CONF-8708114--1

September 1987

BNL--40288

DE88 001556

CHARGE STATE EVOLUTION IN AN ION TRAP IRRADIATED BY VUV SYNCHROTRON RADIATION

M. Meron, B. M. Johnson, K. W. Jones

Brookhaven National Laboratory, Upton, New York 11973 USA

D. A. Church

Texas A&M University, College Station, Texas 77843 USA

Contributed Paper Presented at the
Symposium on Atomic Spectroscopy and Highly-Ionized Atoms
(SASHIA)
Hickory Ridge Conference Centre, Lisle, Illinois
August 16-21, 1987

The submitted manuscript has been authored under contract DE-AC02-76CH00016 with the Office Basic Energy Sciences, US Department of Energy. Accordingly, the US Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so for US Government purposes.

CHARGE STATE EVOLUTION IN AN ION TRAP IRRADIATED BY VUV SYNCHROTRON RADIATION*

M. Meron, B. M. Johnson, K. W. Jones
Brookhaven National Laboratory, Upton, New York 11973 USA

D. A. Church
Texas A&M University, College Station, Texas 77843 USA

The energies of photons obtainable from the VUV ring at the National Synchrotron Light Source (NSLS) are ideally suited for high-efficiency ionization of atomic outer-shell electrons. Given the high fluxes of photons available on a wiggler beam line, multiple photoionization in an ion trap can be easily achieved within times short compared to typical ion storage times in the trap. Measurements of the time evolution of ion populations in such a trap can yield ionic photoionization cross sections and charge-exchange interaction rates for ion-atom or ion-ion collisions. The various processes governing this time evolution are discussed and model calculations illustrating the relative importance of these processes under different conditions are presented.

*Research supported by Fundamental Interactions Branch, Division of Chemical Sciences, Office of Basic Energy Sciences, US Department of Energy under Contract No. DE-AC02-76CH00016 (MM,BMJ,KWJ) and National Science Foundation (DAC).

1. INTRODUCTION

Among the various ionization processes identified and studied in atomic physics, photoionization is the "cleanest" one, capable of producing the desired ionized states while introducing a minimal amount of momentum spread and secondary excitation. This property would make photoionization one of the most useful tools for studying atomic structures, if not for the limitations imposed by low available fluxes of high energy photons. The advent of modern synchrotron light sources, however, has reduced these limitations.

The VUV ring ring at the National Synchrotron Light Source (NSLS) in Brookhaven, for example, routinely delivers photon beams at energies from a few eV to above 1 keV, with sufficient fluxes to serve as a very efficient ionizing agent [1]. Moreover, at low enough target densities the achievable photoionization rates compare favorably with charge recombination rates. This fact has important implications when an ion trap is being used to confine the target ions, as it allows for significant consecutive photoionization within typical storage times. Therefore, a large population of highly charged ions can be accumulated in the trap and either studied in situ or extracted, thus turning the trap into an ion source. This concept has been given the name PHOBIS (PHOton Beam Ion Source) [2-4].

The buildup of a multiply charged ion population in a trap is a complex process, driven by the competition between photoionization and recombination, and modified by the properties of the specific trap that is being used. Still, using some simplifying assumptions, the time development of this process can be mathematically modeled, and the evolution of charge-state distributions can be calculated. Preliminary calculations of this kind were done in the past for high energy (up to 20-30 keV) photon beams from the X-ray ring at the NSLS [2-4]. In these simplified calculations the dominant process was assumed to be inner-shell photoionization followed by multiple autoionisation. This is a major charge buildup mechanism at the X-ray ring energies, but much less so at the VUV ring. Recombination processes, which are the ones determining the ultimate obtainable charge states, were ignored in the previous calculations.

This paper presents calculations for the time dependance of ion production in a generic ion trap irradiated by photons from the VUV ring at the NSLS. Recombination effects are taken into account as fully as possible given the limited amount of existing information about rate coefficients at low energies. Also, for the first time, possible nonlinear effects caused by depletion of the neutral atoms in the trap at high ionization rates are considered. Multiple photoionization is neglected, being of very small importance at the VUV ring energies for all but the lightest elements. In spite of this our calculations indicate that when the main interest is in outer shell photoionization, results comparable to those at the X-ray ring can be obtained.

2. GENERAL CONSIDERATIONS

The case studied here is charge buildup in an ion trap irradiated by synchrotron radiation. Two different sources at the VUV ring (NSLS) are considered: a UV arc (bending magnet) and a UV wiggler, as at beam lines U11 and U13, respectively. The energy distributions of photon flux from both sources [1] are shown in fig. 1. While differing significantly in intensity both sources display a similar sharp drop at energies above 1 keV.

The target used in the calculations is Ne which has been chosen as a typical representative of low Z targets with many outer shell electrons. Neon is, in fact, a likely candidate for an actual measurement, being a monatomic gas without the added complications of molecular bonds.

