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FUTURE DIRECTIONS FOR BEAM-FOIL SPECTROSCOPY

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FUTURE DIRECTIONS FOR BEAM-FOIL SPECTROSCOPY

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The beam-foil source has proved to be so useful for the study of atomic energy levels that it is almost trivial to propose a variety of new experiments involving new elements, higher energies, a broader wavelength range, shorter time intervals, pulsed beams, different targets, and new configurations in geometry or external fields. However, what is perhaps not so trivial is to propose experiments for which there is a specific purpose, experiments from which a novel kind of information might be expected. It is from this latter point of view that I shall talk about experiments which seem to offer unusual opportunities to learn new things about atoms.

To start, Fig. 1 shows the type of beam-foil spectra well-known to everyone here. These spectra were taken with iron,¹ accelerated in one of the Brookhaven tandem Van de Graaffs. At the bottom are data taken at an energy of 16 MeV; at the top are some results at 110 MeV. My colleagues in this experiment were Jack Leavitt (Arizona), Keith Jones and Dan Pisano (Brookhaven), and Ted Kruse (Rutgers).

While there are interesting changes in the spectra as the energy is raised, I wish to concentrate on the spectral analysis. Many of these lines are tentatively identified as originating in transitions between Bohr-type levels, that is, levels which, except for the central charge, are hydrogenic in character, having large values of n , of l , and of orbit radius. I take as a convenient definition of a hydrogenic level one which is degenerate in l .

Preliminary analysis of the 16-MeV data suggests that hydrogenic lines from Fe X through Fe XVI are all present. Thus, Fig. 2 illustrates the transitions which appear to derive from Fe XVI.

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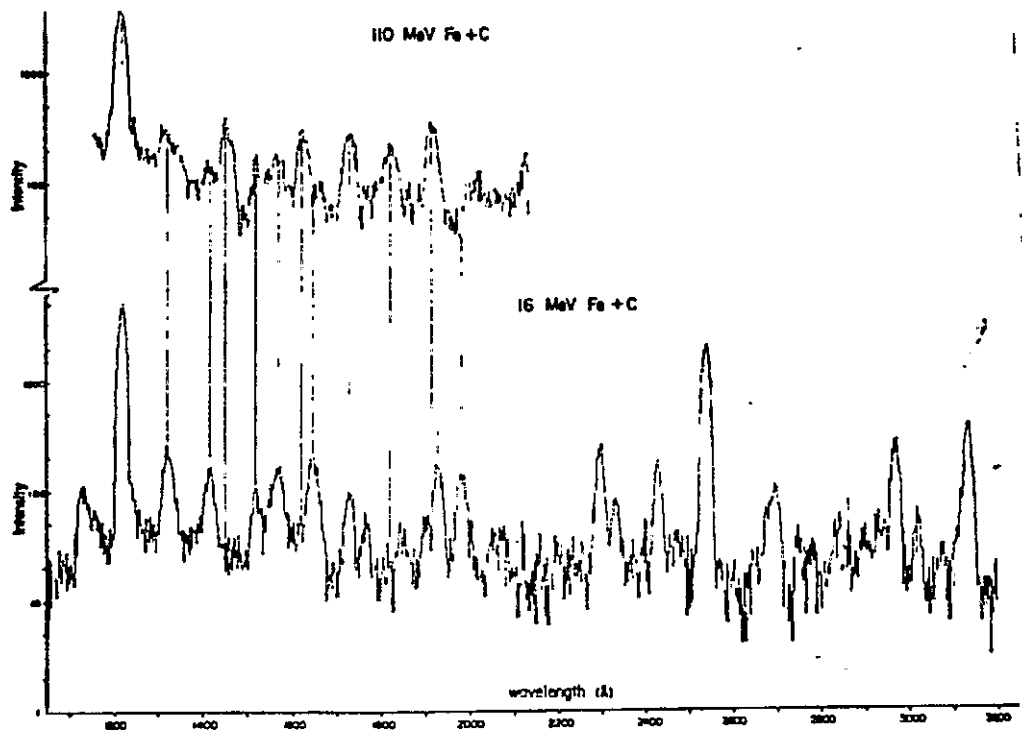


Fig. 1. Beam-foil spectra of iron at 16 MeV (lower) and 110 MeV (lower) on carbon. The vertical lines indicate where maxima in the curves line up.

The solid lines indicate transitions which are probably present in the data; the dashed line indicates one which should have been seen but wasn't. Clearly, the identification of our data with Fe XVI as the source is very good, for we are missing only one of the many transitions expected in our wavelength range.

Now the Bohr radius of orbit n is given by

$$r_n = \frac{n^2}{Z} r_0 \quad (1)$$

where r_0 is the Bohr radius for the ground state of hydrogen. For $Z=16$ and $n=16$, we find $r_n \sim 8\text{\AA}$. Such an orbit, which is considerably larger in radius than the atom-atom spacing ($\sim 2\text{\AA}$) in the foil, is unexpected, but it is often claimed, as I have just claimed, that the foil-excited ions commonly emerge in such states. A significant question, then, is whether one has properly interpreted the origin of these so-called hydrogenic lines.

