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ABSTRACT

When accelerated protons, deuterons, and alpha particles of the order of 100 Mev energy strike thin aluminum foils (0.25 - 0.5 mil), sizable percentages of the Na^{24} formed are ejected by the recoil process. In the higher energy range the percentage of Na^{24} ejected from the foil decreases with increase in energy of the incident particle. This behavior is first shown semiquantitatively to be a consequence of constant nuclear excitation in contrast to compound nucleus formation. Calculations are made for the expected loss of Na^{24} assuming compound nucleus formation along with randomly distributed ejection of particles in the production of Na^{24} and the agreement is found satisfactory for 70 Mev protons and 80 Mev alpha particles but not for higher energies. Calculations are also made for the case of constant nuclear excitation with the incident particle of degraded energy leaving in the forward direction and satisfactory agreement with the experimental values are found here over the entire energy range studied.

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In the measurement of cross sections for reactions induced by particles of energy of the order of 100 Mev it has been the practice to employ as a monitor of the beam intensity an aluminum foil in which is produced Na^{24}. This particular system is chosen because the half-life of Na^{24} (14.9 hr) is convenient, its decay scheme is known, there is no conflicting radioactivity from other spallation products, and the excitation functions for the reactions over the energy interval of interest are quite flat. If thin aluminum foils are employed ($\sim 10^{-3}$ inch or less), an appreciable percentage of the Na^{24} produced will leave the foil as a result of the momentum imparted by the projectile. In the course of measuring this quantity it was observed that the mean range of the recoils in the forward direction (percentage loss from the foils) goes through a maximum with increasing particle energy. The interpretation of this phenomenon to be advanced is concerned with the transition with increasing energy through the region of compound nucleus formation into that of increasing nuclear transparency. For the purpose of comparison, calculations are made for the expected loss of Na^{24} from the aluminum foils if the full particle energy is imparted to the compound nucleus.

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EXPERIMENTAL

All irradiations were made with the internal beam of the 184-inch cyclotron and the radial position of the probe target was used to select the particle energy. A clamp arrangement held 4 to 6 thin aluminum foils sandwiched between 10 mil thick graphite or 5 mil polystyrene sheets either as a group or with the carbonaceous layers interspersed. Since the internal beam strikes predominantly the leading edge of the foil stack, care was taken that the edges of the individual foils in the stack were flush with each other so that all were subject to the same beam intensity.

After irradiations of 7-30 minutes the separate foils were weighed, mounted, and the radioactivity followed after a cooling period of about 15 hours. In the aluminum foils and in those carbonaceous foils which were situated in the forward direction of the beam the decay curves for the succeeding 50 or more hours showed almost pure 15 hour periods and the amounts of activity were initially $>10^4$ disintegrations per minute. In time the decay curves tailed into a long lived component, presumably Na^{22} in large part. In the types of foils mentioned there was no difficulty in comparing yields of Na^{24} for recoils in the forward direction. Only in those graphite and polystyrene sheets on the side of the first aluminum foil at which the beam entered was the activity of Na^{24} so low that resolution from activities induced in impurities was difficult. With respect to such impurities, polystyrene presented a lower background than graphite. Activity from Na^{24} in these foils would represent recoils in the backward direction.

The general observation in comparing Na^{24} contents in a stack of foils was that the first aluminum foil was lower than succeeding adjacent aluminum foils and that the deficiency in the first foil was close in magnitude to the amount caught on the graphite or polystyrene adjacent to the last aluminum foil. The obvious explanation is that Na^{24} is produced in primary nuclear reactions and that the momentum distribution received by struck aluminum nuclei is strongly peaked in the forward direction. As would be expected, the Na^{24} found on the graphite or polystyrene in the backward direction was small but, as will be elaborated later, these amounts are significant to some of the considerations.

