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Principals and Problems in Neutron Nuclear Data Evaluation

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PRINCIPLES AND PROBLEMS IN NEUTRON NUCLEAR DATA EVALUATION

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In the last ten years the evaluation of nuclear data has evolved as an own branch of applied nuclear science. Particularly the evaluation of low energy neutron nuclear data, with which we are dealing here, has attained an eminent importance. In the course of the development the term "evaluation" has become the common name to denote an activity which consists in the establishment of a complete chain of cross sections or other nuclear data characterizing a certain reaction with a certain nuclide in a given energy range. This activity proceeds in several steps, beginning with the compilation of all available experimental references and data concerned, and continuing with a critical judgment and comparison of this information with the aim of elaborating it into a complete, unequivocal set of so called "best" or "recommended" data. "Complete" means in a larger sense, that no gaps are left, in a more specific sense, that the energy dependence of a cross section is reproduced in an "almost monochromatic" way allowing a simple interpolation between adjacent data points.

The requirement of completeness involves the recourse to nuclear theory and systematics in the case of gaps and inconsistencies of the experimental data. Naturally evaluation does also comprise, with a similar requirement of completeness, discontinuous and parametric data like level schemes and resolved resonance parameters. In view of the fact, that the gathering of experimental or theoretical data with the aim of a comparison with a new theory or a new experiment is a very old and basic scientific job, the immediate question arises which reasons have made the evaluation particularly of neutron nuclear data almost a field of its own with such specific stringent terms of reference. A brief historical review will give an answer to this question.

Evaluation, as it has been defined above, is primarily and intimately connected with the development of nuclear reactors. It never would have attained such importance, if, as at the beginning of the reactor development, only thermal reactors would have been designed. The physical behaviour of these reactors is mainly governed by the reactions the neutrons undergo in the small thermal energy range. Apart from the rather complex thermal inelastic scattering interactions, the energy dependence of the cross sections is rather simple, the important capture cross section for example following almost always the simple 1/v-law. In principle, few group calculations with adaptation to integral parameters were thus sufficient to predict the neutronic behaviour of a thermal reactor; the gathering of the few necessary data could easily be done by the reactor physicists themselves. This was the somewhat simplified picture in the early stages of the reactor development.

With the beginning of the design of fast and intermediate breeder reactors and with the rapid development of ever larger and faster electronic computers this situation changed completely. In small fast critical assemblies and metal fuelled fast reactors neutrons concentrate on keV and MeV energies. Large dilute oxide or carbide fuelled intermediate power breeders contain neutrons down to the eV region. For shielding calculations the MeV range of neutron energies, for the calculation of reactor neutron energy spectra and for safety considerations like the calculation of Doppler, steam and sodium void coefficients the intimate knowledge of the resonance properties of almost all reactor materials, in particular of the heaviest ones, became of urgent interest. In parallel the computer development allowed and forced the development of advanced reactor theory programs like one and two-dimensional multigroup diffusion and transport codes and detailed Monte Carlo programs. These comprehensive programs enabled the reactor theory to a much more detailed and precise description of the nuclear properties of reactors, and in particular also of thermal reactors, than previously. The usefulness of these programs, the reliability of their results depended to the largest extent upon the detailed and reliable knowledge of the microscopic nuclear data involved and their effective utilization in these programs. Immediately the original rather easy task of gathering some thermal nuclear data enlarged to the much more complicated and comprehensive task of gathering microscopic neutron data for all occurring nuclear interactions in the

much larger energy range - following nuclear physics terms we called it "low energy" range above - from about 0 to 10 MeV and more; as the reactor neutrons do not leave out any energy subrange or any reaction in this whole region, the gathering of the data had to be complete with regard to the energies and the reactions covered for a given nuclide. Thus the principal requirements on such collections of neutron nuclear data outlined in the beginning of this paper evolved in a quite natural way.

