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*High Resolution Measurements and  
Matrix Analysis of the  
Capture and Fission Cross Sections of  
 $^{239}\text{Np} + n$  from 1 to 600 eV*



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**LA-9756-MS**

**UC-34c**  
**Issued: May 1983**

# **High-Resolution Measurements and R-Matrix Analysis of the Total and Fission Cross Sections of $^{237}\text{Np} + \text{n}$ from 1 to 600 eV**

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HIGH-RESOLUTION MEASUREMENTS AND R-MATRIX ANALYSIS  
OF THE TOTAL AND FISSION CROSS SECTIONS  
OF  $^{237}\text{Np} + \text{n}$  FROM 1 to 600 eV

by

G. F. Auchampaugh, M. S. Moore, J. D. Moses, R. O. Nelson,  
C. E. Olsen, R. C. Extermann, N. W. Hill, and J. A. Harvey

ABSTRACT

High-resolution measurements of the total and fission cross sections of  $^{237}\text{Np} + \text{n}$  have been made using the pulsed-neutron facilities at the Oak Ridge Electron Linear Accelerator and at the Los Alamos Meson Physics Facility. The samples were cooled to liquid-nitrogen temperature. This report presents the total and fission cross sections from 1 to 600 eV as well as the parameters obtained from an R-matrix fit of these data.

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I. INTRODUCTION

Questions have been raised<sup>1</sup> concerning the nature of the coupling of Class II states to the Class I states in the subthreshold fission of  $^{238}\text{Np}$  that have not been adequately answered with existing data.<sup>2,3</sup> Although high-resolution fission-cross-section data exist, there are no total-cross-section data of comparable resolution. Total-cross-section data are very important for locating all Class I resonances and the Class II resonance near a Class II state. We have undertaken high-resolution measurements, with samples cooled to liquid-nitrogen temperature, of both the fission and the total cross sections of  $^{237}\text{Np}$  to provide higher quality data to test current models of Class II-to-Class I coupling and decay.

This report presents the experimental details of the neptunium measurements, the results of the R-matrix, multilevel analysis of the total and fission cross sections, and a tabulation of the resonance parameters. A discussion of the significance of these data and how well they fit various coupling models is presented elsewhere.<sup>4</sup>

II. EXPERIMENTAL DETAILS AND  
NORMALIZATION

A. Total-Cross-Section Measurements

Data were collected in two separate runs. In the first run at the Oak Ridge Electron Linear Accelerator (ORELA), two 12.7-mm-thick lithium-glass detectors were used: the first was edge-mounted on an RCA 8854 phototube and placed in the flight path at 78.017 m; the second was face-mounted and placed 78.203 m from the neutron target. The samples of  $^{237}\text{Np}$  metal were 9.906 mm in diameter and  $(25.83 \pm 0.17)$  and  $(168.73 \pm 0.68) \times 10^{18}$  atoms/mm<sup>2</sup> thick. We cooled the samples to liquid-nitrogen temperature ( $\sim 77$  K) to minimize Doppler resolution broadening. The accelerator repetition rate was 95 Hz, and the burst width was 36 ns. Filters of cadmium, 0.762 mm thick, and lead, 6.35 mm thick, eliminated overlap neutrons and reduced the effects of the gamma flash from the target. Useful data were collected for about 170 hours for each sample thickness.

The second run was made some months later, under the same conditions with certain exceptions. Only one

detector, the face-mounted detector at 78.203 m, and one sample, the thicker one, were used. The cadmium filter was replaced by one made of  $^{10}\text{B}$ , 4.5 mg/mm<sup>2</sup> thick, and the accelerator was operated with 35-ns bursts at a repetition rate of 1000 Hz. Data were collected over an 8-day period for about 100 hours.

### B. Fission-Cross-Section Measurements

We measured the fission cross sections at the Weapon Neutron Research (WNR) pulsed spallation neutron facility at Los Alamos. For these measurements, the nominal flight path was 30 m, and the samples were cooled to 84 K. The design and operation of the cryogenic ionization chamber used to measure the fission cross section has been described elsewhere.<sup>5</sup> The chamber contained 1.35 g of neptunium as  $\text{NpO}_2$ , deposited on 0.0127-mm-thick stainless steel backings, 0.042 g of  $^{235}\text{U}$  as  $\text{UO}_2$ , deposited on a similar backing, and 0.008 g of  $^6\text{Li}$  as  $^6\text{LiF}$ , deposited on a titanium backing 0.025 mm thick. The signal from the  $^6\text{Li}$  foil was used to establish the shape of the neutron flux; the signal from the  $^{235}\text{U}$  deposit was used to normalize the fission cross section. The accelerator beam had a burst width of 150 ns. The time digitizer had channel widths of 64 ns and covered the energy range from 1 to  $10^4$  eV.

### C. Fission-Cross-Section Normalization

Our normalization based on relative masses of the  $^{235}\text{U}$  and  $^{237}\text{Np}$  deposits suggested that the cross sections of Plattard et al.<sup>3</sup> are too low by a factor of 3. This conclusion is reinforced by the observation of resonances in a slight contaminant of  $^{239}\text{Pu}$  in our  $^{237}\text{Np}$  fission deposits. Samples of the  $^{237}\text{Np}$  material, from which the deposits were fabricated, were assayed by isotope dilution for  $^{239}\text{Pu}$  content; the results showed that the  $^{237}\text{Np}$  material contained  $571 \pm 29$  parts per million  $^{239}\text{Pu}$ . The expected ratio, if the normalization of Plattard et al.<sup>3</sup> were correct, is 188 parts per million. Finally, to resolve the question beyond any doubt, we made a high-energy measurement of the  $^{237}\text{Np}$ -to- $^{235}\text{U}$  ratio with the cryogenic fission chamber using the  $^{237}\text{Np}$ -to- $^{235}\text{U}$  cross-section ratio measured by Behrens<sup>6</sup> as the normalization standard. The results of this measurement showed that the normalization of the resonance data of Plattard et al.<sup>3</sup> is too low by a factor of  $2.91 \pm 0.08$ . Our normalization agrees within about 10% with that of Jiaocetti et al.<sup>7</sup> for

the strongest resonances in the 40-eV cluster and for other resonances where their sensitivity to gamma radiation is not important.

### III. R-MATRIX ANALYSIS

We analyzed the total and fission cross sections using the multilevel, R-matrix, search code MULTI.<sup>8</sup> Both cross sections were fitted simultaneously up to 600 eV. Particular attention was given to the regions around the 40- and 120-eV Class II states. A minimum of effort was spent fitting the region above 160 eV; thus, the quality of the fit above 160 eV is not as good as that at lower energies.

The success of a shape fit depends on adequately parameterizing the resolution function for the measurement. The resolution function must include the thermal motion of the atoms in the sample; neutron generation time in the moderator surrounding the target,  $t_m$ ; flight path uncertainties,  $\ell_f$ ; electron or proton burst width,  $t_b$ ; and the time per channel used to record the data. A careful analysis of the low-energy resonances in neptunium, where the Doppler width dominates the other components of the resolution width, revealed that a Gaussian function with a Doppler width corresponding to an effective temperature of 100 K adequately described the shape of the resonances. The e-folding width for the other components of the resolution width are given in Table I. The channel width was chosen to be at least one-third smaller than the total resolution width and was not included in the resolution function.

A better fit to the shape of the fission resonances at 120 eV was achieved by accounting for the exponential decay of the neutron pulse emitted from the moderator. Consequently, above 70 eV the resolution function for the fission data consisted of a convolution of a Gaussian

TABLE I. The e-Folding Widths for the Gaussian and Exponential Resolution Functions

$\Delta E$ (eV)	Total			Fission		
	$t_b$ (ns)	$\ell_f$ (cm)		$t_b$ (ns)	$\ell_f$ (cm)	$t_m$ (ns)
1-70	21.0	3.3		126.0	3.3	0.0
70-580	21.0	3.3		78.0	0.0	110.0

\*The width for the Gaussian function is equal to the quadrature of  $\ell_f$  (converted to time for a particular neutron energy) and  $t_b$ .

function with a width obtained from the quadrature of  $t_b$  and the Doppler width and an exponential function with an e-folding width of  $t_m$ .

The data were fitted using the least squares procedure with the residuals weighted by the statistical error on each point. In general, we varied the fission width, neutron width, and the resonance energy. Below about 60 eV, the capture width was varied for those resonances whose shape was sensitive to the capture width; otherwise it was fixed at 40.8 meV.

A one-fission-channel reduced R-matrix formalism was used to fit the shape of the cross section. The results of Kuiken et al.<sup>9</sup> along with the spin assignments of Keyworth et al.<sup>10</sup> suggested that the Class II state at 40 eV may have more than one K-component, which would require a two-fission-channel formalism. So we made both one- and two-channel fits to the resonances near 40 eV. However, the fit with two fission channels showed no improvement over that with one fission channel even in the valleys between the resonances. The total fission strength appeared to be divided proportionally between the two channels for the major resonances in the 40-eV cluster, which is what we would expect if the Class II state had more than one K-component. The poor quality of the fission data in the resonance valleys and the insensitivity of the fit to the number of fission channels discouraged further investigation with two fission channels.

The analysis of Plattard et al.<sup>3</sup> of the 40-eV cluster suggested a resonance at 39.7 eV with a fission width larger than that of the prominent resonance seen in the fission cross section at 39.9 eV. With our high-resolution total-cross-section data, we can observe the 39.7-eV resonance. However, we find the fission width of the 39.7-eV resonance to be 2.5  $\mu\text{eV}$  instead of 5060  $\mu\text{eV}$  (corrected for the normalization factor 2.91) reported by Plattard et al.<sup>3</sup> and the neutron width to be 100  $\mu\text{eV}$  instead of 62  $\mu\text{eV}$ . Most of the fission strength of the Class II state at 40 eV is contained in the resonance at 39.9 eV.

For the 39.7-eV resonance, we find an anomalously small capture width of about 21 meV that is about a factor of 2 smaller than the average capture width for  $^{237}\text{Np}$ . If the 39.7-eV resonance contains most of the Class II strength, then we would expect its capture width to be mostly Class II and, therefore, different from the capture width of the Class I resonances. But this resonance has essentially no fission strength and, furthermore, may not be a  $J = 3$  resonance. A reanalysis of the polarization data of Keyworth et al.<sup>10</sup> suggests a

resonance near 39.7 eV with a spin opposite to that of the 39.9-eV resonance. However, the fit to the total-cross-section data could not differentiate between the two spin possibilities for the 39.7-eV resonance. Therefore, the spin of this resonance is tentatively assigned  $J = 2$ .

The total- and fission-cross-section data from 1 to 600 eV are presented in Figs. 1 through 22. The data are presented unaveraged except where noted at the top of the figures. The final one-channel fit is represented by the solid line. The resonance parameters are listed in Table II. The uncertainties on the parameters given were obtained from the diagonal elements of the covariance matrix. They do not represent absolute errors but are more representative of the relative errors on the parameters, because systematic errors have not been included in the analysis. Using our high-resolution data and reanalyzing the data of Keyworth et al.,<sup>10</sup> we arrived at the spin assignments given in the last column. We have added some new spin assignments but have not changed those reported by Keyworth et al.<sup>10</sup>

## ACKNOWLEDGMENTS

We gratefully acknowledge the hospitality and assistance of the staff at ORELA, where the total-cross-section measurements were made, as well as the support of W. A. Teasdale, J. M. Anaya, B. L. Jacquez, and the WNR technical staff for the fission measurements. Thanks are also due to J. E. Rein and R. Abernathy for their analysis of the  $^{239}\text{Pu}$  content in the  $^{237}\text{Np}$  material, which provided an independent check of the data normalization.

