IAEA-199

ATOMIC AND MOLECULAR DATA

FOR FUSION

PROCEEDINGS OF AN ADVISORY GROUP MEETING
ON ATOMIC AND MOLECULAR DATA FOR FUSION
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
HELD AT THE UKAEA CULHAM LABORATORY
ABINGDON, UK, 1—5 NOVEMBER 1976

A TECHNICAL DOCUMENT ISSUED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1977

Measurements of Atomic Transition Probabilities in Highly Ionized Atoms by Fast
Ion Beams.

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Abstract

A summary is given of the beam-foil method by which level lifetimes and tran-
sition probabilities can be determined in atoms and ions. Results are presented
for systems of particular interest for fusion research, such as the Li, Be, Na,
Mg, Cu and Zn isoelectronic sequences. The available experimental material is
compared to theoretical transition probabilities.

7. <u>Introduction</u>

In connection with spectroscopic studies of impurities in high temperature plasmas it is of vital interest to know the oscillator strengths for the observed transitions (1,2, 3). It is by no means an easy task to measure oscillator strengths in highly ionized atoms. Recent developments in experimental atomic physics using fast ion beams from accelerators (beam-foil spectroscopy) (4, 5) have given very promising results, however. With the beam-foil method one measures the lifetime τ_i of an excited level which is related to the atomic transition probability A_{if} and oscillator strength f_{fi} as follows

$$\tau_i^{-1} = A_{if} = \frac{6.67 \cdot 10^{15} \cdot k \cdot f_{fi}}{\lambda^2} \tag{1}$$

where λ is the transition wavelength in A and $K = g_f/g_i$ the ratio of the statistical weights of the lower and upper levels. The transition probability in Eq (1) is expressed in s^{-1}; typical values for electric dipole transitions are $10^7 - 10^9$ s^{-1}.

A variety of methods have been developed for experimental determination of lifetimes and transition probabilities. The methods include emission and absorption measurements, the Hanle effect technique, electron excitation using pulsed or modulated beams as well as the beam-foil method. We will here only consider the last mentioned method which is the only one that has been applied to +3 and higher ionization states. All other methods so far known are only useful for f-value studies in neutral or a few times ionized systems. For a review of the field see Ref. (6).

2. Survey of the beam-foil technique.

The beam-foil method has been in use for more than ten years (7, 8) and a large number of atoms and ions have already been studied in this way. The principle is schematically shown in Fig. 1. A beam of fast ions is obtained from a particle accelerator, e.g. a Van de Graaff generator. After magnetic analysis the isotopically pure beam is directed through a thin exciter foil, 500 - 1 000 A in thickness. As a result of collisions with the foil atoms the fast ions can undergo stripping and excitation. The excited atoms decay in flight and the light emitted can be analyzed spectroscopically as shown in Fig. 1. The foil-excited ion beam is thus a spectroscopic light source with several interesting properties.

1) Practically any element can be ionized and accelerated in present-day machines. Beam-foil spectra of more than 60 chemical elements have already been observed. A large interval of ionization degrees are available, ranging from neutrals to some thirty times ionized species. Using a 714 MeV Kr beam from the Berkeley Super-HILAC, Gould and Marrus (9) have thus been able to measure lifetimes in helium-like krypton, Kr XXXV.

2) The beam-foil light source is a time-resolved one. The foil-excited ions travel with a uniform velocity which can be determined to a few per cent accuracy (or better), e.g. using an electrostatic analyzer as shown in Fig. 1. By measuring spectral line intensities, usually by photon-counting techniques, at various distances from the foil one obtains decay curves from which radiative lifetimes can be conveniently extracted. Lifetimes ranging from a few ps to several hundred ns have been measured in this way. The excited ions decay in vacuum and the beam-foil method thus avoids problems such as collisional de-excitation and trapping of resonance radiation.

In the following we shall concentrate on lifetime measurements and results. We have selected transitions in relatively simple systems (1-2 valence electrons) which have appreciable oscillator strengths, i.e. the $\Delta n=0$ resonance lines in the

Li, Be, Na, Mg, Cu and Zn isoelectronic sequences. As shown in Refs (1-3) these are particularly important in the studies of plasma impurities.

3. Lifetime measurements.

In the simplest case the level lifetime τ_i can be obtained from the relation

$$I(X) = I(0) \exp(-x/v\tau_i) \qquad (2)$$

where I(x) is the intensity at a distance x from the foil and v the velocity of the ions. However, the beam foil excitation is not a selective one, instead many levels are usually populated. This leads to cascading processes, by which the levels under study are repopulated from higher-ly ng states. Experimentally one obtains decay curves that do not follow the simple equation (2) but must be approximated by a sum of several exponentials. Examples of such decay curves are shown in Fig. 2. The cascading introduces experimental uncertainties and demands very good counting statistics for unambiguous extraction of lifetimes. Fortunately such problems have been studied for quite some time and a number of remedies are available which reduce the uncertainties even in the case of heavy cascading (10). In most cases modern beam foil experiments now give f-values with 5-10% uncertainties. This fact can be checked by comparing beam foil data for neutral atoms to f-values obtained from Hanle-effect measurements. The latter method here gives uncertainties of only a few percent.

The lower limit for lifetime measurements is given by the spatial resolution along the foil-excited beam, which is approximately 0.1 mm Using high-energy beams from powerful accelerators (e.g. Tandem Van de Graaffs, cyclotrons or heavy-ion linear accelerators) as well as good optical imaging methods it is now possible to measure lifetimes as short as a few ps (11, 12). Such lifetimes are not unusual in very highly ionized systems.

Parallel to this development on the experimental side there are important refine- ments in theoret cal calculations of oscillator strengths. Many such results are found in the tables issued by Wiese et al. (13). The Coulomb-approximation method

of Bates and Damgaard (14) gives reliable f-values in simple systems. Lindgdrd and Nielsen (15) have recently applied a numerical Coulomb-approximation by which a large number of f-values (even for transitions between low levels) have been calculated. The problems of electron correlation have been successfully tackled by several authors, e.g. Weiss (16), Sinanoglu (17), Nicolaides and Beck (18), and Hibbert (19).

Configuration-interaction studies, applicable to heavier elements, have also been reported by Froese Fischer (20) and Biémont (21). A Z-expansion method, developed by Dalgarno and coworkers (22) permits f-value calculations for whole isoelectronic sequences. It is also worth emphasizing that it is now possible to calculate rigorous error bounds for theoretical f-values (23).

Relativistic effects on f-values have recently received much attention. Kim and Desclaux (24) have thus studied highly ionized members of the Li and Be isoelectronic sequences. Weiss (25) reports a similar study for Be-like ions while Sinanoglu and Luken (26) investigate the B sequence. Some of these results will be commented below.

When comparing experimental and theoretical f-values it is often enlightening to search for systematic trends and regularities in the available material. Wiese and Weiss (13) have investigated such regularities, e.g. with respect to isoelectronic and homologous systems as well as spectral series. The regularities within isoelectronic sequences are of special interest in the present context.

According to perturbation theory the oscillator strength for a given transition, e.g. $2s\ ^2S - 2p\ ^2P$ in the Li isoelectronic sequence (Li I, Be II, B III etc.) can be written as a power series in $1/Z$, i.e.

$$f = f_0 + f_1(1/Z) + f_2(1/Z^2) + \qquad (3)$$

where f_0 is the hydrogenic value, being equal to zero for $\Delta n=0$ transitions. It is instructive to graphically display f-values vs $1/Z$. As shown by Smith and Wiese (13) it is possible to estimate unknown f-values by this technique. In the present case we shall study such curves for the resonance lines in the Li I, Be I, Na I, Mg I, Cu I and

Zn I isoelectronic sequences. Here beam foil measurements have provided a wealth of experimental data which can be compared to theoretical values. Another reason for selecting these cases is that transitions with appreciable f-values (in the 0.1-1 range), such as the above-mentioned resonance lines in high Z systems, are very important in quantitative studies of impurity concentrations in Tokamak plasmas (1, 2).

