Photofission and Photoneutron Emission in Uranium*

ROBERT B. DUFFIELD, *University of Illinois, Urbana, Illinois*

AND

JOHN R. HUIZENGA, *Argonne National Laboratory, Lemont, Illinois*

(Received November 24, 1952)

The yields of the two reactions $U^{238}(\gamma,n)U^{237}$ and $U^{238}(\gamma$, fission) produced by bremsstrahlung have been measured as a function of the maximum energy of the x-ray spectrum. The ratio of the number of fissions to the number of U^{237} atoms produced decreased from 0.390 at 8 Mev to 0.275 at 12 Mev and then increased to 0.465 at 22 Mev. These data are interpreted to show that for maximum x-ray energies in excess of 12 Mev, a fraction of the U^{237} atoms formed by the (γ,n) reaction are left with sufficient excitation energy that fission or secondary neutron emission can occur. The cross sections for the reactions as a function of gamma-ray energy have been calculated from the yield data.

I. INTRODUCTION

MEASUREMENTS have been made by Price and Kerst[1] of the total photoneutron intensity from various elements excited by 18, 22, and 320 Mev bremsstrahlung. Measurements have also been made at 330 Mev by Terwilliger, Jones, and Jarmie.[2] Price and Kerst found the number of neutrons per mole per roentgen to vary smoothly (approximately as Z^2) for all the elements except that thorium and uranium gave too many neutrons by factors of 1.36 and 1.76 at 22 Mev. The additional neutrons were attributed to photofission and it was calculated that if the number of neutrons emitted per fission was 2.5, an excited uranium nucleus underwent fission with a probability of approximately 0.5 under their irradiation conditions.

A result at variance with this has been reported by Goward *et al.*[3] Using 23-Mev bremsstrahlung, they found that one milligram of uranium gave 24.2±2 fissions per r and 255±25 neutrons per r, indicating a much lower fission branching ratio. The fission yield was determined by particle counting; the neutron yield by measuring the activity induced in a manganese sulfate solution as compared with that produced by a standard Ra—Be source.

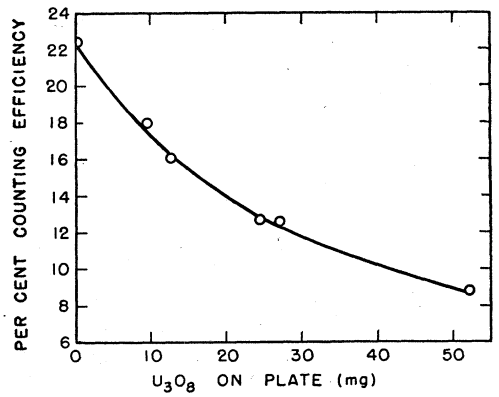

FIG. 1. The counting efficiency for U^{237} in the counter used *vs* sample thickness.

The photofission cross section of uranium for the 17-Mev Li(p,n) gamma-rays was measured by Charbonnier, Scherrer, and Wäffler.[4] They counted the number of fission tracks produced in irradiated uranium-loaded nuclear track emulsions. They obtained a result of 0.046±0.015 barn.

We have measured the yields of the two reactions $U^{238}(\gamma,n)U^{237}$ and $U^{238}(\gamma$, fission) produced by bremsstrahlung from a betatron at energies from 8 to 22 Mev. The yield of the first reaction has been determined by isolating and counting the U^{237} formed and the yield of the second by isolating and counting radioactive fission products. The absolute yields of the two reactions measured in this way depend on an accurate calibration of the counters used. For U^{237}, this calibration was done by measuring the amount of alpha-emitting Np^{237} formed by decay of the U^{237}. For the fission products it was done by measuring the activity of fission products resulting from a known number of neutron-produced fissions. We believe that the absolute values so obtained are correct to a few percent.

II. EXPERIMENTAL

The uranium samples used were depleted of U^{235} and were in the form of U_3O_8. Each sample consisting of approximately 0.325 g of U_3O_8, was irradiated in the external x-ray beam from the University of Illinois 22-Mev betatron. After irradiation, this material was divided, part being used for the U^{237} determination and part for the fission product analysis. The beam was monitored using a standard Victoreen thimble inside an aluminum block of wall thickness 4 cm.