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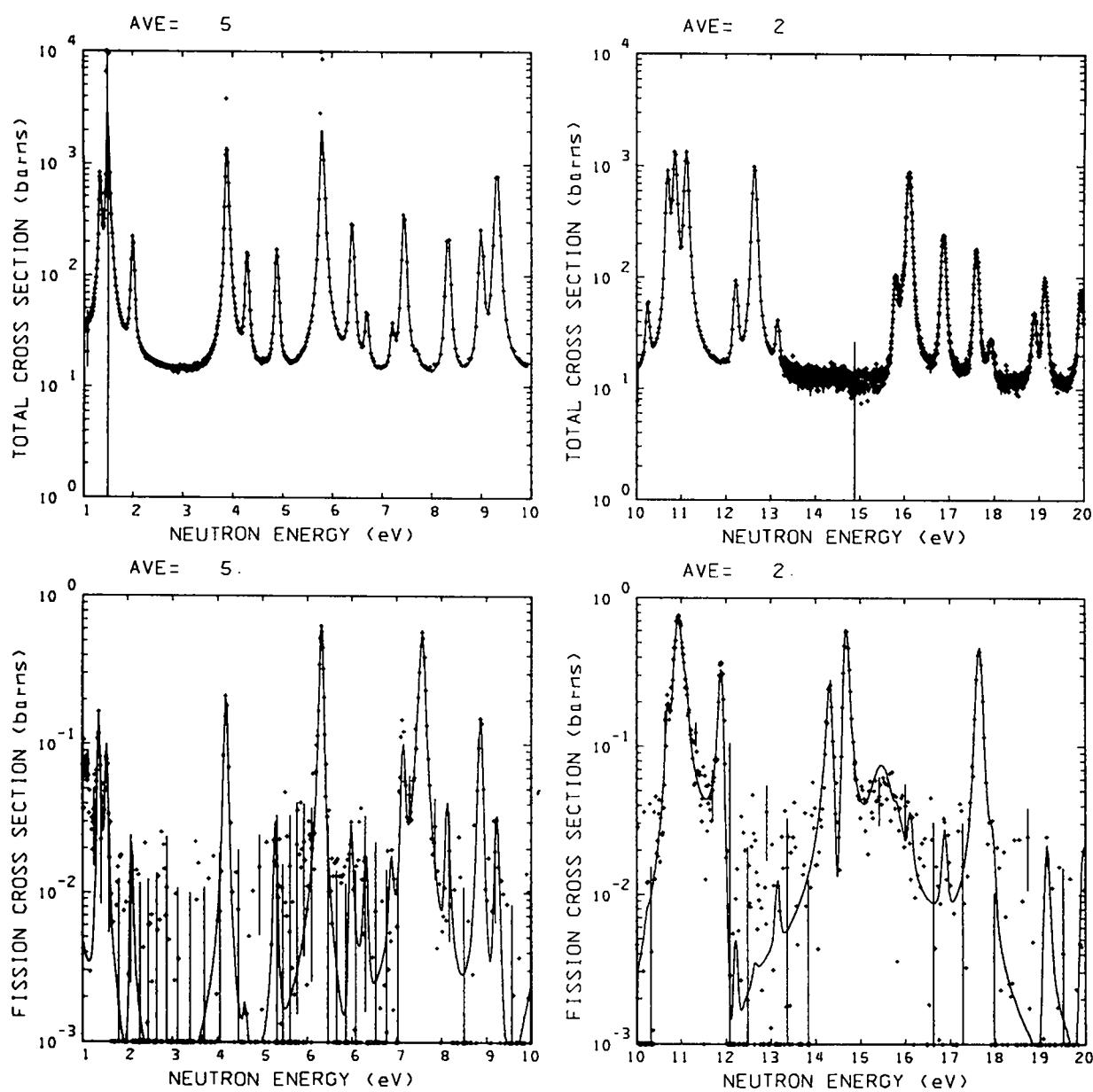


Fig. 1. Total and fission cross sections from 1 to 20 eV. AVE represents the number of points averaged. The statistical errors are represented by vertical lines.

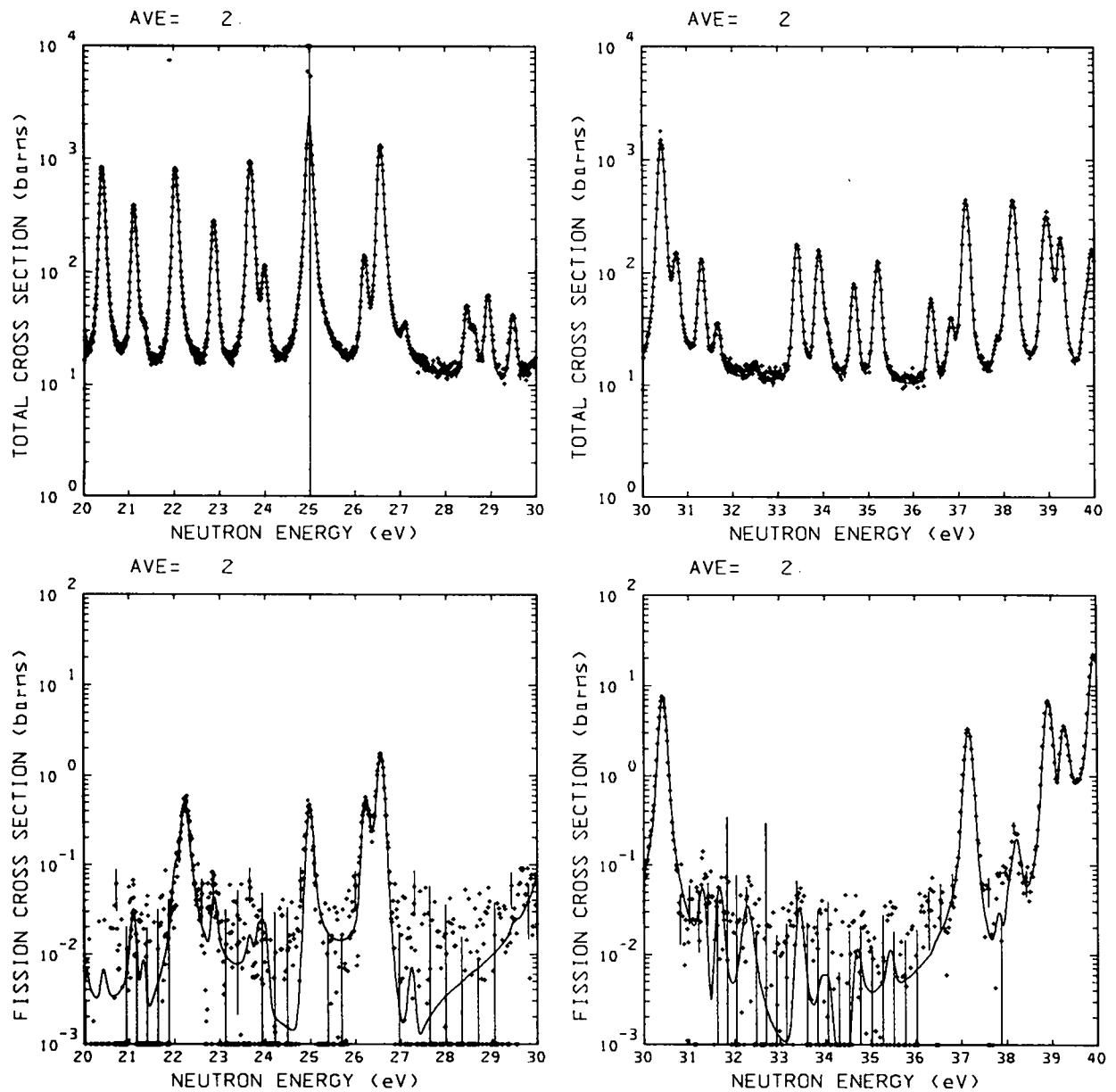


Fig. 2. Total and fission cross sections from 20 to 40 eV. AVE represents the number of points averaged. The statistical errors are represented by vertical lines.

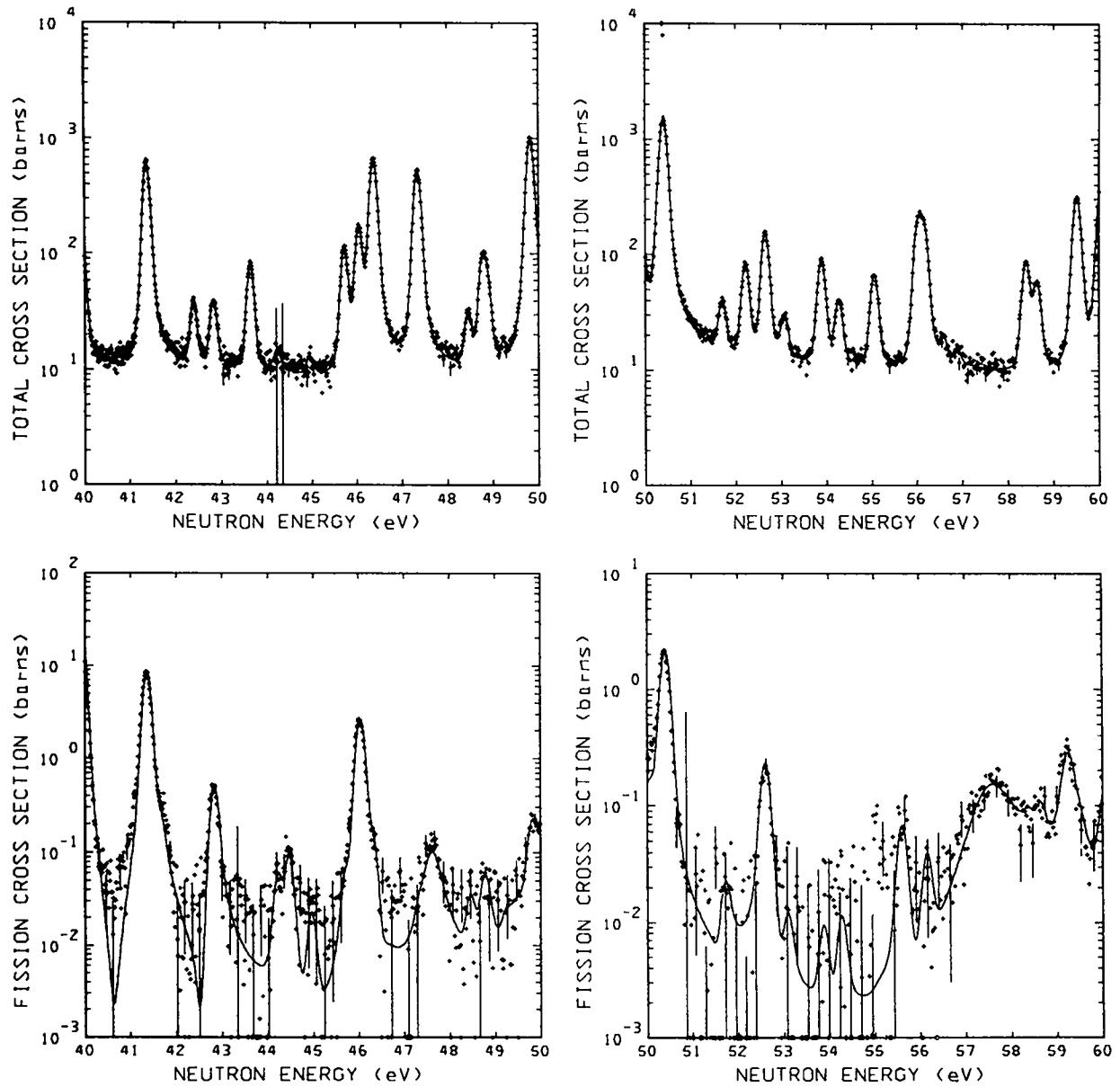


Fig. 3. Total and fission cross sections from 40 to 60 eV. The statistical errors are represented by vertical lines.

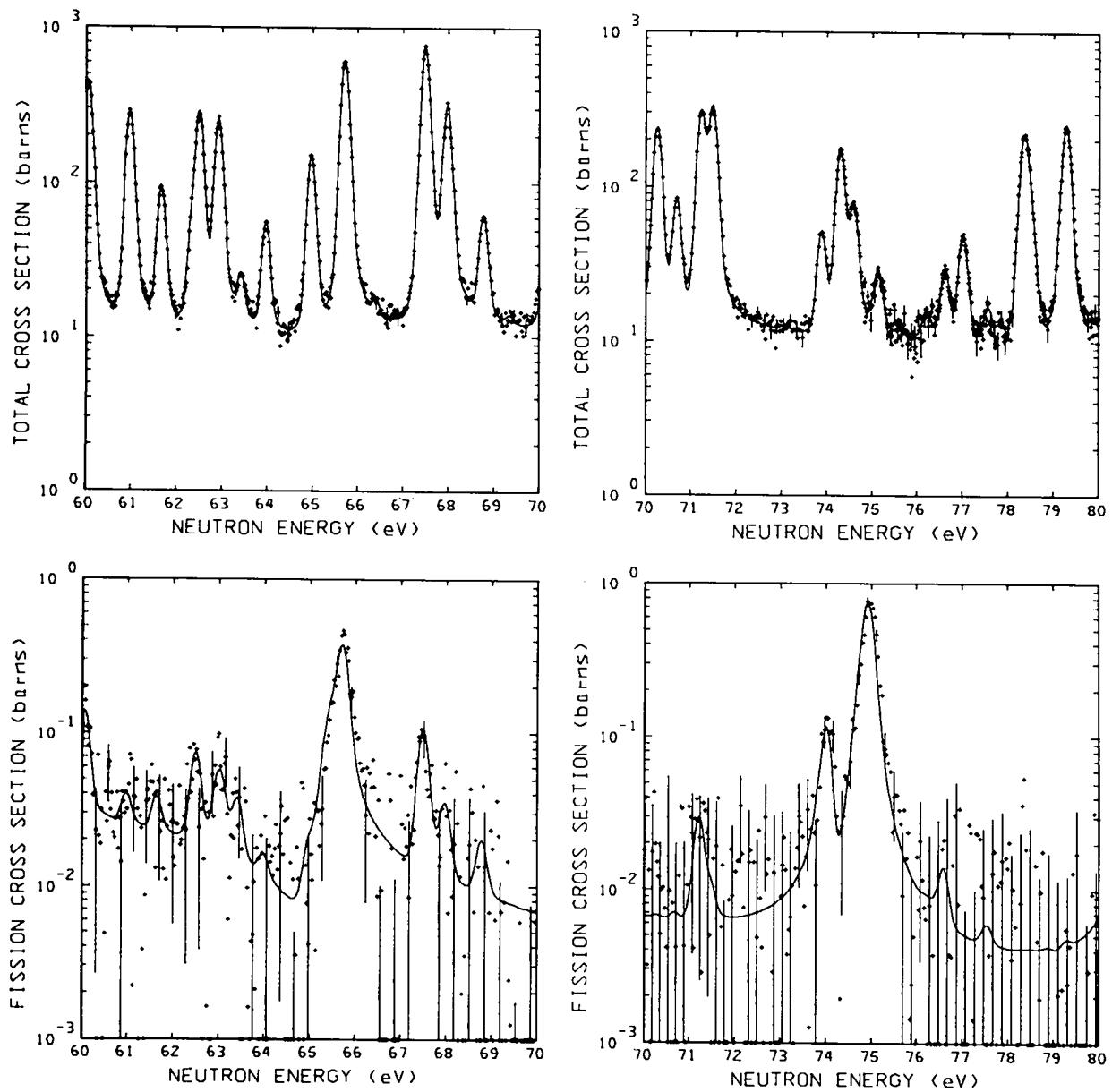


Fig. 4. Total and fission cross sections from 60 to 80 eV. The statistical errors are represented by vertical lines.