The following discussion shows that beam foil experiments have not yet reached these very highly ionized species which are observed in plasmas, e.g. Fe XXIV. Here only theoretical f-values are available at present. However, as already pointed out by Wi'ese and Younger (3) beam foil studies of low Z ends of isoelectronic sequences are of great value in checking various theoretical approaches. In view of present developments in accelerator technology we can safely expect extensions of beam foil work to very high ionization states.

Li I sequence.

The $2p\ ^2P$ level lifetime in Li-like systems has been measured for Li I to Na IX by beam foil methods. The results are generally in good agreement with various theoretical calculations. Recently Martin and Wiese (27) have made a thorough analysis of the oscillator strengths in the Li I sequence and much information about theoretical and experimental f-values for various transitions in Li-like ions can be obtained from their paper. In the present context lithium like metal ions are of special significance. In Fig. 3 we show the theoretical lifetimes for levels in Fe XXIV, calculated by the Coulomb approximation (28). We note a very typical trend, the $n=3$ and $n=4$ terms have very short lifetimes while the first excited level, $2p\ ^2P$ has an order of magnitude longer lifetime. Since the Coulomb approximation calculations take advantage of experimental excitation energies and quantum defects relativistic effects on f-values are partly accounted for. However, a more specific study of such effects on the $2s$-$2p$ transitions in the Li I sequence can be found in Refs. (24) and (27). These calculations show that the relativistic and non-relativistic multiplet f-values are rather close to each other for $Z<30$. However, the $2s\ ^2S_{1/2}$-$2p\ ^2P_{3/2}$ f-value starts to show an increase from

the nonrelativistic f-value already for lower Z systems and the difference is very important e.g. for Mo XL.

Be I sequence.

The f-value information for the $2s^2\,^1S-2s2p\,^1P$ resonance transition is displayed in Fig. 4 . The theoretical results for the low Z end are taken from Refs (13), (20) and (23) while those for Z>10 have been calculated by Nussbaumer (29). In all calculations considered here configuration interaction has been taken into account. This is very important in systems with two valence electrons. Hartree-Fock calculations in the single-configuration approximation thus yield f-values which are typically a factor of two too high in this sequence. Only one beam foil value is given for all ions in Fig. 4, the selection being based on a critical investigation of the available data. The B II case merits further investigations, however it is not of crucial importance in the present context. For higher Z theory and beam foil measurements are in reasonable agreement although the latter f-values systematically tend to be smaller. Here also, additional experiments are needed, preferably for higher Z, to confirm the present trend. With the help of Tandem accelerators equipped with sputtering ion sources further lifetime studies, e.g. for Mg IX or Si XI are quite possible.

As already mentioned, relativistic calculations for the Be sequence are discussed in Refs (24) and (25). The conclusion is that in the case of the $2s^2\,^1S-2s2p\,^1P$ transition the relativistic f-values start to deviate from the nonrelativistic ones around Fe XXIII and then - for higher Z - the difference grows drastically.

Na I sequence

Much interest has recently been focussed on the f-values for the $3s\,^2S - 3p\,^2P$ resonance multiplet in this sequence. The data are shown in Fig. 5. For low Z the beam foil results are in good accord with the theoretical values

of Froese Fischer (20), shown in the figure, whereas small but significant deviations exist for S VI, Cl VII and - in particular - Ar VIII. In the theoretical work (20) both configuration mixing and core polarization were included and the calculated f-values are therefore estimated to possess a high accuracy with uncertainties around a few per cent. Crossley et al. (30) have investigated the experimental situation, particularly with respect to cascading problems. Simulated beam-foil decay curves for the $3p\,^2P$ term have been constructed assuming that more than 20 upper levels were populated at the beam-foil excitation. (The inclusion of an even higher number of upper levels does not change the picture.) The curves, computed using calculated lifetimes (28), and realistic excitation models, are displayed in Fig. 6. (Note that the abscissa is in lifetime units). This figure shows, as already concluded in Ref. (30) that cascading should not be a major source of error in e.g. Ar VIII, because decompositions of the corresponding decay curve should give a principal component very close to the "true" $3p\,^2P$ lifetime. The reason for the discrepancy is still unknown, it is possible that spectral line blends might have complicated the experimental situation. It is also interesting to point out that the agreement between various theoretical results for the $3s\,^2S - 3p\,^2P$ transition probability is not always good, as e.g. noted in Ref. (30). Wiese and Younger (3) also analyze the relativistic contributions to this sequence f-values and find them to be insignificant for $Z < 30$. Even for Mo XXXII the relativistic part of the f-value is rather small.

Mg I sequence

Here, also, we are interested in the resonance transition, $3s^2\,^1S - 3s3p\,^1P$ (Fig. 7.). Beam-foil data, supplied by various authors, here cover the spectra of Mg I - Ar VII. The theoretical values are from the model-potential calculations of Victor et al. (31) and - for higher Z - from the 1/Z expansion procedure of Crossley and Dalgarno (32). Because of configuration interaction, similar to that in the homologous Be I sequence, theoretical calculations are here much more laborious than in the case of Na-like ions. Figure 7 makes clear

that here we have a problem similar to that mentioned above, theory and experiment do not agree too well in Cl and Ar. Work is in progress to construct decay curves (33), similar to those in Fig. 6 and explore to what extent cascades might have been responsible for the experimental uncertainties and whether systematic errors thus were overlooked. It is worth mentioning that Berry et al. (34) recently measured lifetimes for a number of levels in Cl VI and Ar VII (Mg I sequence) as well as Cl VII and Ar VIII (Na I sequence) and in practically all cases their results agree with theory within the estimated uncertainties. The $3p\ ^2P$ and $3s3p\ ^1P$ levels were unfortunately not studied by Berry et al. (34) but it should be obvious that new measurements of these lifetimes, preferably for as high Z as possible, would be extremely valuable. In this way it ought to be possible e.g. to draw more definite conclusions as to the reliability of the theoretical values in the region of fusion interest.

Cu I sequence

The f-value trends for the $4s\ ^2S - 4p\ ^2P$ resonance line in this sequence have been thoroughly discussed by Wiese and Younger (3) who noted appreciable discrepancies between the experimental and theoretical data for Cu I - Kr VIII. The former came from beam foil measurements (35,36) whereas the latter were based on Hartree-Fock (HF) calculations with relativistic corrections. More recently, Froese Fischer (37) has reanalyzed the Cu I sequence using a multiconfiguration Hartree-Fock (MCHF) approach. Her results, together with the HF f-values and beam foil data are shown in Fig. 8. Lindgård and Nielsen ('28) have also calculated these f-values and their results are rather close, although somewhat higher, than the MCHF ones.

It is obvious from the figure that the present situation is not very satisfactory and renewed efforts are thus worthwhile. We have now initiated a new beam foil study of this sequence. In Zn II the lifetimes for the $4p\ ^2P$, $4d\ ^2D$ and $5s\ ^2S$ terms have been determined (38) and the preliminary analysis is consistent with a somewhat higher f-value than that shown in Fig. 8. Also the

Ga III case is being studied (39), here one of the conclusions is that line blending - certain Ga II and Ga III multiplets are very close in wavelength - may seriously complicate lifetime measurements by beam foil methods. A new study of these spectra, using classical atomic spectroscopy (40), has therefore been initiated. Hinnov (41) has observed two lines, 373.8 and 423.5 A, in connection with a spectroscopic study at the Princeton ST Tokamak, and classified these as the $4s\ ^2S - 4p\ ^2P$ doublet in Cu-like Mo (Mo XIV). It would of course be extremely important to determine the corresponding oscillator strengths, and this ought to be possible by the beam foil method.

As a guide to further experiments we have constructed simulated decay curves, so far limited to Cu I - Se VI, using the calculated lifetimes of Ref. (28). The result is shown in Fig. 9. A most interesting effect is the initial rise of the intensity for Ga III and higher ions, explainable by growing-in processes from higher levels with shorter lifetime than $4p\ ^2P$.