After all, the evidence for such an interpretation is only the wavelength measurement and the Bohr-type calculation. By proper

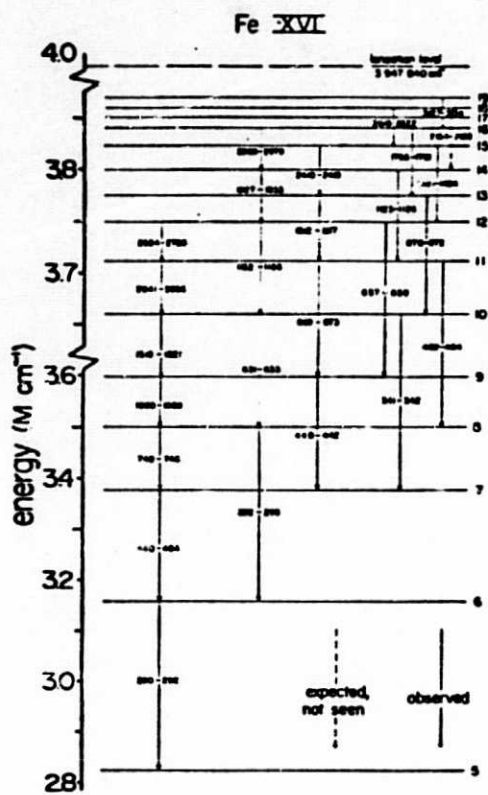


Fig. 2. Transitions in Fe XVI. Observed transitions (solid), expected but unobserved transitions (dashed). Transition wavelengths are indicated.

choice of Δn and the core charge, one can approximate virtually any wavelength. An approximation is all that is needed, since the linewidth for the data of Fig. 1 was 15\AA , large enough to permit serious errors of identification. Linewidths of that or even much larger values are often used in BFS.

In the foregoing remarks I relied on Bohr theory to justify the high net charge used in identifying the spectral lines. Other evidence which may be used to substantiate that choice includes the charge distribution of iron ions emerging from thin foils; the mean charge, at the energies we used, was +13 or more. More convincing is the information displayed in Fig. 3. Here we see data on 35 MeV Fe as obtained² with a grazing-incidence spectrometer installed on the Oak Ridge tandem Van de Graaff. This work was done by Jones, Pisano, Kruse, and myself, in collaboration with Paul Griffin, David Pegg, and Ivan Sellin, of ORNL. Here we were able to see transitions involving deep-lying states, where degeneracies of l , and even of j , could be resolved. Also, the linewidth was as narrow as 0.75\AA ,