The time development of the trapped ion population in this case is driven mainly by two processes : 1) consecutive single outer shell photoionization, and 2) charge recombination through single electron capture from the background neutral gas. Multiple photoionization caused by inner shell photoionization followed by an Auger decay can be neglected here, as the photon flux drops sharply at energies above the Ne K-shell binding energy. Multiple electron capture from neutral atoms can also be neglected, being a low probability event comparing to single capture. Finally, charge exchange between the captured ions can be ignored, as the relevant cross sections tend to zero due to the strong Coulomb repulsion between the ions.

There are two additional processes which become significant when the charge buildup process proceeds long enough for ion densities to become comparable to or greater than the original neutral gas density. One of them is a "pump" effect caused by the depletion of the neutral fraction (both through direct photoionization and through recombination). When the neutral density (i.e gas pressure) outside the trap is maintained constant, this depletion causes additional neutral atoms to flow into the trap. These neutrals are also ionized, and the process keeps repeating, thus building ion densities much higher than the original neutral density. As the pumping process is characterized by a finite "pumping speed" it may not be capable to fully make up for the loss of neutrals through ionization. In this case the neutral population will get progressively lower, thus reducing the rate of recombination and pushing the charge distribution toward higher charge states.

The other process, which to some extent counteracts the first one, is spillover of ions from the trap when the charge density approaches the storage limit of the trap. As both the storage limit and the actual spillover process strongly depend on the actual trap used, this process will be ignored here. In other words it is assumed that the storage capacity of the trap is unlimited. This assumption may sound unrealistic, as it yields impossibly high values for the total amount of charge created over long storage times. However, our main interest is in charge distributions where the spillover of charge has two different effects which tend to cancel each other. On the one hand ion spillover reduces the ion to neutral ratio thus enhancing the relative weight of recombination and pushing the charge state distribution downward. On the other hand, in most traps the trapping potential increases with ionic charge and therefore low charge states are the first ones to be ejected, shifting the charge distribution upward. While the relative weight of both effects is hard to assess, it is to be expected that at low enough target densities they will offset each other.

3. CALCULATIONS

As mentioned in the previous section, the time development of the ion population in a trap, in the present case, is driven mainly by single outer-shell photoionization and by charge recombination through single electron capture from the background neutral gas.

The time rate of change in the number of ions at charge state k, N_k, due to photoionization (photo) is given by:

$$\left.\frac{dN_k}{dt}\right|_{photo} = \frac{N_{k-1}}{\tau_{k-1}} - \frac{N_k}{\tau_k} \tag{1}$$

The photoionization time constants, τ_k are defined by [2]:

$$\frac{1}{\tau_k} = \int_{E_k}^{\infty} J(E)\sigma_{k,k+1}(E)dE \tag{2}$$

where E_k is the ionization energy for charge state k, $\sigma_{k,k+1}(E)$ is the photoionization cross section for charge state k at energy E, and J(E) is the photon flux density at energy E, averaged over the trap area.

The time constants τ_k are obviously dependent both on the specific photon source that's being used and on the trap geometry, through J(E). Values of $1/\tau_k$ for a Ne target and for the two photon sources which are being considered here, i.e. a bending magnet and wiggler on the VUV ring, are given in Table 1. The trap dimensions were assumed to be identical to those of a Penning trap which has been used in recent trapping experiments on the NSLS X-ray ring [3,5]. The ionization energies E_k were taken from the tables of Carlson et al. [6]. The photoionization cross sections $\sigma_{q,q+1}$ are derived from the values given by Henke et al. [7] through renormalization to the number of electrons available for ionization. The τ_q values are expected to be accurate to within 20%.

The time rate of change in N_k due to recombination (rec) is given by :

$$\left.\frac{dN_k}{dt}\right|_{rec} = \left(- \frac{N_k}{\theta_k} + \frac{N_{k+1}}{\theta_{k+1}}\right) \frac{N_o}{N} \qquad (3)$$

Here N_o is the number of neutral atoms in the trap and N is the number of atoms that would be in the trap volume at given pressure with the photon beam off. The multiplicative factor N_o/N reflects the decrease in recombination when the neutrals are depleted. The recombination time constants, θ_k are given by :

$$\frac{1}{\theta_k} = \rho \langle \sigma v \rangle \qquad (4)$$

where ρ is the original neutral target density, σ is the capture cross section and v is the collision velocity. The averaging in the rate coefficient, $\langle \sigma v \rangle$, is over the velocity distribution in the trap. Unfortunately just a few values of rate coeeficients have been determined, either experimentally or theoretically, and the quoted accuracy for these few is rarely better than 50%. To simplify the calculations we used the theoretical results of Hasted [8] which predict, for low velocity ions, rate coefficients independent of velocity and linear in ionic charge. Substitution of those results into eq. (4) yields

$$\frac{1}{\theta_k} = 5\cdot10^7\ k\ P(\text{torr})\ \sec^{-1} \tag{5}$$

Apart from pathological cases caused by an accidental match or mismatch of energy levels, the values of θ_k obtained using eq. (5) are in a reasonable agreement with recent measurements [5]. Still, they should not be expected to be accurate to better than factor 2.

