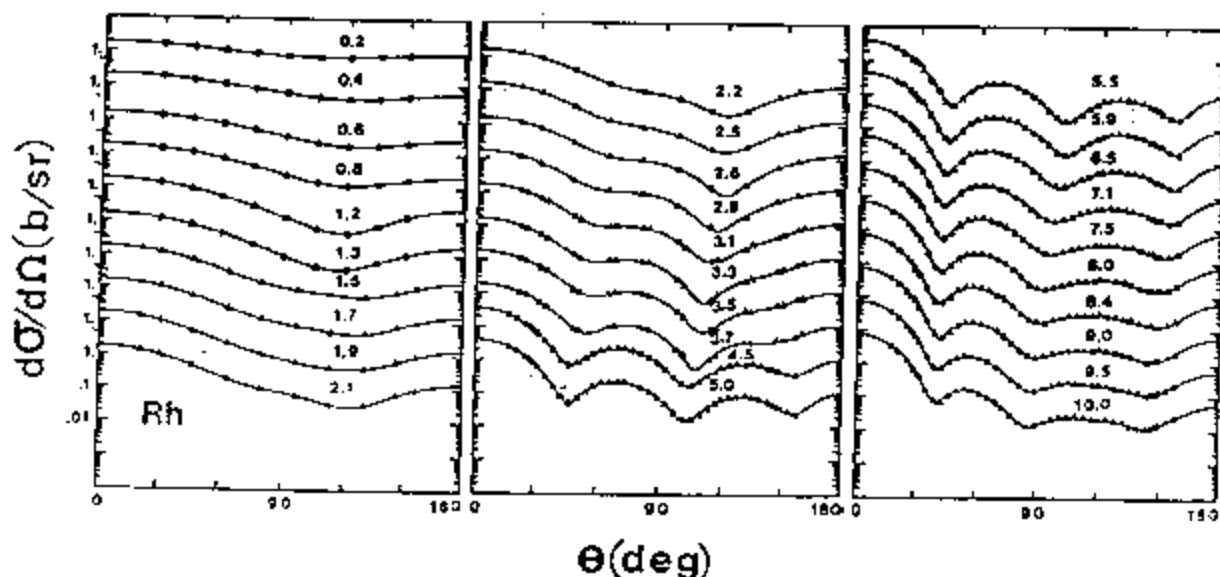


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ANL/NDM-150
NEUTRONS AND ANTIMONY
NEUTRONIC EVALUATIONS OF ^{121}Sb AND ^{123}Sb
by
Alan B. Smith and Andreas Fessler
July, 2000



ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

Operated by THE UNIVERSITY OF CHICAGO

for the U. S. DEPARTMENT OF ENERGY

under Contract W-31-109-Eng-38

ANL/NDM-150

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Argonne, Illinois

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Keywords:

Comprehensive ENDF/B-6 formatted neutronic evaluations of ^{121}Sb and ^{123}Sb .

*This work supported by the United States Department of Energy under contract W-31-109-Eng-38, and by the Nuclear and Energy Engineering Program, College of Engineering and Mines, The University of Arizona, Tucson Arizona.

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NEUTRONS AND ANTIMONY

NEUTRONIC EVALUATIONS OF ^{121}Sb AND ^{123}Sb Abstract

The new experimental results and recent extensive model development discussed in the companion report ANL/NDM-149 [Smi00], the new resonance parameterization of ref. [Mug99] and experimental results and models available in the literature, are used to construct neutronic evaluations for ^{121}Sb and ^{123}Sb in the ENDF/B-6 formats. These are comprehensive evaluations extending from thermal energies to 30 MeV, and include all reactions and processes commonly used in applied neutronic calculations. Comparisons are made with ENDF/B-6 MAT 5125 and 5131 files [ENDF].

1. Introduction

The objective of the present endeavor was an upgrade of the prior ENDF/B-6 evaluations for the two naturally-occurring isotopes of elemental antimony, ^{121}Sb and ^{123}Sb . The corresponding ENDF/B-6 evaluations (Mats 5125 and 5131) [ENDF] are a part of the fission-product files and are very old, largely dating back to the early seventies or before. New information, as described in the companion report ANL/NDM-149 [Smi00] and ref. [Mug99], makes possible considerable improvement in these evaluations. Many of the same methods and results can be used to extend the evaluation process to the other and unstable isotopes of antimony. That broadened scope is not part of the present work, but the stage is set for such endeavors. The present endeavors are "neutronic" evaluations, representing the neutron interaction with the two isotopic targets to incident energies of up to 30 MeV. Such a scope is suitable for a large majority of neutronic applications, extending from the thermal fission reactors to fusion-energy concepts. The present evaluations do not address charged-particle emission spectra, photon emission spectra, nor very minor reactions having cross sections of a mb or less. Estimated uncertainties are qualitatively stated in subsequent sections of this report where they are realistic. Both of the present evaluations are in the ENDF/B-6 formats, and both have successfully passed routine checking procedures. The corresponding numerical data files have been forwarded to the National Nuclear Data Center, Brookhaven National Laboratory. Those interested in numerical values should inquire at that Center.

2. Resonance parameters

Resolved and unresolved resonance parameters were explicitly provided by Dr. Said F. Mughabghab and his co-workers [Mug99]. Their very contemporary study uses a Bayesian analysis to determine L assignments. J assignments for resonances without experimentally-determined spins are randomly made following the $2J+1$ law. A bound level was not required for ^{121}Sb as the radiative width of the first positive-energy resonance was slightly modified to achieve a good description of the thermal capture cross section of 5.9 b. A bound level with a thermal capture contribution of 0.97 b was used for ^{123}Sb to provide a total thermal-capture cross section of 4.1 b. A potential-scattering radius of 6.1 fm was chosen for both isotopes so as to fit the coherent scattering amplitude of the natural element. The resolved resonance parameters are followed by unresolved resonance parameters consisting of s-, p- and d-wave contributions for the possible spin values. The unresolved average resonance parameters are given in Table 2-1.

Table 2-1. Average unresolved resonance parameters. Widths, Γ , are in meV and spacings, D , in eV.

	$\Gamma_{\gamma 0}$	$\Gamma_{\gamma 1}^*$	$\Gamma_{\gamma 2}$	S_0	S_1^*	S_2^*	D_0^{**}
^{121}Sb	91.0	50.0	91.0	0.34	3.0	1.5	15.0
^{123}Sb	97.0	50.0	97.0	0.28	3.0	1.5	27.9

* From systematics of ref. [MDH81], Part B, Figs. 5 and 7.

** Porter-Thomas analysis. An energy dependent level spacing is considered with parameters adopted from Mughabghab and Dunford [MD98].

For ^{121}Sb the resolved resonance description extends up to 2.5261 keV, followed by the unresolved resonance description extending on up to 37.4 keV, the threshold of the first inelastic-scattering group. The comparable limit of the resonance description of ENDF/B-6 is ≈ 0.922 keV and there is no unresolved contribution. The corresponding energies for the present ^{123}Sb evaluation are 2.4800 keV and 161.6 keV. The ENDF/B-6 ^{123}Sb resonance region extends to 1.5096 keV with no unresolved resonances. Thus the present evaluations provide information in an important region which is not available from

ENDF/B-6. Small adjustments were made to assure a reasonably smooth match between the resonance representations and the energy averaged cross sections of file 3.

3. Continuum total cross sections

Evaluated energy-averaged isotopic neutron total cross sections were calculated using the dispersive optical potential of Table 5-1 of the companion report ANL/NDM-149 [Smi00]. There are essentially no experimental isotopic total cross sections to test the isotopic evaluations. However, there is a reasonable body of elemental measured total cross sections as cited in the ref. list. The two present isotopic evaluations were combined to construct the equivalent evaluated elemental total cross sections. These elemental evaluated results are compared with the experimental values in Fig. 3-1. The agreement between the evaluated and measured cross sections is remarkably good, generally only several-percent differences from ≈ 0.1 to 30 MeV. These differences are of the same order as the uncertainties associated with the individual measured values alone. Thus, it is reasonable to estimate the uncertainties associated with the continuum evaluated total cross sections at several percent. Any significant improvement will require some very excellent new measurements, preferably using isotopic samples.

The present evaluated total cross sections differ from those of ENDF/B-6 by amounts considerably in excess of reasonable uncertainties, as is illustrated by the ^{123}Sb example of Fig. 3-2. There are similar discrepancies for ^{121}Sb . Of course, these differences in the two sets of total-cross-section evaluations will cascade through other aspects of the files. The ENDF/B-6 total cross sections seem to have been calculated using a potential that is, arguably, inappropriate at higher energies.

4. Elastic-scattering cross sections

The evaluated elastic-scattering cross sections of ^{121}Sb and ^{123}Sb were constructed from the LQ potential defined in Table 4-1 of the companion report ANL/NDM-149. The calculations followed the procedures as described in ref. [Smi00], and included compound-nucleus contributions where appropriate. It has been shown that this potential and associated calculational procedures lead to very good descriptions of the available experimental evidence, and that the results are physically reasonable, as discussed in detail in ref. [Smi00]. The angle-integrated elastic-scattering cross sections are presented in the 3-2 file section and the relative angular distributions are expressed in section 4-2 as legendre-polynomial expansions. The angle-integrated cross-section magnitudes were slightly