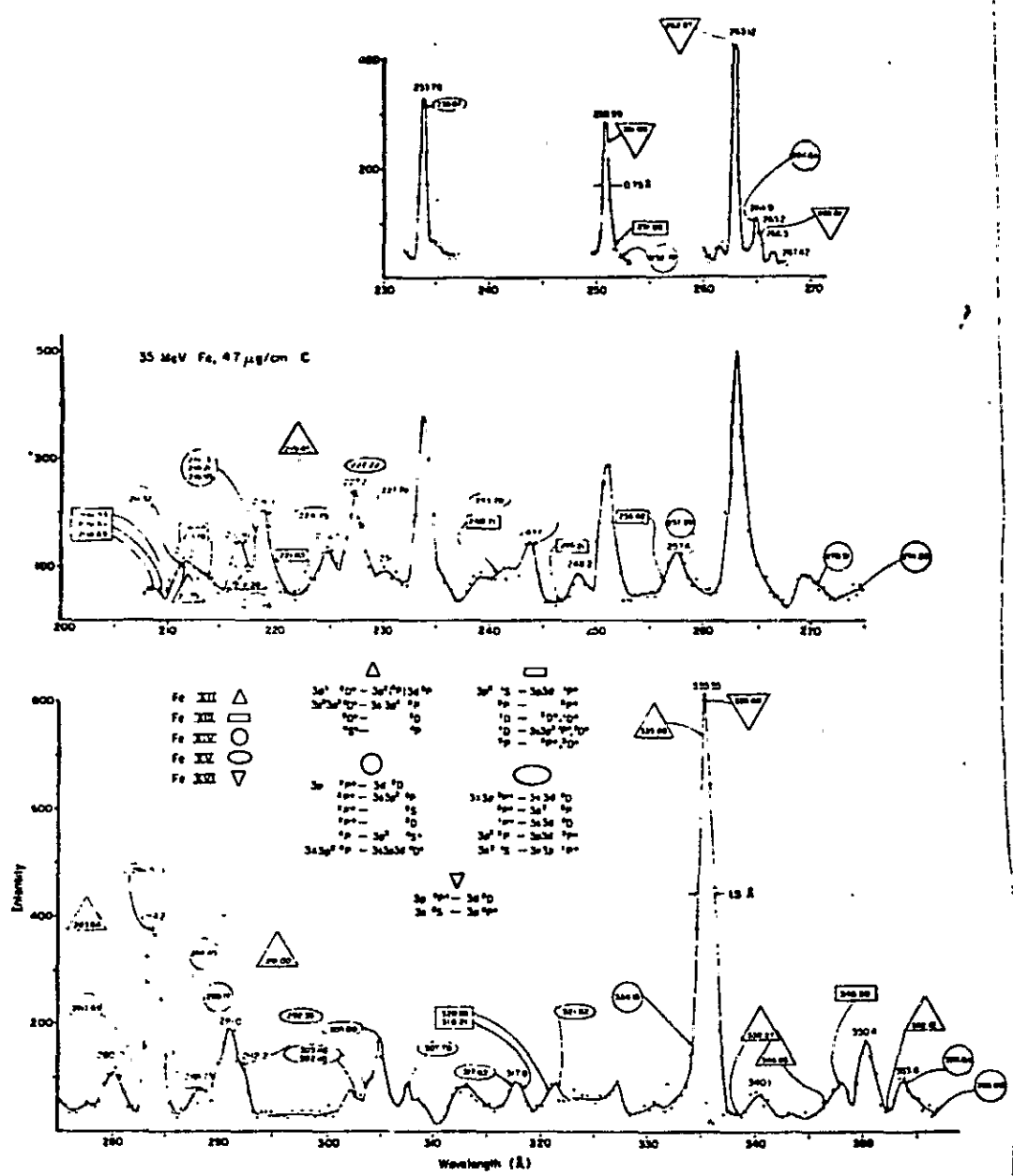


Fig. 3. Beam-foil spectra of iron at 35 MeV. The topmost figure shows high resolution data to be compared with the middle figure. The bottom figure shows an extension of the wavelength range.

so the spectroscopic identifications could be made with confidence. A number of lines can be attributed to Fe XVI, and there doesn't seem to be any reason to doubt that the lines detected in these two experiments indeed originated in high stages of ionization of iron.

Table 1. Transition probabilities and their reciprocals for some states in hydrogen. The average mean lives are calculated assuming statistical equilibrium. After Capriotti (Ref. 3).

n, ℓ	$A(x 10^{-5})$ (sec)	A^{-1} (nsec)	$gA(x 10^{-5})$ (sec)	$\bar{\tau} \equiv n^2 / \sum gA$ (nsec) ²
6s	18.7	536	18.7	
p	244.7	41	734.1	
d	83.9	119	419.4	
f	41.6	240	291.5	192.8
g	24.8	404	222.8	
h	16.4	609	180.6	
12s	3.0	3,337	3.0	
p	31.1	321	93.3	
d	10.8	925	54.1	
f	5.4	1,848	37.9	
g	3.3	3,102	29.0	
h	1.7	5,721	19.2	
i	1.5	6,630	19.6	4,452
k	1.1	8,908	16.8	
l	0.9	11,523	14.8	
m	0.7	14,462	13.1	
n	0.6	17,708	11.9	
o	0.5	21,357	10.8	

Thus, if any of the lines are hydrogenic, the orbits are as large as I have said.

However, there is an immediate reason for suspecting that the lines displayed in Fig. 1 are actually not accounted for by the hydrogenic assumption. In Table 1, we list some transition probabilities for hydrogen, as calculated by Capriotti.³ Table 1 lists the transition probabilities for each of the ℓ -states, the reciprocals of those A -values, and the average mean life of each n -level calculated on the assumption of statistical equilibrium.

Now the average mean life varies as Z^{-4} so that for the choice of $Z=16$, we see that even for $n=12$, the average mean life is a mere 0.07 nsec. Thus, even at 110 MeV, where the speed of an iron ion is $\sim 2 \times 10^9$ cm/sec, the mean decay length is only 1.4 mm. Yet, quite remarkably, the particle beam was visible to the eye over the total viewing length of some 10 cm. Unfortunately, time did not permit us to make a detailed spectroscopic study as a function of distance, but it is qualitatively clear that we deal with states with lifetimes far, far too long for the hydrogenic interpretation to be

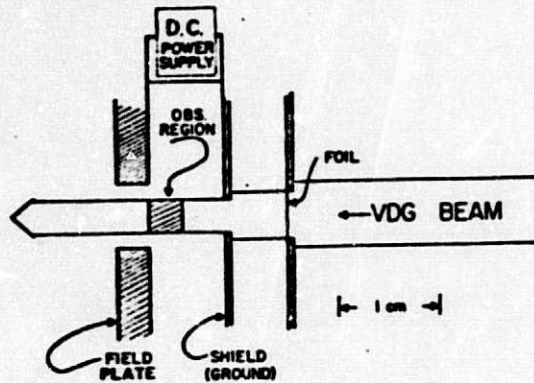


Fig. 4. Schematic arrangement for line-quench studies.

fully satisfactory. At least some of the states we excited may have a different origin, perhaps in some forbidden transitions.

The hydrogenic aspect of BFS merits a thorough investigation. What is needed, in part, is a non-spectroscopic test of the hydrogenic character of the suspect lines. Let me discuss one such test, which I proposed, and which Professor Leavitt has tried in the laboratory.