The comparisons of recoil loss with different projectiles and different energies were quite straightforward since the foils proved to be of uniform thickness and the activity in aluminum foils beyond the first were accordingly quite uniform. In one series of circumstances this was not the case. For alpha particles in the energy range 80-160 Mev, the momentum imparted was sufficient to produce some recoils of range greater than the thickness of a 0.25 mil aluminum foil to the extent that the second foil in a stack had lower activity than succeeding foils by 2-5 percent. In such cases foils beyond the second were averaged to supply the norm. For the same reason the Na^{24} caught in graphite or polystyrene received contributions from an aluminum foil beyond the adjacent one and was therefore greater than that lost by the individual foil.

RESULTS AND DISCUSSION

Tables I, II, and III show the data obtained for protons, deuterons, and alpha particles, respectively. It can be seen in Table I that the percentage loss of Na^{24} from the aluminum foils decreases with increase in proton energy above 70 Mev and has undoubtedly gone through a maximum although at the lowest energy measured (60 Mev) the values are not much lower than at the maximum. The existence of a maximum is seen more readily in the experiments with deuterons and alpha particles (Tables II and III). It will also be noted that the maxima are progressively higher for deuterons and alpha particles which might be expected from the higher momenta of these particles. Some of the data in the alpha particle experiment (e.g., those at 90 Mev) are not considered reliable because of the probable admixture of deuterons in the beam. Because of the relative ease of deuterium ionization, those alpha particle irradiations which followed closely the use of the cyclotron with deuterons probably had large numbers of deuterons in the beams.

That a maximum in the range of the recoils should exist can be seen from a simple approximate calculation. It may be assumed that Na^{24} is produced only in those encounters in which a certain level of nuclear excitation, E, results and that the incident particle of energy E_o is degraded to energy $E_o - E$ and leaves the nucleus without changing direction. The component of the forward momentum of the excited Al^{27} due to momentum conservation in this encounter will be:

$$\left|\vec{P}\right| = \sqrt{2M_iE_o} - \sqrt{2M_i(E_o - E)}$$

$$= E\sqrt{2M_i}/(\sqrt{E_o} + \sqrt{E_o - E}) \qquad (1)$$

where M_i = mass of incident particle. It is clear that $\left|\vec{P}\right|$ will decrease as E_o increases as long as E is assumed to be nearly constant.

Another component of the momentum, $\left|\vec{P_1}\right|$, which the degrading nucleus ($Al^{27} \longrightarrow Na^{24}$) will absorb is the resultant of the emission of nucleons from the excited nucleus. We may assume that the direction of emission is random so that this component will have equal orientation in all directions. Since we have assumed a fairly definite excitation energy, E, for those atoms of Al^{27} which will result in Na^{24}, $\left|\vec{P_1}\right|$ is independent of incident

Table I. Recoil losses of Na^{24} in 0.25 mil aluminum foils irradiated with protons.

Experiment number	Proton energy	Percent loss of Na^{24}	Percent of Na^{24} caught on graphite or polystyrene	
			forward	backward
1	60	27.8	28.0	<2.0
2	60	26.4	28.3	<2.2
3	70	29.2	29.0	<1.5
4	80	27.3	25.4[a]	<0.6[a]
5	80	28.5	25.1[a]	<0.4[a]
6	90	25.8[b]	26.4[b]	
7	90	26.8	26.2	<2.0
8	140	20.9	19.9	
9	180	18.2	18.2[a]	~1.7[a]
10	180	18.6	18.0[a]	~1.5[a]
11	220	17.6	17.6	
12	280	17.5	16.4	
13	340	15.2[b]	15.2[b]	<<4.0[b]
14	340	15.0	15.7	<<4.0
15	340	16.3	17.1	<<4.0
16	340	15.5	14.4[a]	~2.6[a]
17	340	13.9	14.5[a]	~2.8[a]

[a]Values obtained with polystyrene catching sheet; others with graphite.

[b]Those values marked were obtained from experiments on 0.5 mil aluminum foils by doubling the observed percent loss.

Table II. Recoil losses of Na^{24} in 0.25 mil aluminum foils irradiated with deuterons.