The last process that is quantitatively included in our calculations is the replenishment of neutral atoms in the trap through the "pumping" action described above. The flow rate into the trap is given by

$$\left.\frac{dN_o}{dt}\right|_{flow} = \frac{1}{\tau}\left(N - N_o \right) \tag{6}$$

where N_o and N have the same meaning as in eq. (3). The flow time constant $1/\tau$ is given by the ratio between the combined pumping conductivity of the trap apertures, and between the total gas capacity of the trap volume. A simple kinetic theory calculation for a gas at temperature T yields

$$\frac{1}{\tau} = \left(\frac{kT}{2\pi M} \right)^{1/2} \frac{S}{V} \tag{7}$$

where k is the Boltzman constant, M is the molecular mass, S is the total pumping area of the trap and V is the trap volume. For the present case $1/\tau$ was set at $10^4\ \sec^{-1}$, based on the actual geometry of the Penning trap used [5]. It should be mentioned that the calculational approaches used in the past of either assuming no replenishment of neutrals, or assuming a constant neutral density, correspond to choosing $1/\tau = 0$ or $1/\tau = \infty$, respectively.

The master equation for the time development of ion population in a trap irradiated by synchrotron radiation can now be written by combining the processes described by eq. (1), (3) and (6) together. After dividing all the N_k-s by the original number of neutrals, N, the following set of equations is obtained

$$\frac{dy_o}{dt} = \frac{1-y_o}{\tau} - \frac{y_o}{\tau_o} + \frac{y_o y_1}{\theta_1} - \sum_1^n \frac{y_o y_k}{\theta_k} \tag{8.0}$$

$$\frac{dy_1}{dt} = \frac{y_o}{\tau_o} - \frac{y_1}{\tau_1} - \frac{y_o y_1}{\theta_1} + \frac{y_o y_2}{\theta_2} + \sum_1^n \frac{y_o y_k}{\theta_k} \tag{8.1}$$

$$\vdots$$

$$\frac{dy_k}{dt} = \frac{y_{k-1}}{\tau_{k-1}} - \frac{y_k}{\tau_k} - \frac{y_o y_k}{\theta_k} + \frac{y_o y_{k+1}}{\theta_{k+1}} \tag{8.k}$$

$$\vdots$$

$$\frac{dy_n}{dt} = \frac{y_{n-1}}{\tau_{n-1}} - \frac{y_o y_n}{\theta_n} \tag{8.n}$$

where n is the highest charge state possible in the system. The y_k-s, defined by $y_k = N_k/N$ are proportional to the charge state fractions q_k which can be recovered at any stage of the calculation by normalizing the y_k-s to unity. The additional sum terms in eq. (8.0) and (8.1) reflect the fact that each recombination event, while lowering the charge state of the ion involved, also changes one neutral atom into a singly charged ion.

It should be noted that the recombination terms in the equation system above are nonlinear. This leads to a rather complex behavior of the solutions in regions of fast changing y_0, i.e. in cases where the ionization rate is sufficient to cause a significant depletion of the neutral density in the trap.

4. DISCUSSION

The master equation system, eq. (8.0-n), can be solved easily using standard numerical integration techniques. Before discussing the results for real cases, however, it is instructive to examine a set of partial solutions which are displayed in fig. 2. The charge state distributions displayed in fig. 2a-c were all calculated for the case of an ion trap filled with Ne at a pressure of 10^{-8} Torr and irradiated by photons from an arc (bending magnet) source at the UV ring, assuming an electron current of 500 ma in the ring. However, some terms in the master equation were omitted in the first two solutions (fig. 2a and 2b) in order to illustrate the role played by the various effects considered.

The charge state distribution shown in fig. 2a was obtained assuming no recombination and no neutral replenishment ($1/\tau = 0$). As could be expected it displayes the "traditional" picture of consecutive charge fractions rising, reaching a maximum and declining to make room for the next one, untill the maximal possible charge state (8 in this case) is obtained.

By bringing back recombination and assuming no depletion of neutrals ($1/\tau = \infty$), the time evolution displayed in fig. 2b was obtained. Here the time evolution starts the same as in fig. 2a, but after a time of the order of one second a balance between photoionization and recombination is obtained and the charge state distribution reaches equilibrium.