Still one might at first sight believe, that this gathering of data could not be so large a task and that nuclear theory calculations would give necessary and sufficient information. Unfortunately, however, as is well known, no unified nuclear theory exists, which allows the reliable prediction of all neutron nuclear interaction probabilities in the range from 0 to 10 MeV. In case it would exist, one could conceive a coupling together of nuclear and reactor theory programs, the output of the former being the input of the latter, as a satisfactory solution of the whole data problem. Instead only nuclear models with a validity restricted to certain energy ranges, reactions and nuclides and parametric theories are available, which, for such accurate and reliable predictions of nuclear parameters and cross sections, as are needed in reactor calculations, can in most cases only be applied with reasonable success, if some or all parameters entering the theory are taken from experiment.

Thus, if this purely theoretical way does not exist or at best is only successful (and then no longer pure) with the help of experimental parameters, one might think of a purely experimental way in constructing one or a few experimental apparatus and measure with these facilities once for all the necessary data in the whole energy range of reactor neutrons with the desired high accuracy and a throughout perfect energy resolution. Apparently nobody will ever invent such an ideal machine which, if available, would immediately solve the whole problem: the machine could work on line with the reactor computer programs and feed its data directly into these. Returning to reality one finds that just the opposite is true. A large variety of experimental facilities like choppers, Van de Graaff

machines, linear accelerators and others is available almost all over the world, each type of machine being successfully applied only in certain energy ranges and for certain nuclear reactions. Actually, last not least, the development, refinement and rapid distribution of these machines was due to the data requirements from reactor physics side laid down e.g. in the well-known neutron nuclear data request lists of the EANDC. These machines now produce almost exponentially increasing large amounts of data, which have first to undergo the rather complicated and lengthy procedures of mutual comparison, critical evaluation and selection, tabulation and computer storage, before being successfully fed and elaborated in the reactor theory programs. Even so the purely experimental way does not suffice for yielding all necessary data. Unavoidable gaps exist due the limitedness of the machines in energy resolution, energy ranges and reactions to be covered. Avoidable gaps exist due to the fact, that not all needed data have hitherto been measured. Finally more or less severe discrepancies and inconsistencies between different data sets are frequently encountered, reflecting the large difficulties connected with the experimental apparatus and measurement techniques, discrepancies which often can not be solved on the experimental basis alone. However, reactor neutrons obviously do not know of these gaps and discrepancies, and the natural way, to get these difficulties solved, if not physical imagination and ingenium can help, is the recourse to estimates based on nuclear systematics and nuclear model calculations with the partial or full use of "best" guesses of the nuclear parameters involved.

This brief view back immediately makes not only clear the principles, but simultaneously gives a first idea of the difficulties and problems involved in every evaluation. An evaluation physicist has to be aware in a systematic way of all experimental techniques and data concerned. At the same time he should be familiar with current nuclear models, theories and computer programs concerned. The emphasis he places on certain parts of his work is governed by the needs and the importance imposed particularly from the reactor physics side; so he should be aware of the current main problems in reactor physics and be in close contact with the re-

actor physicists. Finally he is always faced with a variety of computer organizational problems. Obviously neither the experimental or theoretical nuclear physicist nor the reactor physicist can perform the work of an evaluation physicist beside their own job; for this it has become much too large. The main problem therefore for an evaluation physicist is to be always up to date in, and aware of, all these aspects of his work. This explains our first assertion that evaluation has become a branch in itself in the domain of applied nuclear physics; it also makes clear, that it can only be efficient in a team work and in close cooperation with reactor and experimental and theoretical nuclear physicists.

Now we turn more extensively to some of those problems encountered during the evaluation itself and, which were already mentioned above. Technical problems connected with the computer storage of evaluated data and the establishment of nuclear data files, although forming an essential and hard part of every evaluation, will be left out of our considerations, and the interest focussed on physical problems.

Discrepancies and inconsistencies between different experimental data sets and their solution represent the largest problem in almost every evaluation regardless of neutron energy, reaction and nucleus concerned. Naturally no two measurements are made under exactly the same conditions, but their results should be compatible with each other and, if reduced to the same experimental conditions, come into agreement, at least within the range of the mutual uncertainties. However, it frequently occurs, that the results of two and more measurements, in spite of the corrections applied, differ systematically by a larger amount than the uncertainties of each individual measurement, showing that the sources of systematic errors have not or not completely been removed. One of the most easily recognizable reasons for such systematic discrepancies is different normalization; this is particularly often encountered in capture, (n,p) and (n,α) reactions. In principle this deficiency can be removed by measuring and/or evaluating ac-