By definition, a hydrogenic level exhibits a high degree of ℓ -degeneracy, especially when n is large. Thus, each spectroscopic term decays with a broad mixture of mean lives, the extreme range being 14 at $n=5$, 39 at $n=6$, and 67 at $n=12$. Now the application of an external electric field has the effect of mixing the several ℓ -states, with the result that the average mean life is reduced, often sharply so. This is the basis for the test of hydrogenic character.

In Fig. 4 we see a schematic representation of the experiment.⁴ The intensity of the suspect line is measured at some point downstream from the foil. An electric field is applied parallel to the particle velocity so as not to deflect the beam. The field is strong enough to induce Stark-mixing of the ℓ -levels, which reduces the lifetime. For measurements made well downstream, the line intensity declines, but it rises for measurements made at the entrance to the field.

Leavitt has used this method, first, on krypton, and then, with an improved geometry, on neon. He has kindly provided Fig. 5 from some of his unpublished work.⁴ On the upper part, we see a spectral scan, the spectrometer being a 1-meter normal-incidence instrument. This scan was taken with zero external field. On the lower part is the same spectral region as studied with an applied field of 28 kV/cm. Clearly, the line at 2976Å has been quenched, relative to

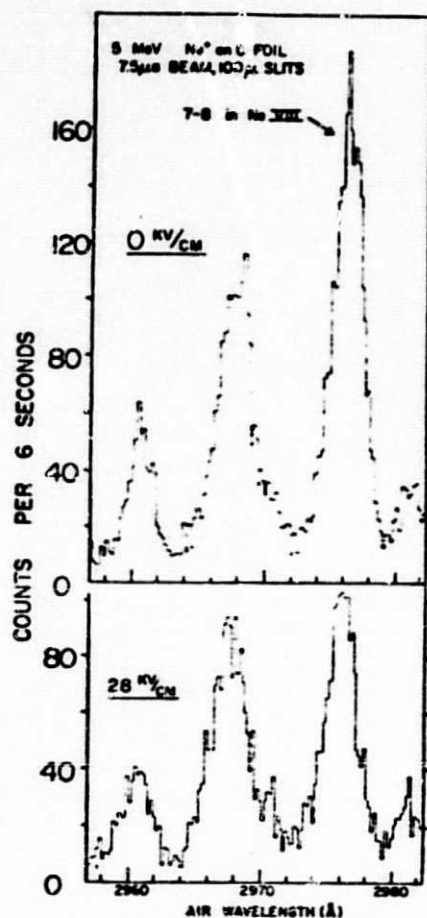


Fig. 5. Beam-foil spectra of neon (top) without electric field, and (bottom) with electric field.

the others. This line, which spectroscopy alone had identified as coming from $n=8$ in Ne VIII, is hereby proved to originate in an ℓ -degenerate state.

Figure 6 shows a case in O VI. The lines near 3434Å are attributed to hydrogenic transitions, as illustrated. The method was slightly changed for this case. Instead of taking complete spectral scans with field off and on, Leavitt merely measured the peak intensity of each of these lines for field off and field on. The line labeled 6d-7f actually rose in intensity by 46% for a field strength of 23 kV/cm, while both the other lines declined by 24%. Thus the hydrogenic identification is substantiated.

This test is rather simple from the experimental point of view, and I believe that future work on spectra should include this