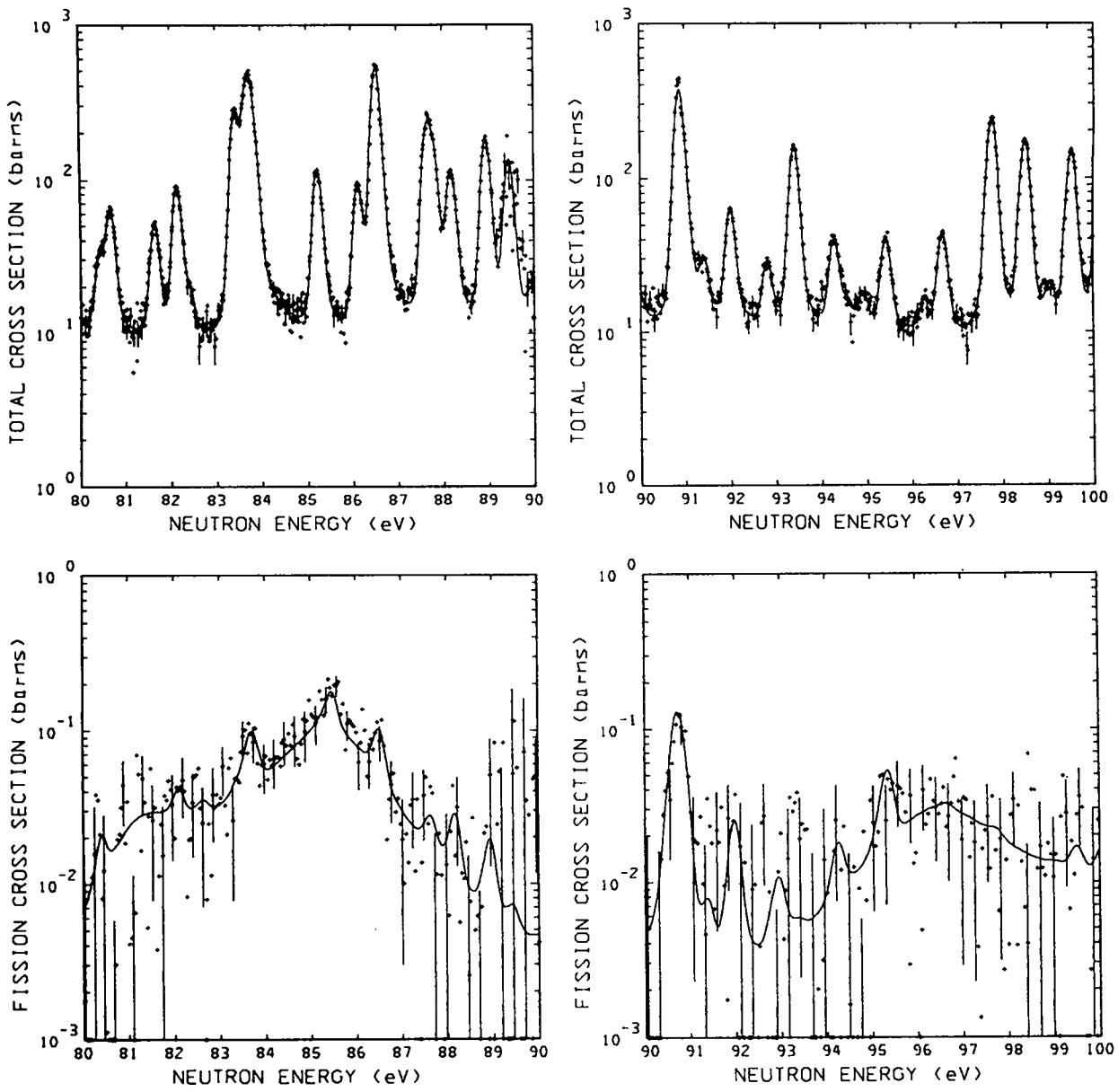


Fig. 5. Total and fission cross sections from 80 to 100 eV. The statistical errors are represented by vertical lines.

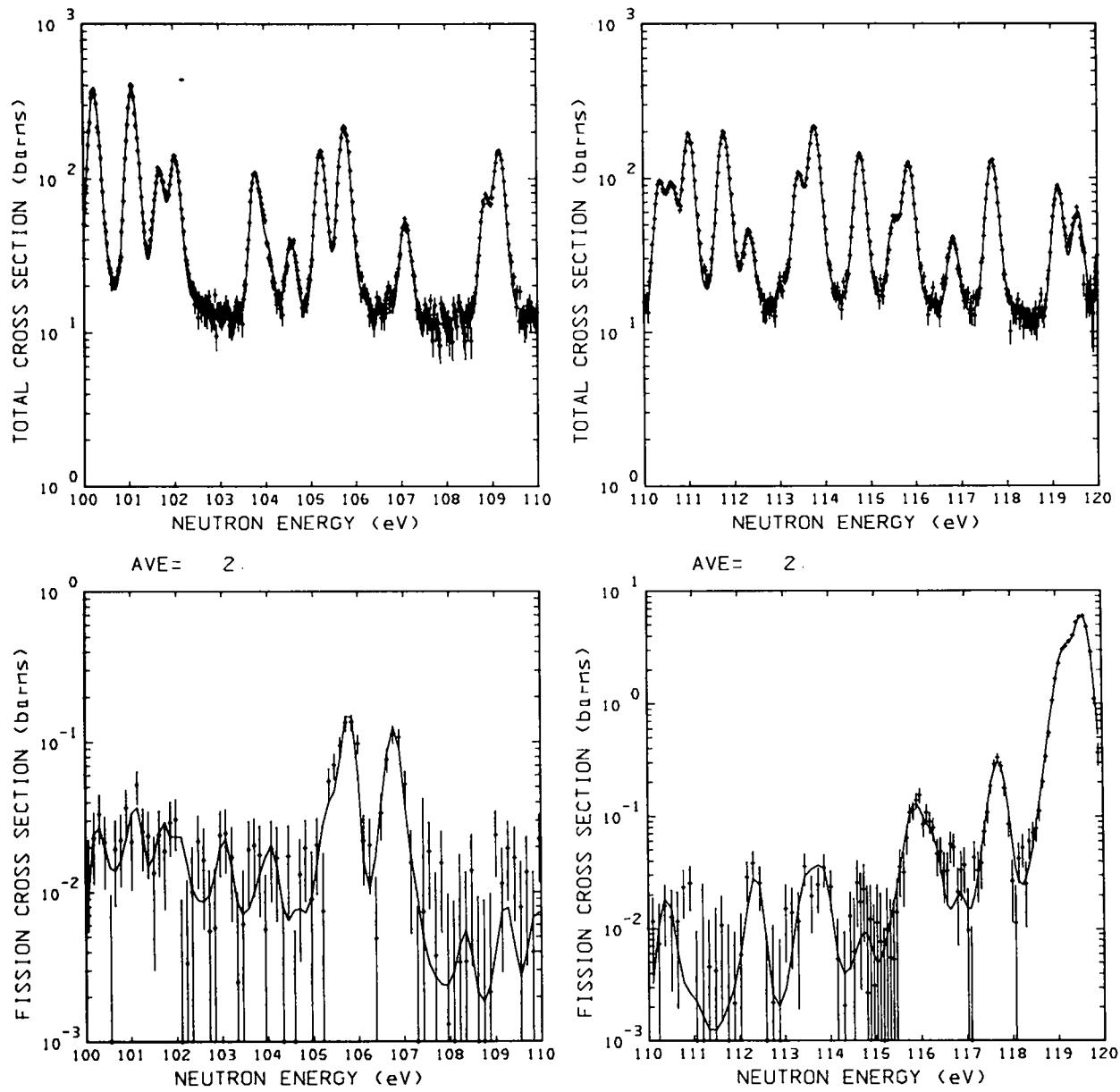


Fig. 6. Total and fission cross sections from 100 to 120 eV. AVE represents the number of points averaged. The statistical errors are represented by vertical lines.

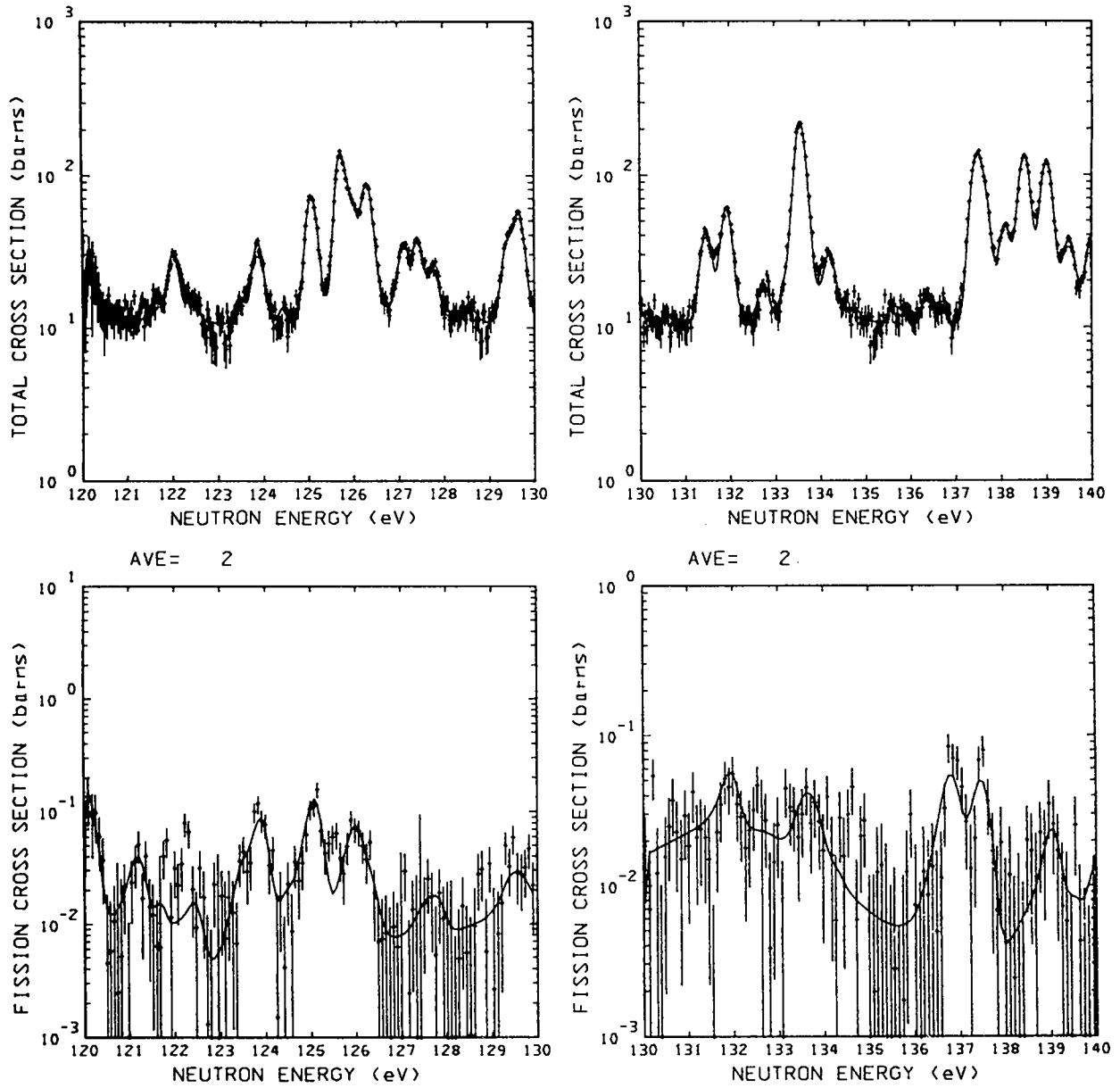


Fig. 7. Total and fission cross sections from 120 to 140 eV. AVE represents the number of points averaged. The statistical errors are represented by vertical lines.

