In Fig. 1 we have also plotted the equation for the diffusion coefficient of Au in lithium [2] and that for "self-diffusion" [3] (i. e. Li^6-tracer in lithium).

The striking feature of Fig. 1 is the small diffusivity observed in the present work, as compared with the very rapid diffusion rate for the univalent tracer. This is in qualitative contradiction to electrostatic theories [4, 5] according to which a polyvalent impurity should tend to diffuse faster than an univalent one.

	D_0 ($cm^2\,sec^{-1}$)	Q ($kcal \cdot mol^{-1}$)	Ref.
Li in Li	0.12 ± 0.05	12.62 ± 0.21	3
Au in Li	0.21 ± 0.08	10.99 ± 0.18	2
In in Li	0.39 ± 0.25	15.87 ± 0.36	

Table 2. Impurity diffusion data in lithium.

According to recent evidence [6] it is possible for certain impurities in polyvalent matrices to dissolve interstitially rather than substitutionally. The tendency for partially interstititial solution is especially marked for univalent impurities while the tendency for substitutional solution grows with increasing valency.

In our experiment the low activation energy and high diffusion coefficient for Au in Li might well indicate an interstitial-like diffusion mechanism. The open structure of the alkali metals may favour this. The three-valent In tracer, on the other hand, may be dissolved mainly substitutionally and so diffuse by a slower mechanism. This work thus indicates that the diffusion mechanism in lithium might possibly depend on the solution mode.

On the other hand, the possibility of a highly relaxed vacancy mechanism does not appear to be completely ruled out. A polyvalent impurity, especially one with a large ionic radius, may tend to neutralise the net negative charge of the vacancy at the same time as it may fill the collective density deficit which defines the defect.

This research has been supported by the Swedish Council for Applied Research. I wish to acknowledge stimulating discussions with Docent A. Lodding and Dr. P. Thernquist.

[1] J. N. Mundy, A. Ott, A. Lodding, and L. Löwenberg, to be published.
[2] A. Ott, Z. Naturforsch., in press.
[3] A. Ott, J. N. Mundy, L. Löwenberg, and A. Lodding, Z. Naturforsch. **23 a**, 771 [1968].
[4] D. Lazarus, Phys. Rev. **93**, 973 [1954].
[5] A. D. LeClaire, Phil. Mag. **7**, 141 [1962].
[6] T. R. Anthony and D. Turnbull, Appl. Phys. Letters **8**, 120 [1966].

Heavy Isotopes of Protactinium

N. Trautmann*, R. Denig, N. Kaffrell, and G. Herrmann

Institut für Anorganische Chemie und Kernchemie der Universität Mainz, Germany

(Z. Naturforsch. **23 a**, 2127—2130 [1968]; received 15 October 1968)

We have identified a new isotope of protactinium, 2.3-min ^{238}Pa, in bombardments of ^{238}U with 14-MeV neutrons and have investigated its decay properties. The nuclides 9.1-min ^{237}Pa, 9.1-min ^{236}Pa and 24-min ^{235}Pa, produced by irradiations of uranium isotopes with 14-MeV neutrons and 100-MeV bremsstrahlung, were also studied since only scanty information on their properties has been published up to now.

Our results are summarized in this note. Discussions of the complex decay schemes have to be postponed until γ-γ and β-γ coincidence spectra are available. Unfortunately, the source strengths achieved by us so far made it impossible to apply coincidence techniques.

Experimental. The targets, all in the form of uranyl nitrate, consisted of 99.3% ^{238}U (i. e. natural uranium), 99.8% ^{236}U or 90% ^{235}U plus 10% ^{238}U. Bombardments with 14-MeV neutrons from the T-D reaction were performed using the Cockcroft-Walton accelerator at this institute; fluxes up to $4 \cdot 10^{10}$ neutrons/cm^2sec were available. Irradiations in a bremsstrahlung spectrum of 100 MeV endpoint energy were carried out at the Mainz electron linear accelerator.