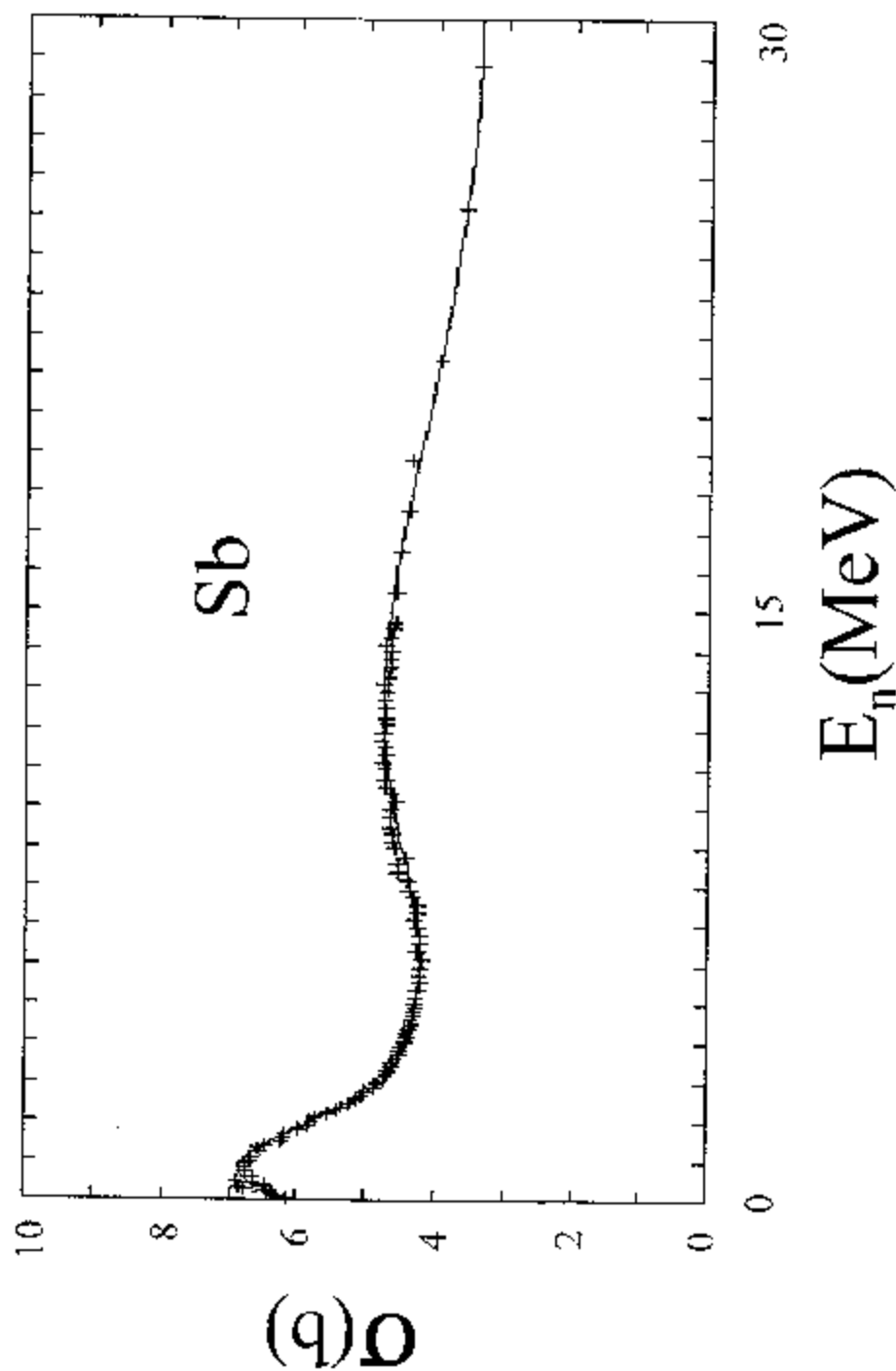


Fig 3-1. Elemental antimony total cross sections. The "+" symbols denote experimental values and the curve the elemental result obtained by combining the present two isotopic evaluations.

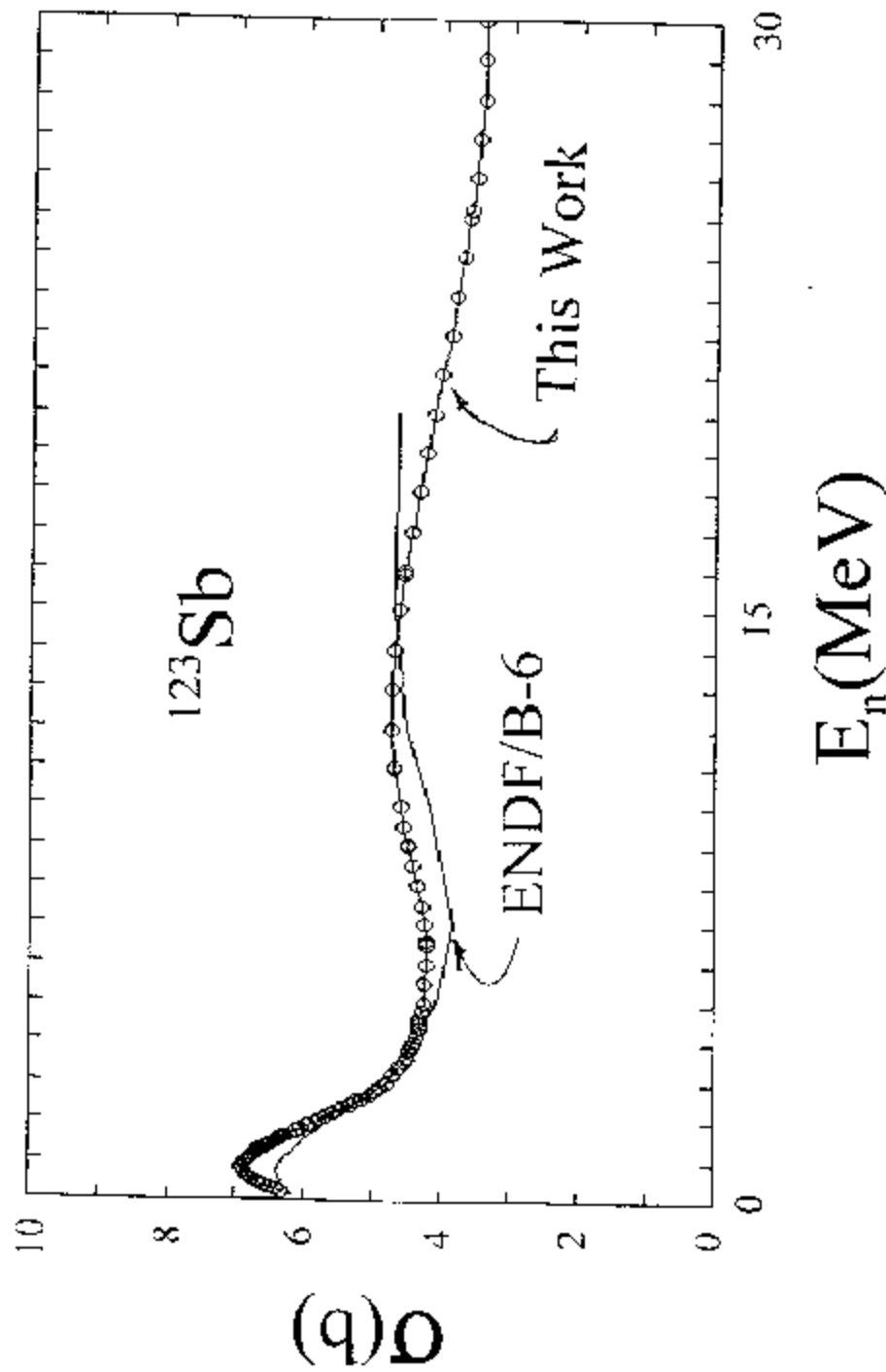


Fig. 3-2. Comparison of ^{123}Sb total-cross-section evaluations. The simple curve indicates the ENDF/B-6 results, and the curve with circular symbols the present evaluation.

adjusted to assure that the sums of the various partial cross sections are consistent with the above total cross sections. The adjustments were generally small. Both of the files were assayed to assure that the elastic-scattering representations were consistent with Wick's Limit" [Wic43]. An illustrative example of the elastic-scattering evaluations is given by the ^{121}Sb results of Fig. 4-1. The estimated uncertainties in the present evaluated elastic-scattering cross sections are 3-4% below ≈ 10 MeV where the elemental experimental data base is reasonably good. At the higher energies the uncertainties become larger but still they are grossly smaller than the discrepancies with the ENDF/B-6 results which are nearly a factor of two larger at ≈ 20 MeV. Furthermore, at higher energies the elastic scattering cross sections are increasingly controlled by Wick's Limit which is fixed by the reasonably known total cross sections. Fig. 4-2 illustrates a few of the elastic-scattering differential cross sections of the present ^{121}Sb evaluation. Similar results were obtained for ^{123}Sb . As the energy increases the anisotropies become very large. ENDF/B-6 specifies isotropic elastic scattering at all energies, clearly an inappropriate description. It is unlikely that the present elastic-scattering evaluations can be substantively improved without some good-quality elastic-scattering measurements spreading over the ≈ 10 to 30 MeV range. The lack of such measurements is a common problem throughout the periodic table.

5. Inelastic-scattering cross sections

The evaluated inelastic scattering due to the excitation of discrete levels used the same structures defined in the companion report, ANL/NDM-149. The excitation energies, spins and parities were taken from the Nuclear Data Sheets [NDS]. Ten levels (including the g.s.) were used for ^{121}Sb up to excitation energies of 1.322 MeV, and eleven in ^{123}Sb up to 1.511 MeV. The respective cross sections and angular distributions of the emitted neutrons were calculated using the LQ potential given in Table 4-1 of the companion report [Smi00]. These calculations were based upon the assumption of compound-nucleus processes. In addition, a small direct-reaction contribution was included using the vibrational core-coupling model described in ref. [Smi00]. The total result of coupling the $5/2^+$ and $7/2^+$ single-particle levels to the respective tin cores, assuming the vibrational interaction, was calculated and then spread over the 10 contributing states weighting the various contributions by $(2J+1)$. This is a very crude approximation but it does introduce the small direct-reaction contribution into the evaluated discrete inelastic-scattering process. As such it brings a qualitative consistency with the experimental results at higher energies where the compound-nucleus processes are deficient. In