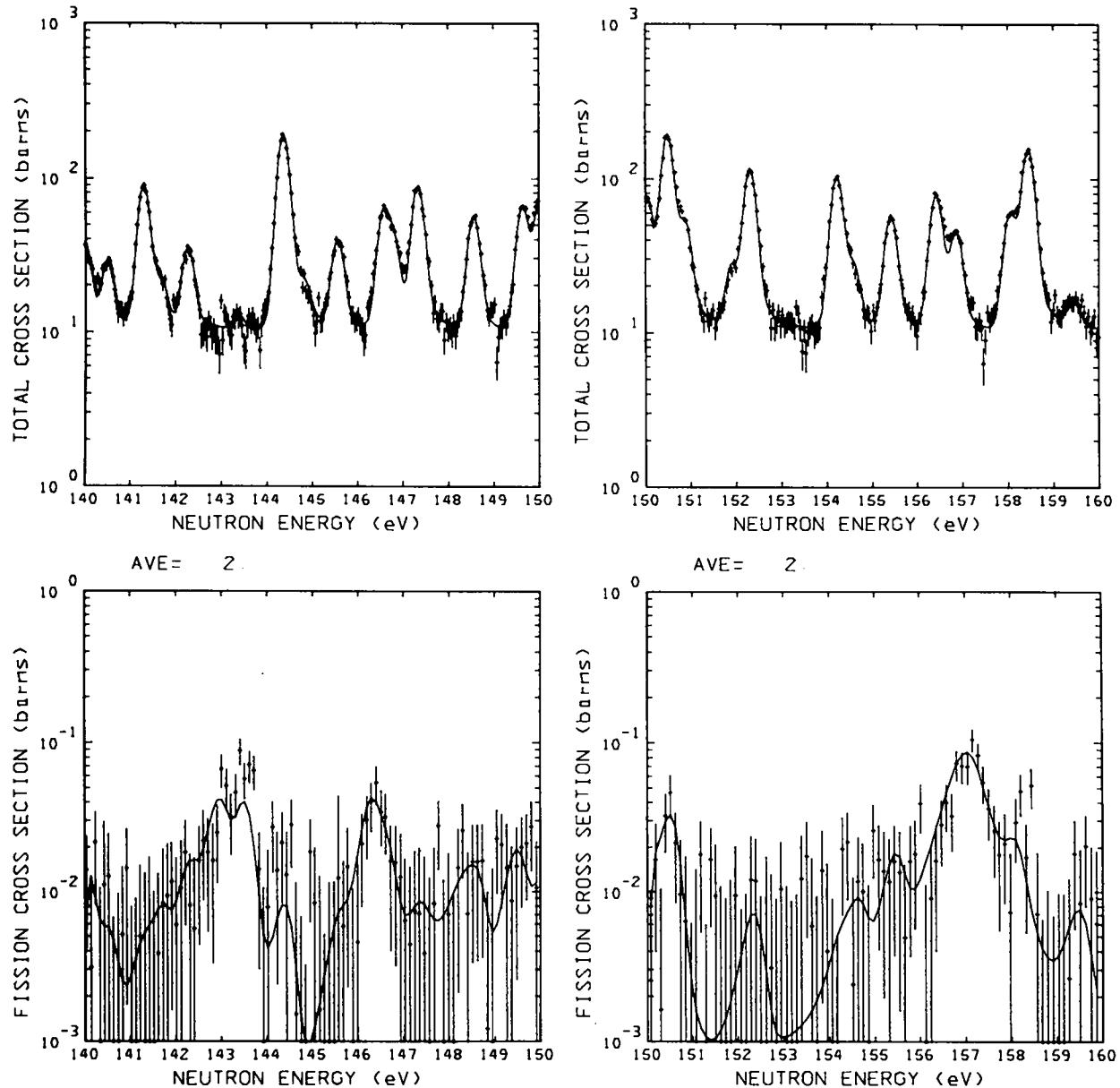


Fig. 8. Total and fission cross sections from 140 to 160 eV. AVE represents the number of points averaged. The statistical errors are represented by vertical lines.

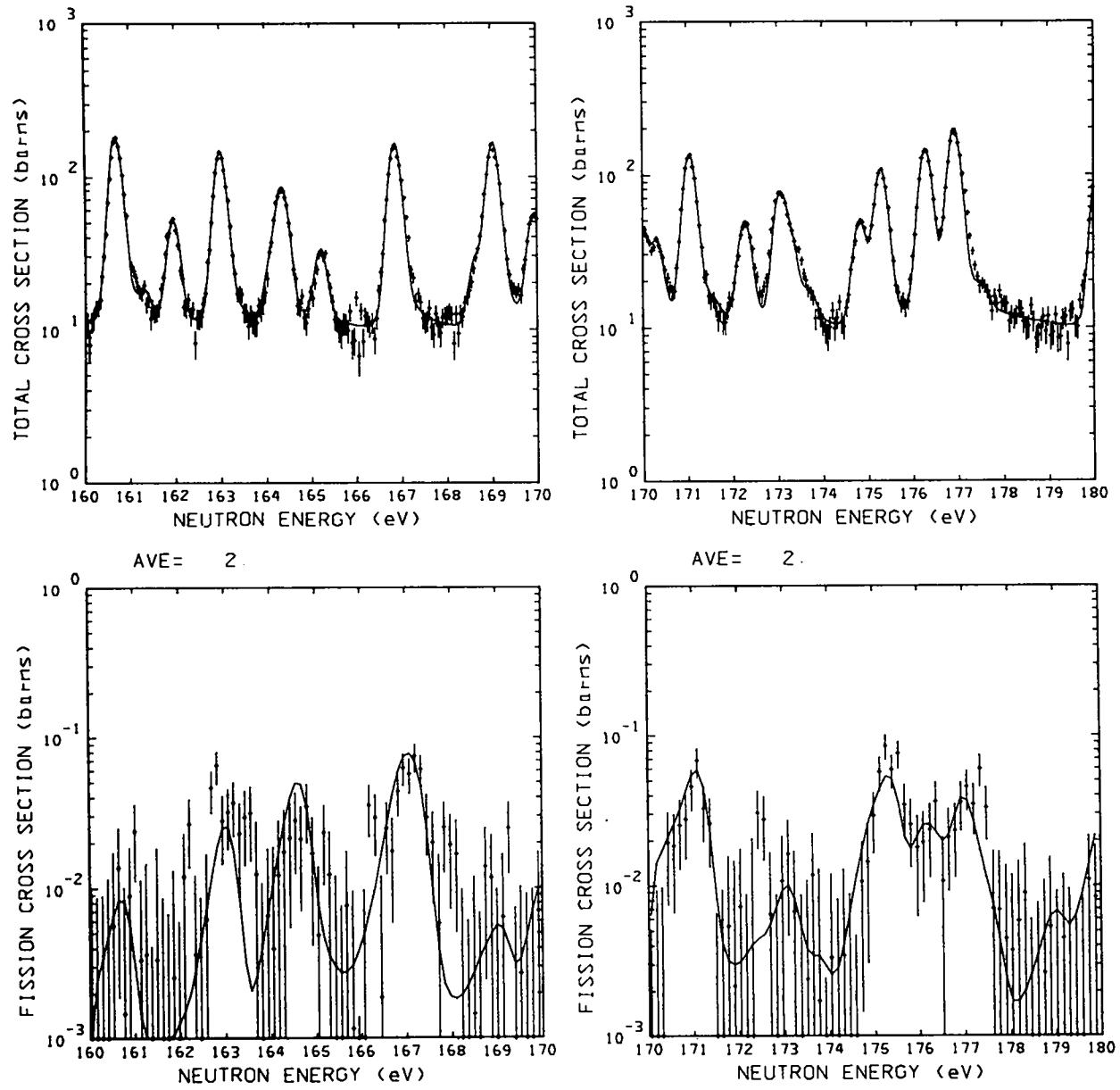


Fig. 9. Total and fission cross sections from 160 to 180 eV. AVE represents the number of points averaged. The statistical errors are represented by vertical lines.

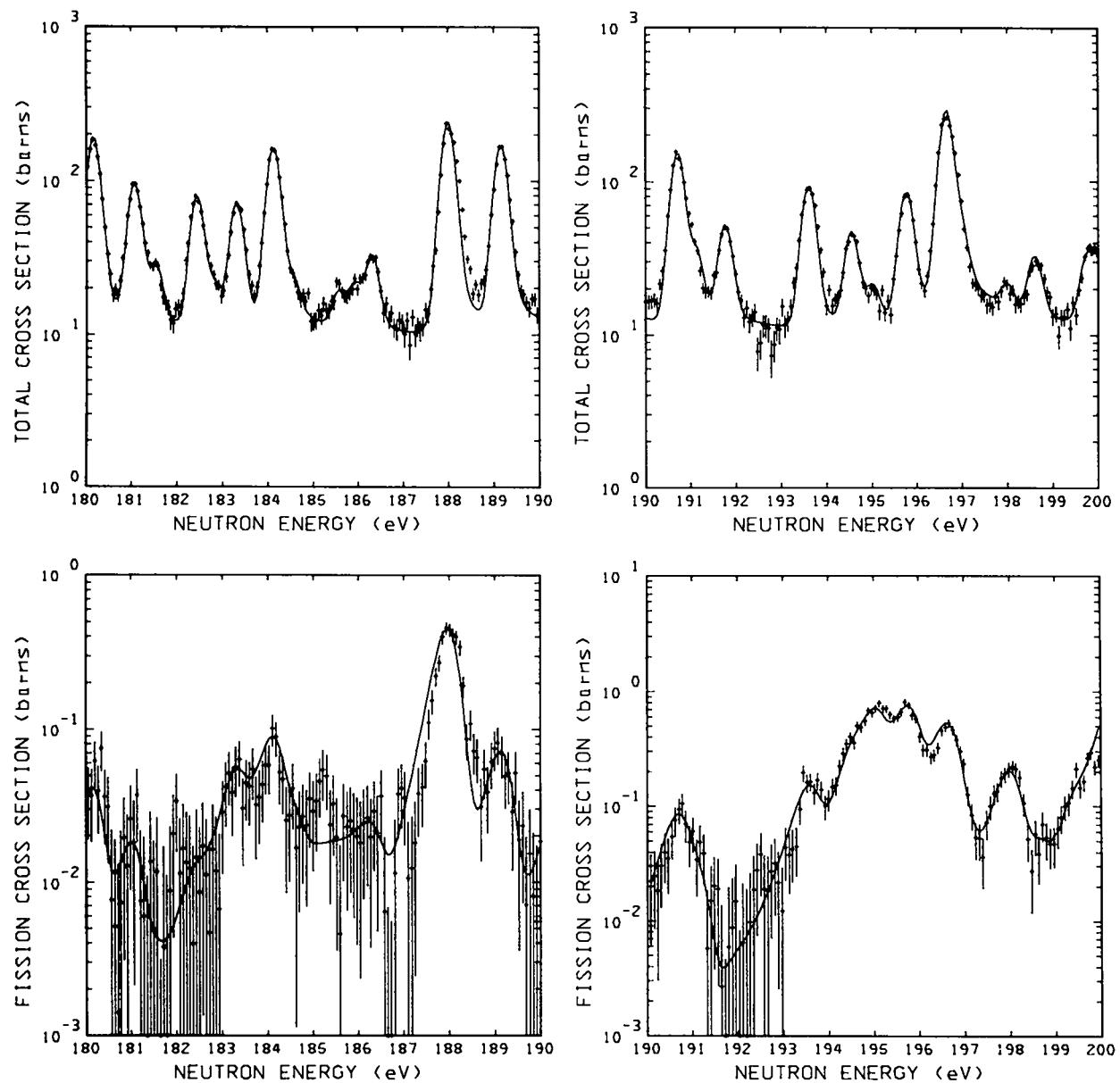


Fig. 10. Total and fission cross sections from 180 to 200 eV. The statistical errors are represented by vertical lines.