Finally, fig. 2c shows the complete solution including recombination and a realistic neutral flow rate ($1/\tau = 10^4$ sec^{-1}). Over the first 20 seconds of storage time the picture is identical to fig. 2b, i.e. an equilibrium between photoionization and recombination is obtained. However, during all this time charge continues to accumulate in the trap and the demand for neutral atoms for recombination keeps rising, until at some point the neutral flow can no longer cover the losses. At this stage the equilibrium breaks down and the charge distribution starts evolving toward higher and higher charge states in a fashion similar to that shown in fig. 2a.

These different modes of time evolution are displayed again in fig. 2d which shows the average charge state for the three cases discussed above, as a function of storage time. As can be seen, there is practically no difference between the three curves untill $t \approx 2$ sec. is reached. At this point the curves corresponding to 2b and 2c level off while the one corresponding to 2a keeps climbing. Around $t = 30$ sec. the curve corresponding to 2c, which seemed already to reach equilibrium, suddenly changes direction and starts climbing very much like curve 2a.

Fig. 3a-d display the calculated time evolution of charge state fractions in an ion trap filled with Ne in four different cases corresponding to combinations of two pressures (10^{-8} torr and 10^{-10} torr) and two photon sources (an arc source and a wiggler at the UV ring). In all four cases an electron current of 500 ma in the ring is assumed. Obviously the photoionization rates are higher in the W (wiggler) than in the A (arc) cases while recombination is stronger in the 8 (10^{-8} torr) than in the 10 (10^{-10} torr) cases. The flow rate coefficient $1/\tau$ remains the same as before, being independent both of pressure and of photon flux. All four sets of results display essentially the same behavior as seen previously in fig. 2c, which is duplicated here as 3a. An initial period of fast evolution toward higher charge states is followed by a temporary quasi-equilibrium which lasts untill the neutral population is depleted and then breaks down, and the climb of the distribution upward is resumed. What changes from one case to another, however, is the position of the quasi-equilibrium, both in terms of storage time and of the distribution of charge states when it occurs.

In fig. 4 the time evolution of the average charge state is displayed for all the cases shown in fig. 3. A noteworthy feature here is the initial climb rate of the W curves which is, as could be expected, much faster than that of the A curves. Given the fact that practical storage times in ion traps are of the order of seconds, fig. 4 clearly indicates that high ionization experiments should be performed on wiggler beam lines. Another noteworthy feature (related to the previous discussion) is that the A and W curves merge both at short storage times, when recombination is not yet important, and at very long times when it ceases to be effective due to the depletion of neutrals.

Fig. 5 shows the evolution of total charge density in the trap. While fig. 4 creates the impression that, for short storage times, the results obtained are pressure independent, this is only true for the average charge state, and not for the total amount of ions created. As was discussed before, in a real case the charge density will not climb indefinitely as it does in fig. 5, but will level off once it reaches the charge storage limit of the trap (typically around 10^7-10^8 e/cm^3). Naively it might be assumed that once this limit has been reached an equilibrium charge state distribution would be maintained. This is not true, however, as the charge spillover rate will be different for different charge states and not related at all to the rate at which they are created. Therefore, the evolution will continue, but at a rate which will be trap dependent and generally different from the one predicted by our model.

5. CONCLUSIONS

A previous model of the time dependence of ion production in an ion trap irradiated by synchrotron radiation has been refined by including the effects of charge recombination, and of finite flow of neutral atoms into the trap. The improved model has been applied to target pressure and photon flux combinations corresponding to actual operational parameters of the VUV ring at the NSLS in Brookhaven. The results of the calculations illustrate the dependence of the ion production process on both the interplay between ionization and recombination and the flow of neutrals into the trap.

The results also clearly indicate that large quantities of trapped highly charged ions can be produced on existing beam lines at the NSLS. This ability makes possible a host of new experiments, and therefore should be of great interest to the atomic physics community.

REFERENCES

[1]. NSLS Users Manual: Guide to the VUV and X-Ray Beam Lines, Eds. N. F. Gmur and S. M. White-DePace, Brookhaven National Laboratory informal report (1986).

[2]. K.W. Jones, B.M. Johnson, and M. Meron, Phys. Lett. 97A (1983) 377.

[3]. D.A. Church, K.W. Jones, B.M. Johnson, M. Meron, and I.A. Sellin, J. Phys. B: At. Mol. Phys. 17 (1984) L401.

[4]. B.M. Johnson, K.W. Jones, and M. Meron, Nucl. Instr. and Meth. B10/11 (1985) 313.

[5]. D.A Church, S.D. Kravis, I.A. Sellin, C.S. O, J.C. Levin, R.T. Short, M. Meron, B.M. Johnson and K.W. Jones, (accepted) Phys. Rev. A, Rapid Communications.

[6]. T.A. Carlson, C.W. Nestor, Jr., N. Wasserman and J.D. McDowell, At. Data, 2 (1970) 63.