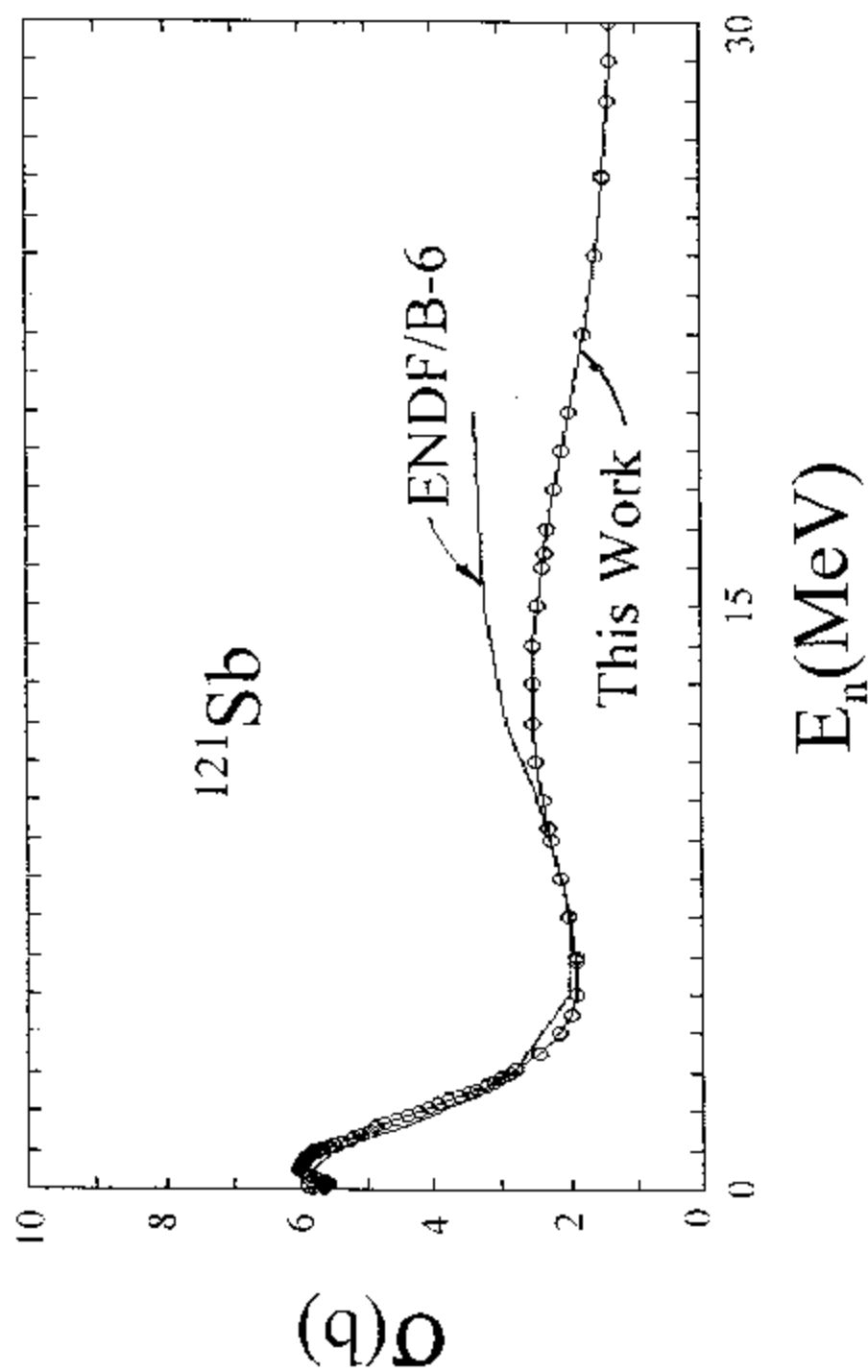


Fig. 4-1. Comparison of ^{121}Sb evaluated elastic-scattering cross sections. The simple curve indicates the ENDF/B-6 result and the curve with circular symbols that of the present work.

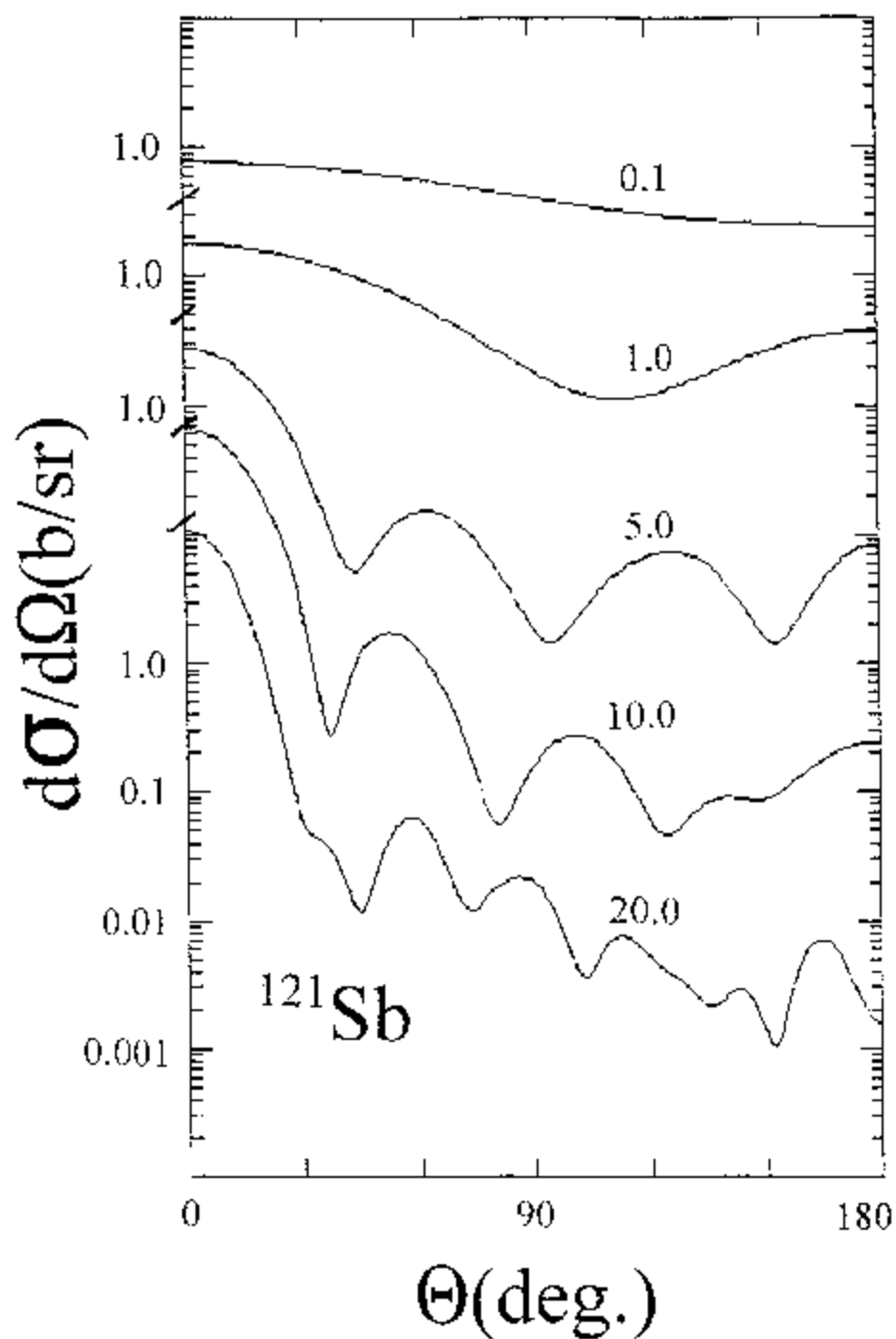


Fig. 4-2. Some illustrative ^{121}Sb evaluated differential elastic scattering cross sections from the present work. Numerical values indicate incident energies.

most applications the impact of the direct reactions will be negligible as the respective direct cross-section components are quite small. The cumulative contributions of the discrete inelastic cross sections is illustrated in Fig. 5-1.

The angular distributions of neutrons emitted due to the excitation of discrete levels are expressed as legendre-polynomial expansions. At low energies these are symmetric about 90° as the processes are very largely due to compound-nucleus processes. However, as the energy increases into the MeV range there is a pronounced and growing angular dependence due to direct-reaction contributions. This is illustrated in Fig. 5-2, relevant to the excitation of the 0.947 MeV level in ^{121}Sb . The experimental knowledge of the isotopic contributions is non-existent, and not very definitive for the element as discussed in refs. [Smi00], [SH67] and [SGW82]. With these inelastic-scattering uncertainties, it is speculative to more specifically estimate the uncertainties in the evaluated discrete-inelastic cross sections.

The continuum inelastic scattering was initially calculated using the statistical model code STAPRE [Uhl+76] with a direct-reaction component and the LQ potential of ref. [Smi00], in consort with calculations determining the contributions to other reaction channels, as described below. The calculated result was then slightly adjusted to assure consistency with other aspects of the file. The emitted neutron spectrum has an angle-energy correlation, particularly at higher energies. There is no experimental knowledge of such correlations in antimony. Furthermore, in the large majority of applications of antimony data (e.g. fission-reactor neutronics) continuum neutron emission is assumed to be isotropic and the emission spectra are averaged over all angles. In addition, the inclusion of continuum angle-energy correlations and the presentation in file-6 formats greatly extends the size of the evaluations. In view of these considerations, the present evaluations provide angle-averaged spectra and assume isotropy of continuum-neutron emission. That representation is sufficient for the vast majority of users, and more economical to handle. The resulting inelastic-continuum evaluations are illustrated by the ^{121}Sb result shown in Fig. 5-1. The energy dependence is physically reasonable, with the large compound-nucleus contribution falling to a direct-reaction tail at higher energies as competition with other channels (e.g., $(n,2n')$) sets in. There is a small up-turn in the continuum cross section between 25 and 30 MeV. The neglect of very weak reaction channels such as that of the $(n;n',t)$ reaction, as described below, contributes to this effect. Since there are no experimental results for verification of the