curate standard data and renormalization of the original experimental results, an important task, to which much work is already devoted. In the majority of the cases, however, the reasons for such systematic discrepancies are much more difficult to find out being then most probably intimately connected with the experimental apparatus and techniques, ambiguities in the interpretation of the measured raw data and so on. A famous example is the discrepancy between the, Livermore on one side and the Saclay and Harwell on the other side, resonance fission cross section measurements on U^{235}; long discussions among the experts concerned and an own subcommittee of the EANDC were necessary to solve this discrepancy. One of the immediate consequences of such unsolved discrepancies is that the uncertainties of the evaluated data, which are a more or less sophisticated average through such discrepant measurements, are larger than the accuracies asserted to be achieved in the underlying experiments.

Another kind of inconsistency is rather often encountered in the range of overlapping resonances in the keV range of neutron energies, reflecting particularly the difficulties of the accurate determination of the neutron flux in this energy region: measurements with broader energy resolution and worse statistics show larger cross section fluctuations than those with finer energy resolution and better statistics. A prominent example for this kind of inconsistency are the discrepant fission cross section measurements on Pu^{239} in the lower keV range. Unfortunately the parametric character of the nuclear theory already invoked before in general prohibits an unequivocal solution of those discrepancies.

We illustrate some of the above discrepancies on two typical cases, i.e. the evaluation of capture cross section measurements on Mo and Fe in the keV energy range. The first example has already once been discussed by the author at another IAEA conference, i.e. the Seminar on the Physics of Fast and Intermediate Reactors in Vienna in 1961 [1]; we choose again this example, because most of the then open discrepancies could meanwhile be resolved. On the contrary, the Fe keV capture cross section represents at present one of the worst examples of very large unsolved discrepancies.

Figure 1 shows the presently available σ_γ measurements on Mo in the energy range between 1 keV and 1 MeV; no measurements are available above 1 MeV (the reference numbers in figures 1 and 2 are those used in reference [2]). In 1961, apart from the 30 and 65 keV values of Gibbons et al. [Mo - C57] and the 24, 220 and 830 keV results of Belanova [Mo - C51] the following more comprehensive measurements were available: Block et al. [Mo - C62] from below 1 keV to 6 keV; Staviskii, Shapar [Mo - C67] from 50 keV to 1 MeV and Diven et al. [Mo - C63] from 175 keV to 1 MeV. We noted then, that Staviskii's results differed consistently from those of Gibbons and Diven by + 50 to 100 %. This discrepancy could then not be resolved; an average curve through these discrepant results and through the measurements of Block et al. [Mo - C62] was therefore recommended. Afterwards in order to solve the discrepancy semi-empirical statistical theory calculations were carried out by d'Auria and Schmidt [3] for all Mo isotopes in the range between 1 keV and 1 MeV and added to get σ_γ for natural Mo.

In these calculations statistical s-wave resonance parameters determined from measured resolved s-wave resonance parameters were used and spin, energy and, if not defined from resonance measurements, atomic weight dependences of the average level spacings determined from the Fermi gas model of the nucleus. The transmission coefficients for elastic and inelastic scattering of neutrons with higher orbital angular momenta than 0 were computed from the simple complex square well potential of Feshbach et al. [4]. Finally statistical fluctuation factors and inelastic scattering competition to all knwon levels below 1 MeV were taken into account. Without going into the numerical details, the results of these calculations were much better compatible with the data of Gibbons and Diven et al. than with those of Staviskii and Shapar. These results later on were confirmed, when it became apparent, that the discrepancy was solely due to a wrong standard value used by the Russian authors. Originally they normalized their σ_γ data to an I^{127} capture cross section value of 400 mb at 200 keV. A later comparison with the particularly extensive σ_γ measurements