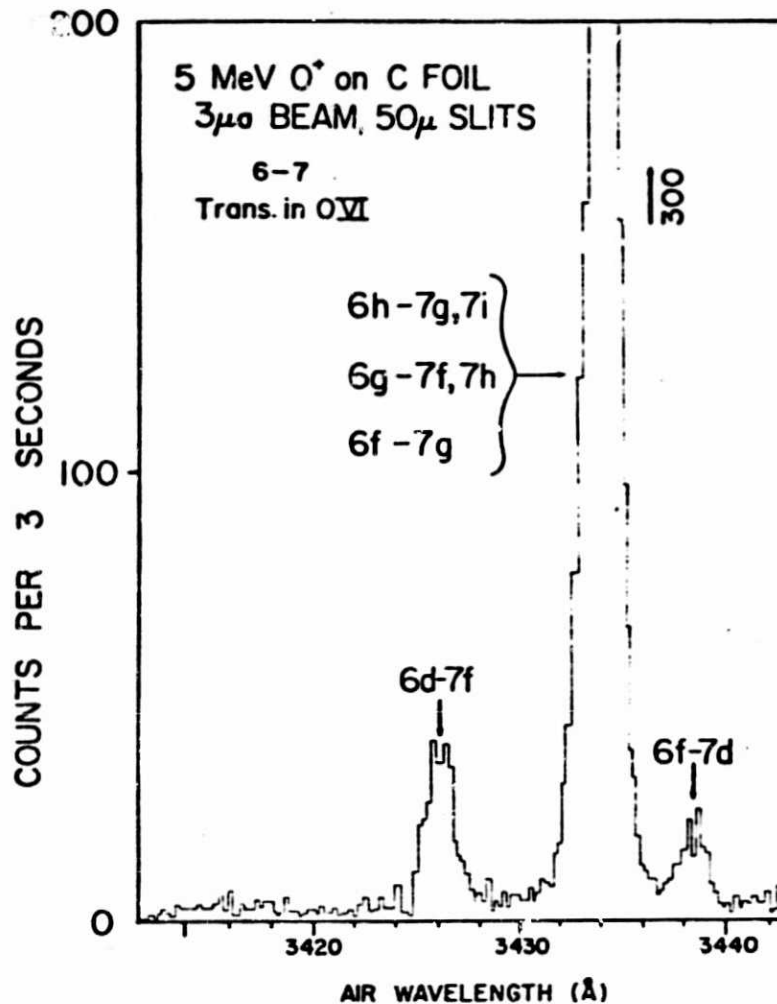


Fig. 6. Beam-foil spectrum of oxygen showing hydrogenic transition in OVI .

measurement. Furthermore, such observations bear on another important subject, namely, the measurement of level lifetimes.

The standard type of experiment often gives results such as are displayed in Fig. 7. These data were presented by Roberts, Andersen, and Spørensen at the 1972 Beam-Foil Conference.⁵ That group obtained excellent lifetime data over a wide range of line intensities for the decays of two different levels in $Ti II$. You will note that the level Z^4G^0 exhibits a strictly exponential decay over a factor of 300 in yield, whereas e^4G obviously does not decay exponentially. The explanation of the latter curve is that the level of interest is populated by cascades as well as by the direct beam-foil interaction.

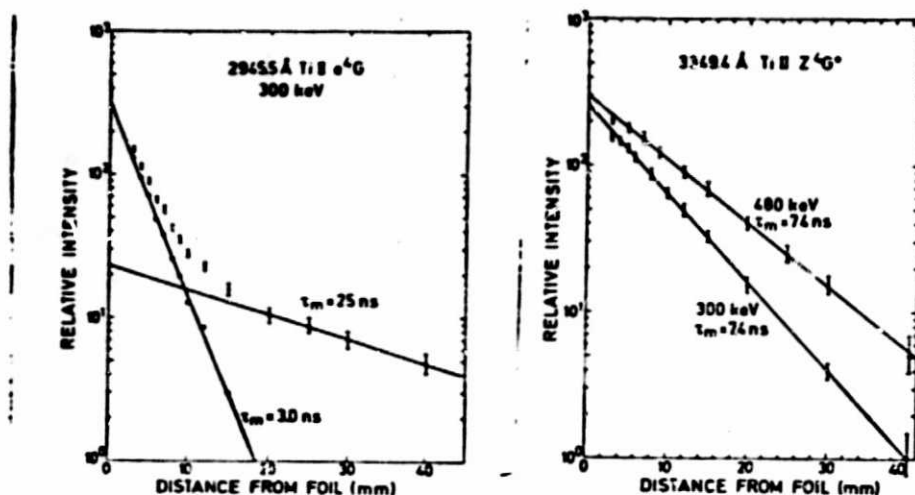


Fig. 7. Beam-foil lifetime data for two different levels in Ti II. After Roberts et al. (Ref. 5).

However, if we look at Fig. 8, we see the quartet energy levels for Ti II and the transitions which were used to produce Fig. 7.

What you notice is that it is the lower level, Z^4G^0 , into which at least one cascade definitely occurs, which displays a strictly exponential decay, whereas the upper level, e^4G , into which no cascading lines have ever been reported, decays in a non-

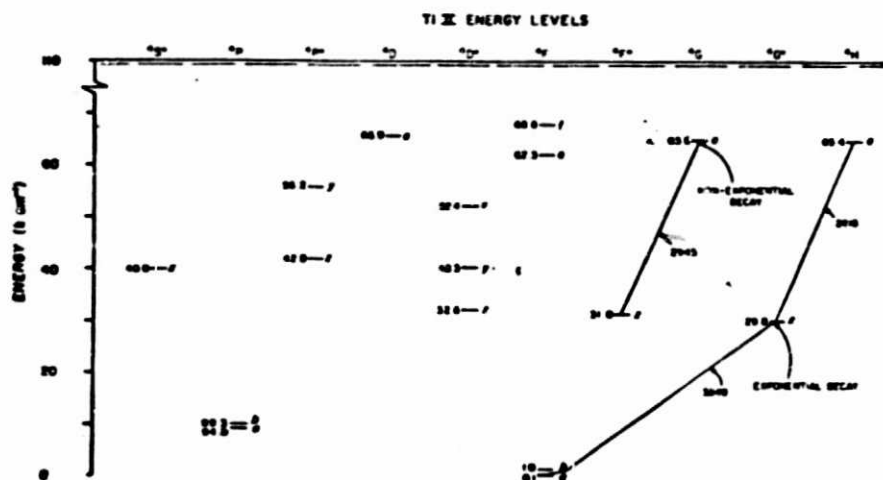


Fig. 8. Energy levels for Ti II.