Experiment number	Deuteron energy	Percent loss of Na^{24}	Percent of Na^{24} caught on graphite or polystyrene	
			forward	backward
18	40	22.2		
19	60	25.8	28.0	<2.5
20	60	24.0[a]	25.2[a]	<2.6[a]
21	80	32.4	34.1	<2.6
22	80	34.0[a]	29.6[a]	<2.6[a]
23	100	32.0	32.2[b]	~1.0[b]
24	100	34.6[a]	31.0[a,b]	~1.4[a,b]
25	120	30.8	28.2[b]	~1.0
26	120	32.4[a]	27.0[a,b]	~1.0[a,b]
27	140	31.0[a]		
28	160	27.0	27.4[b]	~1.3[b]
29	160	27.4	26.8[b]	~1.3[b]
30	190	25.0	23.8[b]	~1.3[b]
31	190	24.8	24.6[b]	~1.2[b]
32	194	26.6	26.8	
33	194	24.8[a]		
34	194	25.4[a]		
35	194	25.6[c]		<2.0[c]

[a]Values obtained from experiments on 0.5 mil aluminum foils by doubling the observed values.

[b]Values obtained with polystyrene catching sheet; others with graphite.

[c]From experiments on 1.0 mil foil by quadrupling observed values.

Table III. Recoil losses of Na^{24} in 0.25 mil aluminum foils irradiated with alpha particles.

Experiment number	Alpha particle energy	Percent loss of Na^{24}	Percent of Na^{24} caught on graphite	
			forward	backward
36	60	30.7	31.5	<2.1
37	80	57.8	64.8	<1.7
38	80	59.0	65.2	<1.7
39	90	38.0	36.2	<1.7
40	90	36.0	36.9	<1.8
41	90	51.3	--	--
42	100	59.7	64.6	<1.0
43	100	58.8[a]	60.4[a]	<1.0[a]
44	110	60.3	67.6	<1.0
45	110	61.9	68.5	<1.0
46	120	58.7	62.3	<1.1
47	120	65.4[a]	60.0[a]	<1.2[a]
48	120	60.3	68.1	<1.9
49	120	58.7	66.5	<1.9
50	130	56.4	66.0	<1.7
51	130	56.8	67.4	<1.7
52	140	55.0[a]		
53	140	54.2[a]	52.0[a]	
54	140	54.2	59.3	<1.3
55	140	55.6[a]	56.4[a]	<1.4[a]
56	160	50.5	59.3	<1.9
57	160	51.3	60.7	<1.9
58	190	46.2	55.7	<2.8
59	190	48.2	54.3	<2.0
60	220	35.0	34.1	<1.8
61	220	34.8	35.1	<1.8
62	280	35.6	41.9	<<6.0
63	280	35.9	43.1	<2.9
64	340	33.2[a]		<3.0[a]
65	340	29.8[a]		<3.0[a]
66	340	33.4[a]		<3.0[a]
67	340	27.7	29.8	<2.5
68	340	26.7	31.0	<2.2
69	370	26.0[a]		<1.8[a]
70	370	28.0[a]		<2.0[a]
71	370	29.8[a]		<2.4[a]
72	380	24.2		

[a]Values obtained from 0.5 mil aluminum foils by doubling the observed values.

particle energy, E_o. The most favorable means of observing this effect is the measurement of the recoils in the backward direction. Since the momentum in the forward direction $|\vec{P}|$ decreases with increase in energy, the percentage of Na^{24} which leaves the foil in the backward direction should increase. The best data illustrating this effect is taken from the proton irradiations in which it can be seen (Table I) that with 80 Mev protons <0.5 percent of Na^{24} is found on the polystyrene foil in front of the aluminum stack, 1.5 percent for 180 Mev protons, and 2.7 percent for 340 Mev protons. The reason that the data taken with polystyrene are considered pertinent and not those from the graphite is that the graphite contained some impurity which prevented accurate resolution of the Na^{24} activity. The decay of Na^{24} in the polystyrene could be followed with much greater precision.