Zn I sequence.

The f-values for the $4s^2\ ^1S-4s4p\ ^1P$ resonance line are shown in Fig. 10. Two sets of theoretical values are available. The open circles are from the calculations of Warner (42) who used Thomas-Fermi-Dirac wavefunctions while the crosses are from Coulomb-approximation work (28). The latter data agree better with the experimental f-values, from beam foil studies of Sørensen (35) and Livingston (36) although a discrepancy still exists. We have now remeasured the Zn I f-value, finding good agreement with Refs. (28) and (35), while an analysis of a new Ga II study is in progress.

In view of the importance of configuration mixing, e.g. between $4s^2$ and $4p^2\ ^1S$ in the ground state and $4s4p$ and $4p4d\ ^1P$ in the excited state (c.f. the Be I and Mg I sequences) calculations based on C I would be desirable for this sequence.

We finally note that the $4s^2\ ^1S-4s4p\ ^1P$ transition in Mo XIII (341.0 A) has been identified by Hinnov (41).

4. Summary and conclusion

We have given a short survey of the beam foil method by which transition probabilities in highly ionized atoms can be determined. A wealth of such data, even for transitions n the far UV and soft X-ray regions, has been accumulated, of which we here have discussed f-values for $\Delta n=0$ resonance lines in the Li I, Be I, Na I, Mg I, Cu I and Zn I isoelectronic sequences.

As emphasized e.g. in Ref. (5) modern beam foil work has in many cases yielded f-values that are much smaller than the results of conventional HF calculations, sometimes order-of-magnitude differences exist. More extensive calculations in which electron correlations are included have usually led to very good agreement with the experimental f-values. This is also illustrated in Figs. 4, 5, 7 and 8. However, also in such cases we note that the theoretical and experimental f-values tend to depart from each other when Z increases in an isoelectronic sequence, indeed this occurs before relativistic effects on f-values start to play a role. This is a serious problem which has to be solved before safe conclusions can be drawn about f-values in very h ghly ionized systems, e.g. Li-like Mo.

One possible explanation for shortcomings on the experimental side is cascading into the level under study. However, our simulated decay curves for the first excited level in the Na I and Cu I sequences (Figs. 6 and 8, respectively) show that the cascading problems become less serious when Z increases. After an initial rise the constructed curves follow rather closely the expected np ^{2}P (n=3, 4) decay time. The reason for this is also shown in Fig. 3 where the 2p ^{2}P level survives all n=3 and n=4 levels. The fact that such resonance lines with $\Delta n=0$ have much lower transition rates than $\Delta n=1$ lines in highly ionized systems should also permit lifetime measurements with the beam foil method of cases such as the 2p level in Fig. 3.

With new powerful heavy-ion accelerators (43) we can therefore expect important breakthroughs in lifetime measurements in highly ionized species within the near future. As an illustration of this, Fig. 11 shows a beautiful beam foil spectrum of iron, obtained at the Oak Ridge tandem accelerator by Bashkin and coworkers (44). The 3s ^{2}S-3p ^{2}P doublet in Fe XVI, at 335.4 and 360.8 A and the 3s ^{2} ^{1}S-3s3p ^{1}P

resonance line in Fe XV, at 284.1 A are clearly observable, together with many other important transitions in highly ionized iron. Lifetime measurements, which already are in progress (44) might well resolve some of the present difficulties, discussed above.

In this review we have mainly discussed f-value studies of importance for determining impurity concentrations in high temperature plasmas. It is equally important to have good knowledge of the atomic energy levels and wavelengths (1, 2, 46). Several groups are actively engaged in this work and much important material of relevance to plasma diagnostics has been obtained in recent years.

We have already mentioned that our new lifetime program has initiated a study of ionized Ga. Additional investigations of the Cu I and Zn I sequences are also being undertaken at the University of Lund because the data available at present' are often very unsatisfactory. In addition to the inherent interest in obtaining accurate level energies and ionization limits, such material is also of importance in beam-foil investigations of lifetimes and theoretical calculations of oscillator strengths.

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Figure captions

Fig. 1.   Experimental arrangement for beam-foil studies of lifetimes.

Fig. 2.   Three examples of decay curves, encountered in lifetime measurements
using the beam-foil technique. In (a) cascading is virtually absent
while (b) and (c) show the effects of repopulation from a shorter-
lived (b) or a longer-lived (c) higher level.

Fig. 3.   Theoretical lifetimes (in ps) for $n=2$, 3 and 4 terms in Fe XXIV (Li I
sequence), calculated in the Coulomb approximation (28). The lifetimes
for $n=3$ and $n=4$ terms are   too short to be measurable by beam-
foil. However, the $2p$ lifetime could probably be studied experimentally.
Thus if Fe-ions are accelerated to, say, 300 MeV and sent through a foil,
the excitation of the $2p$ ^2P level in Fe XXIV is quite probable. A life-
time of 20 ps would then correspond to a 1/e decay length of 0.7 mm,
about 10 times longer than the spatial resolution in modern beam-foil
work (11, 12).

Fig. 4.   Comparison between theoretical and experimental oscillator strengths
for the $2s^2$ ^1S-$2s2p$ ^1P transition in the Be I sequence. The experimen-
tal points were critically selected from various beam-foil papers, see
e.g. Refs. (23) and (29).

Fig. 5.   Comparison between theoretical and experimental oscillator strengths
for the $3s$ ^2S-$3p$ ^2P transition in the Na I sequence. The data sources
are found in Refs. (3), (20) and (30).

Fig. 6.   Simulated decay curves for the $3p$ ^2P level in the Na I sequence. More
than 20 cascading levels were included in the analysis and initial
populations according to $(2\ell+1)(n^x)^{-3}$ were assumed, ℓ and n^x being
the azimuthal quantum number and effective quantum number respectively.
Theoretical lifetimes (28) were also used. Note the initial intensity

rise in e.g. Ar VIII, explainable by cascading from higher levels with lifetimes shorter than that for $3p\ ^2P$.

Fig. 7.   Comparison between theoretical and experimental oscillator strengths for the $3s^{2}\ ^1S-3s3p\ ^1P$ transition in the Mg I sequence. The data sources are discussed in the text and Ref. (45).

Fig. 8.   Comparison between theoretical and experimental oscillatorstrengths for the $4s\ ^2S-4p\ ^2P$ transition in the Cu I sequence. The data sources are Refs. (3),(35), (36) and (37).

Fig. 9.   Simulated decay curves for the $4p\ ^2P$ level in the Cu I sequence, obtained in the same way as the curves in Fig. 6.

Fig. 10.   Comparison between experimental and theoretical oscillator strengths for the $4s^{2}\ ^1S-4s4p\ ^1P$ transition in the Zn I sequence. The data are from Refs. (28),(35), (36) and (42).