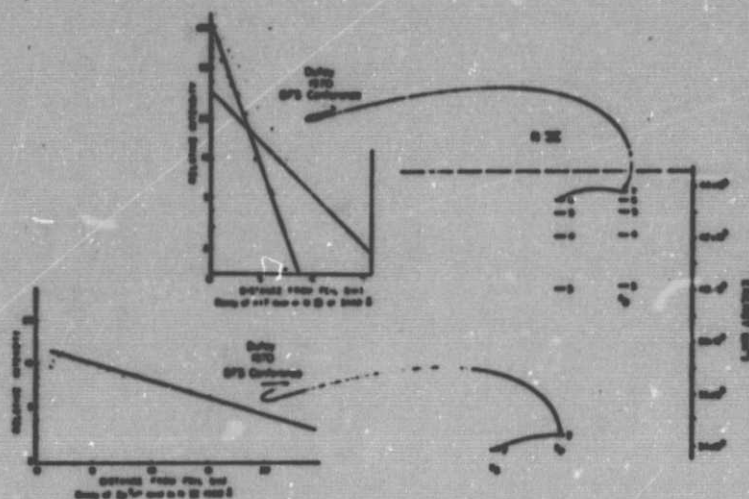


Fig. 9. Beam-foil decay data for two levels in N VI. After Dufay et al., Ref. 6.

exponential manner. Thus the lifetime data are not in accord with the spectroscopic data.

Now it seems to me that the upper level cannot possibly be significantly affected by cascades. For one thing, it is fairly close to the ionization limit; for another, there are no observed terms which could decay into it. Of course, the decay data are definitely non-exponential, and this anomaly, which occurs fairly often in BFS lifetime work, must be explained.

Even more striking examples may be given. Figure 9 shows part of the level system of N VI, and also two decay curves.⁶ Notice that the $2p\ 3P^0$ level shows exponential decay, despite the fact that it is fed by cascade from many higher levels. Yet the decay of $n=7$, which can hardly include any cascade contributions, shows substantial curvature, which is attributed to cascades. In Fig. 10, we see some decay data⁷ for C II and the level systems involved; the same phenomenon occurs. The lower level, into which many levels decay, shows no curvature, whereas the upper level, which is even autoionizing so that cascades into it are most unlikely, is curved. Dufay's data⁸ for N V appear in Fig. 11, along with the relevant level scheme. The level under study is populated by cascades from numerous others, only a few of which are indicated, but the decay curve is exponential over a range of three mean lives. On the other hand, the decay of $5f\ 2F^0$ is curved, a fact which is attributed to cascades.

One finds many similar situations in beam-foil papers.⁹ Levels

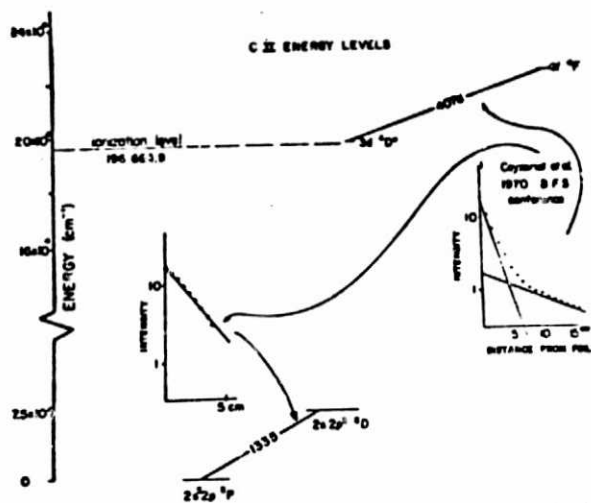


Fig. 10. Beam-foil decay curve for two levels in C II. After Ceyzeriat et al. (Ref. 7).

with low excitation which are known to be populated in cascade exhibit little cascade effect, whereas high-lying levels often show considerable curvature which is ascribed to cascades. Let me propose a solution to this problem. If the level in question is hydrogenic, we have many ℓ -states with widely differing lifetimes. Therefore, the decay curve is a composite of many exponentials, the exact form

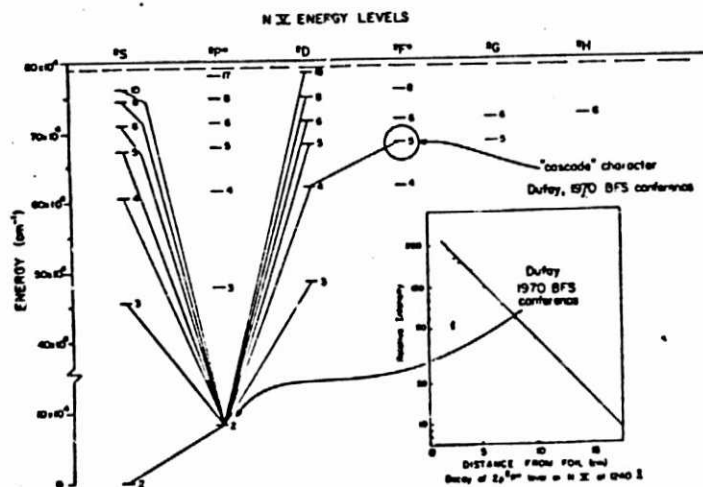


Fig. 11. Beam-foil decay data for a level in N V. After Dufay et al. (Ref. 8).

depending on the number and relative populations of those states, as well as on their specific mean lives. Thus there is a blend, completely unresolvable by means of standard optical spectroscopy, and there is substantial curvature in the decay, but there are no cascades.