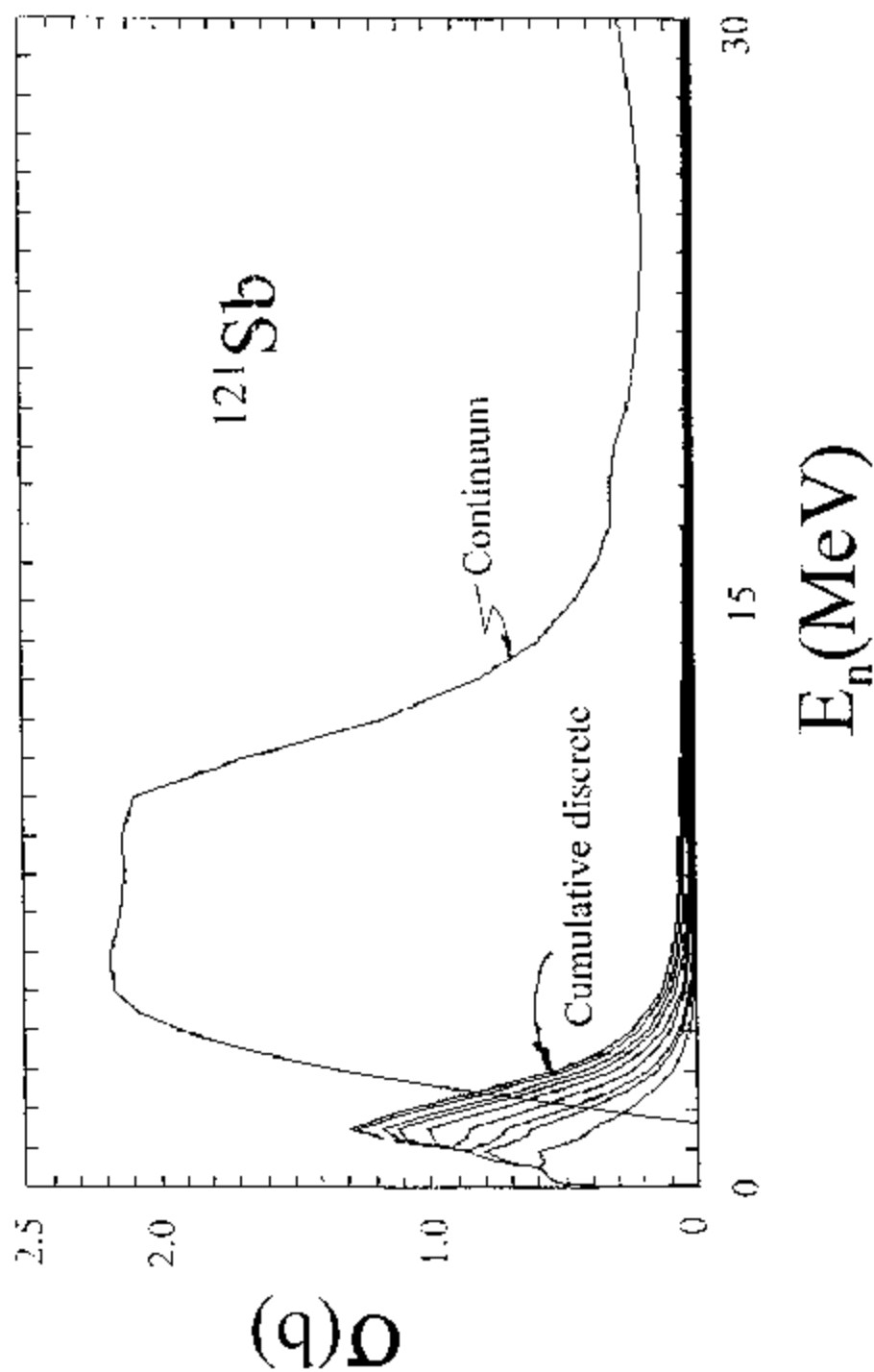


Fig. 5-1. Illustrative evaluated inelastic-scattering cross sections of ^{121}Sb . At the left is plotted the cumulative sum of the discrete components. The continuum contribution is also shown over most of the figure.

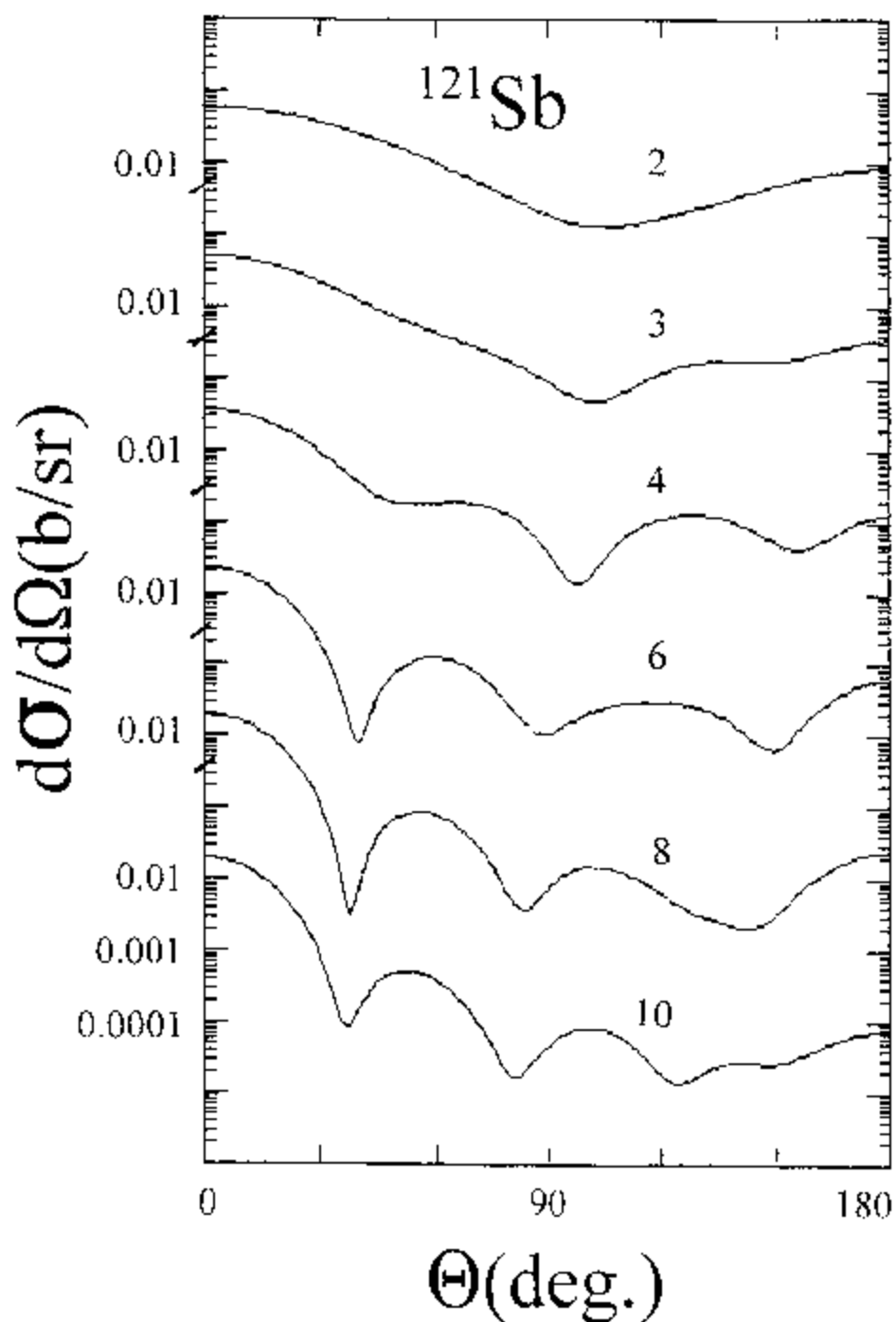


Fig. 5-2. Illustrative inelastic-neutron angular distributions for the excitation of the 0.947 MeV level of ^{121}Sb . The incident energies are numerically given in MeV.

antimony continuum-inelastic calculations it is difficult to estimate the uncertainty to be associated with the corresponding evaluated quantities. A reasonable estimate may be 10% at energies where the magnitude is large, increasing as the cross section magnitude falls.

Of course, the above components sum to the total inelastic-scattering cross section. Fig. 5-3 illustrates the result for the case of ^{121}Sb . Above several MeV the present evaluated total-inelastic cross sections are grossly different from those given in ENDF/B-6.

6. Radiative capture cross sections

There is some scattered experimental knowledge of the radiative capture cross sections of the two stable antimony isotopes, as cited in the relevant portion of the reference list. These isotopic values were largely obtained using activation techniques. There is more experimental information for radiative capture in the element, some of it from seemingly reliable tank measurements. With this weak experimental data base, the evaluations were constructed from calculations, subjectively adjusted to "best" describe the elemental experimental information. The calculations used the Brink-Axel formalism ([Bri55],[Axe62],[Lyn69]), implemented with the spherical optical-statistical-model code ABAREX, as described in ref. [Law98]. These calculations give an energy-dependent shape of the cross section, and the normalization is adjusted by varying the s-wave capture strength. After the calculations gave reasonable isotopic results they were combined to obtain the elemental capture cross sections. There remains a lot of scatter in the elemental experimental results but the comparisons with the calculated values are far better than those for the isotopes alone. The whole procedure was iterated several times, ending with the results illustrated in Figs. 6-1 and 6-2. The elemental experimental results are reasonably described up to the maximum energy of experimental availability (≈ 4 MeV). At lower energies several of the larger data sets are simply discrepant and the calculations can only strike a reasonable average. At very low energies minor adjustments were made to assure that the continuum capture cross sections reasonably match to the averages constructed from the resonance values. The subjective uncertainties in these capture evaluations are rather large, perhaps 10 \rightarrow 25% below 5 MeV. By 10 MeV the capture cross sections become very small and at higher energies they become very uncertain. There is no experimental information and the models used in the present evaluations do not include contributions from pygmy resonances, etc. These higher energy uncertainties should be of little note in most neutronic applications. The present ^{121}Sb capture evaluation is very similar to that of ENDF/B-6, as illustrated in Fig. 6-3. Strangely, there are large differences between the two