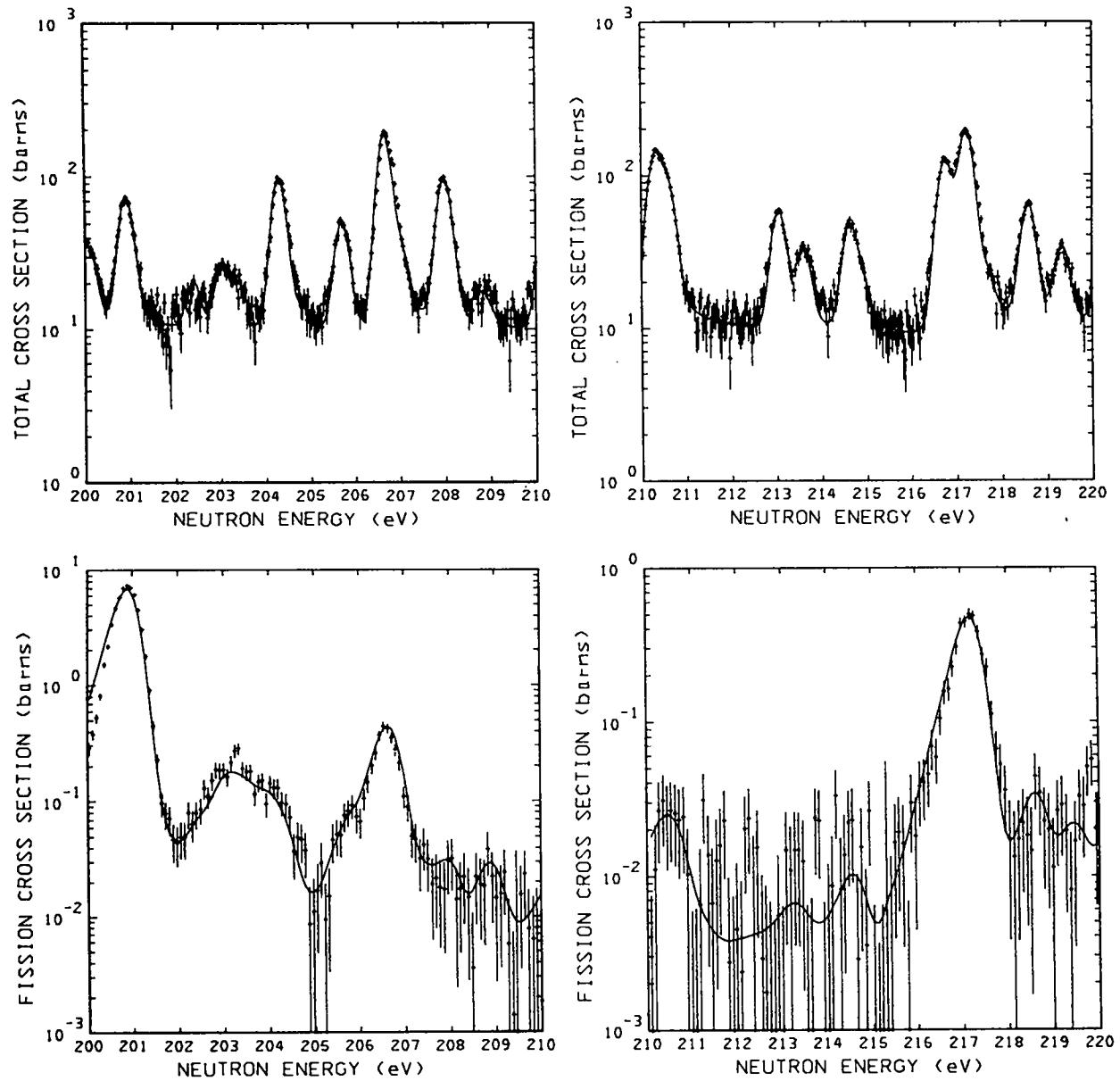


Fig. 11. Total and fission cross sections from 200 to 220 eV. The statistical errors are represented by vertical lines.

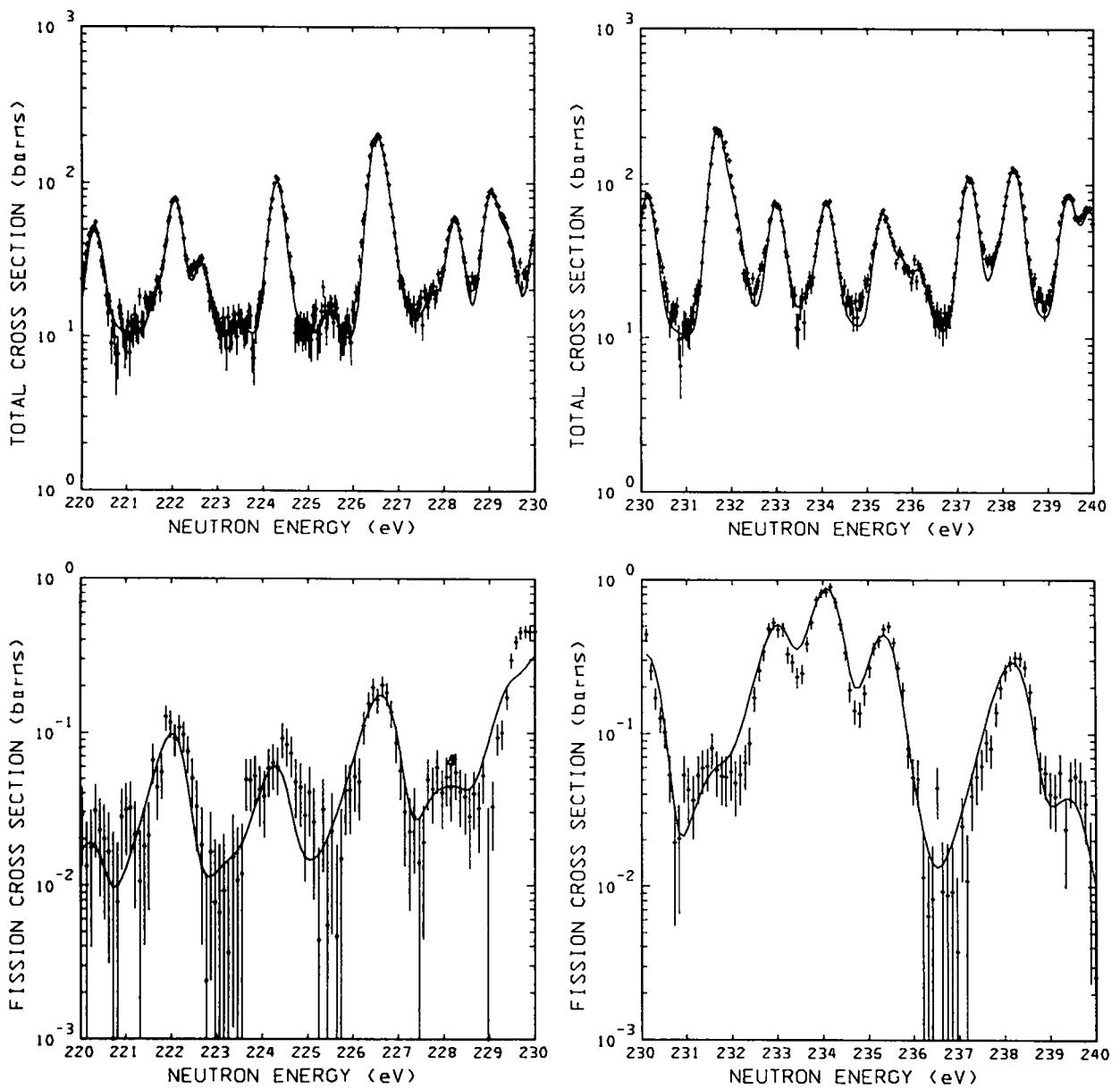


Fig. 12. Total and fission cross sections from 220 to 240 eV. The statistical errors are represented by vertical lines.

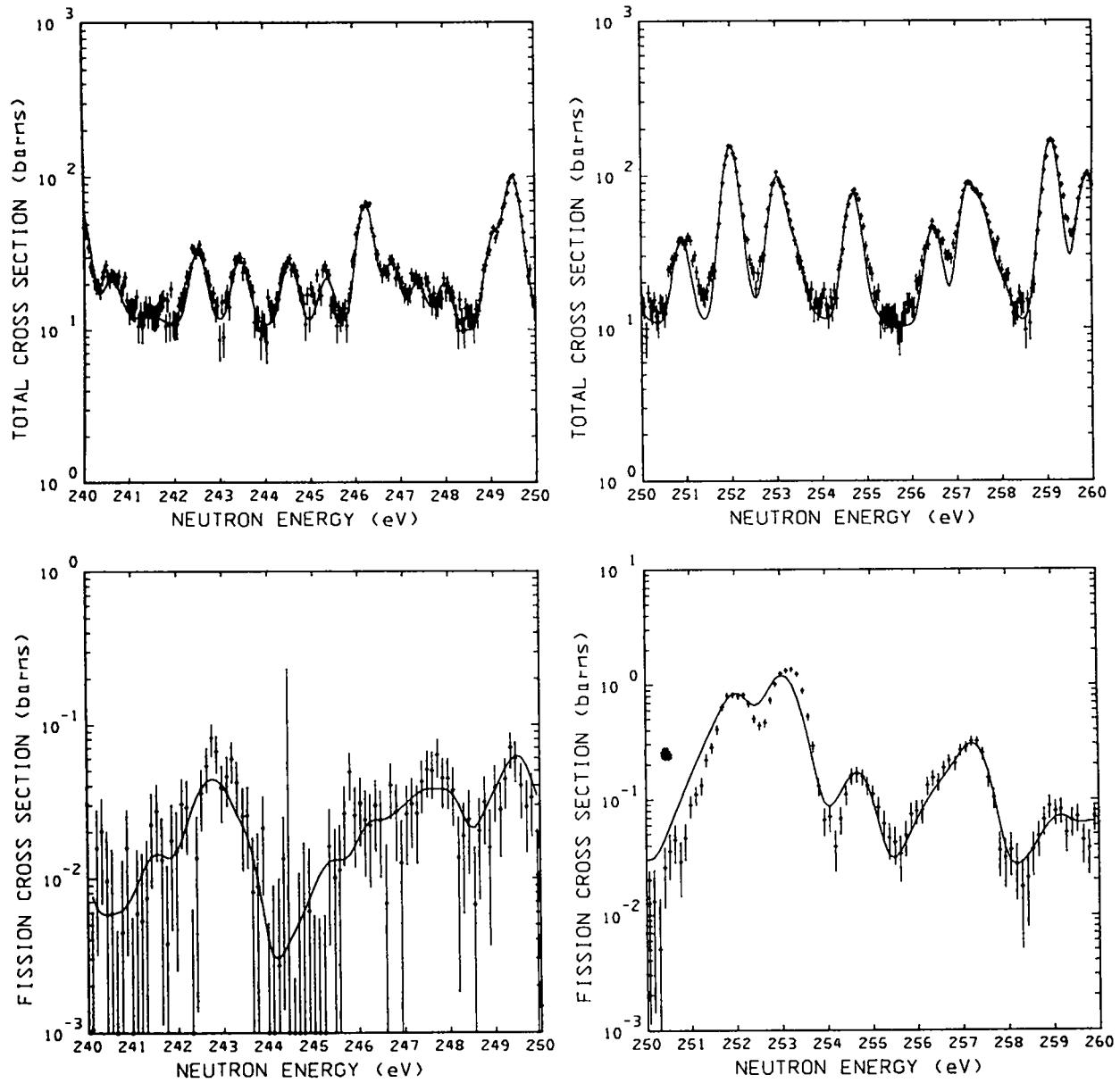


Fig. 13. Total and fission cross sections from 240 to 260 eV. The statistical errors are represented by vertical lines.

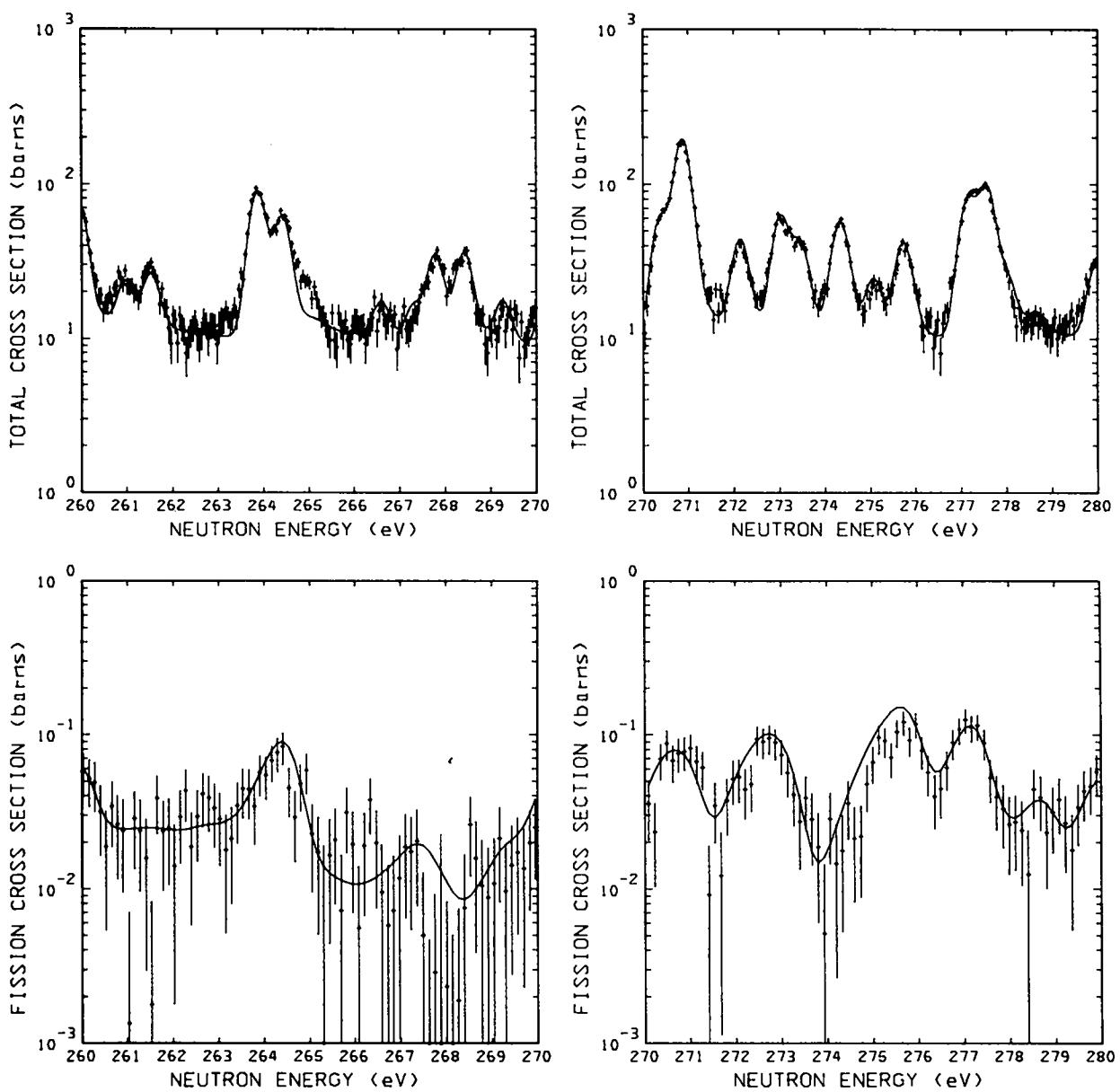


Fig. 14. Total and fission cross sections from 260 to 280 eV. The statistical errors are represented by vertical lines.

