission of photoelectron waves [3,4]). For core-level photoionization of these molecules, an additional complication appears due to the presence of the gerade and ungerade 1σ bound orbitals with a slight energy gap between them. This gap is 0.1 eV and is even smaller than the vibrational splitting of ~0.3 eV.

The experiment was carried out at the high-resolution soft X-ray photochemistry beamline **BL27SU**. The light source of the beamline is a figure-8 undulator which provides linearly polarized radiation. The photoelectron spectra were recorded using a high resolution Scienta SES-2002 analyzer. The analyzer bandwidth was set to ~31 meV. The overall bandwidth was determined separately by measuring Xe 5p photoelectrons at the same monochromator and analyzer settings.

Figure 1 shows the N 1s photoelectron spectrum

recorded at photon energy of 831 eV. The experimental spectra were decomposed by least-squares curve fitting into 1σ_g and 1σ_u components with the individual vibrational progressions.

The partial cross section ratios of σ_g/σ_u for the photoionization of the 1σ_g and 1σ_u shells corresponding to the 0 → 0 and 0 → 1 vibrational transitions and are plotted in Fig. 2 as a function of the photoelectron momentum *k* in atomic unit (a.u.) in order to display clearly the oscillations. The results of previous measurements [5] are also included. The *ab initio* calculations using RPA method are compared with measurement.

At low momenta *k* < 1.5 a.u. the ratios are defined by the σ* shape resonance, and only at *k* > 1.5 a.u. the CF oscillations become apparent. There is a good qualitative agreement between the theory and the experiment for both vibrational transitions.

The main difference between the 1σ_g and 1σ_u shells is connected with the symmetry of these states. From the dipole selection rules, only odd partial waves contribute to the photoionization of the 1σ_g shell, and only even partial waves to the 1σ_u shell. The interference modulation is mainly connected with the onset of transitions to the states of increasing orbital angular momentum at increasing photon energies, while the different parity of the photoelectron partial waves for the 1σ_g and 1σ_u shells causes the shift of interference modulation for these shells by π. Due to this, the cross section ratio 1σ_g/1σ_u observed experimentally is greatly enhanced, although generally the interference modulation is expected to be relatively small. And the sum of the contributions from the 1σ_g and 1σ_u shells will not display any CF

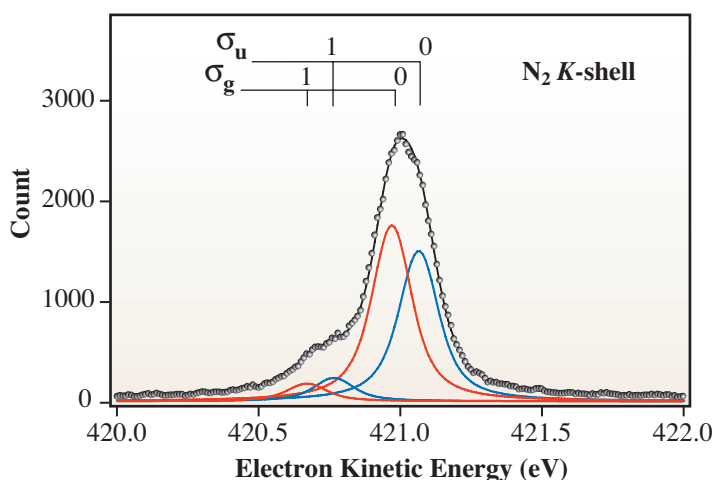


Fig. 1. Photoelectron spectrum at photon energy of 831 eV, parallel to the polarization vector.

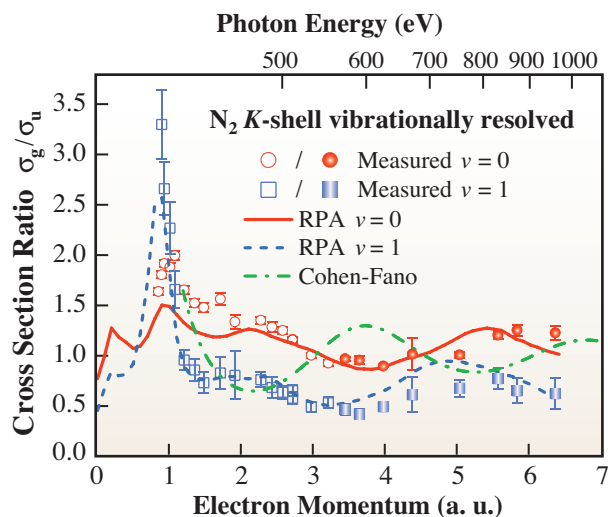


Fig. 2. Comparison of experimental cross section ratios with *ab initio* calculations.

oscillations.

From another point of view, at high electron kinetic energy with the plane wave approximation, the original CF interference pattern is shown in Fig. 2 as the dash-dotted line. Apparently, the phase of the interference pattern is shifted from both the experimental and *ab initio* ones.

To explain this disagreement, we take the sum over the vibrational components for the experimental spectra and obtain the ratios as shown in Fig. 3. By

the multiple scattering theory, the Cohen-Fano interference pattern is shifted by twice the phase of the photoelectron scattering by the neighboring N atom. Taking the reported value of phase shift, we obtain the dash-dotted line in Fig. 3. The agreement with the experiment is improved in comparison with the original Cohen-Fano approach but it is still far from being fair. We can obtain the improved value of the scattering phase using a least-squares fitting, shown as the solid line in Fig. 3.

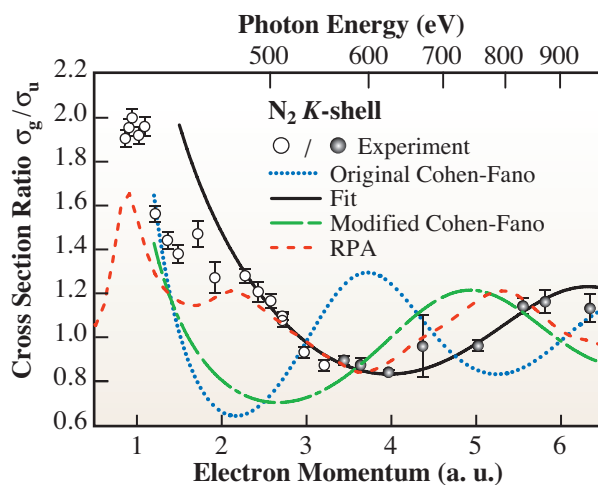


Fig. 3. Ratios of $1\sigma_g$ and $1\sigma_u$ partial photoionization cross sections of N_2 summed over the vibrational components.

X.-J. Liu*, N.A. Cherepkov, F. Gel'mukhanov and K. Ueda

Institute of Multidisciplinary Research for Advanced Materials, Tohoku University

*E-mail: xiliu@mail.tagen.tohoku.ac.jp

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