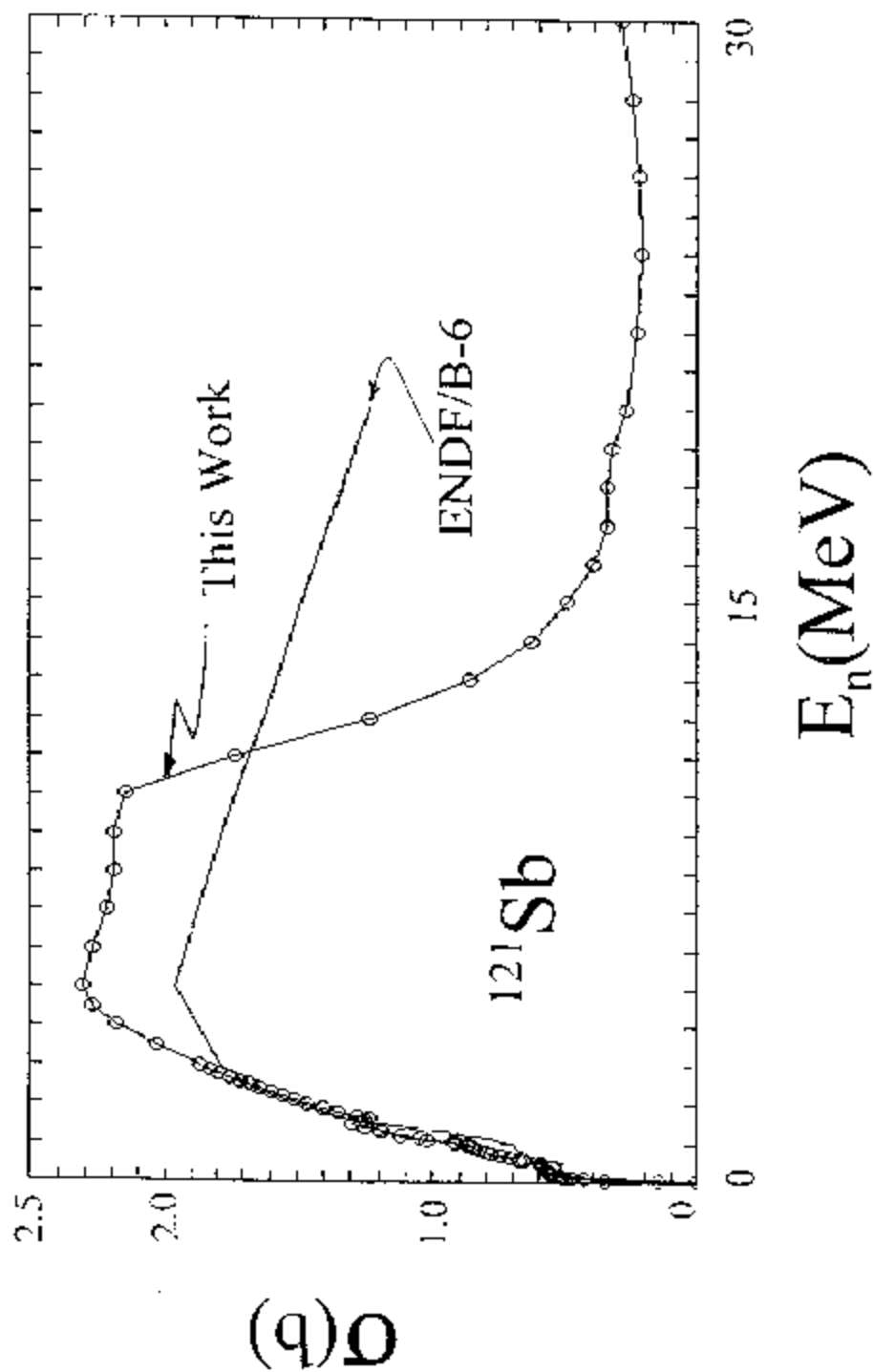


Fig. 5-3. Comparisons of evaluated total inelastic-scattering cross sections of ^{121}Sb . The simple curve indicates the ENDF/B-6 result and the curve with circular symbols the present evaluation.

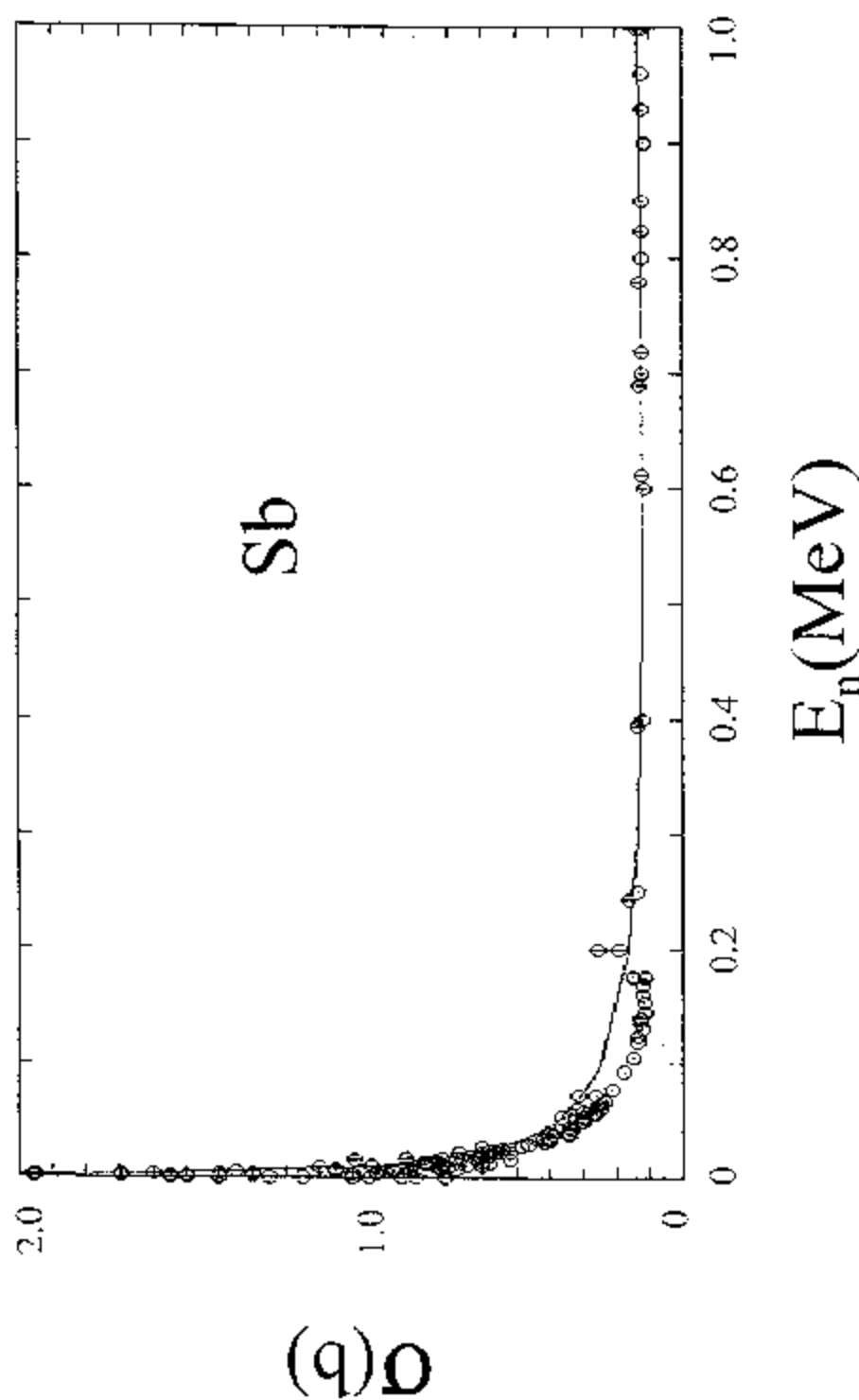


Fig. 6-1. Comparison of measured (symbols) and calculated (curve) radiative-capture cross sections of element antimony over the 0 - 1.0 MeV energy range, as outlined in the text. The calculated results are the evaluations above the first inelastic thresholds.

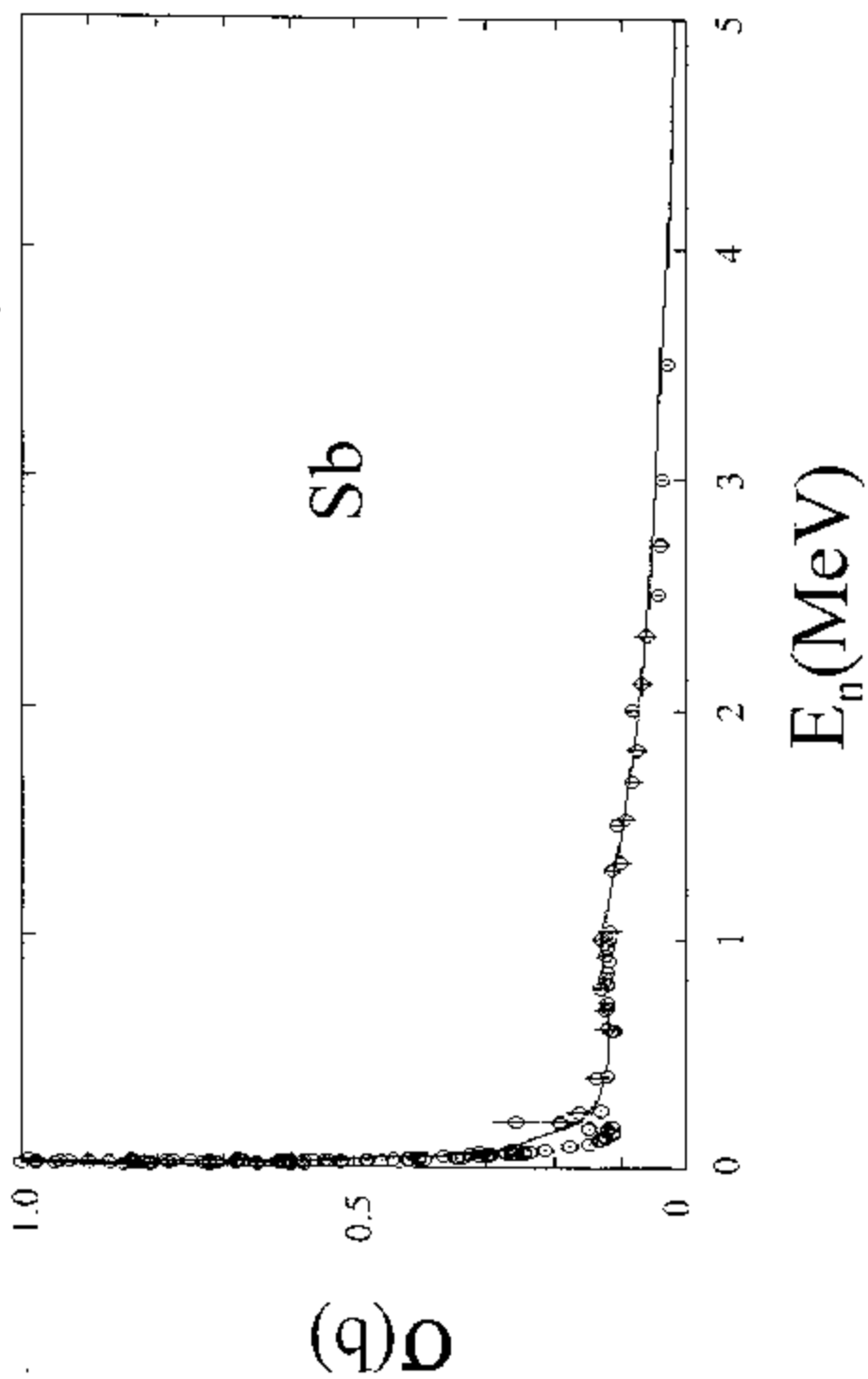


Fig. 6-2. The same as Fig. 6-1 except extended to cover the energy range $0 \rightarrow 5.0$ Mev.