Fig. 11.   Beam-foil spectrum of iron in the grazing-incidence region. Ions of Fe were accelerated to 35 MeV in the Oak Ridge tandem accelerator and the wavelength spectrum was analyzed with a 2.2 m grazing-incidence monochromator (44).
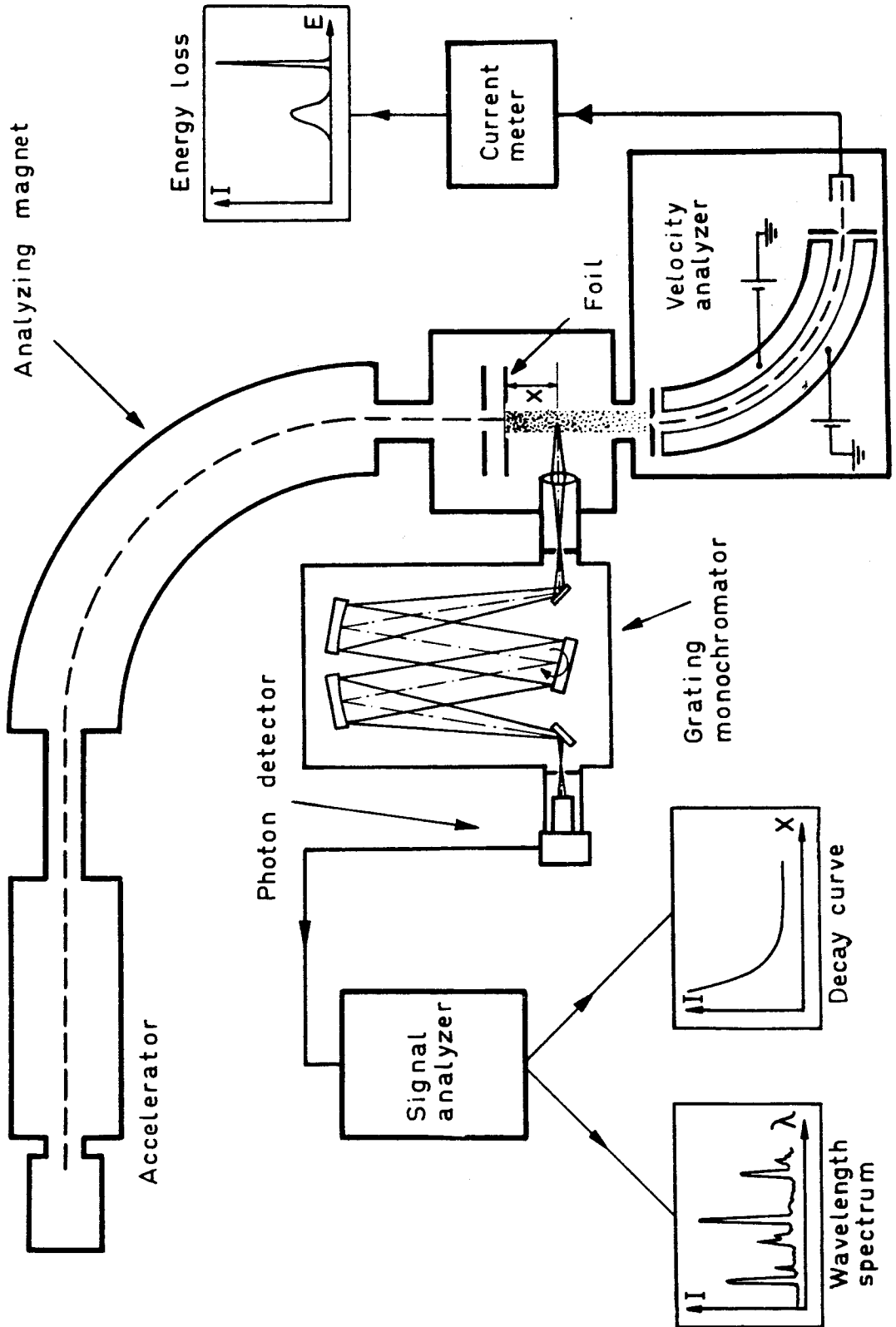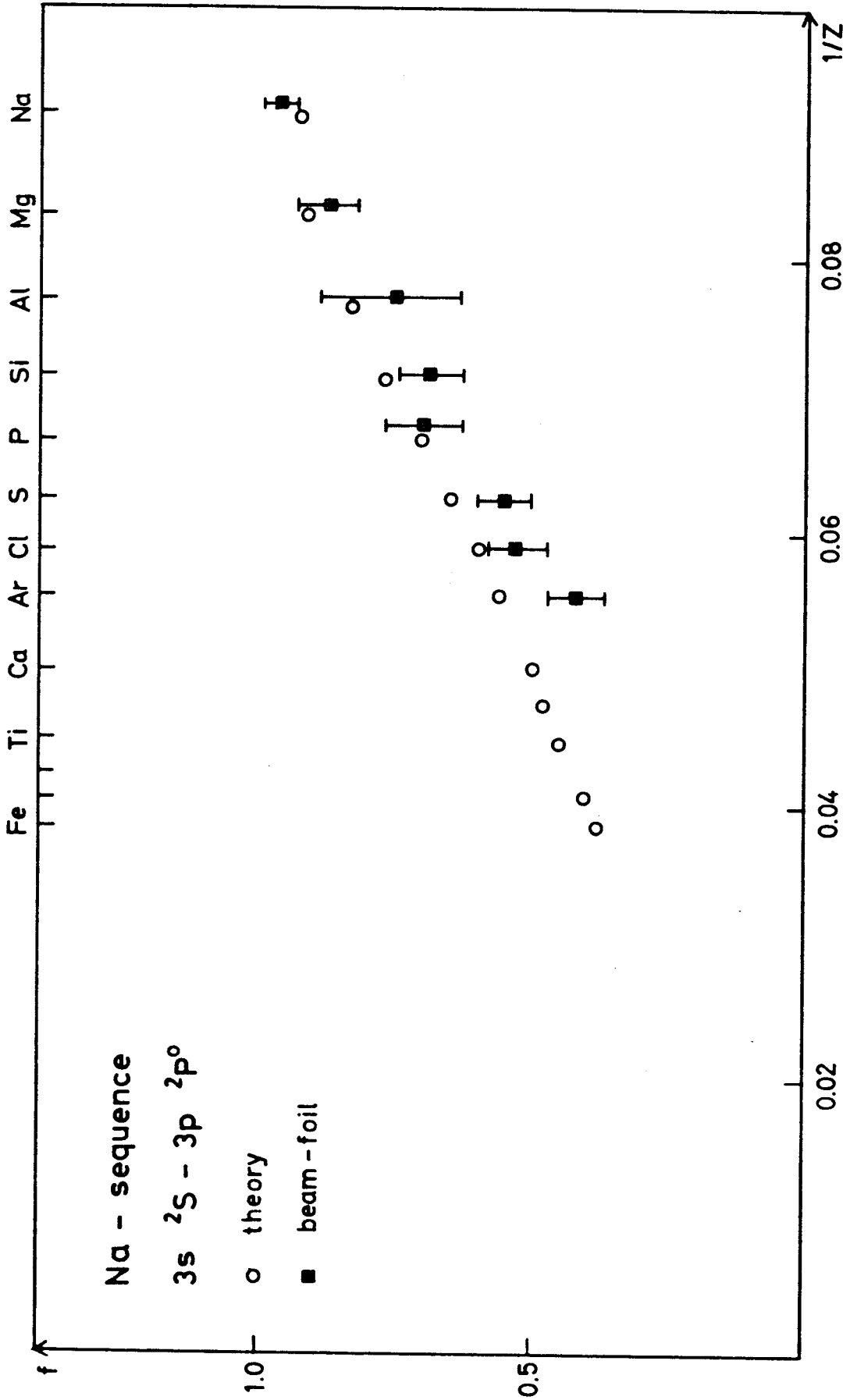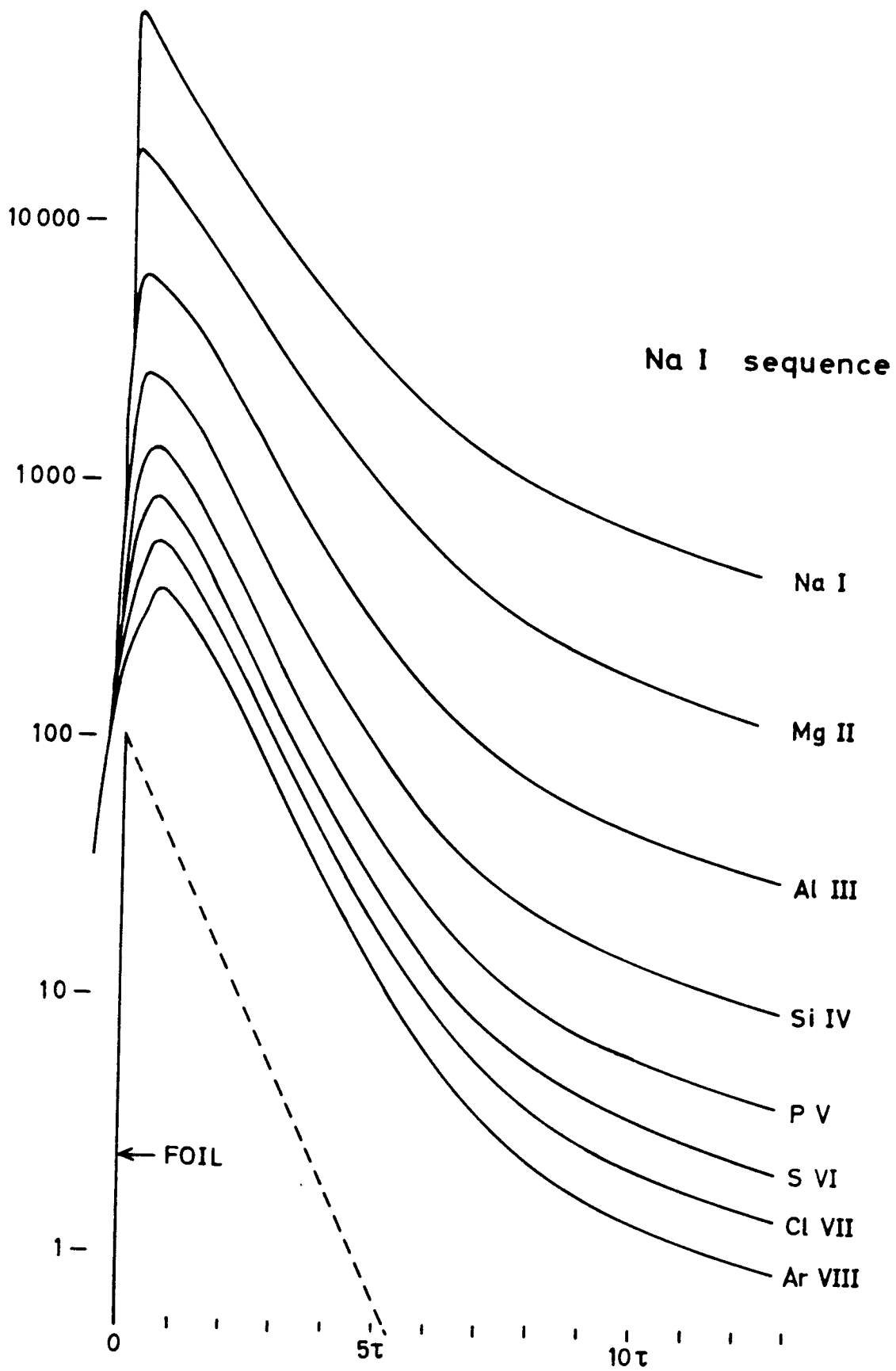
Figure 1
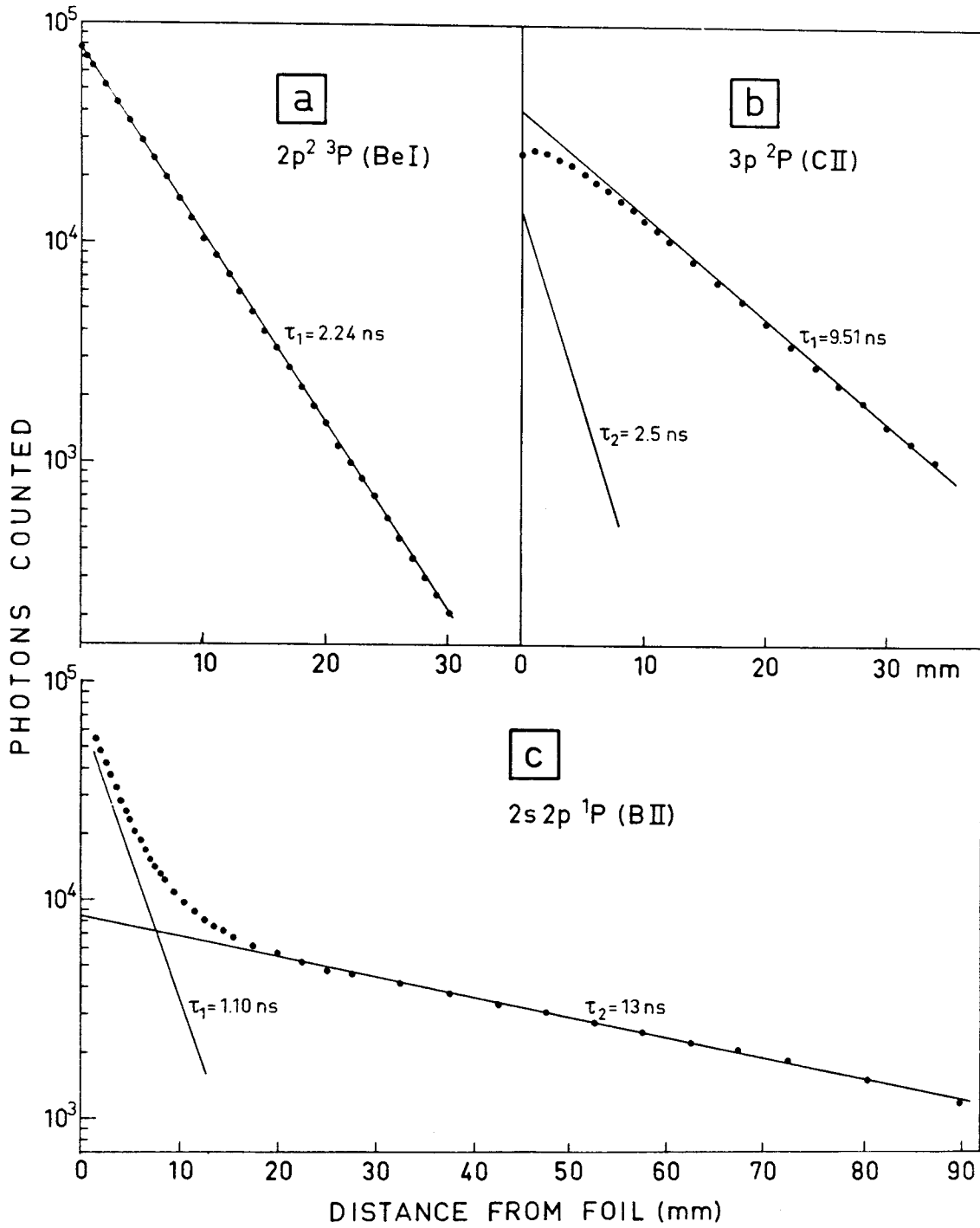
Figure 2