[7]. B.L. Henke et al., At. Data and Nuc. Data Tables, 27 (1982) 3.

[8]. J.B. Hasted, Physics of Atomic Collisions, p.478 (Butterworths, London 1964).

TABLE 1. Ne photoionization time constants $1/\tau_k$ for arc (A) and wiggler (W) photon sources on the VUV ring at the NSLS in Brookhaven, assuming 500 ma of electrons in the ring.

k	A: arc $1/\tau_k$ (sec^{-1})	W: wiggler $1/\tau_k$ (sec^{-1})
0	1.191	61.8
1	0.517	25.6
2	0.215	10.2
3	0.098	4.41
4	0.047	1.98
5	0.023	0.889
6	0.009	0.235
7	0.005	0.078
8[a]	0.000	0.000

a) As mentioned in the text, K-shell photoionization has been ignored.

FIGURE CAPTIONS

Fig. 1. Photon flux as function of energy from an arc (A) and wiggler (W) sources at the VUV ring, NSLS [1].

Fig. 2. Charge evolution as a function of storage time, calculated for a Ne filled trap at 10^{-8} torr, irradiated by an arc source (curve A in fig. 1). The results are for (a) no recombination and no neutral flow, (b) recombination and unlimited neutral flow and (c) recombination and realistic neutral flow. The curves in (d) show the average charge states corresponding to the cases considered in a (dash), b (chain-dot) and c (solid).

Fig. 3. Calculated charge state distributions as a function of time in a Ne filled ion trap for the following conditions:

a) 10^{-8} torr, arc source (Ne8A).

b) 10^{-8} torr, wiggler source (Ne8W).

c) 10^{-10} torr, arc source (Ne10A).

d) 10^{-10} torr, wiggler source (Ne10W).

Fig. 4. Average charge state for the four cases shown in fig. 3. as function of storage time.

Fig. 5. Total charge stored in the trap for the four cases shown in fig. 3.

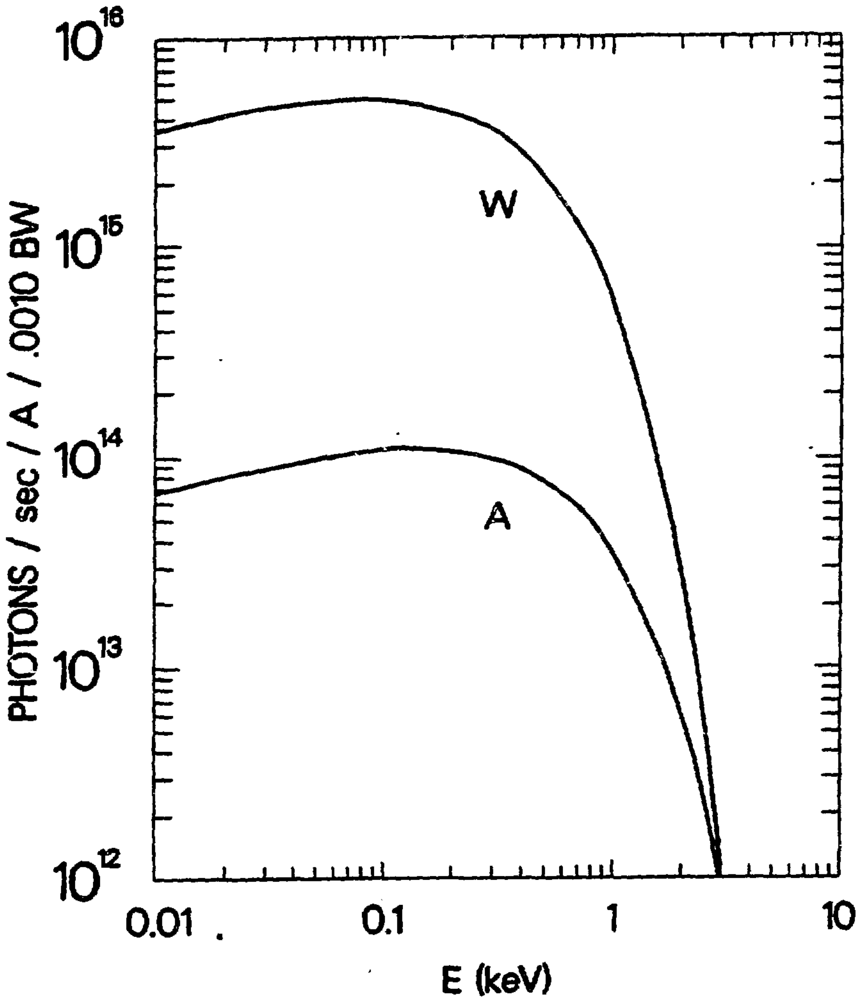

PHOTONS / sec / A / .0010 BW
10^{16}
10^{15}
10^{14}
10^{13}
10^{12}
W
A
0.01
0.1
1
10
E (keV)