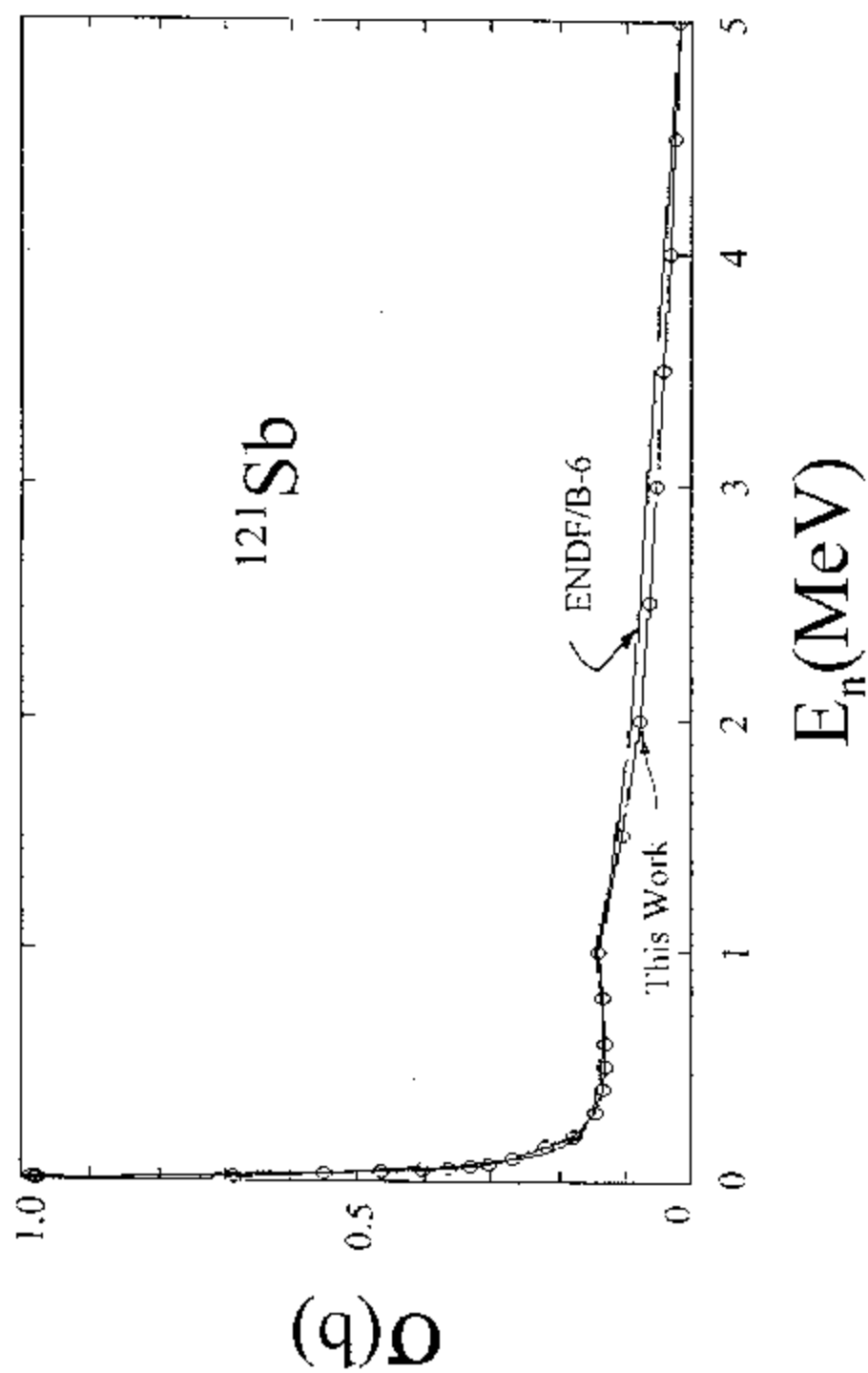


Fig. 6-3. Comparison of ^{121}Sb neutron capture evaluations. The simple curve indicates the ENDF/B-6 result and the curve with circular symbols the present evaluation.

evaluations for the ^{123}Sb isotope, as illustrated in Fig. 6-4.

7. (n,2n'), (n,3n') and (n,4n') cross sections

The threshold energies for these three reactions with the two isotopes of antimony are summarized in Table 7-1. These values were taken from the Livermore threshold compilation [How90].

Table 7-1. (n,2n'), (n,3n') and (n,4n') reaction thresholds in MeV.

Reaction	^{121}Sb	^{123}Sb
(n,2n')	9.316	9.039
(n,3n')	16.384	15.902
(n,4n')	26.051	25.216

The respective cross sections were calculated with the code STAPRE [Uhl+76], using calculational parameters adjusted to optimize the description of reported experimental results. The latter are confined to (n,2n') measurements. A few of these were tank measurements giving the total (n,2n') cross section, and there were a number of activation measurements based upon ground- and isomeric-state activities. These experimental results are cited in the corresponding section of the reference list. The calculated (n,2n'), (n,3n') and (n,4n') cross sections are illustrated in Fig. 7-1, together with comparisons with measured (n,2n') results. The (n,2n') evaluation uncertainties in regions of prominent cross sections are estimated to be $\approx 10\%$, increasing in regions where the cross sections are smaller. The uncertainties associated with the (n,3n') and (n,4n') processes are more speculative as there is no experimental information to test the accuracy of the respective evaluations. There are no (n,2n'), (n,3n') or (n,4n') components in the corresponding ENDF/B-6 evaluations.

The calculations also provided neutron emission spectra for each of these processes. These were averaged over angle and expressed as 5-16, 5-17 and 5-37 files, assuming isotropy of emission. The spectra are relatively "soft" as illustrated in Fig 7-2.

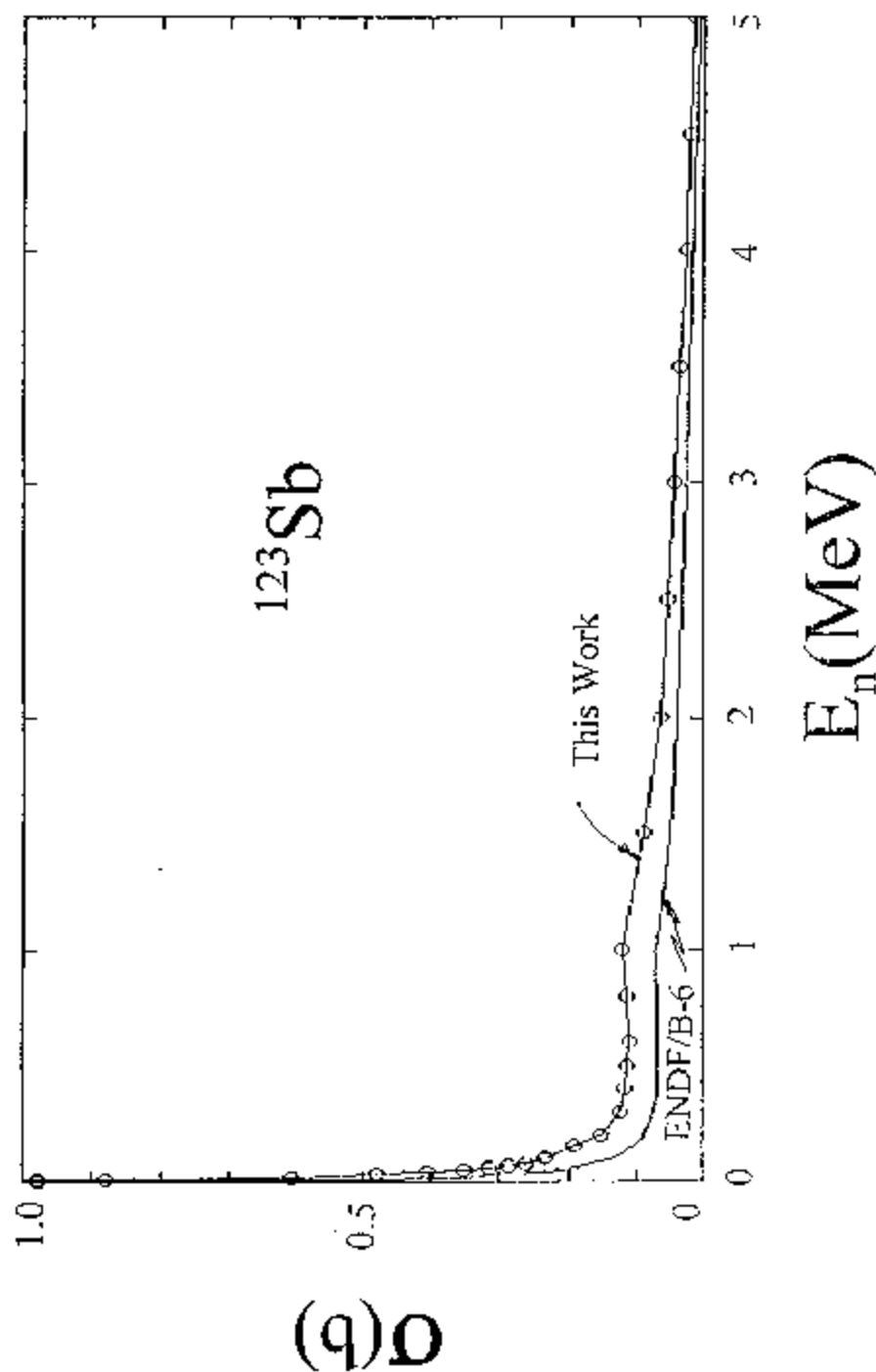


Fig. 6-4. Comparison of ^{123}Sb neutron capture evaluations. The simple curve indicates the ENDF/B-6 representation and the curve with circular symbols the present evaluation.

