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DIFFUSION OF Pb_{214} IN NUCLEAR EMULSION

by

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INTRODUCTION

In this paper a preliminary study of Pb_{214} diffusion in nuclear emulsion is made.

Many α-particle stars which originate from Rn_{222} disintegration in nuclear emulsion appear in the form of two branch stars (α-particles from Rn_{222} and Po_{218}) plus an isolated track (α-particle from Po_{214})[1]. This should result from diffusion of one or both of Pb_{214} and Bi_{214} nuclides because the Po_{214} half-life (15×10^{-4} sec) is too short to allow for its diffusion.

In this paper we analyse these separated stars. Our conclusions can be summarized as follows:

1) The observed separations are due essentially to diffusion of Pb_{214} although the possibility of a small diffusion of Bi_{214} is not excluded.

2) About 50% of Pb_{214} atoms that originate from the Po_{218} in nuclear emulsion do not diffuse in our conditions of work.

3) The order of magnitude of the Pb_{214} diffusion coefficient at 0º C in our conditons of work is 10^{-12} C.G.S.

4) The Pb_{214} diffusion coefficient increases with increase of temperature as should be expected.

5) The diffusion coefficient seems to decrease with the age of the emulsion used.

I. NATURE OF THE MIGRATING ELEMENT

A G-5 nuclear emulsion was placed in the upper part of an aluminum tube 14 cm high and 3,5 cm in diameter. In the lower part of the tube there was about 600 mg of a pulverized Uranium mineral (250 mg of radium per ton)[2] upon which a lead plate 1 cm thick was placed. This plate, having thin perforations, protected the emulsion from the action of the γ-rays but allowed the radon to go easily to the upper part of the tube. The emulsion face of the nuclear plate was protected by a stainless steel plate in order to lessen the background of α-particles originating from disintegrations in air. The radon could penetrate the emulsion through a thin air layer between the emulsion and the steel plate. Finally the tube was closed with a rubber plug and protected with paraffin. The same procedure was adopted for the exposure of C-2 plates, to be referred to in Part II.

The exposure of the G-5 plate had a duration of three days at a temperature around 12° C. After development the 3-branch Radon stars were observed as well as the separated ones[1]. In order to find out which of the elements Pb_{214} and Bi_{214} diffuses, an analysis of the electrons associated with the 3-branch, 2-branch stars and the isolated α near the 2-branch stars was made. The results are given in Table I for 150 3-branch stars and 150 separated ones.

TABLE I

No. of betas	3-branches	2-branches	isolated
0	45	143	47
1	75	7	71
2	29	0	32
3	1	0	0

About 5% of the observed cases were considered doubtful since, due to occasional spots or background betas corssing the origin of the α tracks it was difficult to establish where the β-rays originated; these were not included in Table I. It should be noticed that in our conditions of work the emulsion did not register minimum ionization electrons - therefore only the low energy part of the spectra of the beta emitters (Pb_{214} and Bi_{214}) and the low energy conversion electrons were observed.

Table I shows that the distribution of electrons originating from the isolated α-tracks is statistically identical to the distribution of electrons in the 3-branch stars, the 2-branch ones having practically no electrons associated. Thus we should conclude that almost all electron emitters are associated with the isolated track.

Therefore, if we notice that:

a) The α-disintegrations of Rn_{222}, Po_{218} and Po_{214} are not followed by γ-emission which could produce conversion electrons[3];

b) The electron-emitter Pb_{214} should produce more observable electrons than Bi_{214} in our conditions of work[4,5,6]; the results of Table I indicate that Pb_{214} is the diffusing element, Bi_{214} having a much smaller diffusion coefficient (their half-lives are of the same order of magnitude). Indeed, if Bi_{214} had a significant diffusion the electrons from Pb_{214} would be lost from the isolated α due to Po_{214} and the distribution of the electrons associated with this track would be different from that corresponding to the 3-branch stars.

Finally we should mention that the electrons associated with the 2-branch stars can be explained as due to the small diffusion of Po_{218} already mentioned by Piccioto[5]. Indeed we have found that 3% of the separated stars (both for the G-5 and C-2 emulsions) have the longer track due to Po_{214} in the 2-branch star[7] Only three of those cases could not be explained in this way as they had an electron associated with the 2-branch star and another one with the isolated track.

II. STUDY OF THE DIFFUSION OF Pb_{214}

1. Experiment.

An Ilford C-2 plate 1"x3" and 100μ thick was divided in two parts, each of them was placed in a tube in the same conditions as described in Part I. The emulsion was four months old and had been stocked at 12° C, 100% humidity.