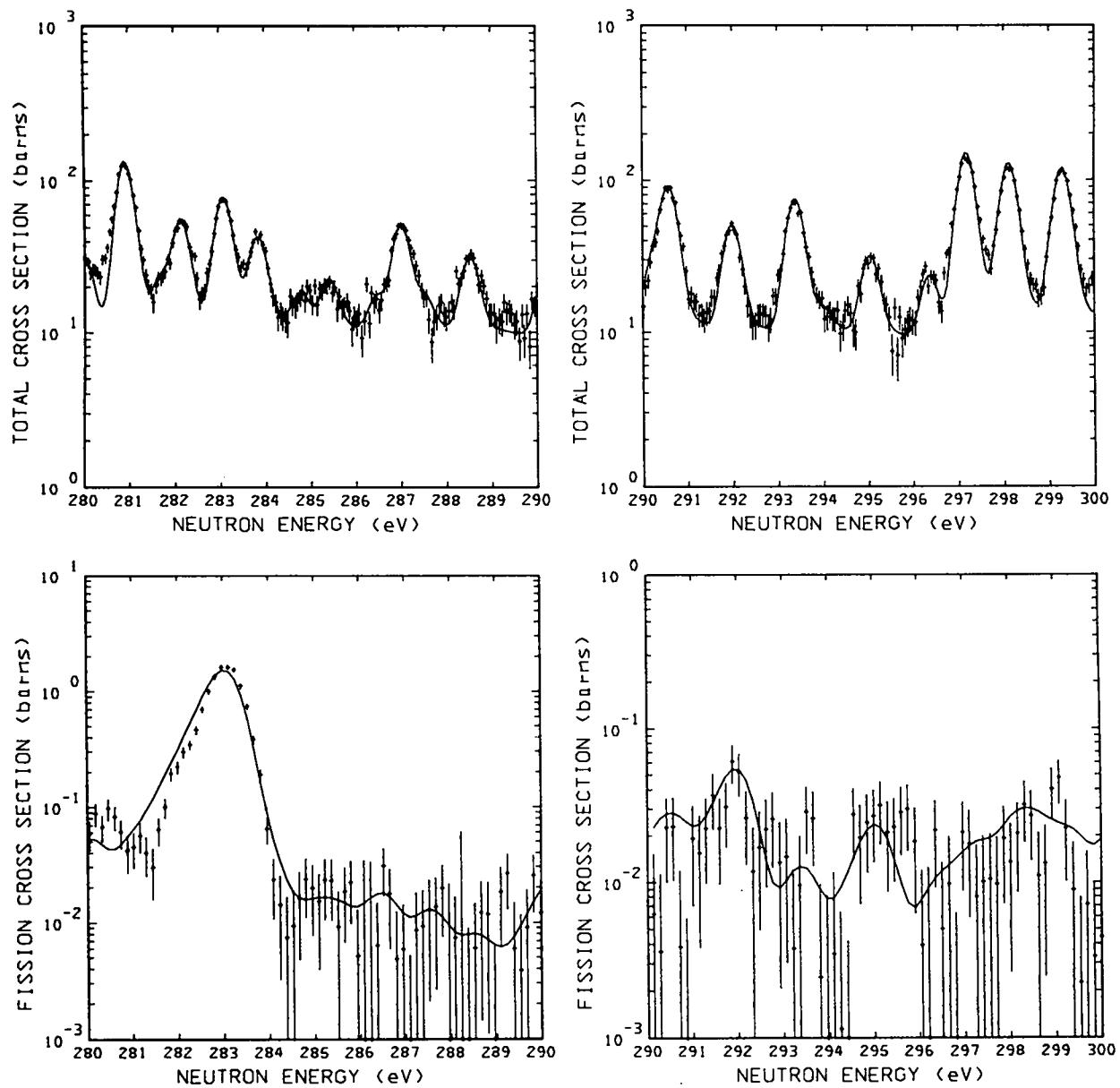


Fig. 15. Total and fission cross sections from 280 to 300 eV. The statistical errors are represented by vertical lines.

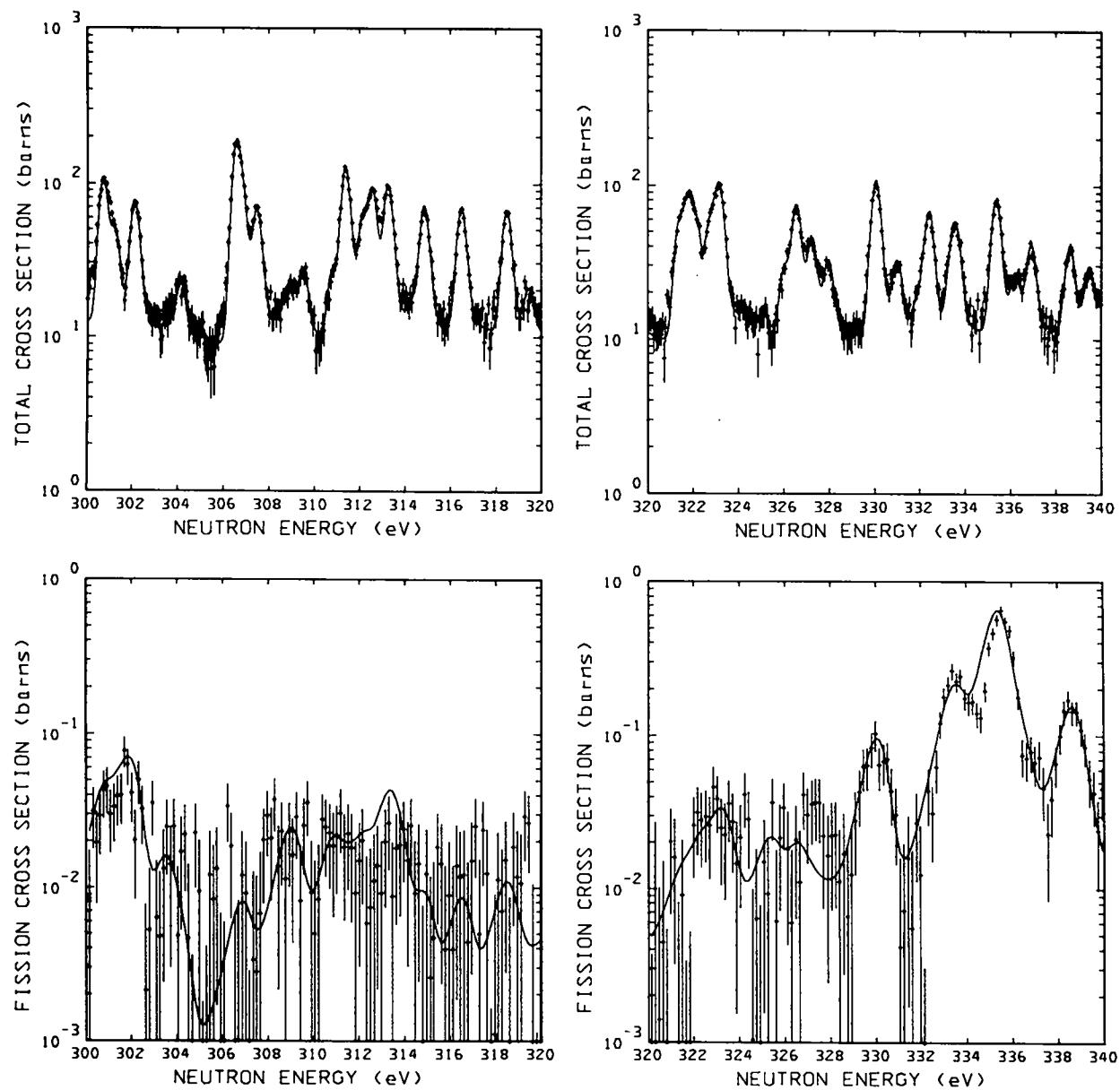


Fig. 16. Total and fission cross sections from 300 to 340 eV. The statistical errors are represented by vertical lines.

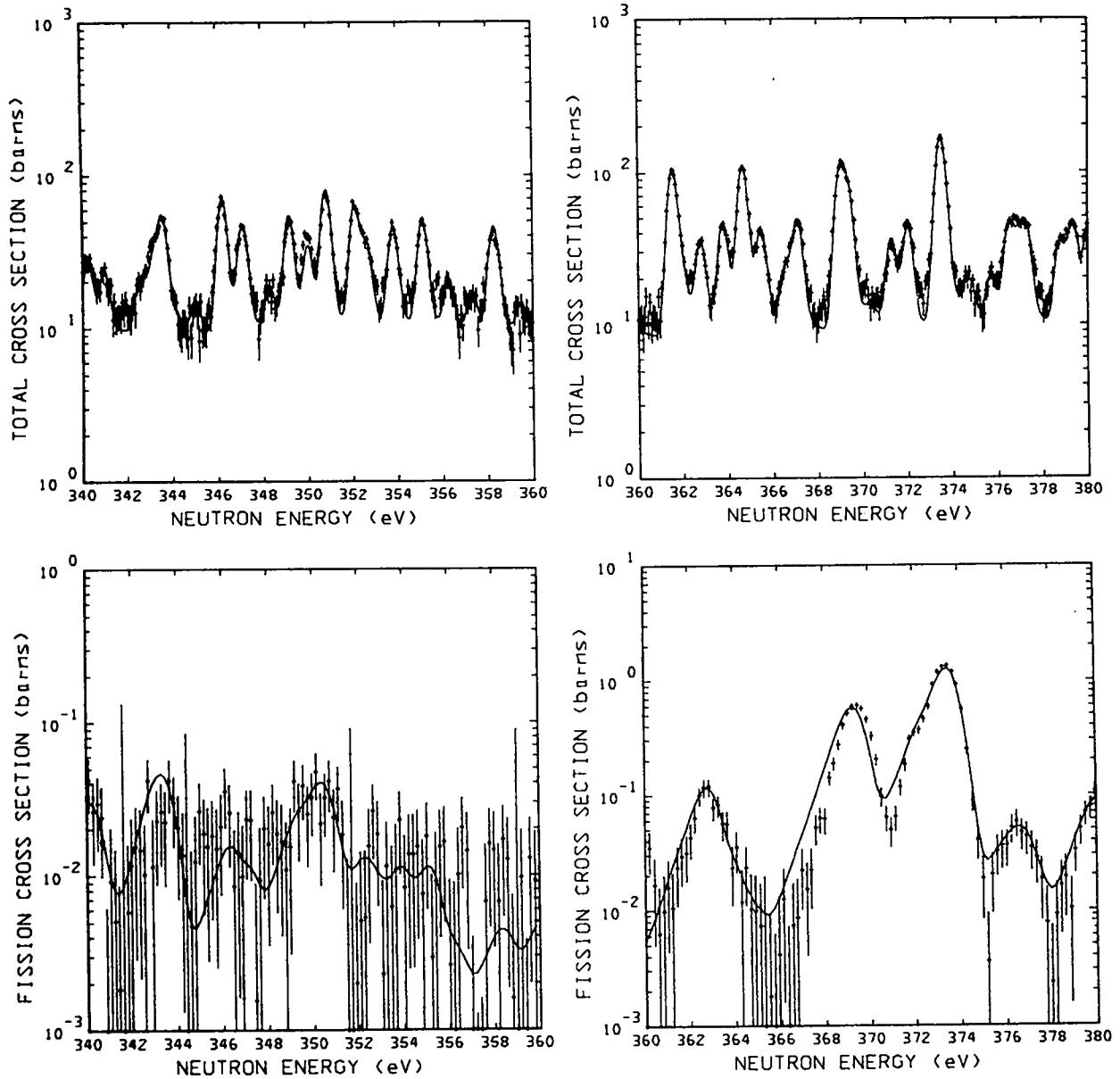


Fig. 17. Total and fission cross sections from 340 to 380 eV. The statistical errors are represented by vertical lines.