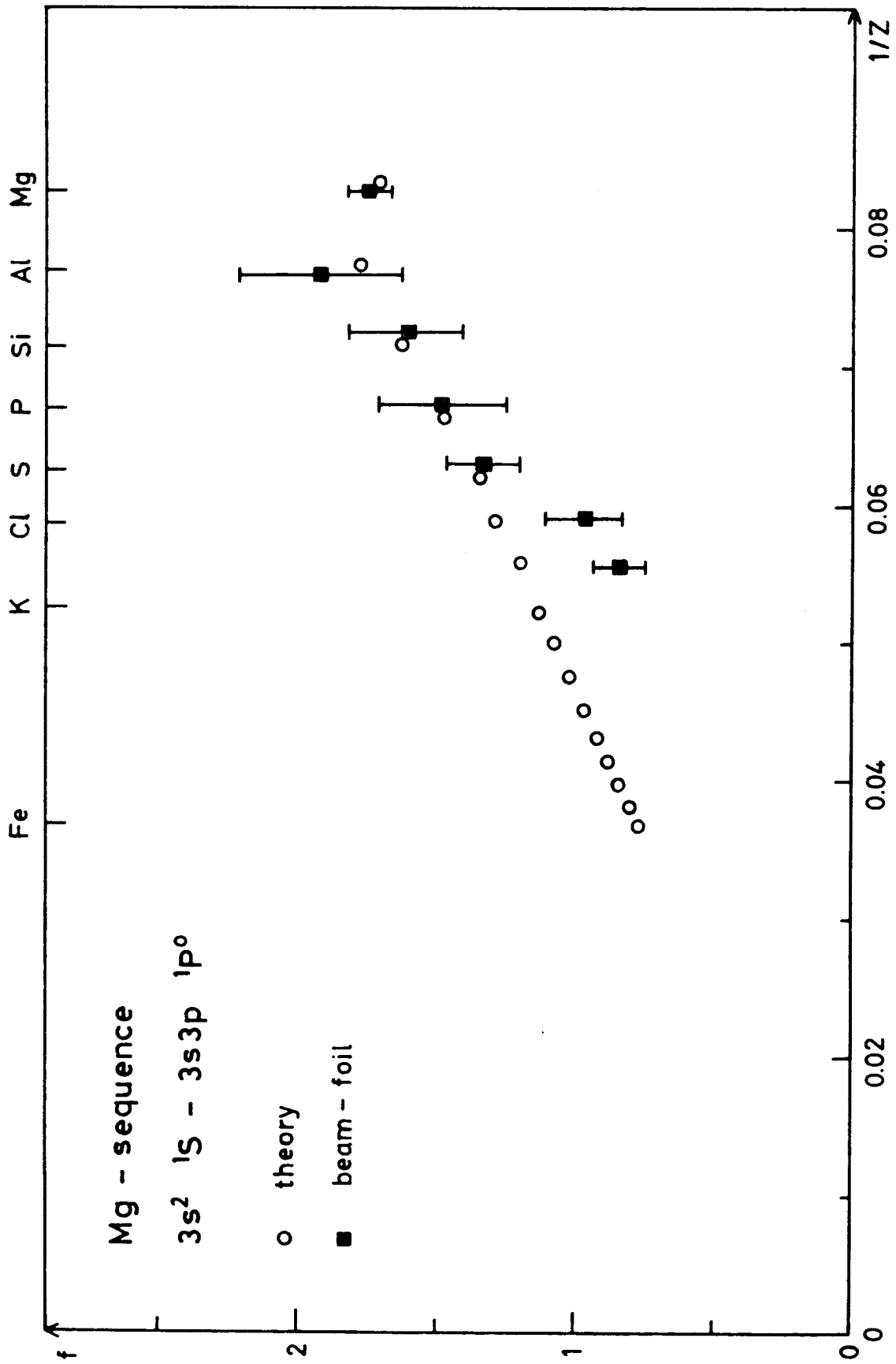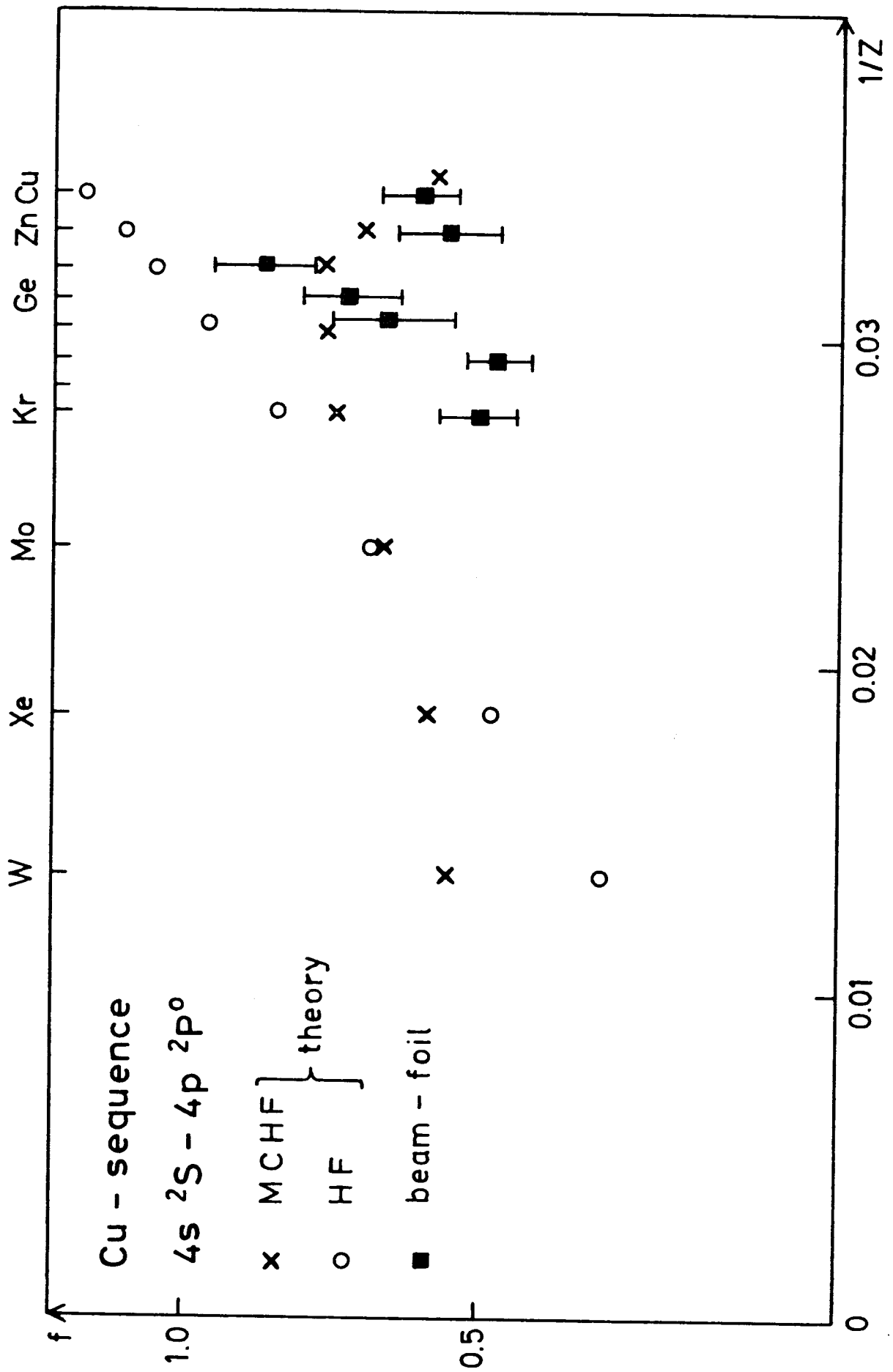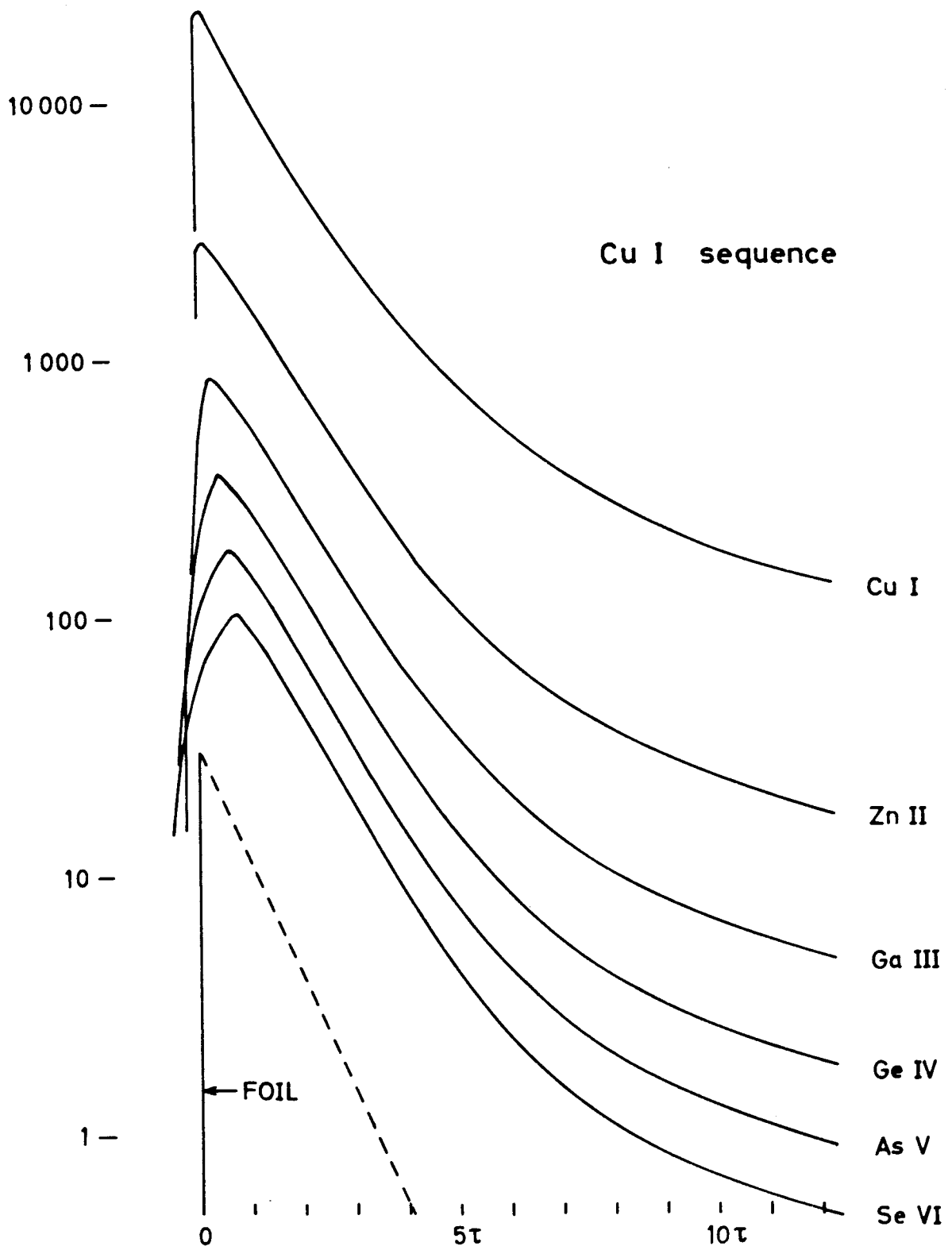