One of these tubes was placed in a Dewar bottle with a water and ice mixture at 0° C; the other was placed in a bath, controlled by a thermostat at 30° C. After 3 days exposure in these conditions the plates were developed. 45 days after, this experiment was repeated with another C-2 plate of the same box. In this case, however, the two parts of the plate were weighted, both before and after the exposure, in order to determine the variation in water content. This water content did not change for the plate kept at 0° C. The plate kept at 30° C did lose, however, an amount of water corresponding to 0,25 mg per square centimeter of emulsion surface.

2. Observations.

a) As a preliminary study all four plates referred to in Section I were scanned in order to find the proportion of stars visibly affected by diffusion (2-branch stars + isolated α) to the total number of stars. The visibly separated stars correspond to those cases in which the distance of the 2-branch star vertex from the nearest extremity of the isolated track is larger than $0,5\mu$. For each plate a total number of 500 stars were observed using 100 x 12 magnification. The following results were obtained:

First exposure:

Plate A (0° C) - 25%
Plate B (30° C) - 47% (1)

Second exposure:

Plate a (0° C) - 27%
Plate b (30° C) - 46% (2)

It was also observed that the average value of the separation distances should correspond to a few micra.

These results give already an indication that, in this case also, a significant proportion of Pb_{214} atoms are retained in the point where they originate, not being able to diffuse, as has been previously found in Rn[8]. The decomposition of the 3-branch stars into those for which Pb_{214} does not diffuse and those for which there is a small diffusion will result from the analysis given in Section 4.

b) For each plate 200 visibly separated stars were selected in order to determine the distribution of separation distances.

As the determination of the real distance between the vertex of the two-branch star and the nearest extremity of the α-track (separation distance) involves measurement of depth and of the shrinkage factor we decided to express our results in terms of the horizontal projection of the separation distance. Whenever the vertex of the star was at a distance smaller than 5μ (in the developed emulsion) from the free surface or from the glass, the event was rejected. This is satisfactory since the maximum observed projection is 8.5 micra. In Table II are shown the results, normalized to a total number of 100 cases for each plate. The distances were measured with 100 x 12 magnification for which 1 division of the scale corresponded to 0,5μ.

The cases with projections smaller than 1 division correspond to visibly separated stars with real distances larger than 0,5μ, where the separation can be decided by differences in depth.

3. Diffusion coefficient of Pb_{214} in nuclear emulsion.

We wish to determine, from the results of Table II, the values of the diffusion coefficient of Pb_{214}. In view of the experimental conditions;

a) Thickness of the emulsion much larger than the average diffusion distance;

b) Exclusion of stars with vertex near the emulsion surfaces;

c) Exposure time much larger than the half-life of Pb_{214} (as well as of Po_{218}, Bi_{214} and Po_{214});

we may use the following idealization of our problem;

A large number, N_o, of atoms of a radioactive element (mean life τ), initially concentrated at some point, diffuses in an indefinite medium (diffusion constant D) and stops at the disintegration point (the daughters do not diffuse). What distribution is to be expected

TABLE II

1 Scale Div. = 0,5

Projection in scale divisions	Plate A	Plate B	Plate a	Plate b
	Number of Cases			
0 - 1	7	6	15	2
1 - 2	40	17,5	56,5	24
2 - 3	26,5	13,5	19,5	25
3 - 4	14,5	11,5	6,5	15
4 - 5	5,5	10,5	2	11,5
5 - 6	2	8,5	0	6,5
6 - 7	2,5	2,5	0,5	4
7 - 8	1,5	5,5	0	5
8 - 9	0	4	0	4
9 - 10	0	2,5	0	0,5
10 - 11	0	2,5	0	0,5
11 - 12	0,5	4	0	0
12 - 13	0	2,5	0	1
13 - 14	0	2	0	0,5
14 - 15	0	0,5	0	0,5
15 - 16	0	1	0	0
16 - 17	0	0,5	0	0
17	0	0	0	0

for the projected distances between the original position of the diffusing atoms and the daughters, after a time much greater than the lifetime?

Tiomno has shown[10] that the number of such atoms with projected distances (on a plane) between r and r + dr is given by

$$dN = N_o \lambda^2 r K_o (\lambda r) \, dr \qquad (3)$$

where

$$\lambda^2 = (D \tau)^{-1} \qquad (4)$$

and $K_o(x)$ is the K-Bessel function of order zero[11]. He has also shown that the number of daughter atoms with projected distances larger than a value r_o is given by:

$$N(r_o) = N_o \lambda r_o K_1(\lambda r_o) \qquad (5)$$

where $K_1(x)$ is the K-Bessel function of first order[9].