on I^{127} in the keV range due to Bame and Cubitt [5] showed, that this value was by as much as a factor of 1.6 too high. Lowering their standard value by this factor to 245 mb and renormalizing their data Staviskii and Shapar got very good agreement with the results of Diven, as can be noted from figure 1, in which already the corrected Russian results are inserted. However, the good agreement now attained between these two measurements does not necessarily involve the correctness of these data on an absolute scale. This, however, is rather confidentially ascertained by the fact, that both measurements agree in spite of normalization to different standards, and that in particular the U^{235} capture and absorption cross section values Diven used for normalization appear to be reliable being compatible to within a few % with the best presently available measurements on U^{235} [2]. A rather large difference, however, still remains between Staviskii's and Gibbons' [Mo - C57] results between 30 and 65 keV and is so far unexplained; this disagreement is the worse, as Gibbons' σ_γ values are normalized to reliable In σ_γ standards (763 mb at 30 keV and 448 mb at 65 keV). The high 200 keV value of Leipunski et al. [Mo - C81] inserted unchanged in figure 1 is due to the same wrong I^{127} standard as used by Staviskii and Shapar. The results of Belanova [Mo - C51] are believed to be rather unreliable; in her method σ_γ is essentially obtained as the difference of total and scattering cross sections, and, as these are about equally large, great errors are necessarily involved in the difference. This explains the large discrepancy of Belanova's 830 keV value to the other experiments and renders the good agreement of the 24 and 220 keV values with the other experiments purely incidental; for other elements like Cr for example Belanova's results at all three energies differ from other data by an order of magnitude.

Below 50 keV two measurement series are now available due to Kapchigashev and Popov [Mo - C74] and to Mitzel and Plendl [Mo - C75], both being performed with the neutron-slowing-down-in-a-lead-pile method. In spite of the similar method used and the rather good agreement between both measurements below 1 keV, above 1 keV Mitzel and Plendl's results are systematically lower than those of Kapchi-

gashev and Popov by up to 60 % at 30 keV. The decision, which of both measurements is correct, is made almost irrelevant, since the errors involved in both measurements are of the order of the difference between both measurements, i.e. 30 % and more: the method involves the estimate of σ_γ from the difference of the combined γ-spectra of Pb and the investigated material and of the γ-spectrum of Pb alone and, as σ_γ is already rather small above 1 keV, the cross section values in the keV range obtained by the lead pile method necessarily become rather unreliable. The only argument in favour of Kapchigashev's results is that they join better to the renormalized results of Staviskii and Shapar thought of as reliable than those of Mitzel and Plendl. Both lead pile measurements, however, concordantly show, outside their experimental errors and in compatibility with the calculations mentioned above, that the earlier measurements of Block et al. [Mo - C62] below 6 keV are by more than a factor of two too high. Also this discrepancy can be explained: it is due to the lack of corrections for multiple scattering before capture and resonance self shielding in Block's measurement which both render the measured capture cross section too high; particularly the latter correction has been applied in the two lead pile experiments and is responsible for most of the discrepancy. On the basis of the foregoing arguments it is understandable why the curve called "presently recommended" in figure 1 has been chosen as a smooth average through the results of Kapchigashev and Popov, Staviskii and Shapar and Diven et al.

For the Fe capture cross section in the keV range even more discrepancies are encountered and less of them solved than for Mo. Figure 2 shows the available experimental data. The region 100 keV to 1 MeV does not interest us here; apparently the different measurements are compatible in this range. The region of discrepancies between most of the available experiments extends from 100 keV down to about 100 eV. We begin with a comparison of the two lead pile measurements of Isakov et al. [Fe - R57] and again of Mitzel and Plendl [Fe - R80]. As in the case of Mo, both measurements were performed under essentially the same experimental conditions. In spite of this similarity in the method, two characteristic differences are

