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Journal Title: Journal de Physique Colloques
Journal Volume: 48
Journal Issue: C9
Journal Month/Date:
Journal Year: 1987
Pages: 951+
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RESONANCES IN NEAR-THRESHOLD X-RAY PHOTOABSORPTION OF INNER SHELLS

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Résumé - Nous avons mesuré au Laboratoire de Brookhaven (NSLS) et à Stanford (SSRL) des sections efficaces absolues de photoabsorption au moyen de rayonnements de synchrotron près du seuil et pour un large éventail d'énergie (20 eV - 3 keV). Des données de transmissions recueillies pour des cibles multi-couches aux propriétés bien définies nous ont permis de déterminer les sections efficaces absolues avec une incertitude globale de l'ordre de 10% et pour une résolution sur l'énergie meilleure que 0.2%. Nous présentons quelques exemplaires de nos résultats.

Abstract - Synchrotron radiation measurements of near-threshold and broad-range (20 eV - 3 keV) absolute photoabsorption cross sections were made at the Brookhaven National Laboratory (NSLS) and at Stanford (SSRL). Transmission data for well-characterized multilayer foils provided absolute cross sections with 10% overall uncertainties and better than 0.2% energy resolution. Several examples of our results are presented.

The interaction of electromagnetic radiation with matter is of fundamental significance, as well as of great importance in numerous applications. In the soft x-ray range, photoabsorption is the dominant interaction process. Yet our knowledge of the photoabsorption cross sections in the soft x-ray range, over the bulk of the periodic table, is demonstrably poor[1]. Thus, at the Lawrence Livermore National Laboratory, we have embarked on a program to measure reliable absolute photoabsorption cross sections for a broad range of elements. In this paper, we report on some details of our experiment, along with some examples of our results.

The radiation sources used for these measurements were the Stanford Synchrotron Radiation Laboratory (SSRL) and the Brookhaven National Synchrotron Light Source (NSLS) electron storage rings which allowed us to make measurements from 20 eV to several keV. Using well-characterized ultra-thin targets in transmission, and continuously tunable monoenergetic beams of soft x-ray radiation, absolute cross section measurements were made with error bars of approximately $\pm 10\%$.

Key to reducing cross section uncertainties from factors of two or more to 10%, were the developments of monoenergetic VUV and soft x-ray synchrotron beam lines and the fabrication of extremely stable, pure, multilayer targets. Also, significant backgrounds were greatly reduced by taking redundant measurements to verify the self-consistency of our data. To this end, we had available several samples per element (in varying thicknesses), a dozen prefilterers, up to three mirror/grating combinations per monochromator, overlapping wavelength regions, and a total of four different monochromators at NSLS and SSRL.

The use of judiciously selected prefilterers in conjunction with various mirror and grating combinations allows us to greatly reduce significant backgrounds from high order harmonics and scattered light. These backgrounds limit the useful continuous energy region for a given unfiltered mirror/grating combination. Errors generated by not taking this into account have introduced discrepancies of a factor of two or more in previous measurements.

Further, we developed extremely stable, strong, free-standing foils with carbon surface layers and interlayers of varying thicknesses from 50 Å to 500 Å. The surface layers of carbon minimize oxidation which has contributed significant errors for previous measurements. The foils were made in pairs, with one foil having only layers of carbon and the other foil having the elemental foil in the multilayer configuration. Layer thicknesses were designed to minimize interference effects from Bragg diffraction. The carbon-only and multilayer targets were made simultaneously and measured separately. The foils were characterized for their purity, weight-per-unit area and elemental thickness using very accurate balances, ion back-scattering and particle-induced x-ray emission.

For transmission measurements, the NSLS U14A plane grating monochromator was used, along with toroidal grating, double multilayer crystal, and spherical grating monochromators at SSRL. In addition, we designed and built instrumentation that enabled us to continuously monitor the incident beam, to interchange up to six prefilterers, and to translate each of several samples into the target position. To measure the transmitted soft x-ray photons at NSLS, we designed a three element photodiode that permitted selection of pure metal cathodes of vapor-deposited gold, and diamond-turned Al or Cr, in order to take advantage of the metallic surface with the optimum response in each spectral region. Photodiode currents were measured with electrometers. The energy scale was calibrated using photoemission along with known absorption edges; our energy resolution was better than 0.2%. A more detailed description of the experiment will be presented elsewhere [2].