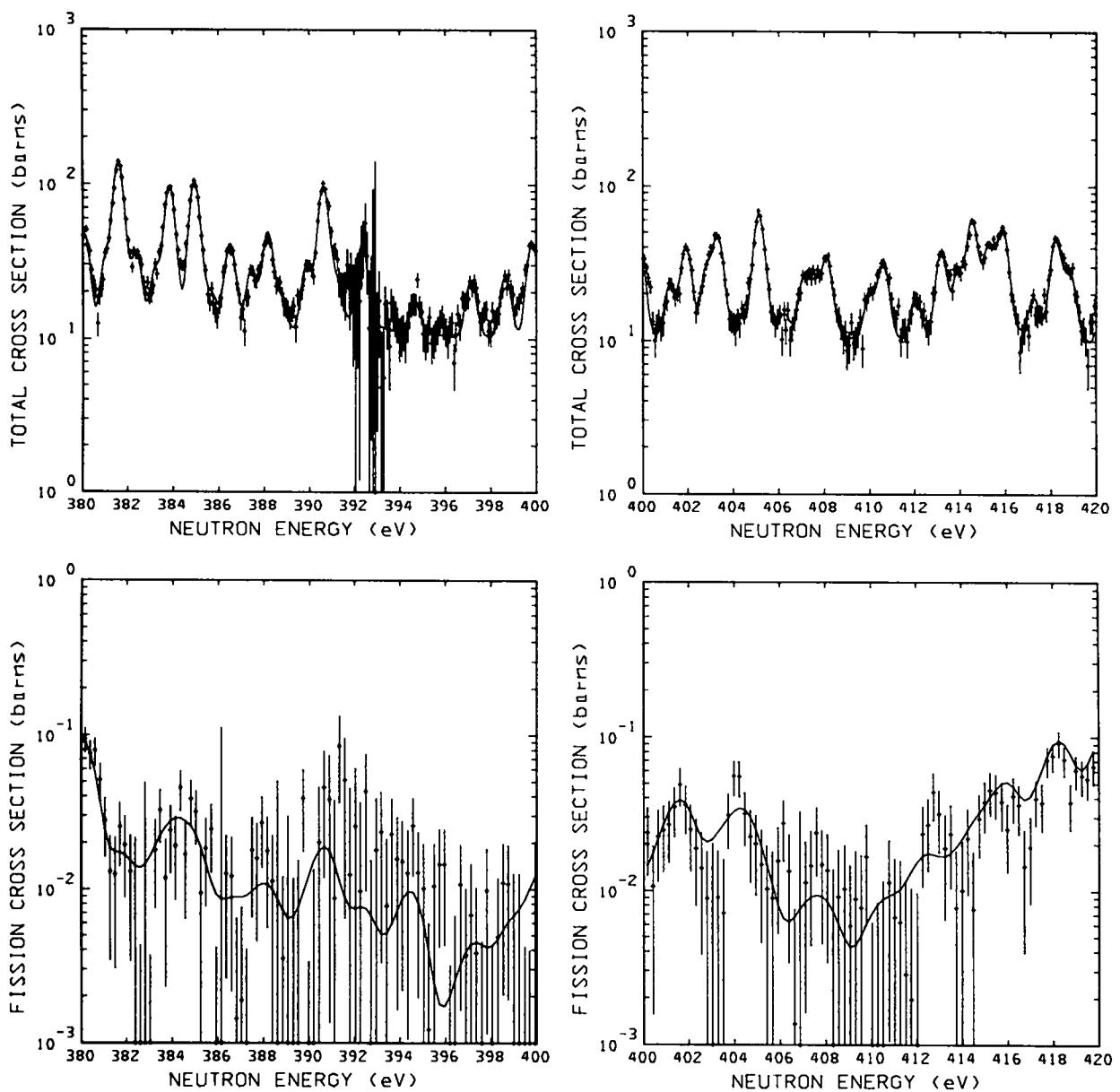


Fig. 18. Total and fission cross sections from 380 to 420 eV. The statistical errors are represented by vertical lines.

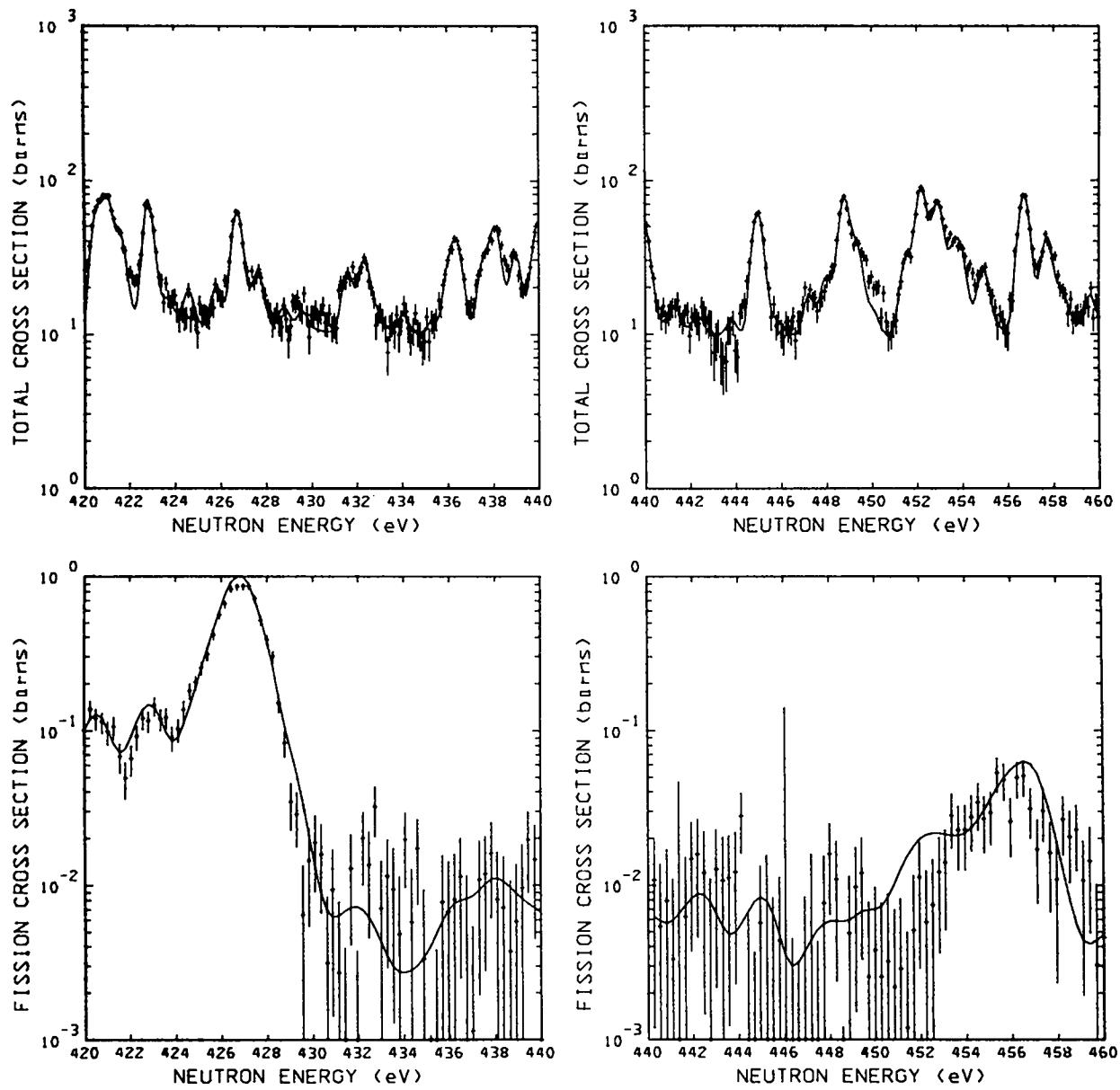


Fig. 19. Total and fission cross sections from 420 to 460 eV. The statistical errors are represented by vertical lines.

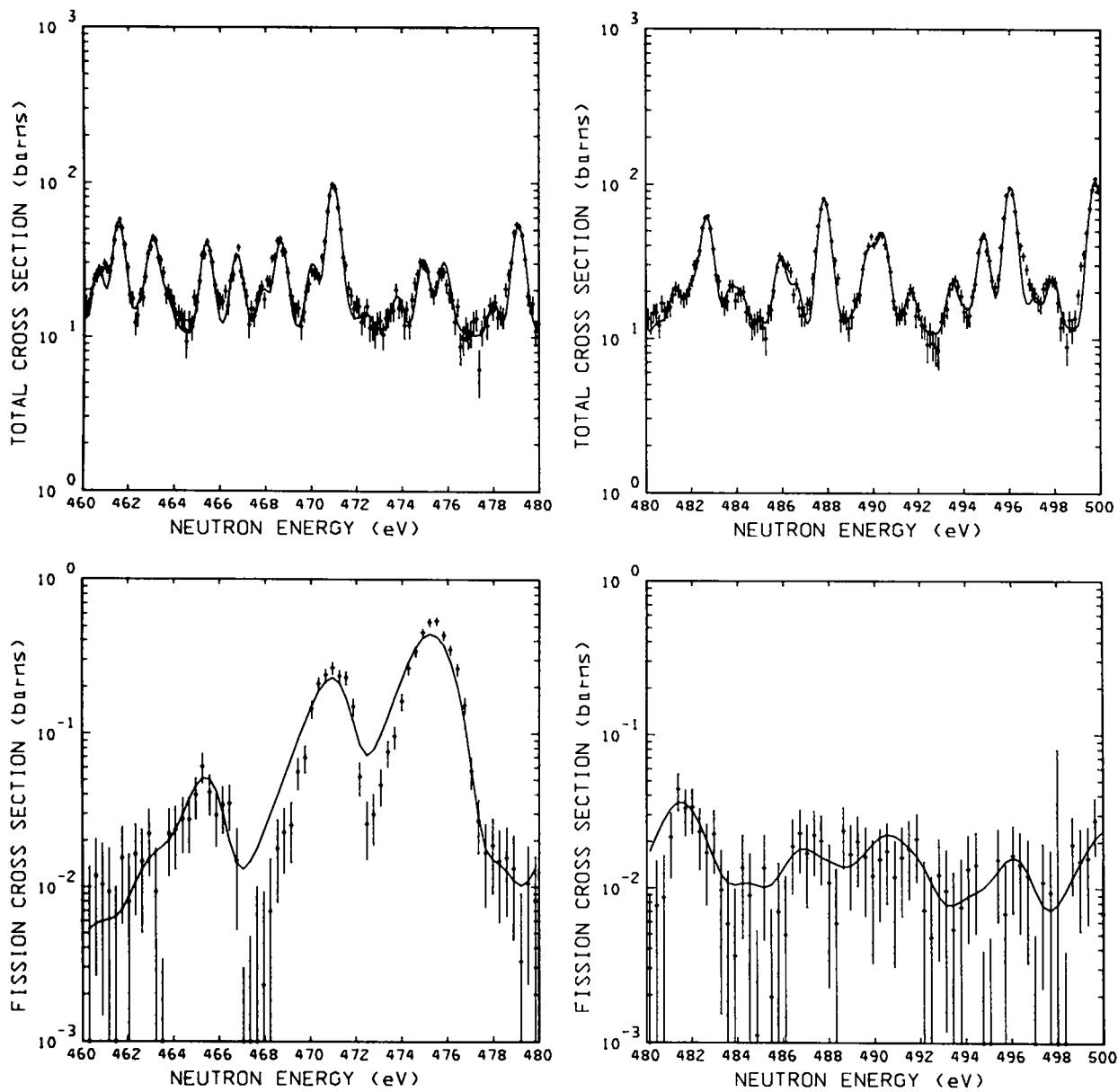


Fig. 20. Total and fission cross sections from 460 to 500 eV. The statistical errors are represented by vertical lines.

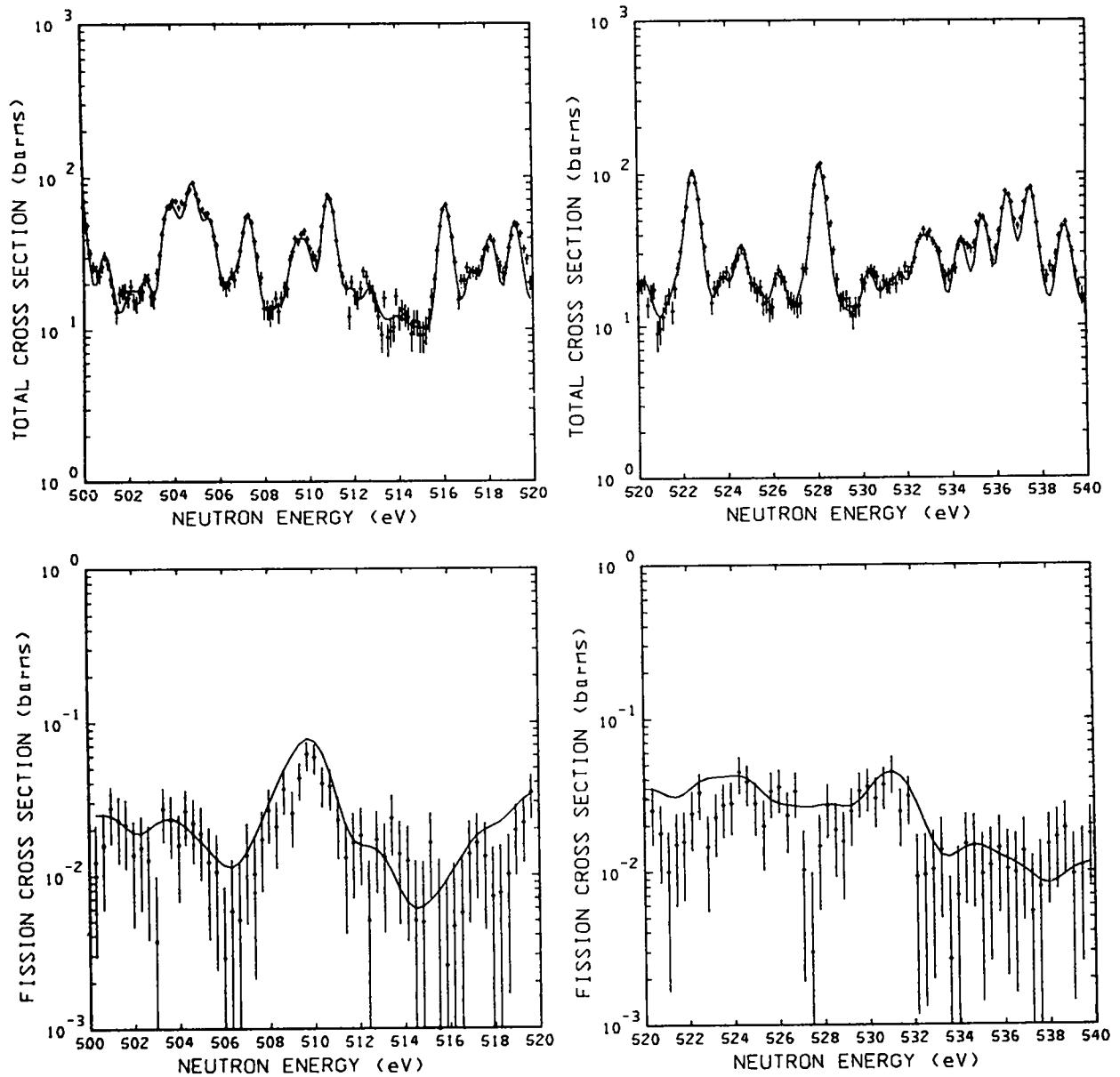


Fig. 21. Total and fission cross sections from 500 to 540 eV. The statistical errors are represented by vertical lines.