The preceding discussion gives qualitative plausibility for the phenomenon of nuclear transparency,[1] according to which the degree of nuclear excitation is not proportional to the projectile particle energy and the excess energy is carried off by the particle largely in the forward direction. It should also be mentioned that the cross section for formation of Na^{24} does not change much in the proton energy interval under consideration.[2,3] The effect of nuclear transparency in diminishing the recoil energy becomes pronounced for projectile energies (protons) in excess of ~100 Mev for aluminum, and it is therefore in this energy region that nuclear reactions cannot be interpreted in terms of compound nucleus formation. For deuterons the decrease in recoils occurs above ~140 Mev and for alpha particles above ~160 Mev.

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[1] R. Serber, Phys. Rev. 72, 1114 (1947).

[2] P. C. Stevenson and R. L. Folger, private communication.

[3] N. M. Hintz, Phys. Rev. 83, 185 (1951).

It is, however, instructive to make some calculations from the data at hand on the ranges of recoil nuclei assuming compound nucleus formation in order to see how far the observed ranges depart from these calculations and at what energy compound nucleus formation can be assumed. Similar calculations may also be made assuming a particular degree of nuclear excitation and some direction of emission for the incident or exchanged particle.

There are two principal aspects to the calculation of the ultimate positions of nuclear residues formed in reactions of the type illustrated here by Na^{24} from Al^{27}. The first is the calculation of the resultant momentum from the initial energy transfer and the ejection of various particles from the excited nucleus and the second is the corresponding range of the Na^{24} which in turn demands consideration of the charge of atoms moving rapidly through matter. The validity of the various assumptions that must be made in these calculations cannot be evaluated separately from the data at hand and can only be judged by the over-all effectiveness in producing a picture consonant with the observed results.

Obviously a number of reactions can be written for the formation of Na^{24} from Al^{27} by each of the projectiles studied. Of these one for each bombardment has been selected for further consideration.

$$Al^{27} + He^4 \longrightarrow (P^{31}) \longrightarrow Na^{24} + 2H^1 + He^4 + n \qquad (2)$$

$$Al^{27} + H^1 \longrightarrow (Si^{28}) \longrightarrow Na^{24} + n + 3H^1 \qquad (3)$$

$$Al^{27} + H^2 \longrightarrow (Si^{29}) \longrightarrow Na^{24} + H^1 + He^4 \qquad (4)$$

For the case of the alpha particle bombardment, the momentum in the forward direction $|\vec{P}|$ is simply

$$|\vec{P}| = M_a v_a (1 - v_a^2/c^2)^{-1/2} \qquad (5)$$

where as before, v_α is the velocity of the incident alpha particle in laboratory system, M_α is its rest mass, and c is the velocity of light.

The momentum component $|\overrightarrow{P_1}|$ resulting from subsequent evaporation of one neutron, one alpha particle, and two protons can be treated like a random walk problem. The momentum $|\overrightarrow{P_1}|$ (oriented with equal probability in all directions) acquired by the Na^{24} due to particle evaporation is then:

$$|\overrightarrow{P_1}| \cong (\Sigma\, p_i^{\,2})^{1/2} \tag{6}$$

where the p_i's are the respective momenta of the evaporated particles. By assuming evaporation under constant nuclear temperature, $|\overrightarrow{P_1}|$ can be approximated in terms of the incident particle energy (E_o), the potential barrier of the compound nucleus toward protons (V), and the average binding energy (B) of nucleons in the nucleus. The expression is:

$$|\overrightarrow{P_1}| \cong \frac{0.97}{\sqrt{2}} \left\{ \frac{M_{Al}27}{M_{Al}27 + M_\alpha} (M_n + 2M_p + M_\alpha)E_o - 3(M_n + 2M_p + M_\alpha)B + 4(M_\alpha - M_n)V \right\}^{1/2} \tag{7}$$

The resultant momentum $|\overrightarrow{R}|$ in its relation to the components, the forward momentum $|\overrightarrow{P}|$, and the "random walk" momentum $|\overrightarrow{P_1}|$ are shown in Fig. 1. From Fig. 1 it is seen that:

$$|\overrightarrow{R}| = (\vec{P}^2 + \vec{P}_1^{\,2} + 2|\vec{P}||\vec{P}_1| \cos \theta)^{1/2} \tag{8}$$

$$\omega = \tan^{-1} \frac{|\overrightarrow{P_1}| \sin \theta}{|\overrightarrow{P}| + |\overrightarrow{P_1}| \cos \theta} \tag{9}$$