* This work was supported by the joint program of the U. S. Atomic Energy Commission and the U. S. Office of Naval Research.

[1] G. A. Price and D. W. Kerst, Phys. Rev. **77**, 806 (1950); **79**, 725 (1950).

[2] Terwilliger, Jones, and Jarmie, Phys. Rev. **82**, 820 (1951).

[3] Goward, Jones, Watson, and Lees, Proc. Phys. Soc. (London) **A64**, 95 (1951).

[4] Charbonnier, Scherrer, and Wäffler, Helv. Phys. Acta **22**, 385 (1949).

1. U^{237} Determination

The uranium was separated from fission products and counted by the procedure previously described.[5] This involved ether extractions, deposition as a thin film, and counting on a thin window Geiger counter. This counter was standardized for absolute beta-counting of U^{237} in the following way. A sample of uranium containing 95 percent U^{236} was irradiated in the thimble of the Argonne heavy water reactor to produce U^{237}. After bombardment the uranium was purified of neptunium and fission products. A known portion of the total purified uranium was then used to prepare several thin plates for beta-counting. The U^{237} in these was diluted with various known weights of normal uranium so that the counting efficiency could be obtained as a function of film thickness.

The remainder of the purified uranium was allowed to decay for about five U^{237} half-lives. The Np^{237} formed by this decay was then extracted and purified (*cf.* Appendix) and its amount determined by alpha-counting in a counter of known geometry. Alpha-pulse analysis[6] indicated that more than 99 percent of the alphas were in the energy range of Np^{237}. Good purification from uranium is essential in this step since the U^{234} and Np^{237} alpha-energies are very nearly the same. However, no correction for U^{234} was necessary since U^{236} was not detected in the alpha-pulse analysis (this uranium sample had a U^{236}/U^{234} alpha-activity ratio greater than ten).

Using 6.75 days as the half-life[7] of U^{237} and 2.2×10^6 years as the half-life[8] of Np^{237}, the counting efficiency of U^{237} was found as a function of sample thickness and the results are given in Fig. 1. These data were used to calculate the number of U^{237} atoms formed in the betatron irradiations.

TABLE I. Number of fissions and U^{237} atoms from U^{238} as a function of betatron energy.

Betatron energy	Roentgens	Total number of fissions per mole of U^{238}	Total number of U^{237} atoms per mole of U^{238}	Fissions per mole of U^{238} per 100 r	U^{237} atoms per mole of U^{238} per 100 r	Fissions per atom of U^{237}
8	···	6.69×10^9	1.72×10^{10}	···	···	0.390
10	5746	9.16×10^9	2.31×10^{10}	1.59×10^8	4.02×10^8	0.396
12	9433	3.03×10^{10}	1.10×10^{11}	3.22×10^8	1.17×10^9	0.275
12	···	1.08×10^{11}	3.91×10^{11}	···	···	0.276
14	8793	5.14×10^{10}	1.49×10^{11}	5.85×10^8	1.69×10^9	0.345
16	10 553	7.81×10^{10}	2.05×10^{11}	7.40×10^8	1.94×10^9	0.381
18	···	6.91×10^{10}	1.74×10^{11}	···	···	0.397
20	9808	8.91×10^{10}	2.00×10^{11}	9.08×10^8	2.04×10^9	0.446
20	···	5.45×10^{11}	1.15×10^{12}	···	···	0.474
20	···	3.62×10^{12}	8.15×10^{12}	···	···	0.444
22	···	7.81×10^{10}	1.68×10^{11}	···	···	0.465

[5] Huizenga, Magnusson, Fields, Studier, and Duffield, Phys. Rev. **82**, 561 (1951).

[6] The pulse analysis was carried out by J. Mech of the Argonne National Laboratory.

[7] J. R. Huizenga and K. Flynn (unpublished data).

[8] L. B. Magnusson and T. J. LaChapelle, J. Am. Chem. Soc. **70**, 3534 (1948).