Now this suggested solution can be tested experimentally. One uses the quenching technique I described earlier. If the line is quenched, the parent term is hydrogenic, the curvature is explained, and cascades disappear as an element in the analysis. I believe that the curvature seen in the cases of C II, N V, and N VI is due to a blend of many different ℓ -states, and not to cascades. It would be most informative if, for example, the case in Ti II were re-examined for field effects. If they occur, it will be necessary to revise the level identification, and we see that a quenching experiment might contribute to a clarification of the quantum character of a level.

A curious situation occurs in O V. Figure 12 illustrates the decay curve¹⁰ for a line at 2941Å. There is unmistakable curvature to the decay. This line was identified as coming from the $n=6$ to $n=5$ transition. Moreover, the line at 4930 was observed and assigned to the $n=7$ to $n=6$ transition. The curvature of $\lambda 2941$ was accounted for in terms of cascades from $n=7$ and possibly higher levels.

There are several separate points to make here. In the first place, the Bohr-type calculation puts the $n=6$ to 5 transition at 2982Å, rather than at 2941Å. Thus the simple-minded Bohr calculation doesn't help identify $\lambda 2941$. However, Bockasten and Johansson¹¹ have

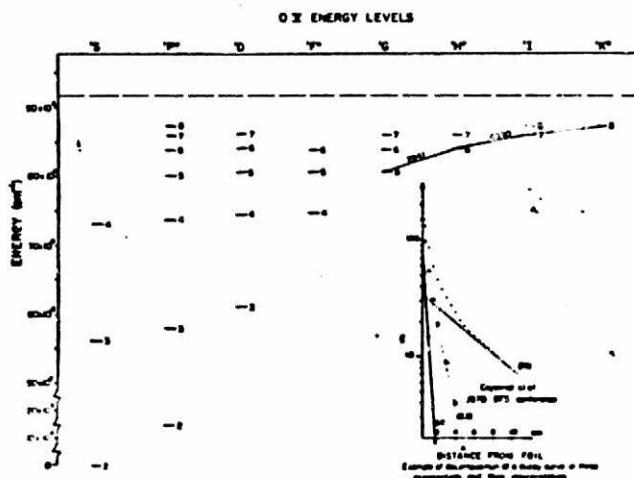


Fig. 12. Beam-foil decay curve for a level in O V. After Ceyzeriat et al. (Ref. 10).

made a careful study of the 0 V spectrum; by taking proper account of configuration interactions, they found that 2941 is indeed the 6 to 5 line.

Now Leavitt also subjected $\lambda 2941$ to the external electric field. The quenching effect was a mere 2%, so small that one cannot state with certainty that there was any quenching at all. How can one account for this negative result? At the same time, one must explain the curvature in the decay of $\lambda 2941$. The answers, I believe, lie in the detailed structure of $n=6$. According to Bockasten and Johansson, the ℓ -levels for $n=5$ and $n=6$ are, respectively, very close in energy, as Table 2 shows. Consequently, the "line" from 6 to 5 includes an unresolved blend of three or four ℓ -state contributions. This gives rise to the curvature in the decay. At the same time, the principal constituents have lifetimes which are not only long, but also about the same. As Table 1 showed, the lifetime of 6f, 6g, and 6i in hydrogen are in the ratio 1 to 1.7 to 2.5, whereas the dramatic difference occurs for 6p; that lifetime, relative to 6f, is 0.17, and is 0.07 relative to 6h. In the case of $n=6$ in 0 V, then, we have 6f, g, and h close enough to be mixed by the field, but with small effect on the lifetime; 6p, on the other hand, is too far away for the small field we tried to have played any significant role in reducing the average mean life.

You will probably have recognized a certain handicap to the field experiments as done to date, namely, because of the open aperture and the utility of having a uniform field, observations had to be made fairly far downstream. Not only did this cost a lot of intensity, but the relative influence of such cascades as might occur becomes large. I believe it would improve matters considerably if the apertures in the field plates were covered with foils, but we have not yet had a chance to try that arrangement. As a matter of fact, certain cases, as $\lambda 2941$ in 0 V, might be better studied at much greater downstream distances; many different things remain to be tried.

In the foregoing remarks, I have emphasized that the decays of low-lying states are often exponential, and that the non-exponential behavior of high-lying states is probably caused by ℓ -degeneracy. We are, then, faced with the possibility that cascades have been emphasized unduly in much of the literature. Yet it is commonly found that level lifetimes, as measured by the beam-foil method, are longer than calculations would have us believe.^{12,13} Wiese, for example, has often urged¹⁴ that experimenters pay particular attention to evaluating the errors in their data, especially when the measured lifetimes are too long. If cascades cannot be invoked to explain the discrepancy between measured and calculated mean lives, what is the correct situation? This is precisely the question which, in my view, must be answered by the experiments of the near future.