As a first example, our measured photoabsorption cross section for amorphous carbon, near the K-edge, is shown in Figure 1, along with a comparison with the theoretical relativistic Hartree-Slater (RHS) result of Scofield [3] and the compilation of Henke [4] as well as an LLNL extension and upgrade of Ref. 4 (CSLOW). From Figure 1 it is clear that the experimental results show a broad threshold feature which is not predicted by RHS theory or included in the latest compilations. Excellent agreement, however, is found both above and below this threshold enhancement. Our tentative explanation is that the enhancement is due to a molecular shape resonance [5], although autoionizing transitions from the 1s state to unoccupied valence bands in the solid are also possible. This point must be investigated further in order to understand the mechanism for this phenomenon. It is interesting to note, however, that even for K-shell photoabsorption for so light an element, as yet unexplained effects occur. Note further, that this represents the first absolute measurement of the photoabsorption cross section of carbon in the vicinity of the K-edge.

As a next example, the photoabsorption cross section for uranium, in the region of the 5d-5f giant resonance, is shown in Figure 2 along with RHS theory [3], the Henke compilation [4], and the local density random phase approximation (LDRPA) calculation of Wendin [6]; the Henke compilation in this region is based on the experimental work of Cukier et al [7]. The outstanding feature of the experimental result is the strong, broad absorption feature maximizing at about 115 eV, along with the smaller one at about 101 eV. Agreement with the LDRPA

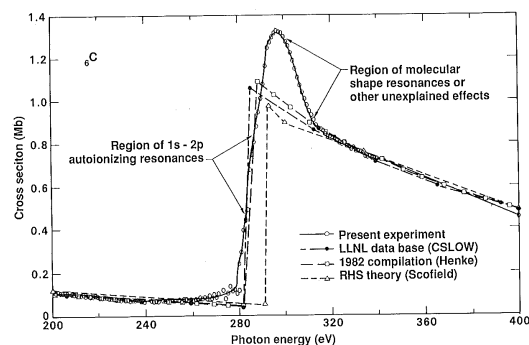


Fig. 1. Experimental photoabsorption cross section for amorphous carbon in the vicinity of the K-edge compared with theoretical and compiled results. See text for details of the latter curves.

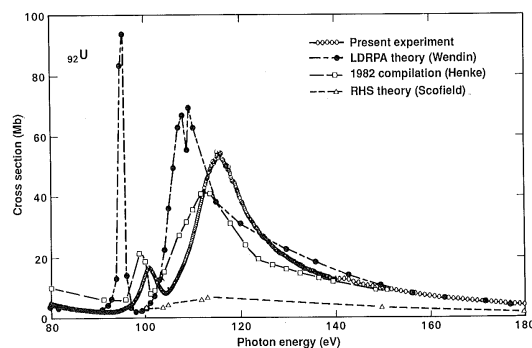


Fig. 2. Experimental photoabsorption cross section for uranium in the region of the 5d-5f resonance compared with theoretical and compiled results. See text for details of the latter curves.

calculation [6] and the experiment of Ref. [7] is good qualitatively, indicating our qualitative understanding of the process, but is still lacking quantitatively; it is, however, clear from Figure 2 that the calculation [6] is in significantly better agreement with the present experimental values than with the earlier measured values [7]. On the other hand, the single particle RHS calculation, which does not include autoionization or other many-body effects, completely fails to reproduce this resonance even qualitatively.

Physically, this structure is due to the very strong $\Delta n = 0$ transitions to an unfilled f-subshell; similar resonances are in evidence in the lanthanides, with unfilled 4f-subshells, and other actinides with unfilled 5f's [1]. The phenomenon was first explained in lanthanum [8]. Basically, we believe that the process hinges on the strength of the 5d-5f transitions, although shape resonance effects could contribute also. The process could proceed as a 5d-5f absorption, followed by an autoionizing decay; or as a 5f-eg absorption, strongly modified by interchannel coupling with the 5d-5f transitions. Owing to the open f-subshell, there are a myriad of possible optically allowed 5d⁴5f⁴ final multiplets; the strength of the interactions between these multiplets makes the calculations very difficult because many-body effects are so important [6]. Furthermore, the fact that the experiment was performed on a solid, while the calculation was done for a free atom, could lead to some discrepancies. However, this should not be a large effect since both the 5d and most of the 5f wave functions are localized and not involved in the binding of the solid.

Finally, we emphasize that only a small selection of our recent photoabsorption measurements using synchrotron radiation are presented here. These results, which include elements as light as Be ($Z = 4$) and as heavy as Th ($Z = 90$), show numerous new structures and enhancements in the vicinity of inner-shell thresholds. Further reports will be presented elsewhere.

This work was supported by the Lawrence Livermore National Laboratory under the auspices of the U. S. Department of Energy Contract No. W-7405-Eng-48, by U. S. Department of Energy (Office of Basic Energy Sciences) Contract No. DE-AC02-76CH00016 and by U. S. Army Research Office Contract No. DAAL 03-86-K-0085.

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