The protactinium isotopes were radiochemically separated from the predominating fission-product activity by partition between diisobutylcarbinol and strong hydrochloric acid containing complex-forming agents [1]. Counting samples were prepared by coprecipitation with ferric hydroxide. The first count could be started within 3.5 to 4 minutes after the end of bombardment. Decontamination from short-lived fission products was tested using samples prepared by thermal-neutron irradiation of ^{235}U in the Mainz research reactor.

The γ-ray spectra of ^{238}Pa and ^{237}Pa were measured with Ge(Li) detectors, those of ^{236}Pa and ^{235}Pa with NaI(Tl) detectors. In addition, a γ-γ sum spectrometer consisting of two NaI(Tl) crystals and an X-ray spectrometer with xenon proportional counter were used. Beta-ray spectra were obtained with plastic scintillators. Only approximate β-ray energies and intensities could be deduced for ^{238}Pa, ^{237}Pa and ^{236}Pa since their spectra are rather complex.

Mass assignments. The β-decay curves of the protactinium fractions show, apart from small admixtures of expected long-lived nuclides, short-lived components of the half-lives and relative activities listed in Table 1. The γ-decay curves are similar except that the 24-min activity decreases sharply or disappears entirely. The 9.1-min components A and B are discernible by their γ-ray spectra.

Under the conditions used, the principal processes resulting in the formation of protactinium isotopes are

* In part from a Dissertation, Mainz 1968.

[1] N. Trautmann, R. Denig, and G. Herrmann, to be published.