It remains now to determine the range in aluminum of Na^{24} with momentum R and from this the fractional loss from a plane foil can be determined by the relation:

$$f = \frac{1}{2t} \int_0^\pi S \cos \omega \sin \theta \, d\theta \tag{10}$$

where f is the fractional loss from the foil, and t the thickness and S the range of the Na^{24}, both in the same units.

Selecting equation (3) as the principal reaction for the proton irradiation, one can obtain similar expressions for $|\vec{P}|$ and $|\vec{P_1}|$.

$$|\vec{P}| = M_p V_p (1 - V_p^2/c^2)^{-1/2} \qquad (11)$$

$$|\vec{P_1}| \cong \frac{0.98}{\sqrt{2}} \left\{ \frac{M_{Al}27}{M_{Al}27 + M_p} (M_n + 3M_p)E_o - 3(M_n + 3M_p)B + 3(M_p - M_n)V \right\}^{1/2} \qquad (12)$$

Returning to the question of the range of the Na^{24} as a function of its energy, the principal uncertainty in the calculation is concerned with the charge on the ion as a function of its velocity. Somewhat different treatments are given by Brunings, Knipp, and Teller (BKT)[4] and by Bohr.[5] Calculations were made according to both using the atomic stopping power for aluminum given by Livingston and Bethe.[6]

In the BKT treatment adopted, a parameter $\Upsilon = V_e/V$ is introduced for determining the charge of the ion in which V_e is the root mean square velocity of the energetically most easily removable electron in the Thomas-Fermi model of the ion and V is the velocity of that ion. In general Υ is not unity and varies slowly with atomic number of the ion and with its velocity. In the Bohr treatment the condition for ionization is assumed to be simply the equality between the velocity of an electron and of the ion.

As will be noted in Table IV, the best agreement with the measurements is obtained using $\Upsilon = 1.4$ for sodium. For comparison, values obtained by

[4]J. Knipp and E. Teller, Phys. Rev. 59, 659 (1941); Brunings, Knipp, and Teller, ibid. 60, 657.

[5]N. Bohr, Kgl. Danske Videnskab. Selskab, Mat. fys. Medd. 18, 8 (1940).

[6]M. S. Livingston and H. A. Bethe, Revs. Modern Phys. 9, 272 (1937).

BKT for C^{12} and Ne^{20} were 1.3 and 1.2, respectively. The somewhat higher value assumed here for Na^{24} would signify greater ease in removal of an electron. A sample of the data used for the numerical integrations employed in determining the range is given in the Appendix.

Table IV shows calculated percentage losses from the aluminum foils for selected energies of protons and alpha particles. Columns 7 and 8 give the calculated values assuming compound nucleus formation (full energy transfer) for the BKT calculation with values of $\gamma = 1.3$ and 1.4. Column 9 is with the Bohr assumption of $\gamma = 1$. With the assumption of compound nucleus formation, obviously the calculated percentage losses increase continuously with energy. For the case of $\gamma = 1.4$ the agreement with experiment is not bad for 60 and 70 Mev protons and 80 Mev alpha particles. At higher energies the results from calculations and experimental values diverge.

Column 10 shows a few calculations assuming constant nuclear excitation rather than compound nucleus formation. This excitation is taken to be 70 Mev for protons and 110 Mev for alpha particles and the further assumption is made that the incident particle, after causing excitation, leaves in the forward direction. This introduces another particle, constantly oriented, in the calculation of $\left|\overrightarrow{P_1}\right|$. Despite these severe approximations it is seen that the calculated values are not in discord with the experiment but that they are uniformly low. Higher values which would be in good agreement with the experiment could be obtained if the emission of the incident particle were given a preferred direction not quite in the forward direction. It is also seen (compare columns 11 and 6) that the Na^{24} atoms which recoil back into the direction from which the incident particles come are roughly the same between calculation and experiment.