Thus we write (3) as

$$dN/dr = N(r_o) \frac{\lambda r K_o(\lambda r)}{\lambda r_o K_1(\lambda r_o)} \qquad (6)$$

Equation (6) has been used to determine the values of λ for Pb_{214} in the several plates using the results of Table II and taking $r_o = 1$ scale division.

The advantage of this comes from the fact that we do not need to know the total number of diffusing atoms.

In Fig. 1 the experimental points for dN/dr are plotted with the probable errors (statistical), indicated, both for plates A and B, for $N(r_o) = 100$. In Fig. 2 they are plotted for plates <u>a</u> and <u>b</u>. By comparison with a set of theoretical curves (6) for several values of λ we have found the best fits of the experimental points and the corresponding values. The continuous curves and the corresponding values indicated in Fig. 1 and Fig. 2 correspond to these best fits. From these values and the mean life of Pb_{214}

$$\tau = 38.7 \text{ min}$$

we obtain the values of the diffusion coefficient of Pb as given in Table III.[12] (Equation 4).

TABLE III

Plate	Temp.	1 scale div.	D x 10^{12} (C. G. S.)
A	0° C	1.25 ± 0.09	1.7 ± 0.25
B	30° C	3.33 ± 0.25	11.7 ± 1.7
a	0° C	0.75 ± 0.06	0.6 ± 0.1
b	30° C	2.0 ± 0.15	4.3 ± 0.6

We see that D increases with the temperature as should be expected. It also depends on the age of the emulsion, only by a proportionality constant

$$\frac{D_A}{D_a} = \frac{D_B}{D_b} \cong 2.7 \pm$$

The values of the diffusion coefficient for Pb_{214} (Table II) should be compared with the value

$$D \cong 10^{-7} \text{ c.g.s.}$$

found by Eichholz and Flack[13] for Thoron. As should be expected Tn, being an inert gas, has a much larger diffusion coefficient.

4. Proportion of non-diffusing Pb_{214} atoms.

From equation (5), knowing the number $N(r_o)$ of atoms which diffused more than r_o (in projection) and the value of λ we obtain the total number N_o of diffusing Pb atoms (r_o = 1 scale division). From results (1), (2) we obtain the numbers N_3 of visually non-separated 3-prong stars (retained Pb atoms + non distinguishable separated stars) corresponding to $N(r_o) = N(1)$. Indeed results (1) and (2) correspond to

$$\frac{N(1) + N_{o-1}}{N(1) + N_{o-1} + N_3}$$

where N_{0-1} are the observed separated stars in the range 0-1. Adding N_{0-1} to N_3 and subtracting $N_o - N(1)$ we find the number of retained Pb atoms (which were not able to diffuse):

$$N_{Ret} = N_3 + N_{0-1} + N(1) - N_o$$

The results are given in Table IV where the proportions k of retained Pb atoms to the total number of cases are also given for the case of 100 visually separated stars.

$$k = \frac{N_{Ret}}{N_o + N_{Ret}}$$

TABLE IV

Plate	N_{0-1}	$N(1) =$ $= N(r_o)$ $r_o = 1$ p.d.	N_o	$\frac{N(1)+N_{0-1}}{N(1)+N_{0-1}+N}$	N_3	N_{Ret}	k
A	7	93	135	0,25	300	275	0,66 ± 0.022
B	6	94	102	0,47	113	111	0,52 ± 0.022
a	15	85	181	0,27	270	189	0,52 ± 0.04
b	2	98	118	0,46	117	99	0.46 ± 0.026

We see that the proportion of retained Pb_{214} atoms is of the order of 50% in our conditions of work

The last column of Table IV gives the fractions k of retained Pb_{214} atoms with probable errors. These results indicate that k increases when the temperature decreases and seem to indicate that k decreases for older plates.

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7. We have also observed occasionally radon stars with the three branches separated.

8. In the case of the diffusion of Rn we have shown in reference (1) that only 10% of the Rn formed in the emulsion did not diffuse thereby originating 4-branch stars. We have called attention to the fact that methods of determination of the Rn content of the emulsion by counting the 4-branch stars assuming that all Rn was retained would lead to large errors. In order to overcome this difficulty Debeauvais, Picciotto and Wilgain[9] have developed a method based on very low temperature exposures which eliminates the Rn diffusion.

   We cannot, however, agree with the statement made by E. Picciotto in his footnote to page 264 of reference (9) that corrections which take into account the loss of Rn were well known as early as 1952. Even now such corrections would not be reliable as the proportion of retained Rn changes very much with the conditions of the emulsion proportions from 5% to 73% having been quoted in the literature[7]. (Note by Elisa Frota-Pessôa).