apparent, a shift in the energy scale of about 1 to 2 keV between both measurements at energies above 2 keV, and, different heights of the peaks observed in both experiments between 100 and 1200 eV. Concerning the first discrepancy, probably the energy scale in the Russian measurements is correct. This is suggested by a comparison of the broad peaks observed in the broadly resolved capture measurements with the more detailed resonance structure as observed in much finer resolved transmission experiments. The transmission experiments reveal a smaller resonance at 6.0 keV in Fe^{57} and a larger resonance at 8.0 keV in Fe^{54}. In the capture measurements one would thus expect to see a large peak centered nearer to 8 keV than to 6 keV, and this is actually only the case for the Russian measurement. The second discrepancy can be unequivocally clarified. The peaks observed below 1 keV are clearly due to Mo, Co and Mn impurities in the samples of both authors; the difference in the amount of these admixtures explains the difference in the observed peak heights. The larger peak height Mitzel and Plendl observe for the 1.2 keV resonance in Fe is also due to a much larger impurity admixture than in the Russian sample of Mn^{55} which has a resonance at 1.08 keV. Thus one is led to the conclusion that there is no resonance in Fe below the 1.2 keV resonance and that the capture cross section there follows an undisturbed 1/v law. This conclusion is furthermore ascertained by the very careful and well resolved capture and transmission studies of Moore et al. [6] in the vicinity of 1 keV, which do not reveal any other resonance than that at 1.2 keV.

Thus the differences between the two lead pile measurements are well understood. The more disturbing are the much larger discrepancies between the lead pile measurements on one side and the Harwell linear accelerator [Fe - R79] and Oak Ridge Van de Graaff [Fe - R82] measurements on the other side in the range from about 1 to 100 keV. The small differences between these two latter measurements are due to differences in isotopic sample composition and do not concern us here. The much smaller shape and much higher peak cross section observed in the Harwell measurement compared to the two lead pile experiments is clearly the result of the much better energy resolution

in the Harwell measurements. Capture areas and resonance parameters deduced from all three experiments for the 1.2 keV resonance are well compatible with each other after due correction for the impurity admixtures in the lead pile measurements. Still the much finer resonance structure observed by Harwell and Oak Ridge above 1 keV is clearly explainable as being due to the much finer energy resolution. Unexplained remains so far the systematic discrepancy by an average factor of about 2 to 3 between Van de Graaff and linear accelerator, and the lead pile results. It is true, that the linear accelerator measurements are still not corrected for multiple neutron scattering before capture; this means that the corresponding σ_γ values plotted in figure 2 are actually too high. However, at best this correction is expected to be important in the large 28 keV resonance in Fe^{56} and does thus not explain the discrepancies below this resonance; furthermore the Oak Ridge measurements are corrected for this effect and are actually smaller in this resonance than the Harwell measurements, but still about twice as large as the lead pile results. From the known capture widths of heavier nuclei than Fe (see e.g. the survey of reference [7]) one would expect capture widths of Fe resonances to be of the order of 0.4 to 0.5 eV, values which actually could be derived from the observed lead pile capture areas, whereas the about 3 times larger Harwell and Oak Ridge capture areas are only compatible with about 3 times larger capture widths. Thus this argument based on nuclear systematics of the capture widths would be in favour of the lead pile results. On the other side some serious doubt is cast on these results by the large uncertainties, already discussed above in the case of Mo, connected with the subtraction of the Pb γ-ray background in the keV range; however, it can not be said, whether the true capture cross section values are larger or smaller than those observed, and the resulting large uncertainties in the lead pile σ_γ do not account for the discrepancy to the other measurements.

A final possibility to decide between the discrepant measurements consists in a comparison of direct determinations of the non-1/v part of the infinite dilute capture resonance integral with values calculated from the experimental capture cross section data. Without going into numerical detail, the result is, that five independ-

ent direct measurements of this quantity are altogether between 2 and 4 times as large as the value calculated from the highest differential, i.e. the Harwell data (for details we refer to reference [2], section III). Our obvious conclusion from the above arguments consists in the preliminary recommendation, valid as long, as these discrepancies are not understood and removed, of a 1/v behaviour below 1 keV and of the Harwell σ_γ results in the range 1 to 100 keV as representing most closely the true energy dependence of σ_γ for Fe; the curve called "presently recommended" in figure 2 shows this compromise.

We elaborated rather extensively on these two examples, to which easily others could be added, because they show better than any general discussion the true bottle-neck of every evaluation. Almost needless to mention the impossibility of the reliable prediction of the physical properties of a fast reactor working with steel as structural material, if such big discrepancies are not removed.