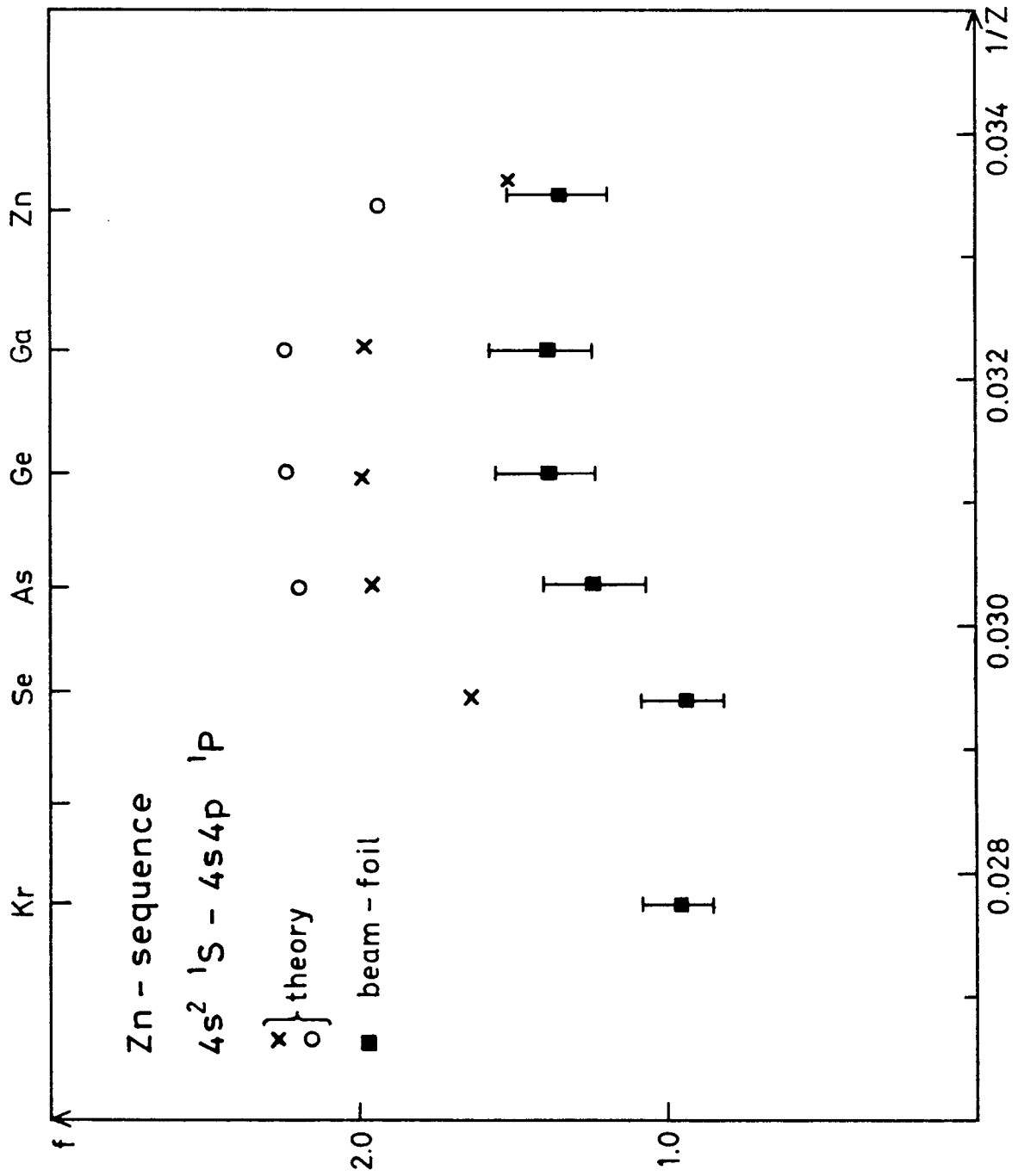# Fe XXIV, theoretical lifetimes (ps)
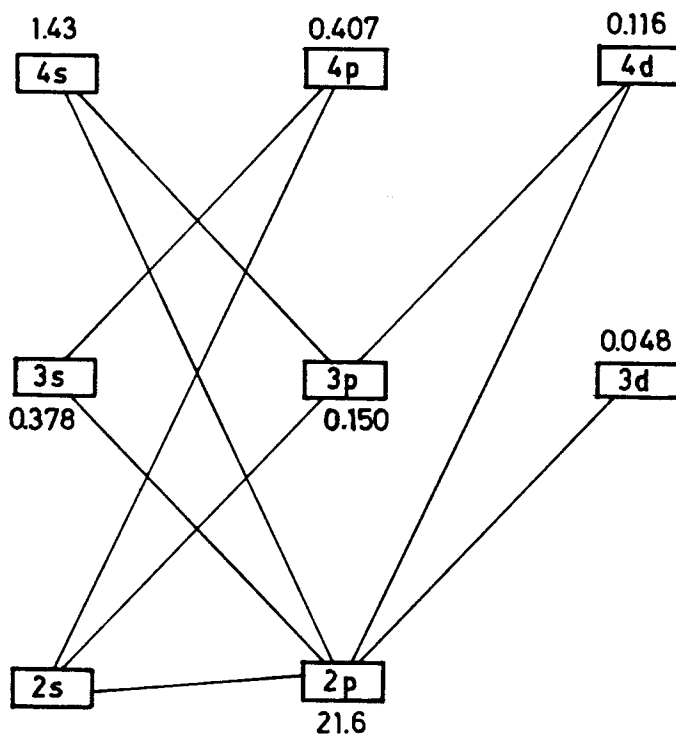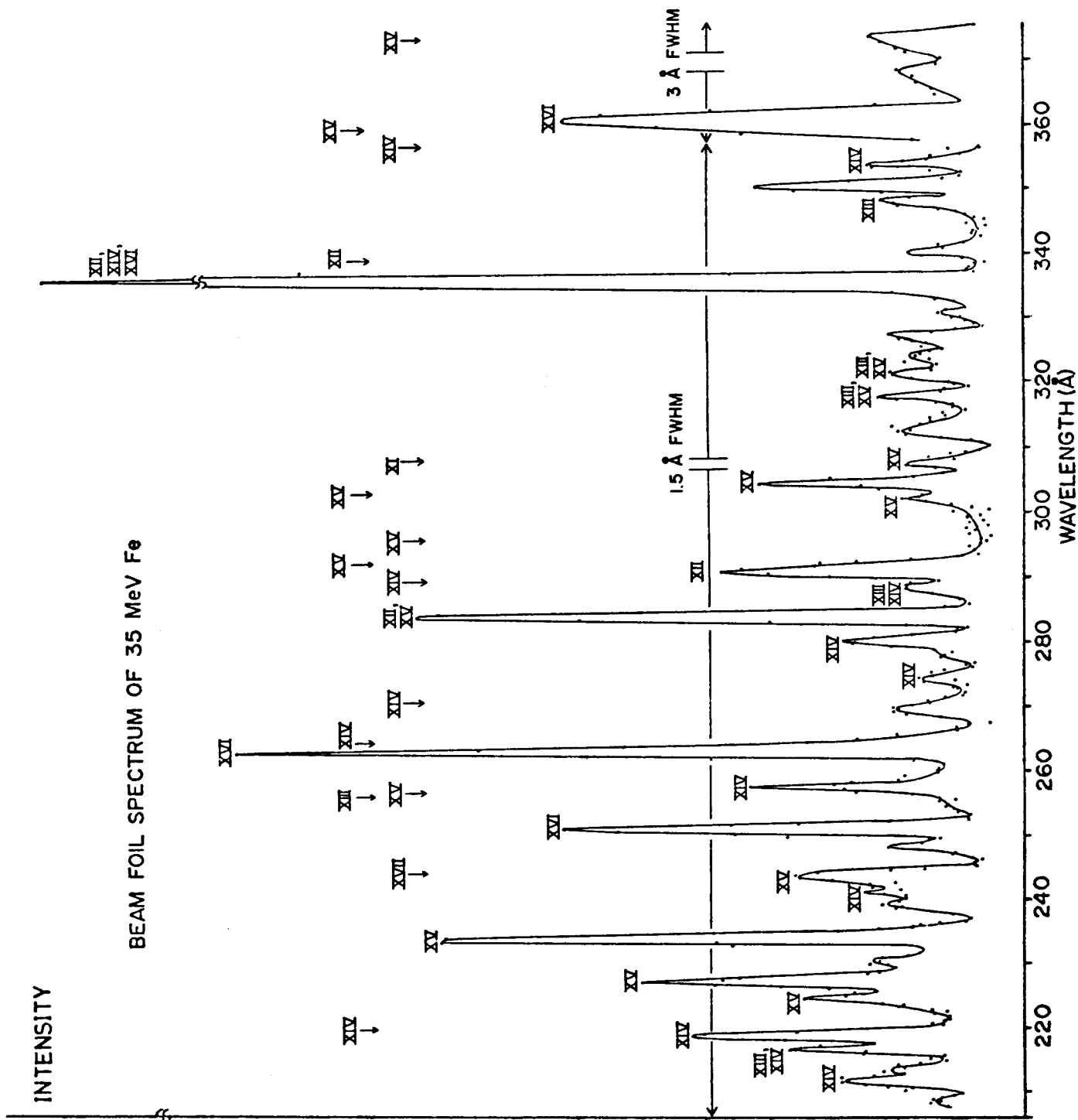