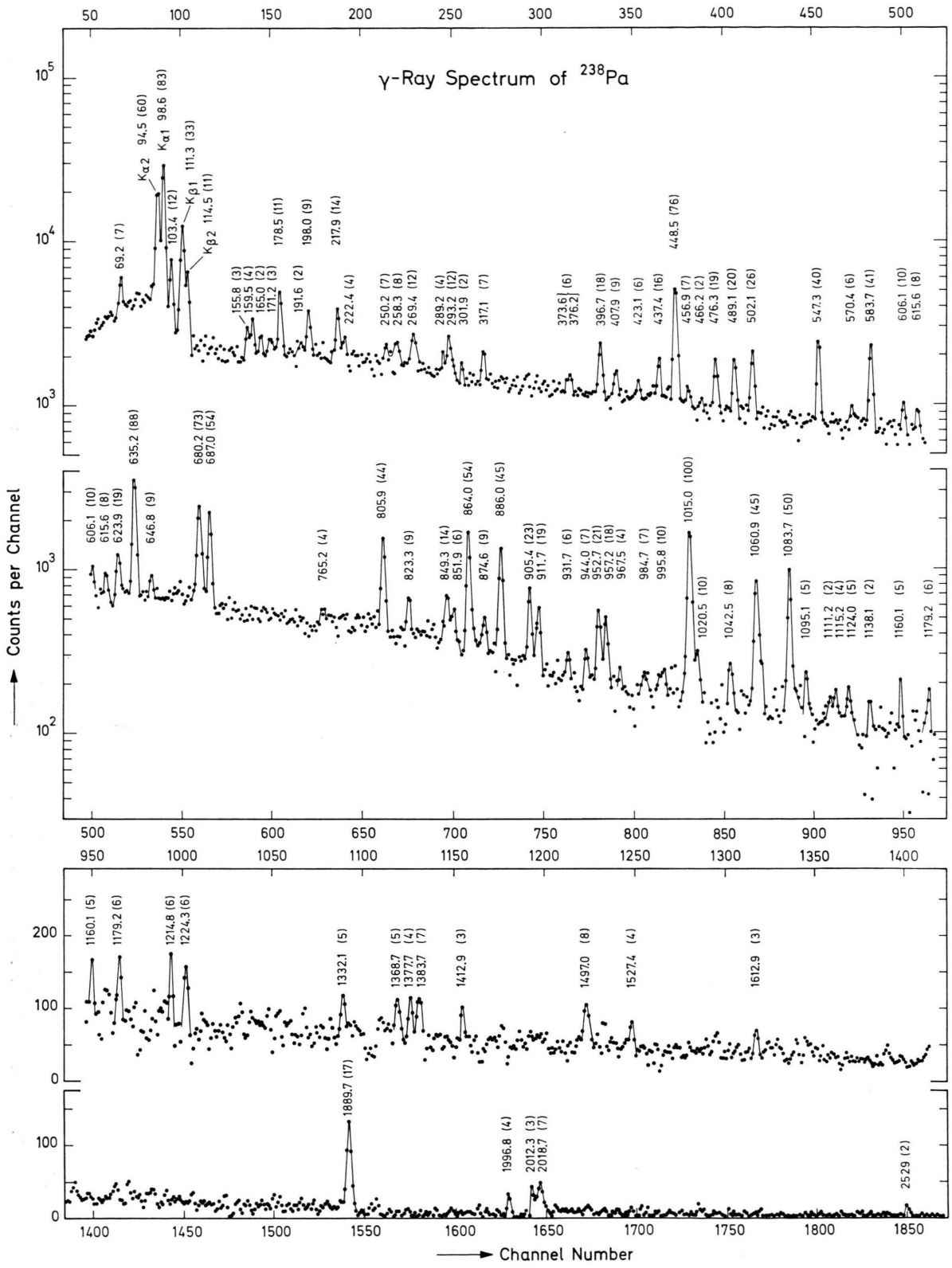

Fig. 1. Gamma-ray spectrum of ^{238}Pa obtained with a 30-ccGe(Li) detector by accumulating the results of 48 runs. Each peak is labelled by its energy (in keV) and intensity (in parentheses, relative to the intensity of the 1015 keV γ-ray).

Half-life	Relative β-activity at saturation in bombardment of ^{238}U +neutrons	^{238}U +γ-rays	^{236}U +neutrons	^{235}U +neutrons	Assignment
2.3 min	100 (n,p)	—	—	≈ 5 *	^{238}Pa
9.1 min (A)	15 (n,pn)	100 (γ,p)	—	—	^{237}Pa
9.1 min (B)	—	≈ 20 (γ,pn)	100 (n,p)	—	^{236}Pa
24 min	15 (n,α) **	40 (γ,p2n)	30 (n,pn)	100 (n,p)	^{235}Pa

* Originates from the 10% ^{238}U admixture. ** Formed via the short-lived ^{235}Th.

Table 1. Short-lived protactinium isotopes observed in bombardments of uranium isotopes with 14-MeV neutrons and 100-MeV bremsstrahlung. In parentheses: production process.

Mass number	Half-life [min]	β-ray energies [MeV] and intensities [%]	γ-ray energies [keV] and intensities [relative units]
238	2.3 ± 0.1	2.9 (5), 2.2 (20), 1.7 (40), 1.2 (35)	see Fig. 1
237	9.1 ± 0.2	2.3 (10), 1.6 (30), 1.1 (60)	K_α (23), K_β (6), 310 (6), 498 (11), 529 (39), 540 (27), 554 (5), 642 (24), 687 (8), 852 (100), 864 (44)
236	9.1 ± 0.3	3.1 (10), 2.0 (50), 1.1 (40)	110 * (25), 225 (≈ 3), 642 (100), 925 (≈ 30), 1090 (≈ 40), higher energies
235	24.2 ± 0.3	1.41 ± 0.05 (97)	75 (5), 115 * (25), 165 (30), 265 (35), 320 (100), 400 to 1270 ** (≈ 100) ***

* Probably K X-rays. ** Broad, unresolved distribution.
*** Total intensity of the γ-ray spectrum: about 3 γ-rays per 100 β-rays.

Table 2. Decay properties of heavy protactinium isotopes.

the (n,p) or (γ,p) reactions. In addition, the (n,pn) and (n,α) reactions — the latter via short-lived thorium intermediates — or the (γ,pn) and (γ,p2n) processes should make significant contributions. These arguments lead to the mass assignments given in Table 1.

Protactinium-238. The γ-ray spectrum of 2.3-min ^{238}Pa is shown in Fig. 1. Eighty-four γ-transitions are observed with energies and intensities as indicated in the figure. The various K X-rays of uranium are present with expected energies and relative intensities. This applies also to the L_α, L_β and L_γ X-rays in spectra measured with the xenon proportional counter. The γ-γ sum spectrum exhibits a complex structure with peaks extending up to 2.0 MeV in energy. The β-ray spectrum shows the groups listed in Table 2.