A second general problem in evaluation, but generally not as severe as the discrepancies, are the gaps in experimental information. In the simplest case one has to do with a smoothly energy dependent cross section, which is well established in the whole energy range concerned except a certain interval in which no data are available; simple graphical interpolation closes such a gap reliably. More open to question is such a graphical or a statistical theory interpolation in a region of overlapping resonance structure; the error of the interpolation is of the order of the variance of the cross section concerned. A gap in the resolved resonance range can principally not be closed adequately, because there is no theory which predicts the position and properties of the resonances; here only the experiment can help, and any interpolation is necessarily pure invention.

Certain limits in the experimental techniques render the experimental investigation of the one or other cross section impossible. In many cases the finite energy resolution in the detection of the

scattered neutrons for example prohibits measurements of the inelastic excitation of well-known separate nuclear levels. In these cases the simple statistical theory developed by Hauser and Feshbach [8] with transmission coefficients from realistic optical potentials is used; it has proven to predict neutron inelastic excitation cross sections within the, however, still rather large uncertainties and inconsistencies of the experimental data. Naturally this does not mean, that the theory is able to replace a good experiment.

Also in other cases nuclear theory can be successfully used for closing gaps, provided that important parameters entering the theory are sufficiently well known. Due apparently to some cancellation effect in the neutron transmission coefficients, the generally very narrow distribution of the capture widths as concluded from resonance experiments and their weak spin and energy dependences, the Hauser-Feshbach theory applied to fast capture cross sections of medium weight and heavy nuclei yields rather reliable results. Above 1 MeV for many nuclei large gaps in experimental capture cross section data exist; at the same time the compound hypothesis underlying the Hauser-Feshbach theory becomes more and more invalid and the direct capture more and more important; the theory for this transition range is still unsufficient. Fortunately, from the practical point of view, for most nuclei the absorption of neutrons by other reactions than capture, mainly by the (n,p) and (n,α) processes, is more important in the MeV range. Unfortunately the experimental information on these latter reaction cross sections is often rather scarce, if at all available, and in addition often discrepant. Although extensive work has been devoted to the refinement and improvement of optical model transmission coefficients for neutrons and charged particles and the level density laws, the predictions of the evaporation theory are still not reliable enough ranging from rather incidental good agreement to complete incompatibility with experiment. One of the reasons is the strong dependence of the evaporation cross section expressions upon such shell structure influenced and still not accurately enough known parameters as the single particle level

density. The usefulness of refined optical models for the prediction of elastic scattering angular distributions and non-elastic scattering cross sections at energies where the compound elastic scattering has died out, is so well-known and has so often been described that we can neglect it here. The practically and fundamentally equally important problem of the correct interpretation of fission resonances is also only mentioned here; the author considered this problem extensively elsewhere [9].

In the third place we wish to draw the attention not so much to a problem, but rather an inconsistency encountered in every evaluated data set, being the consequence of a similar inconsistency in the experimental information used. In a given energy subrange measurements of different neutron cross sections generally differ in the experimental conditions, in particular in the energy resolution. The better resolved cross section might still exhibit something like a resonance structure, whereas the other worse resolved cross section shows a smooth energy dependence. A famous example for this are the very broadly resolved KAPL reactor spectra α measurements and the 10 nsec/m and better resolved Saclay and Harwell fission cross section measurements on U^{235} in the 100 eV to low keV range. The only possibility to render such two measurements consistent with each other is to fold the better resolved measurement with the energy resolution function of the worse resolved measurement. Generally this is not done in order not to loose the detailed information contained in the better resolved measurement. However, if these two cross sections have to be used to determine a third one, because this is not or only with large difficulties attainable to experiment, this third cross section can get large errors, on the average of the order of its variance, depending upon the size of the difference in resolution in the first two cross sections. In our example this is true of the capture cross section of U^{235} determined from σ_f and α. This rather frequent observation has only a consequence for the experiment: in order to get a physically true picture of "derived" cross sections and nuclear data, the experimental conditions, foremost the energy resolution, in the measurement of the "basic" cross sections and nuclear data should be as similar as possible. The more this condition is fulfilled in the

experiments, the more physically meaningful and consistent in all, "basic" and "derived", quantities become the evaluations.