Figure 1

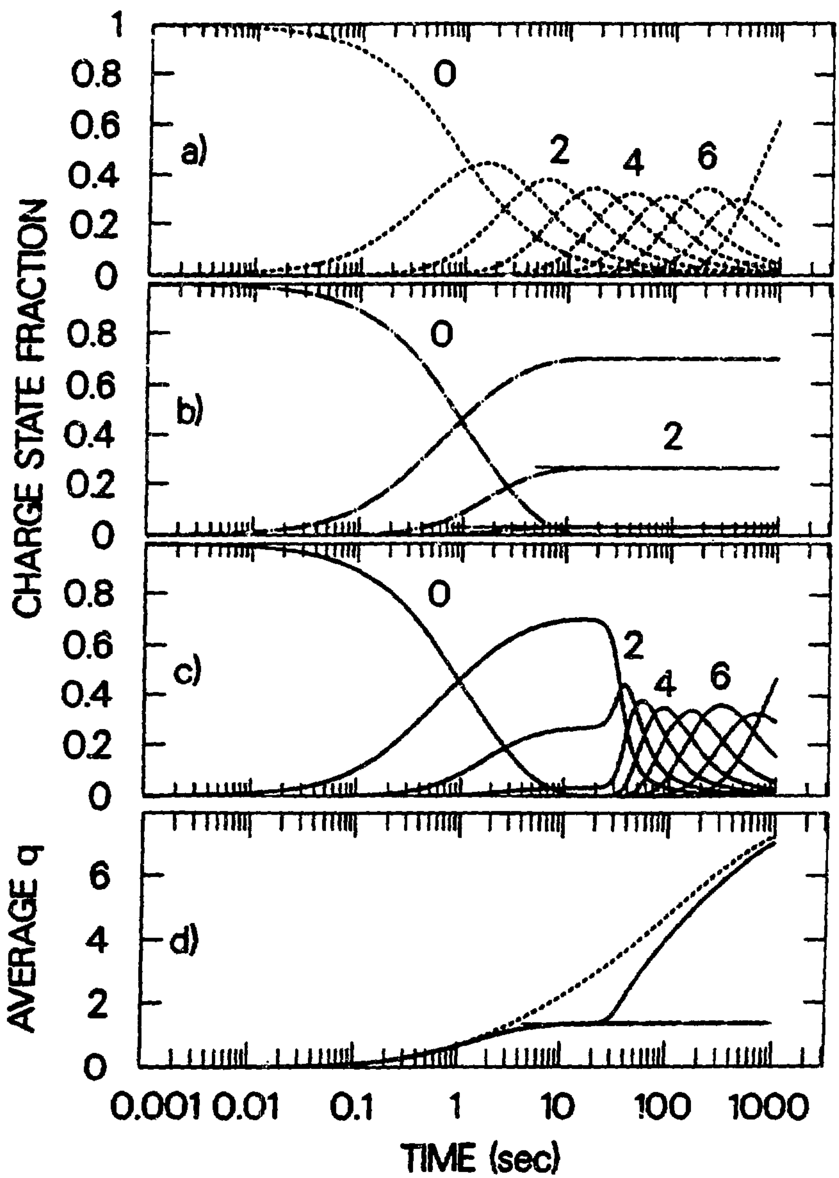