Table IV. Comparison of measured recoil losses with those calculated using the different models.

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)
				Percent of Na^{24} caught on graphite or polystyrene behind the Al foils		Calculated percent loss of Na^{24} of first Al foil			Calculated percent loss of Na^{24} on nuclear transparency model[a]	
						BKT		Bohr		
Nuclear particles	Energy of particle (Mev)	Thickness of Al foils (mils)	Percent loss of Na^{24} from first Al foil (experimental)	forward	backward	$\gamma = 1.3$	$\gamma = 1.4$	$\gamma = 1$	forward	backward
proton	60	0.25	27.1	28.2		33.9	28.5	19.0		
proton	70	0.25	29.2	29.0		35.9	30.4	20.7		
proton	80	0.25	27.9	25.3		37.0	32.5	23	22.0	
proton	140	0.25	20.9	19.9		50	44.2	32	15.1	
proton	180	0.25	18.4	18.1	~1.6	58.5	52.6	37	13.7	1.3
proton	340	0.25	14.7	14.5	~2.7	77	71.0	61.8	11.7	2.2
alpha	80	0.5	29.2	32.5		37.5	34.3	24.4		
alpha	100	0.5	29.4	30.2		45.5	41.3	30.1		
alpha	120	0.5	32.7	30.0		54.5	49.9	36.6		
alpha	140	0.5	27.8	28.2		63.5	58.3	43.8		
alpha	340	0.5	15	15	<1.5	>78	>78	78	12.4	0.8

[a]Calculations based on BKT method with $\gamma = 1.4$; nuclear excitation assumed to be 70 Mev for protons and 110 Mev for alpha particles; incident particle assumed to leave nucleus without direction change.

APPENDIX

A sample numerical integration and the data employed are given in Fig. 2 and Table V. The case shown is for 70 Mev protons. Substituting into equation (11), first of all we find the forward momentum component

$$\left|\vec{P}\right| = 1.97 \times 10^{-14} \text{ g cm sec}^{-1}.$$

Similarly substituting into equation (12) and assuming B ≅ 10 Mev and V ≅ 4 Mev, the randomly oriented momentum $\left|\vec{P_1}\right|$ is 1.42×10^{-14} g cm sec^{-1}. (Actually the values adopted for B and V are not critical.) From these values the resultant momentum $\left|\vec{R}\right|$ can be calculated [Fig. 1 and equation (8)] for a number of values of θ. The corresponding energies of the Na^{24} ions are listed in column 2 of Table V. The calculated ranges (S) of the ions are listed in column 3 and the values of cos ω in column 4. The product (S cos ω sin θ)/2 of column 5 is the range of the Na^{24} in the forward direction weighted by the number of ions at the selected angle. The integration of this product over π radians of θ in Fig. 2 gives the average range in the forward direction of the Na^{24} ions. This value divided by the foil thickness gives the fraction loss from the foil.

Table V. Data used for numerical integration in Figure 2.

(1) θ	(2) Energy of Na^{24} ion (Mev)	(3) Range in Al (S) (mg cm^{-2})	(4) cos ω	(5) (S cos ω sin θ)/2 (mg cm^{-2})
0				0
$\pi/36$	8.90	0.86	0.997	0.038
$\pi/18$	8.83	0.86	0.993	0.074
$\pi/9$	8.65	0.85	0.987	0.144
$\pi/6$	8.32	0.83	0.978	0.203
$\pi/4$	7.62	0.80	0.947	0.268
$\pi/3$	6.74	0.76	0.910	0.299
$5\pi/12$	5.70	0.71	0.862	0.296
$\pi/2$	4.56	0.65	0.811	0.264
$7\pi/12$	3.45	0.59	0.758	0.216
$2\pi/3$	2.40	0.50	0.715	0.155
$3\pi/4$	1.52	0.42	0.691	0.103
$5\pi/6$	0.82	0.32	0.718	0.058
π				0