Several of the strong γ-transitions fit into levels of ^{238}U revealed in Coulomb excitation [2], particularly into the $K^\pi = 0^-$ and 1^- octupole vibrations at 679 and 1105 keV and the associated rotational bands. The following γ-rays deexcite levels of the $K^\pi = 0^-$ band to the ground state band: 680.2 ($I = 1 \rightarrow I = 0$), 635.2 ($1 \rightarrow 2$), 687.0 ($3 \rightarrow 2$) and 583.7 keV ($3 \rightarrow 4$). Thus, energies of 680.2 and 732.0 keV result for the $I = 1$ and $I = 3$ levels in the 0^- band. The γ-rays of 448.5 ($2 \rightarrow 1$), 437.4 ($3 \rightarrow 3$) and 476.3 keV energy ($4 \rightarrow 3$) correspond to transitions from the $K^\pi = 1^-$ into the 0^- band, resulting in energies of 1128.7, 1169.4 and 1208.3 keV for the $I = 2$, 3, 4 levels in the 1^- band. The β-ray and γ-γ sum spectra indicate that ^{238}U levels extending up to about 2.0 MeV are fed in the β-decay of ^{238}Pa.

Protactinium-237. Our results confirm earlier observations of a 10-min ^{237}Pa among the spallation products of uranium by high-energy deuterons [3] and protons [4]. The mass assignment and a half-life value of 10.5 ± 1 min have been determined by repeated milking of the ^{237}U daughter activity [3]. A 39-min ^{237}Pa, previously found and studied in irradiations of ^{238}U with bremsstrahlung [5], was not observed in our work.

The decay properties of ^{237}Pa are summarized in Table 2. Gamma-ray spectra were measured using samples prepared by bombardment of ^{238}U with bremsstrahlung and with 14-MeV neutrons. Both spectra were found to be identical within the statistical accuracy, except that an excessive intensity at 642 keV was caused by ^{236}Pa in the samples from the bremsstrahlung bombardment.

Protactinium-236. As only 30 mg of ^{236}U were available, the spectra obtained for 9.1-min ^{236}Pa were low

[2] F. S. Stephens, B. Elbek, and R. M. Diamond, unpubl. work quoted in R. M. Diamond and F. S. Stephens, Ark. Fysik **36**, 221 [1967].

[3] W. W. T. Crane and G. M. Iddings, Phys. Rev. **95**, 1702 [1954].

[4] B. D. Pate and A. M. Poskanzer, Phys. Rev. **123**, 647 [1961].

[5] K. Takahashi and H. Morinaga, Nucl. Phys. **15**, 664 [1960].

in statistics. Therefore, only the principal properties can be given in Table 2. The energy of 642 keV of the most intense γ-ray, measured as mentioned in the preceding section, agrees with that of a transition occurring in the α-decay of ^{240}Pu and assigned to a K, $I^{\pi} = 2, 2^-$ level in ^{236}U [6].

Previously, a half-life value of 12.5 ± 1.0 min and β-rays of 3.35 ± 0.10 MeV maximum energy have been attributed to ^{236}Pa produced by bombardment of ^{238}U with 26-MeV deuterons [7]. Since no γ-rays have been reported, it remains open whether this activity and the one found in our work are identical.

Protactinium-235. The 24-min ^{235}Pa, identified in the ^{234}Th(n,γ)^{235}Th $\xrightarrow{\beta^-}$ ^{235}Pa, ^{238}U(d,αn)^{235}Pa and ^{238}U(p,α)^{235}Pa reactions [8, 9], is confirmed. Half-life and β-ray energy (Table 2) agree with earlier measurements [9] which gave 23.7 ± 0.5 min and, by absorption techniques, 1.4 MeV. In addition, ^{235}Pa is found to emit a complex γ-ray spectrum of about 3% abundance.

The authors are indebted to Prof. F. Strassmann for his interest in this work, to Dr. G. Luthardt, Dr. P. Patzelt and Mr. H. Herter for providing high fluxes of 14-MeV neutrons and numerous bombardments, to Prof. B. Ziegler and his associates for irradiations with bremsstrahlung, and to the staff of the Mainz research reactor for irradiations with thermal neutrons. They wish to thank the Bundesministerium für wissenschaftliche Forschung for financial support.

