

Measurement of the L Absorption Spectra of Xenon*

TAKESHI WATANABE

Laboratory of Atomic and Solid State Physics and Department of Physics,
Cornell University, Ithaca, New York

(Received 7 October 1964)

The x-ray L_I , L_{II} , and L_{III} absorption spectra of gaseous xenon were measured with a two-crystal x-ray spectrometer. Absolute values of the absorption coefficients were determined on both sides of each edge. It was found that the L_{II} and L_{III} spectra have similar structural characteristics at the edge, each having a resonance absorption peak, while the absorption coefficient at the L_I edge increases smoothly and does not exhibit the absorption peak. The jump ratios were found to be 1.12, 1.38, and 2.60 for the L_I , L_{II} , and L_{III} edges, respectively. An estimate was made of the oscillator strengths for the bound-bound transitions.

I. INTRODUCTION

THE gross features of the L_{III} absorption edge of gaseous xenon have been investigated by Brogren¹ and Barinskii.² Both made some qualitative analyses by considering the similarity between the x-ray excited xenon spectrum and the optically excited cesium spectrum. However, there is a distinct difference in the experimental shapes of these two measurements, and neither of these measurements gives the absolute value of the absorption coefficients which could be quantitatively compared with theoretical calculations.

The study of the L absorption edges of neighboring elements has been carried out by several investigators. Nordfors³ has investigated elements from silver to tellurium, and determined the jump ratio at the L_I , L_{II} , and L_{III} edges for these elements. Parratt,⁴ Meisel and Nefedow,⁵ and Blokhin and Sachenko⁶ have made systematic analyses of the L_{III} state width with empirical formulas, and their results are consistent with each other. However, between the two experimental values of the L_{III} state width estimated by Brogren and Barinskii for xenon there exists a discrepancy which is more than the experimental uncertainty.

The difficulty in a direct measurement of the state width in an absorption spectrum arises from the fact that the width itself is greater than the energy separation between two adjacent empty bound orbitals outside the filled $5p$ orbital, and as a consequence the total absorption spectrum is given as the composite of several (in principle) resonance absorption lines. Moreover, since the initial states of the L_{II} and L_{III} absorption have p -type symmetry, the final state should be either d or s type. Therefore, although the absorption due to the p -to- d transition is usually an order of magnitude

greater than that of the p to s transition, both types of transitions contribute to the total absorption spectrum.

In the present study the mass absorption coefficients were measured near the L_I , L_{II} , and L_{III} edges of xenon with a two-crystal x-ray spectrometer. The jump ratios were obtained at each edge, and they are consistent with the results of Nordfors³ for neighboring elements. With the assumptions that the oscillator density of the continuum matches smoothly the fictitious oscillator density below the absorption edge and that the energies of x-ray excited states can be approximated by those of the optically excited state of cesium, the oscillator strengths of some of the L to bound-state transitions were estimated.

II. EXPERIMENTAL

The apparatus in the present experiment consists of, in addition to the two-crystal spectrometer, a regulated dc voltage supply to the x-ray tube, an x-ray tube current regulator, and a system for measuring x-ray intensities. The details of the instrument can be found elsewhere.⁷ In order to lower the spurious counts due to the secondary x rays from the second crystal, quartz crystals are used with the (1101) reflection planes. The dispersions are 21.10, 24.06, and 27.56''/eV at the L_I , L_{II} , and L_{III} edges, respectively.

The x-ray tube has a platinum target, and continuous x-ray radiation is used in the experiment. The x-ray plate voltage and the x-ray current are about 17 ± 0.02 kV and 50 ± 0.05 mA, respectively. The x-ray tube is separated from the main vacuum tank of the spectrometer by a beryllium foil and the pressure in the tube is kept at less than 10^{-7} mm Hg. With these operational conditions the x-ray intensity, after reflection from the second crystal, is about 150 counts/sec with no absorber.

The Xe absorber, which could be alternately inserted in or removed from the x-ray beam, is located between the x-ray tube and the first crystal. The absorbing path length is 2.54 cm, and the xenon pressure in the cell is controlled during each run. The pressure is kept at 100 mm Hg for the L_I and L_{II} absorption edges, at 60 mm Hg for the L_{III} edge. With a laboratory-made

* Supported by the U. S. Air Force Office of Scientific Research, by the National Science Foundation, and by the Advanced Research Projects Agency through the Materials Science Center at Cornell University.

¹ G. Brogren, *Nova Acta Regiae Soc. Sci. Upsalensis* **14**, No. 4 (1948).

² R. W. Barinskii, *Bull. Acad. Sci. USSR* **25**, 958 (1961) (in English).

³ B. Nordfors, *Arkiv. Fysik* **20**, 25 (1961).

⁴ L. G. Parratt, *Rev. Mod. Phys.* **31**, 616 (1959).

⁵ A. Meisel and W. Nefedow, *Ann. Physik* **7**, 48 (1961).

⁶ M. A. Blokhin and V. P. Sachenko, *Izv. Akad. Nauk. USSR* **21**, 1343 (1957).

⁷ H. W. Schnopper, Ph.D. thesis, Cornell University, 1962 (to be published).