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ILLUSTRATIONS FOR UCRL-1709

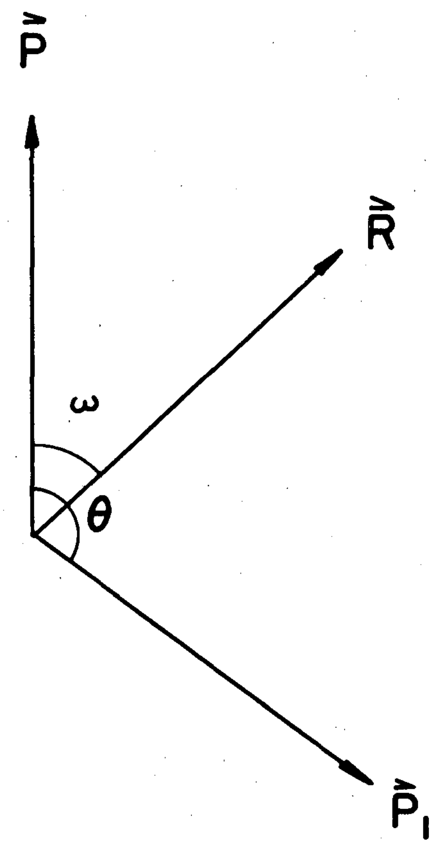

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Fig. 1

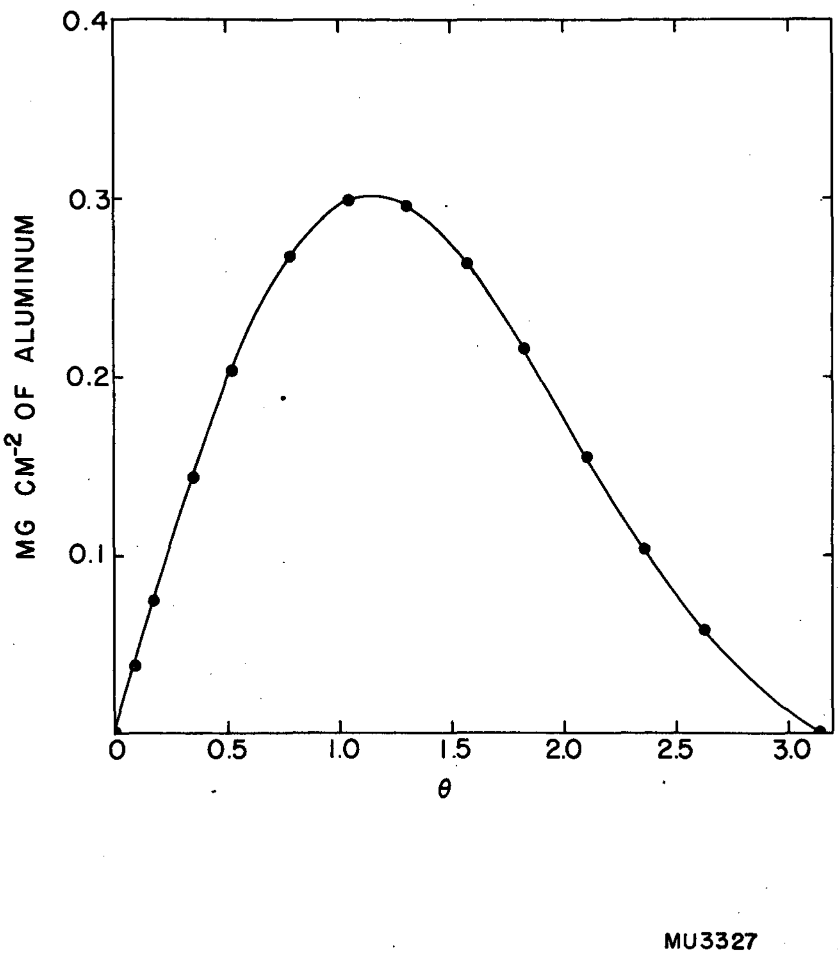

Fig. 2