In the foregoing we discussed some of the main physical problems encountered in the evaluation of neutron cross sections. Upon their solution in each individual case not only the reliability of the evaluated data for the accurate calculation of reactor physical properties, but also their usefulness for the checking of nuclear theories depends. The examples we discussed demonstrate the large difficulties one will be faced with in the trial of systematizing and automatizing the evaluation process. However, in order to keep pace with the still increasing amount of experimental data flowing in and to utilize to the fullest extent the possibilities of the large computers, the automatisation without loss of the individual physical aspects will certainly be one of the main future problems in evaluation.

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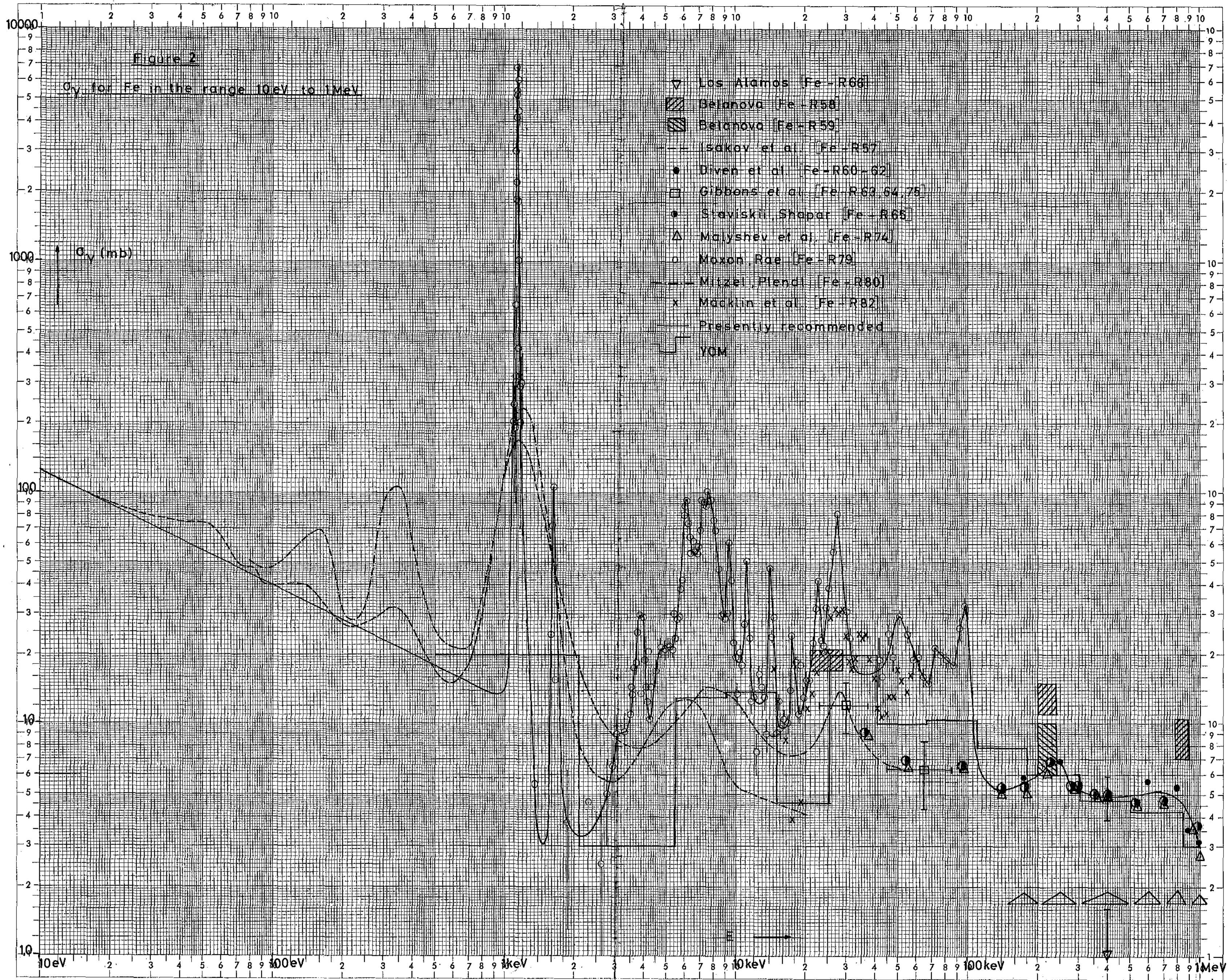
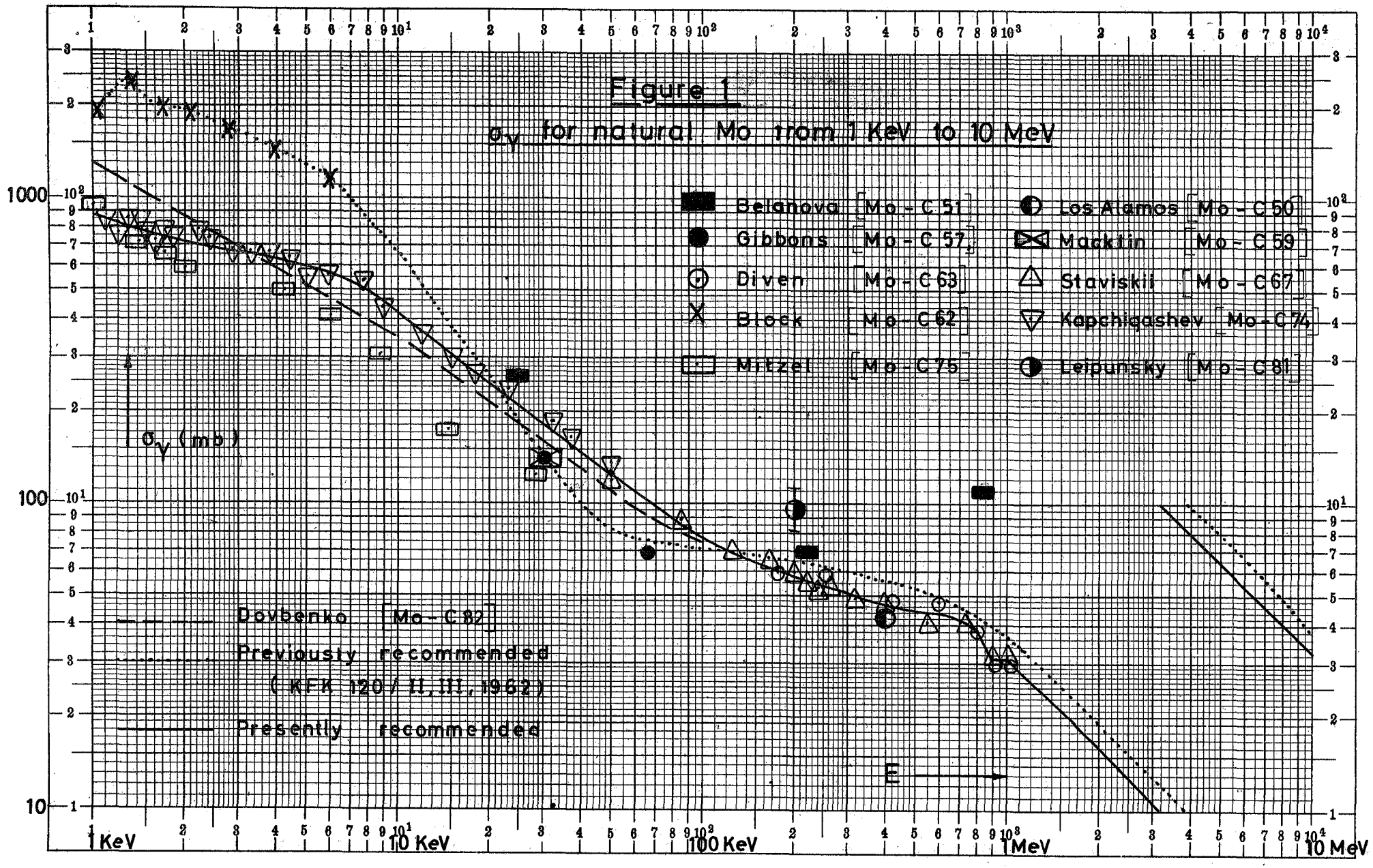


Figure 2

σγ for Fe in the range 10eV to 1MeV



Beide Achsen logar. geteilt von 1 bis 10000 und 1 bis 300, Einheit 62,5 mm