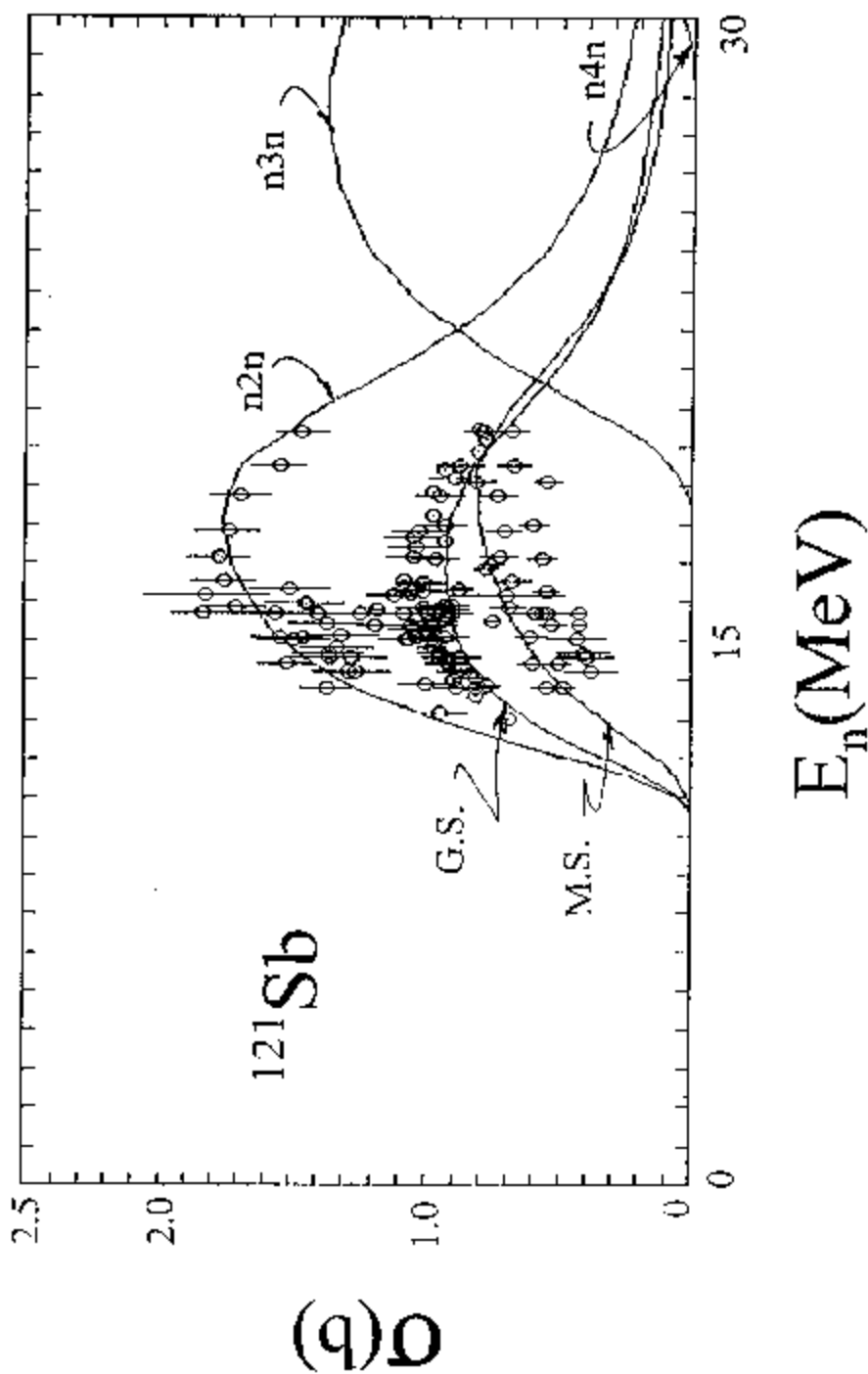


Fig. 7-1. $(n,2n')$, $(n,3n')$ and $(n,4n')$ cross sections of ^{121}Sb . Curves indicate the present evaluated results and symbols measured values. "gs" and "ms" curves refer to calculated $(n,2n')$ values resulting in the activation of the ground- and metastable-states by the $(n,2n')$ process. Symbols correspond to experimental $(n,2n')$ results.

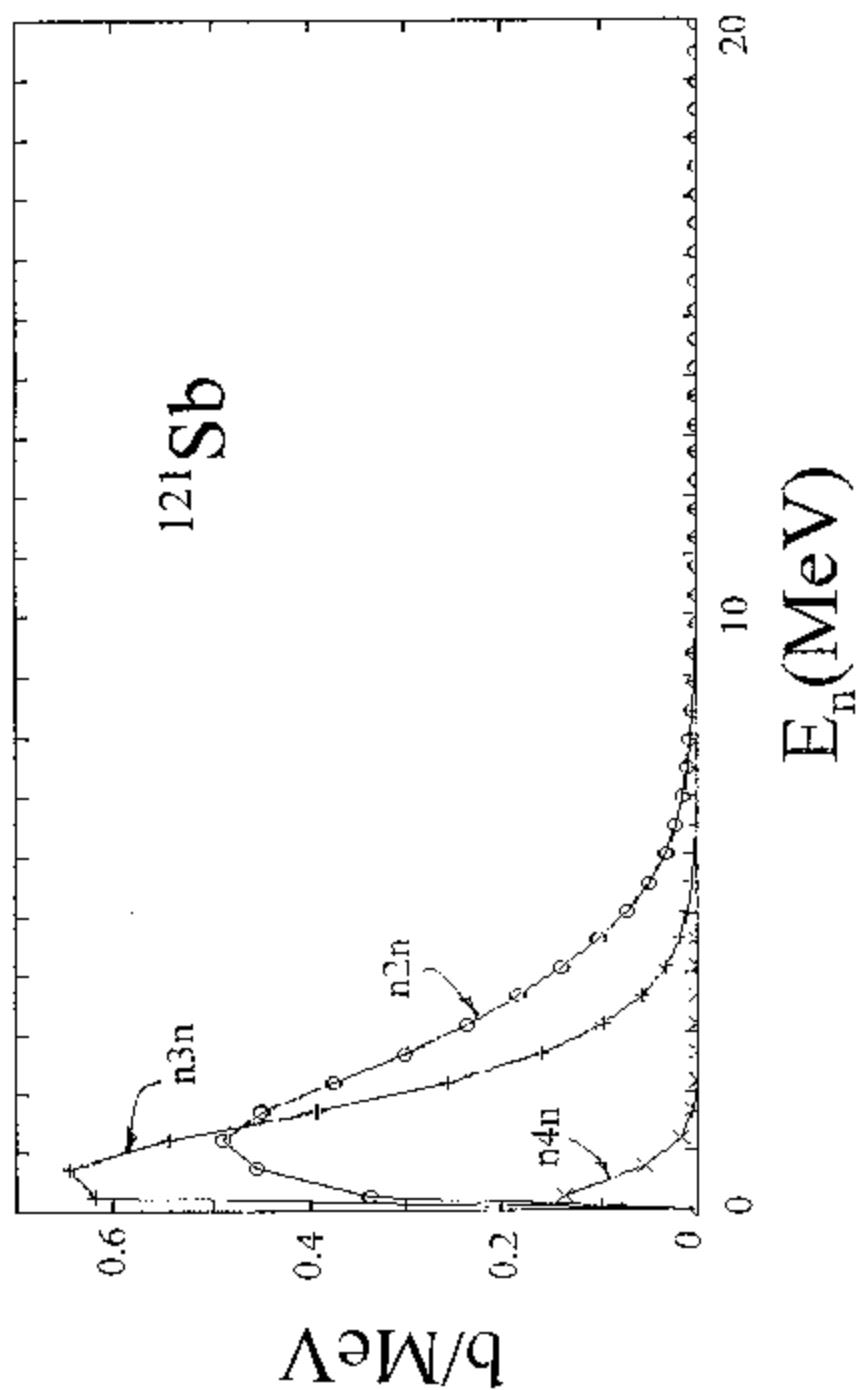


Fig. 7-2. Calculated continuum emission spectra from the $\{n, 2n'\}$, $\{n, 3n'\}$ and $\{n, 4n'\}$ reactions at an incident energy of 30 MeV.

8. Charged-particle-emission cross sections

8-1. (n,p) and (n;n',p) Processes

The thresholds for these two reactions are given in Table 8-1, below [How90].

Table 8-1. Thresholds for the (n,p) and (n;n',p) reactions in ^{121}Sb and ^{123}Sb , in MeV,

Reaction / Isotope---	^{121}Sb	^{123}Sb
(n,p)	-0.396	0.619
(n;n',p)	5.823	6.614

There have been a handful of (n,p) and (n;n',p) measurements, confined to ≈ 14.0 incident energies, with results that are not in particularly good agreement. Therefore, the present two evaluations used STAPRE calculations to determine the properties of the (n,p) and (n;n',p) reactions. An illustrative result for ^{121}Sb is given in Fig. 8-1. The reactions for ^{123}Sb are very similar. The cross sections for both processes are relatively small, e.g. ≈ 35 mb at 20 MeV. The calculations also provided neutron-emission spectra from the (n;n',p) reaction. In the present evaluations these spectra were averaged over angle and assumed to be emitted isotropically. The uncertainties may be large but the present evaluations are an improvement over the comparable ENDF/B-6 which is entirely devoid of these reactions. Uncertainties associated with the present (n,p) and (n;n',p) evaluations are speculative due to the lack of any reasonable experimental information. Significant improvements will probably require some detailed measurements. They are difficult due to the nature of the residual activities (or lack thereof).

8-2. (n, α) and (n;n', α) Processes

The thresholds of the (n, α) and (n;n', α) processes in ^{121}Sb and ^{123}Sb are summarized in Table 8-2.