Table 2. Term values, in cm^{-1} , for $n=5$ and $n=6$ in 0 V.
(After Bockasten and Johansson (Ref. 11)).

Config-uration	Level Symbol	Term Value	Config-uration	Level Symbol	Term Value
2s5s	5s 3S_1	122,589	2s6p	6p 3P_2	80,285
5p	5p 3P_2	115,782		1P_1	79,041
	3P_1	115,798	6d	6d 3D	77,624
	1P_1	116,191		1D_2	76,570
5d	5d 3D_3	112,208.9	6f	6f 3F_4	75,918.0
	3D_2	112,211.9		3F_3	75,937.0
	3D_1	112,213.9		3F_2	75,953.0
	1D_2	110,304.7		1F_3	77,835.5
5f	5f 3F_4	110,044.2	6g	6g 3G_5	76,368.8
	3F_3	110,046.4		3G_4	78,490.9
	3F_2	110,047.3		3G_2	78,533.6
	1F_3	109,740.2		1G_4	78,271.8
5g	5g 3G_5	110,268.4	6h	6h H	76,283.8
	$^3G_{4,3}$	110,272.1	2p6p	6p' 3P_2	79,938
	1G_4	110,268.4		3D_3	79,798
2p5p	5p' 3P_2	114,399		1P_1	79,938
	3D_3	116,072		1D_2	78,220
	1P_1	116,451			
	1D_2	113,119			
5d	5d' 3P_2	110,754			
	3D_3	111,246			
	1D_2	112,340			
	1F_3	109,158			

The cascade possibility must be studied by attempts to identify the cascade line or lines, despite the severe experimental difficulties that generally impede such work. It might be worth mentioning that, in N II, there are several chains of decays, among them, $3s\ ^3P_2 - 3p\ ^3D_3 - 3d\ ^3F_4 - 4f\ ^3G_5$ with successive wavelengths 5651Å, 5006Å, and 4041Å, and also $3s\ ^5P_3 - 3p\ ^5D_4 - 3d\ ^5F_5$ with successive

wavelengths 5536Å and 5178Å, which could be studied with the same grating.¹⁵ In point of fact, this kind of investigation must be complemented by others which are quite demanding. Thus, we all know that cascades have an influence which depends on the relative populations of the level of interest and the feeding levels. Therefore the effort must be made to determine those level populations. Moreover, levels are not populated in statistical equilibrium so the problem is really one of measuring populations of individual ℓ -states.

One can easily extend the variety of experiments needed to clarify the matter of level lifetimes. Since ℓ -state populations must be found, it becomes attractive to do the kind of work carried out by Pipkin and his collaborators on hydrogen,¹⁶ namely, to send the beam through a cavity so as to generate resonance microwave transitions from one ℓ -level to another. Such studies are now being tried in the Arizona laboratory by Drs. Dan Dietrich, William Wing, and Jack Leavitt. Finally, it is increasingly attractive to think of Zeeman experiments in which the magnetic field is parallel to the particle beam. Such work should contribute important information on the ℓ -state components of some excited term. Naturally, these researches could be further amplified by means of the tilted-foil geometry. Associated with the foregoing experiments are others to clarify the nature of the long-lived states observed in the Brookhaven studies on iron. Lifetimes of several nanoseconds in ions having a net charge of +10 or more are too long by at least a factor of 10 to belong to states with ordinary modes of decay.

I mentioned earlier that the orbits of the levels of large n which are apparently detected in the work on iron imply electron orbits perhaps 8Å in radius. Such orbits surely cannot exist inside the foil; whether they are created at the exit surface or in the space just beyond that surface remains to be determined. Information on this subject should also be extractable from the experiments I have outlined.

If we look at the fundamental aspects of BFS, we find that we are just as far as ever from having a good theoretical model for the excitation mechanism. It is particularly disturbing that we have no way of accounting for the relative intensities of spectral lines. Nearly all beam-foil spectral distributions show a few lines which are strikingly prominent. Some of those lines, but by no means all, are sensitive functions of the particle velocity. Work in the next few years should be directed towards the acquisition of good data on this aspect of the beam-foil source, and the quenching experiments have a definite role in that work. Perhaps the theorists would then be stimulated to look closely at the problem of relative line intensities and the related but not identical problem of finding the systematics of level populations.

To summarize my paper, I suggest that experiments for the near

future should treat the related problems of identifying hydrogenic levels, of accounting for the non-exponential decay of levels, and of determining the ℓ -state populations at each value of n . In order of increasing difficulty, these experiments should involve line-quenching with external electric fields, a thorough wavelength scan with apparatus calibrated to give absolute line intensities, microwave excitation of transitions between ℓ -states, and Zeeman experiments.

I wish to thank Dr. Leavitt for having interrupted his own research in order to provide some of the data I showed today. Mr. Bartley Cardon has been most helpful in making calculations, searching the literature, and discussing the general subject. I am also indebted to Dr. Indrek Martinson who, several years ago, emphasized to me that the hydrogenic levels which the beam-foil source seems to produce so prolifically merit close inspection. They do indeed.

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