[6] C. M. Lederer, Univ. of California Report UCRL-11 028 [1963].
[7] G. Wolzak and H. Morinaga, Radiochim. Acta **1**, 225 [1963].
[8] B. G. Harvey and B. I. Parsons, Phys. Rev. **80**, 1098 [1950].
[9] W.W. Meinke and G. T. Seaborg, Phys. Rev. **78**, 475 [1950].

The Immersion Grating: Spectroscopic Advantages and Resemblance to the Echelon Grating

T. Larsson and H. Neuhaus

Institute of Physics, University of Stockholm, Stockholm, Sweden

(Z. Naturforsch. **23 a**, 2130—2132 [1968]; received 6 November 1968)

The function of the immersion grating is explained and special attention is given to the role of the prism, which has been subject to misunderstandings. The analogy between the immersion grating and the echelon grating, used in reflection through the glass, is demonstrated.

The immersion grating consists of two solid components, a reflection grating and a prism. These are held in optical contact by means of the third component, a film of immersion oil, such as monobromnaphthalene. The contribution of each component, as well as the dispersion and resolving power of the system as a whole, have been considered by Hulthén and Neuhaus [1]. The use in spectroscopy of immersion gratings was first suggested by Hulthén [2] and the results of a number of experiments have been reported [1, 3–5].

Recently, this instrument has been described by Stroke [6], in his chapter on Diffraction Gratings in Flügge's Handbuch der Physik. The present note offers some comments, as we consider Stroke's treatment rather misleading. At the same time we draw attention to the close theoretical analogy between the immersion grating and the echelon grating when used as a reflection grating illuminated through the glass. A theoretical explanation of the latter arrangement was given by Mitra [7], who did not, however, point out the above analogy.

Let us first consider a diffraction grating covered with a plane parallel film of a dielectric medium with a high refractive index. It is easily shown [1, 6] that the grating equation is not altered by the presence of the medium. From this it may be concluded: The spectra from a free and an immersed grating, as observed in air, coincide in all orders. Thus the medium *per se* does not contribute to the dispersion or resolving power of the grating. But this does not mean that no spectroscopic advantages are to be won by the immersion arrangement.

However, a better performance from the grating can only be attained by the immersion technique if the grating is blazed. This is the crucial point of the argument, and one that is overlooked by Stroke.

We shall denote the blaze angle of the grating by ε, and throughout the discussion we presume that the grating works in autocollimation, so that the angle of diffraction is approximately equal to the angle of incidence α on the grating. Thus we have for a free grating in air the equation

$$m\,\lambda = 2\,d\,\sin\alpha \qquad (1)$$

and for an immersed grating we may write

$$m_i\,\lambda_i = 2\,d\,\sin\alpha_i\,. \qquad (2)$$

If, in both cases, we work close to the blaze in order to ensure maximum intensity, we have the additional condition

$$\alpha \approx \varepsilon \approx \alpha_i \qquad (3)$$

and noting that $\lambda_i = \lambda/n_i$, where n_i is the refractive index of the dielectric medium, it will be seen immediately that

$$m_i \approx m \cdot n_i\,. \qquad (4)$$

[1] E. Hulthén and H. Neuhaus, Arkiv Fysik **8**, 343 [1954].
[2] E. Hulthén, Proc. London Opt. Conference 1950, p. 111.
[3] E. Hulthén, N. Johansson, and U. Pilsäter, Arkiv Fysik **14**, 31 [1958].
[4] H. Neuhaus, Z. Naturforsch. **21 a**, 2113 [1966].
[5] T. Larsson, H. Neuhaus, and N. Åslund, Ark. Fysik **37**, 141 [1968].
[6] G. W. Stroke, Handbuch der Physik (S. Flügge, Editor), Springer-Verlag, Berlin 1967, Vol. XXIX, p. 457.
[7] S. S. Mitra, J. Opt. Soc. Amer. **50**, 1028 [1960].