TABLE I. Xenon L jump ratios.

	This work	Nordfors
$r(I,II)$	1.12 ± 0.1	1.15
$r(II,III)$	1.32 ± 0.1	1.38
$r(III,M)$	2.36 ± 0.2	...

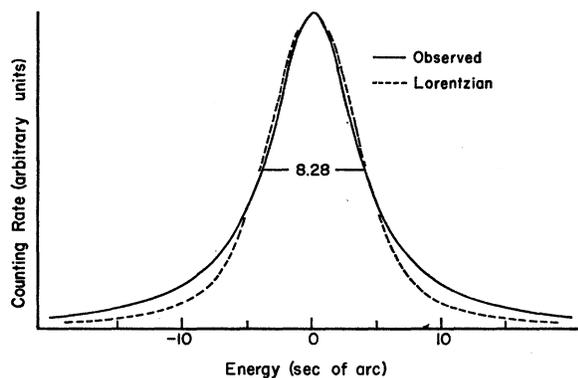
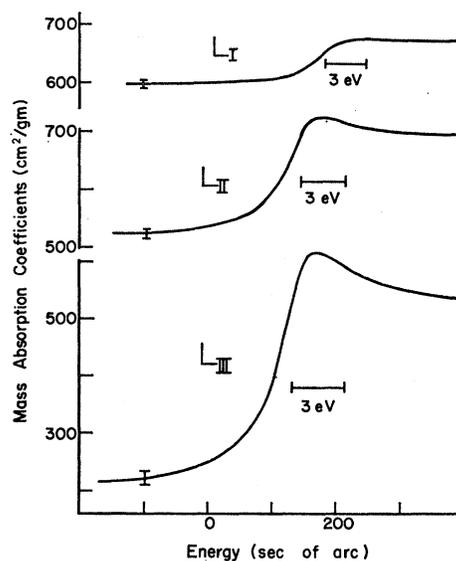
flow proportional counter mentioned below the direct and transmitted x-ray intensities, I_0 and I , are measured alternately. Correction is made for the absorption in the container window (1-mil-thick Be foil), which amounts to about 6% in the present experiment.

A gas flow proportional counter with 90% argon and 10% methane is used as the detector. The counter gas pressure is kept at 660 mm Hg, and with this pressure about 95% of the incident x rays are absorbed in the chosen energy range. The counter voltage is maintained at about 1800 V so that the main portion of the pulse height distribution is between 15 and 35 V. The electronic gain in the linear amplifier is about 6000.

As an approximation for the instrumental window function, the $(1, -1)$ rocking curve is recorded. A typical curve at the L_{II} absorption edge ($\lambda = 2.43 \text{ \AA}$) is shown in Fig. 1, from which the full window width at half-maximum is estimated to be 0.34 eV. For comparison, the Lorentzian curve with the same width and height as the $(1, -1)$ curve is also shown. For the L_I and L_{III} edges the widths, defined here as the energy interval between the $\frac{1}{4}$ and $\frac{3}{4}$ points, were found to be about 0.30 and 0.37 eV, respectively.

The measurements were made every 5" in the vicinity of the L_I , L_{II} , and L_{III} edges with a counting time of 200 sec for each intensity measurement. Alternate measurements of I and I_0 reduced the effects of fluctuations in counting rate due to changes in the counter gas gain caused by temperature change. For each of the three edges three runs were made, and the average values of I_0/I were calculated for each energy setting.

After corrections were made for the absorption in the gas cell window, the mass absorption coefficients were

FIG. 1. The $(1, -1)$ rocking curve at $\lambda = 2.43 \text{ \AA}$.FIG. 2. The xenon L absorption spectra near L_I , L_{II} , and L_{III} edges.

plotted against energy (Fig. 2). The standard error (standard deviation divided by the square root of the number of counts) is indicated by the short vertical bars.

III. ANALYSIS AND DISCUSSION

Jump Ratio

The absorption jump is determined by the ratio of the absorption coefficients just above and below the absorption edge. At the L_I edge the ratio $r(II,I)$ is estimated to be 1.12 ± 0.1 , which is consistent with the values for the neighboring elements.³ At the L_{II} and L_{III} edges the ratios are taken at the flat portions of the spectra, and $r(III,II)$ and $r(M,III)$ are estimated to be 1.32 ± 0.1 and 2.36 ± 0.2 . If the peak values of these spectra were used, the ratios would become 1.38 ± 0.1 and 2.60 ± 0.2 , respectively.

The previous experimental values for the neighboring elements obtained by Nordfors³ are about 5% higher than the present values for the L_I and L_{II} jumps, and about 15% higher for the L_{III} jump. However, his measurements were carried out with solid samples. For comparison his mean values and the present values of $r(II,I)$ and $r(III,II)$ are tabulated in Table I.

Oscillator Strength

In the one-electron model the x-ray absorption by a rare-gas atom is interpreted as the excitation of an inner electron to some bound orbital outside the closed electron core or to an unbound hyperbolic orbital (i.e., to a continuum state). For the resonance absorption to the n th bound orbital an oscillator strength f_n is assigned, and, in principle, there exists an infinite number of oscillator strengths, one for each resonance absorption transition, below the continuum edge.