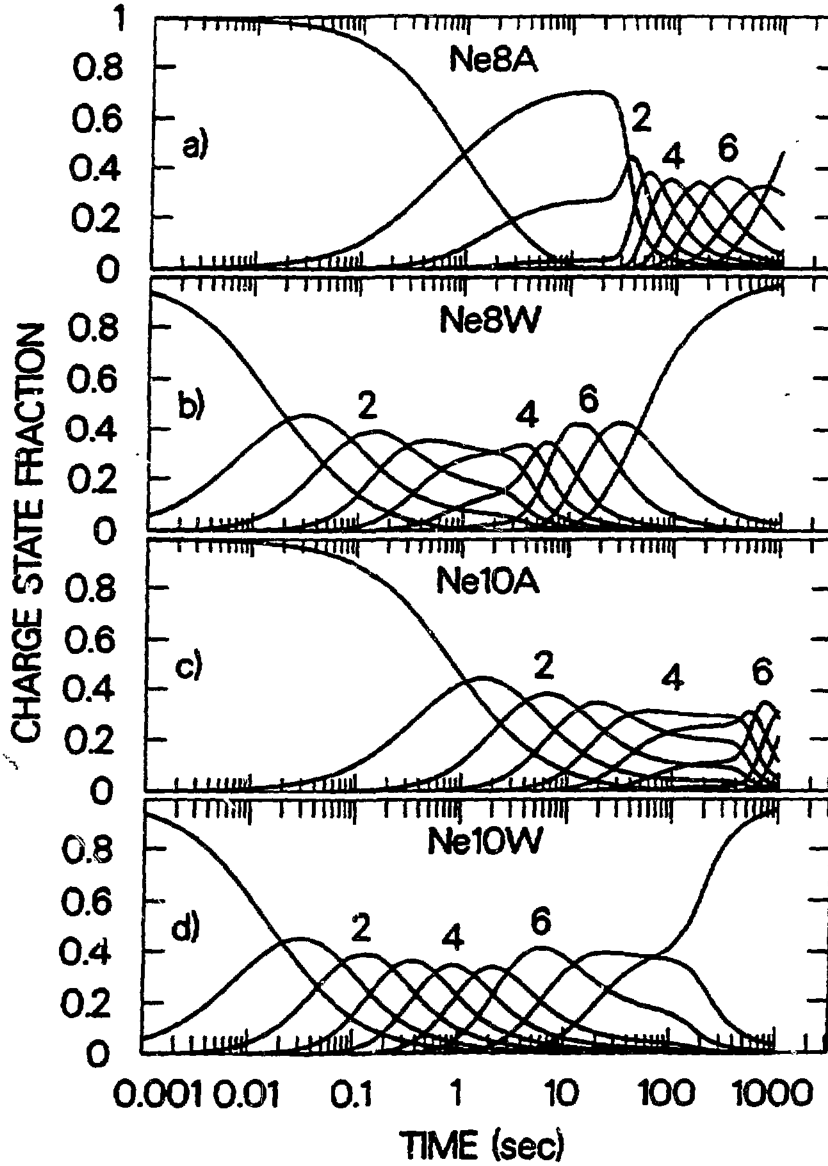
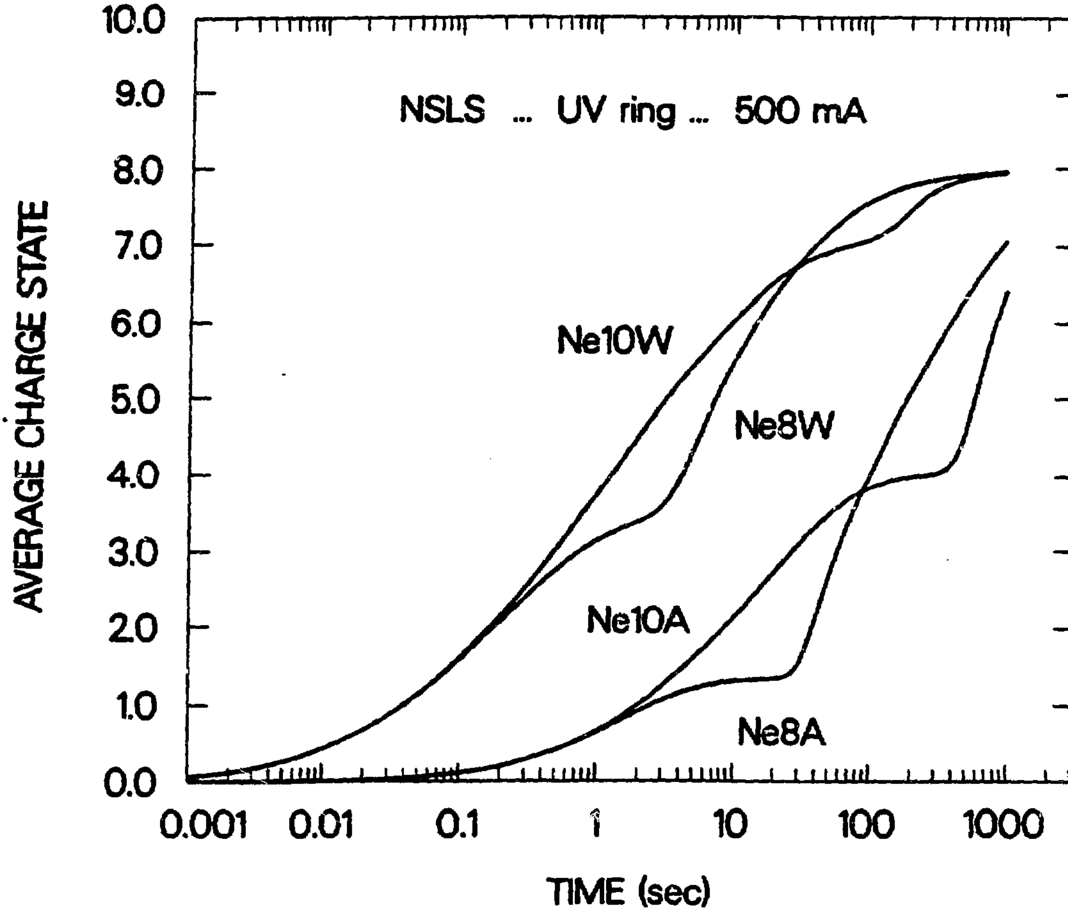