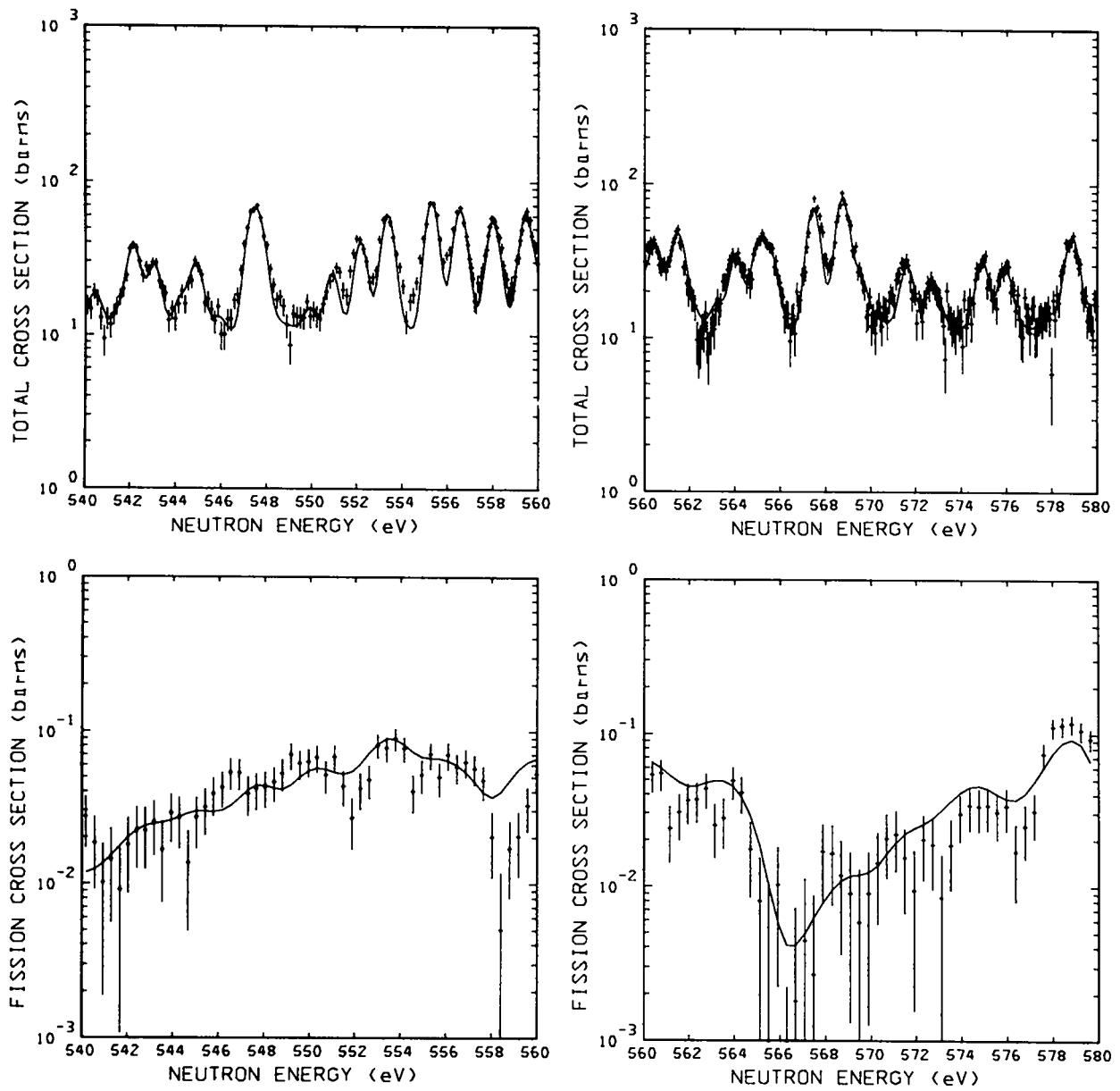
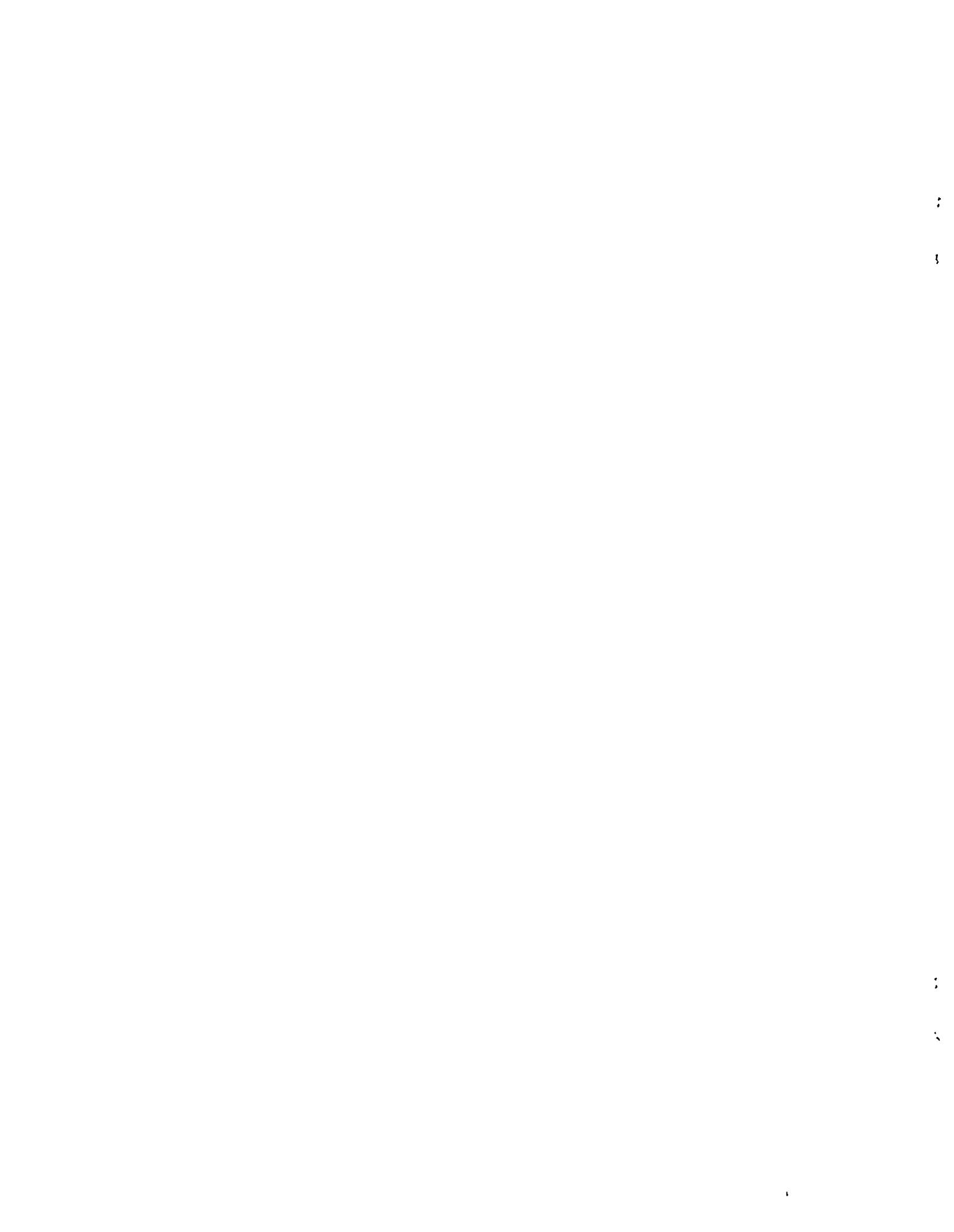


Fig. 22. Total and fission cross sections from 540 to 580 eV. The statistical errors are represented by vertical lines.

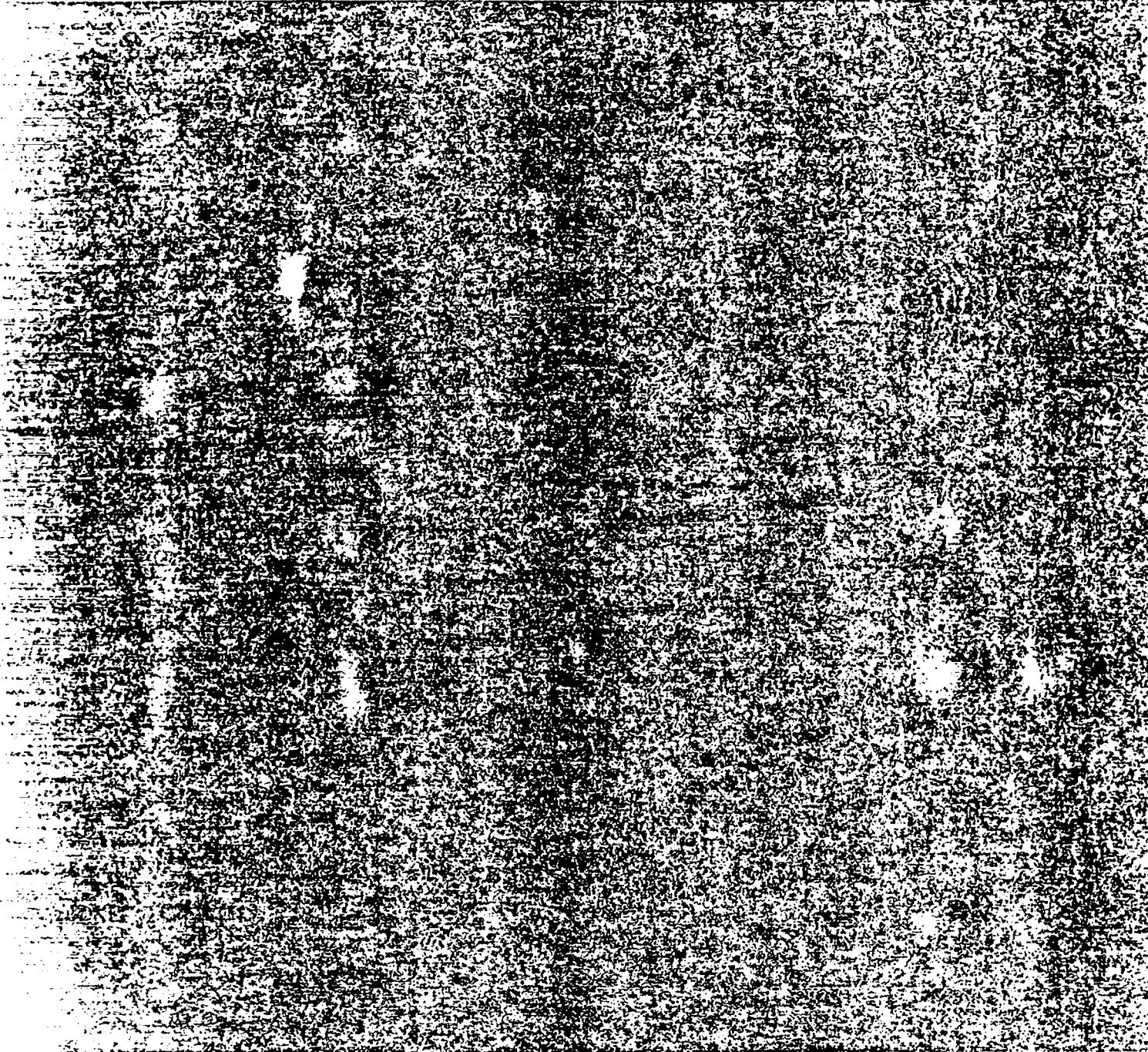


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