TABLE II. ΔE_{np} and f_{np} associated with the L_I absorption.

np	$7p$	$8p$	$9p$	$10p$
ΔE_n	0.0651	0.0265	0.0136	0.0080
f_{np}	0.37×10^{-4}	0.15×10^{-4}	0.79×10^{-5}	0.45×10^{-5}

Introduce the fictitious oscillator density below the edge by

$$\left(\frac{\partial f}{\partial E}\right)_n = \frac{2f_n}{|E_{n-1} - E_{n+1}|} \quad E \leq 0,$$

where E_n is the energy of the n th bound orbital. It can be proved⁸ analytically that for hydrogenic wave functions this quantity matches smoothly the oscillator density of the continuum at the absorption edge, i.e.,

$$\lim_{n \rightarrow \infty} \frac{\partial}{\partial E} \ln \left(\frac{\partial f}{\partial E} \right)_n = \frac{\partial}{\partial E} \ln \left(\frac{\partial f}{\partial E} \right)_{\text{continuum}} \Big|_{E=0^+}.$$

Cooper⁹ and Watanabe¹⁰ showed that this continuity relation is satisfied approximately for an argon atom, and Payne and Levinger¹¹ found that in the nonrelativistic case the oscillator density for the L , M , and Q shells of lead extrapolates smoothly to the continuum value at the series limit given by the Stobbe formula. Although the absorption line is smeared due mainly to the lifetime of the inner hole, if the linewidth is large compared with the energy separation between two neighboring empty orbitals outside the closed xenon core, the continuity relation still holds at the threshold.

In the present case it is assumed that the energies E_n of the x-ray excited states of xenon can be approximated by those of the corresponding optical excited states of cesium.¹² Then, the use of the continuity relation gives the oscillator strength f_n approximately as

$$f_n \simeq 1.24 \times 10^{17} (A/N_0) \mu_m \Delta E_n,$$

where A and N_0 are the atomic weight of xenon and Avogadro's number. ΔE_n is given by

$$\Delta E_n = \frac{1}{2} |E_{n-1} - E_{n+1}| \text{ (in Ry)}.$$

The mass absorption coefficient μ_m for the L_I absorption is estimated to be 70 cm²/g. The final positions of the L_I x-ray excited electron are np orbitals with $n \geq 6$. Therefore, $2\Delta E_{np}$'s are approximated by the energy

difference between $5p^6(n-1)p$ and $5p^6(n+1)p$ configurations of a cesium atom. Some values are listed in Table II. With these values of ΔE_{np} some of the oscillator strengths associated with the L_I absorption are estimated and also listed in Table II.

The L_{II} and L_{III} absorption spectra are given by the composite of $p \rightarrow s$ and $p \rightarrow d$ transitions. Therefore, unless the relative intensity of these transitions is known fairly accurately, the above analysis is not applicable. The use of the hydrogenic wave function for both the initial and final states could give a rough estimate. However, since the calculation is very sensitive to choice of the effective nuclear charge and the validity of the use of this wave function is not certain, the estimate of the oscillator strength associated with L_{II} and L_{III} absorption is not made.

State Width

Based on the assumption that the resonant absorptions can be approximated by shifting the energy position of the edge, the L_I state width is estimated by fitting an arctangent curve to the L_I absorption spectrum near the edge. It is found to be 1.1 ± 0.3 eV, which is close to the value obtained by Brogren. As to the L_{II} and L_{III} state widths, it is difficult to obtain the unique values from these spectra. *Within the limit of the present analysis*, perhaps ± 0.7 eV, they are approximately 2 eV.

IV. CONCLUSION

The mass absorption coefficients of xenon were measured near the L_I , L_{II} , and L_{III} absorption edges. It was found that the L_{II} and L_{III} spectra have a broad resonance structure, while the L_I spectrum feature of the present measurement differs from the one obtained by the counter efficiency method (by Brogren¹) and supports Barinskii's photographic result.

The values of the absorption jump were experimentally determined and compared with those for the neighboring elements. Since the present sample consists entirely of free atoms, the present values should be useful for comparison with the atomic theoretical calculations.

Based on the simplified theoretical assumption, the oscillator strengths for some of the L_I x-ray absorption transitions were estimated. Although the assumption requires further justification, the method applied here is probably the only way to estimate the x-ray oscillator strength of those transitions with a short lifetime.

ACKNOWLEDGMENTS

The author wishes to thank Professor L. G. Parratt, Professor J. A. Krumhansl, and Professor C. Horie for stimulating and useful discussion on the subject.

⁸ Y. Sugiura, J. Phys. Radium 8, 113 (1927).

⁹ J. W. Cooper, Phys. Rev. 128, 681 (1962).

¹⁰ T. Watanabe, Ph.D. thesis, Cornell University, 1964 (to be published).

¹¹ W. B. Payne and J. Levinger, Phys. Rev. 101, 1020 (1956).

¹² C. E. Moore, Natl. Bur. Std. (U.S.) Circ. No. 467, Vol. III (1958).