Figure 3
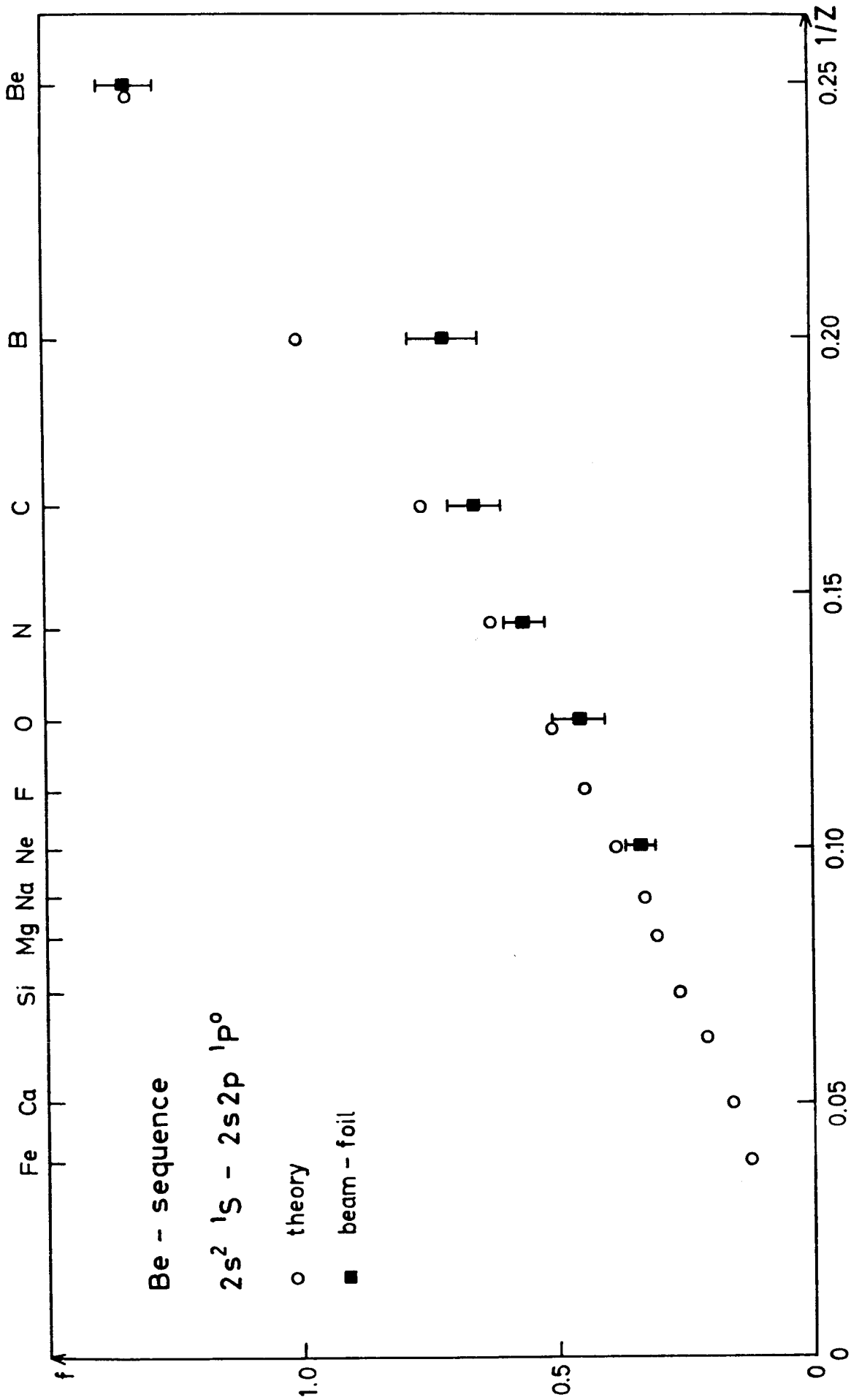
Figure 4

Figure 5

Figure 6

Figure 7

Fig. 8

Figure 9

Figure 10

Figure 11