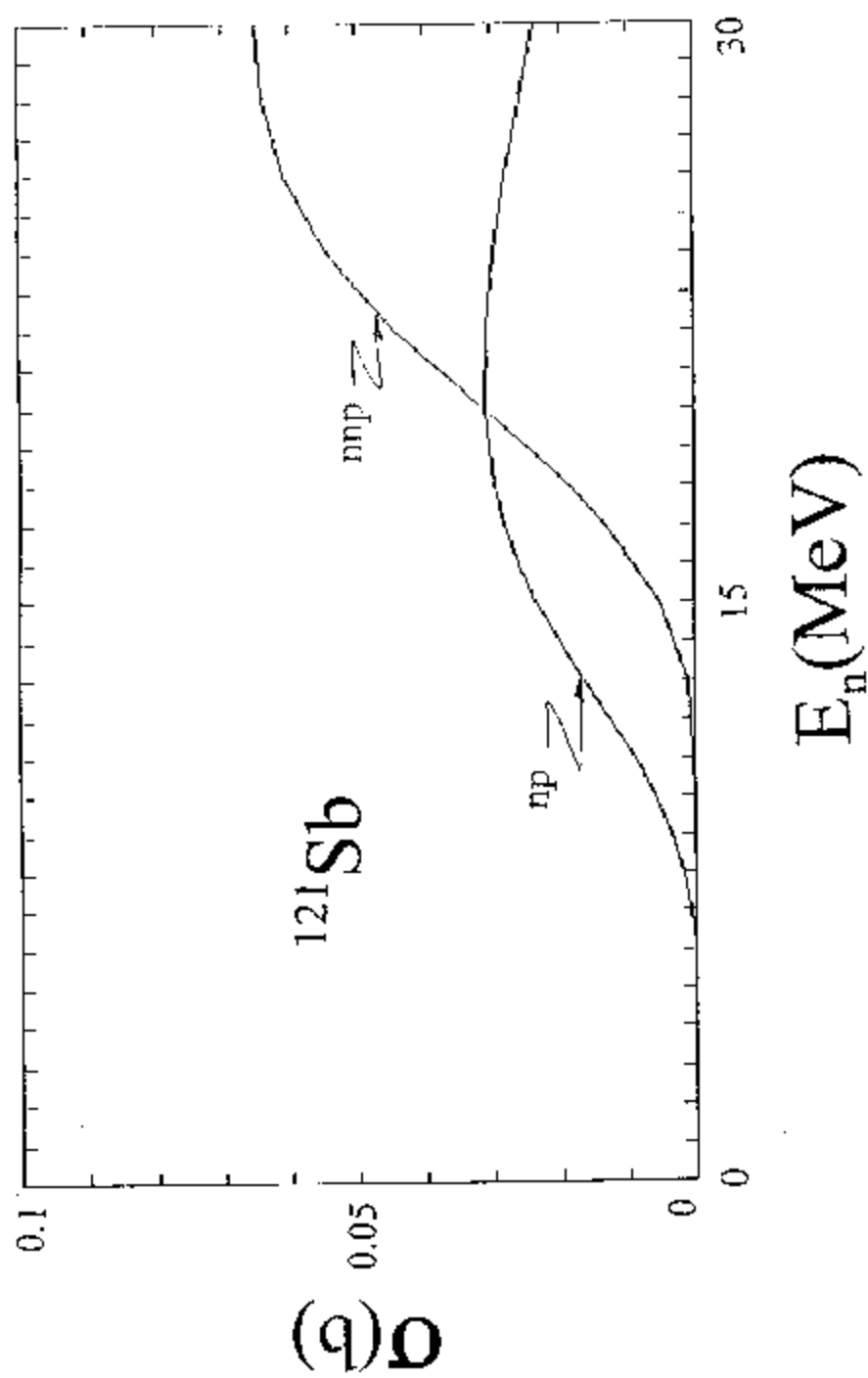


Fig. 8-1. Evaluated (n,p) and $(n;n',p)$ cross sections of ^{121}Sb (curves).

Table 8-2. (n,α) and $(n;n',\alpha)$ thresholds for ^{121}Sb and ^{123}Sb in MeV [How90].

Reaction / Isotope--	^{121}Sb	^{123}Sb
(n,α)	-3.509	-2.129
$(n;n',\alpha)$	3.094	3.944

Apparently, there is no experimental information relevant to any of the these processes. Therefore the evaluations again relied entirely upon the predictions of STAPRE calculations, with results as illustrated by the ^{121}Sb cross sections shown in Fig. 8-2. The ^{123}Sb results are qualitatively similar though with somewhat smaller magnitudes. In either case the (n,α) cross sections do not exceed several mb and those of the $(n;n',\alpha)$ reaction only several tens of mb, and then only at the highest energies. The calculations also provided the neutron emission spectra from the $(n;n',\alpha)$ reactions. They were averaged over angle for the present evaluations, and the emission was assumed to be isotropic. The uncertainties associated with (n,α) and $(n;n',\alpha)$ evaluations are speculative as they are entirely model based. However, the cross sections are small, and at higher energies, and thus will be a minor or negligible consideration in the large majority of neutronic applications. ENDF/B-6 contains no (n,α) or $(n;n',\alpha)$ information.

8-3. Other (n,X) and $(n;n',X)$ Processes

Primary of these processes are the six reactions listed in Table 8-3.

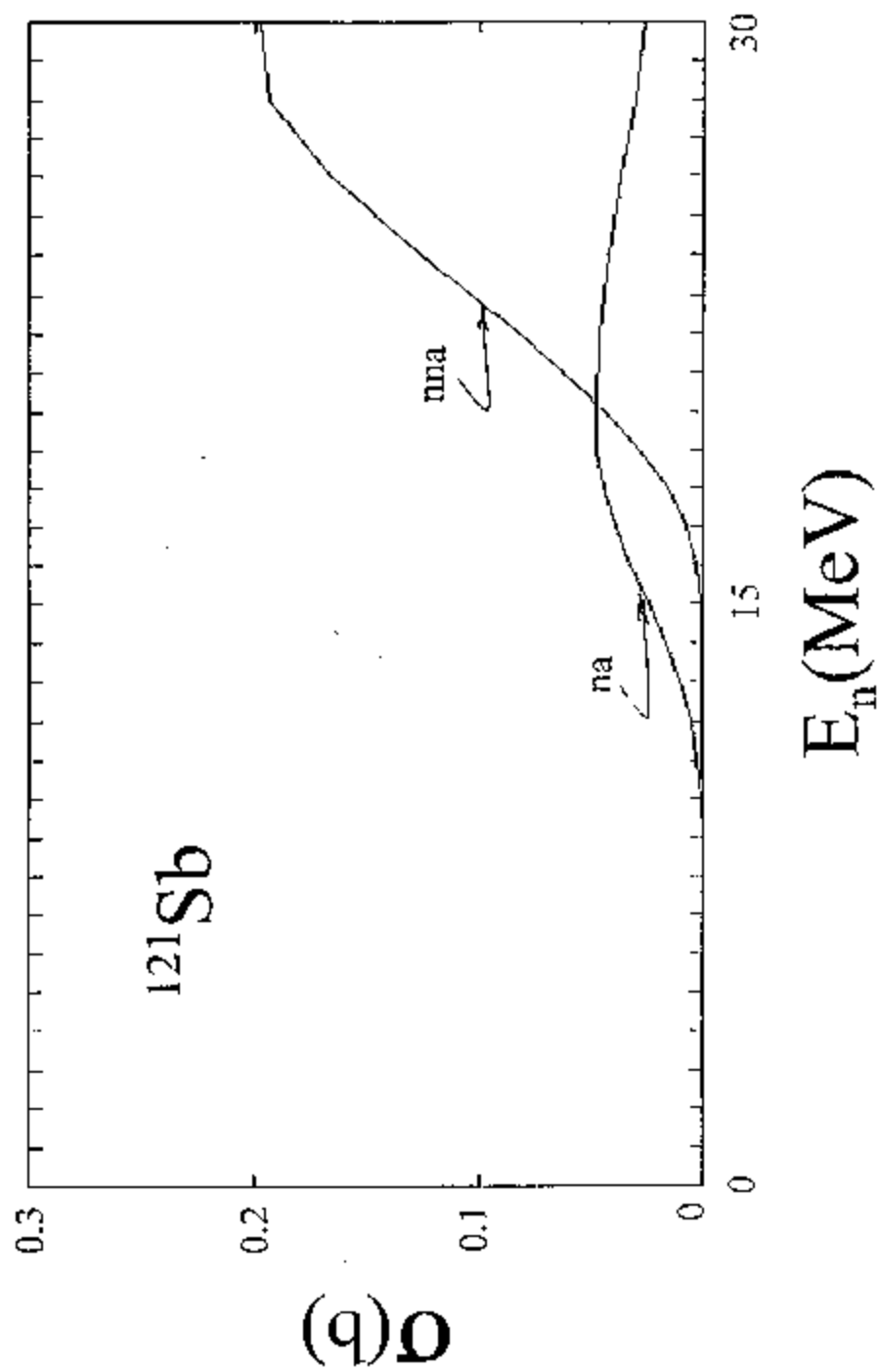


Fig. 8-2. Illustrative (n, α) and $(n; n', \alpha)$ evaluated cross sections of ^{121}Sb .

Table 8-3. Thresholds for other (n,X) and (n;n',X) reactions with ^{121}Sb and ^{123}Sb , in MeV [How90].

Reaction / Isotope----	^{121}Sb	^{123}Sb
(n,d)	3.581	4.372
(n;n',d)	12.763	13.260
(n,t)	6.454	6.952
(n;n',t)	12.992	13.173
(n, ^3He)	8.792	10.320
(n;n', ^3He)	17.217	18.601

The majority of these processes have quite high thresholds. There is essentially no experimental information available for any of them. However published theoretical estimates and other evaluations [JENDL] indicate that all of these processes have very small cross sections-- of the order of a mb or less at an incident energy of 20 MeV. Calculations are complicated by uncertain d, t and ^3He potentials. In view of these factors these processes were ignored in the present evaluations. Their omission will have essentially no effect on the neutronic applications for which the present two evaluations were designed. The specialized user who has a need, for example, for the (n, ^3He) cross sections should consult a specialized evaluation. Guidance can be obtained from the National Nuclear Data Center, Brookhaven National Laboratory or from the JENDL evaluations. ENDF/B-6 is also devoid of these minor (n,X) and (n;n',X) processes.

9. Summary remarks

The present two evaluations reasonably achieve their objectives of significantly upgrading the ENDF/B-6 evaluated neutronic files of ^{121}Sb and ^{123}Sb . They are simple and compact, and are soundly based upon new experimental and analysis information where that is available. Many aspects of these evaluations can be significantly improved only with new experimental work that does not appear likely. There are shortcomings, notably the omission of minor particle-emitting reactions, the neglect of photon emission, and the neglect of angle-energy correlations in neutron continuum emission. These omissions will have essentially no effect on the vast majority of neutronic applications.

10. Suggestions for future work

Basic and applied physics are observational sciences. To substantively improve the above outlined evaluations some careful measurements are suggested. The suggestions are not inclusive and are confined to energies above the resonance region.

- 0 The total cross section should be measured from ≈ 10 keV to 30+ MeV to accuracies of $\leq 1\%$ for both the element and the two naturally-occurring isotopes. Attention should be given to self-shielding at the lower energies. Such measurements are technologically straight forward.
- 0 Half a dozen good quality elastic-scattering distributions are needed between ≈ 10 and 30 MeV in order to firmly establish the higher-energy behavior of the model used in the evaluations. The measurements would be most useful if isotopic targets were used, but even good elemental results would be of considerable value. Such measurements are technologically feasible but difficult.
- 0 Significant improvement in the discrete inelastic-scattering evaluations requires some careful isotopic scattering measurements using direct neutron detection. They are feasible but isotopic measurement samples will be required. A few isotopic double-differential inelastic continuum measurements would be of use to check the overall envelope of the calculations but will not, of course, explicitly define the inelastic components.
- 0 Significant improvements in the evaluated (n,p) , $(n;n',p)$, (n,α) and $(n;n',\alpha)$ cross sections require some careful measurements. They are feasible but difficult, and the respective cross sections are small.
- 0 Experimental knowledge of photon production and emission spectra is uncertain. A few relevant measurements should be undertaken. Without them, the respective evaluations will be only a calculational speculation.

Acknowledgments

The authors are, of course, deeply indebted to Dr. S. Mughabghab for the provision of his most recent resonance parameterizations. They provide major new insight in an important region. The authors are also indebted to Dr. D. Cullen for his suggestions and instructive dialog. Generally, the National Nuclear Data Center, Brookhaven National Laboratory, is a unique resource for experimental information generally, and for instructive comment and assistance.

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