9. M. Debeauvais, E. Picciotto and S. Wilgain, Il Nuovo Cimento, 5, 260, 1957.

10. J. Tiomno (private communication).

11. G. N. Watson, A Treatise on Bessel Functions, pg. 172, Cambridge University Press, (1948).

12. The relative error in $1/\lambda$ was taken equal to $\frac{1}{\sqrt{N}}$ where N is the total number of observed stars with separation distances r larger than 1 scale division.

13. G. G. Eichholz and F. C. Flack, J. Phys. Chem., 19, 363, (1951).

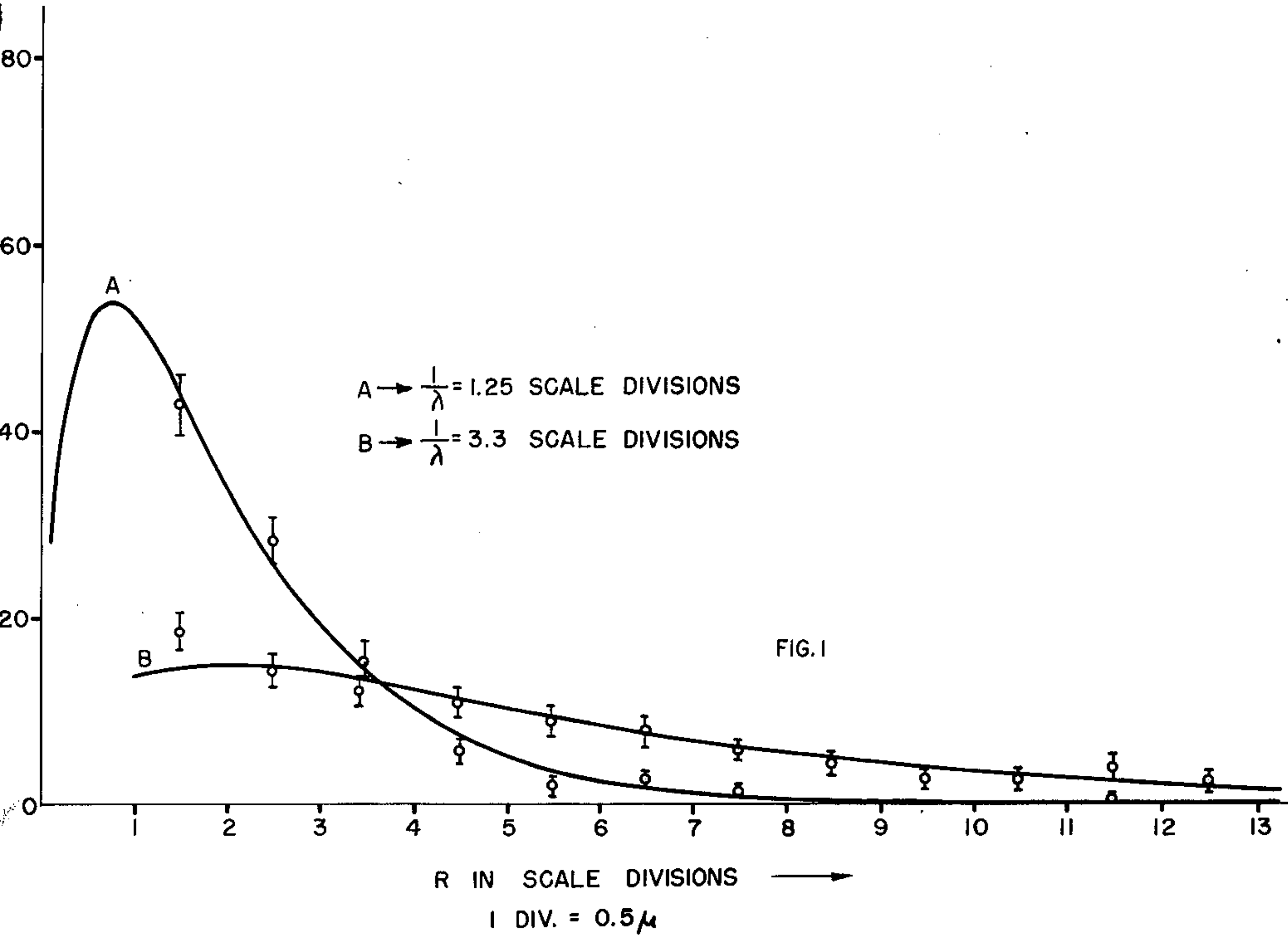
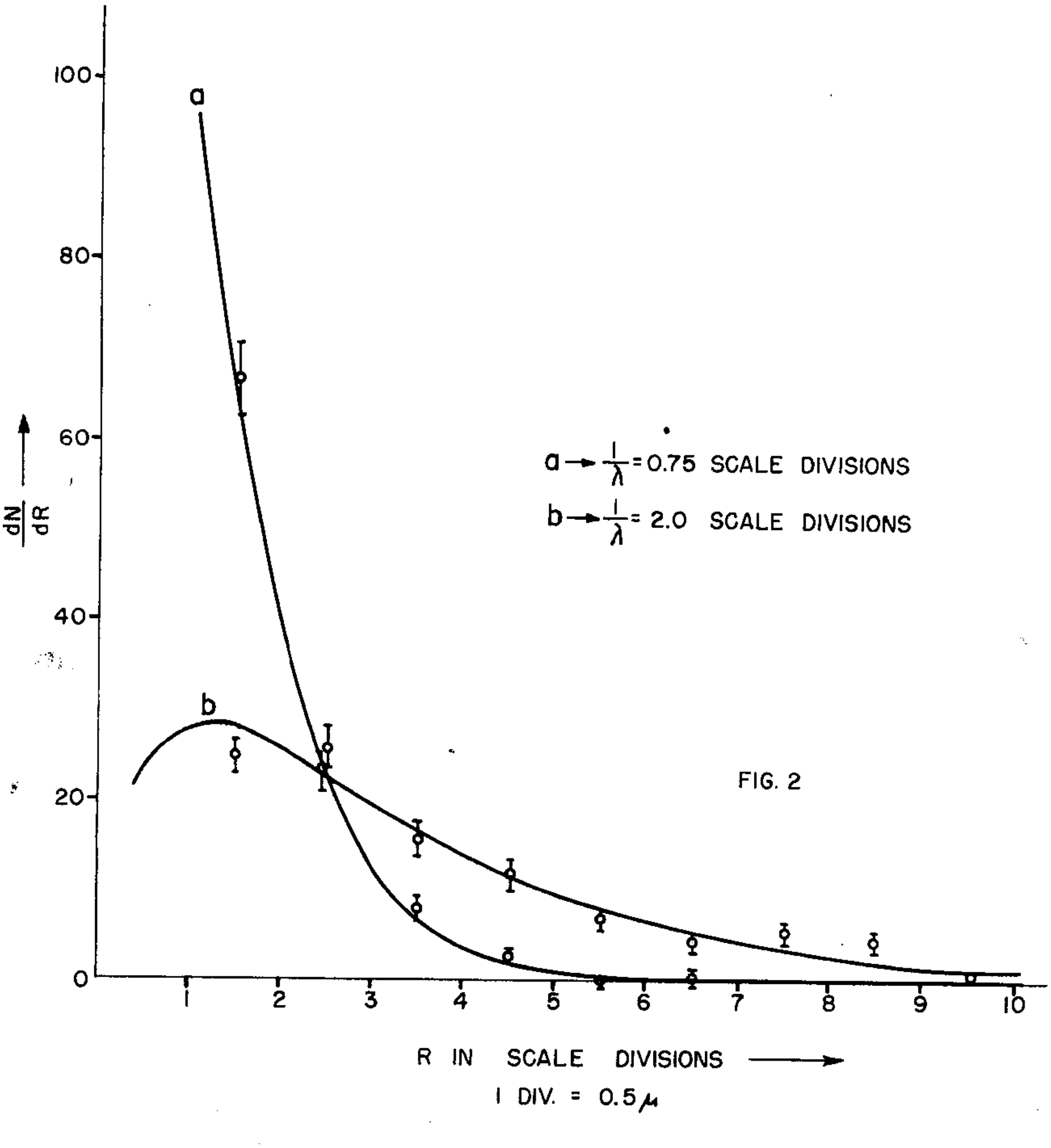

80
60
40
20
0
A
B
A → 1/λ = 1.25 SCALE DIVISIONS
B → 1/λ = 3.3 SCALE DIVISIONS
1
2
3
4
5
6
7
8
9
10
11
12
13
R IN SCALE DIVISIONS →
1 DIV. = 0.5μ

FIG. 1



dN/dR
R IN SCALE DIVISIONS
1 DIV. = 0.5 μ
a
b
a → 1/λ = 0.75 SCALE DIVISIONS
b → 1/λ = 2.0 SCALE DIVISIONS
100
80
60
40
20
0
1
2
3
4
5
6
7
8
9
10

FIG. 2