CHARGE STATE FRACTION
AVERAGE q
TIME (sec)
a)
b)
c)
d)
0
2
4
6
0.001 0.01 0.1 1 10 100 1000
0.8
0.6
0.4
0.2

Figure 2



Ne8A
a)
2
4
6
Ne8W
b)
2
4
6
Ne10A
c)
2
4
6
Ne10W
d)
2
4
6
CHARGE STATE FRACTION
1
0.8
0.6
0.4
0.2
0
0.001 0.01 0.1 1 10 100 1000
TIME (sec)

Figure 3



NSLS ... UV ring ... 500 mA
Ne10W
Ne8W
Ne10A
Ne8A
AVERAGE CHARGE STATE
TIME (sec)
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0.0
0.001
0.01
0.1
1
10
100
1000

Figure 4

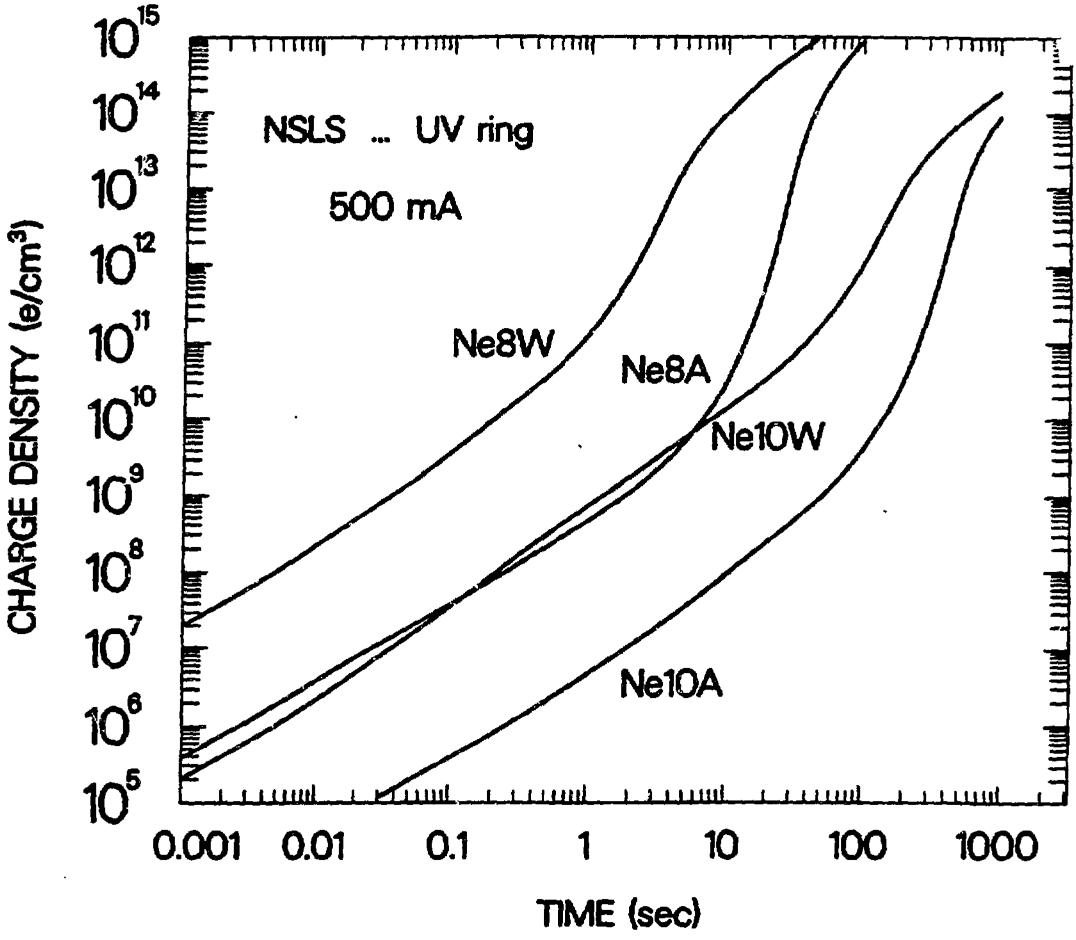

NSLS ... UV ring
500 mA
Ne8W
Ne8A
Ne10W
Ne10A
CHARGE DENSITY (e/cm³)
10^15
10^14
10^13
10^12
10^11
10^10
10^9
10^8
10^7
10^6
10^5
0.001
0.01
0.1
1
10
100
1000
TIME (sec)

Figure 5