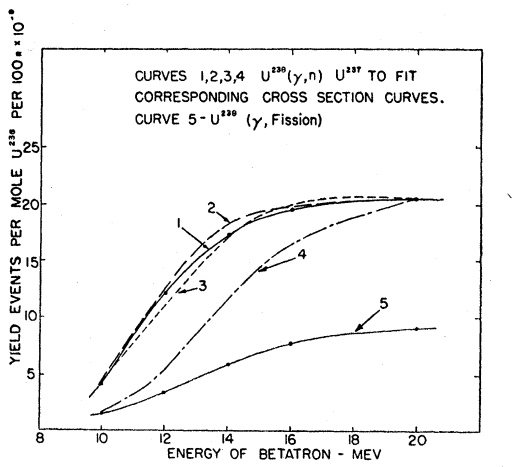

FIG. 2. The yields of the reactions $U^{238}(\gamma,n)U^{237}$ and $U^{238}(\gamma,$ fission) *vs* betatron energy. 1 and 5 are the experimental yield curves for the reactions $U^{238}(\gamma,n)U^{237}$ and $U^{238}(\gamma,$ fission), respectively. Curves 2, 3, and 4 are hypothetical $U^{238}(\gamma,n)U^{237}$ yields resulting from the corresponding assumed cross-section curves of Fig. 5.

2. Fission Determination

The number of fissions occurring in each irradiated sample was determined by measuring the number of atoms of Ba^{139}, Mo^{99}, and Ba^{140} which were made during the irradiation. These fission products were separated chemically from the uranium using well-known procedures[9] and the amounts determined by counting the activity.

The standardization of the Geiger counter used was done in cooperation with R. W. Spence of the Los Alamos Scientific Laboratory. The procedure consisted in measuring the number of counts of a given fission product in a sample of U^{235} in which a known number of slow neutron-produced fissions had occurred. The latter

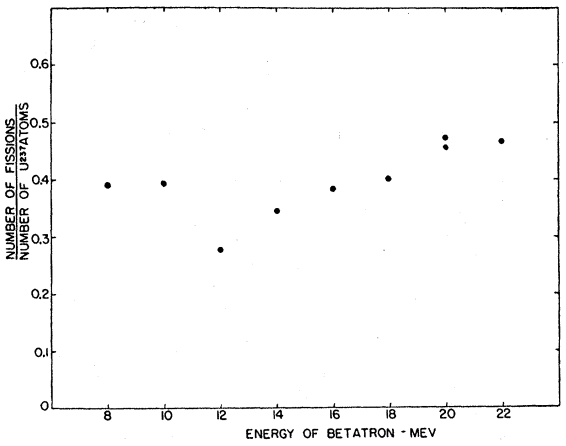

FIG. 3. The ratio, the number of fissions occurring to the number of U^{237} atoms formed by bremsstrahlung *vs* betatron energy.

[9] *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

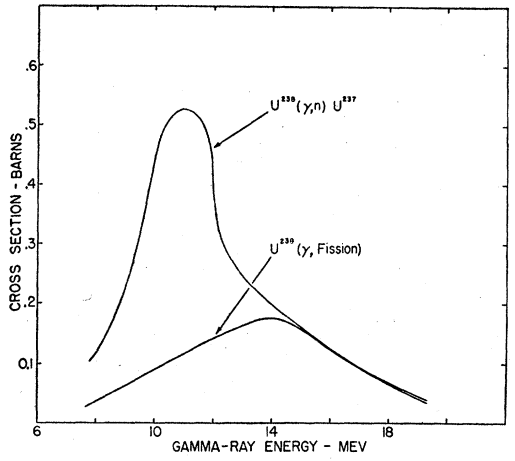

FIG. 4. The cross sections for the reactions $U^{238}(\gamma,n)U^{237}$ and $U^{238}(\gamma,$ fission) *vs* the energy of the gamma-ray.

number of fissions was determined by particle counting in an ionization chamber.

This standardization was checked at Argonne National Laboratory by an independent measurement of the number of fissions occurring in one of the 20-Mev runs. The two numbers differed by 2.5 percent.

III. RESULTS

The data are summarized in Table I and shown graphically in Figs. 2 and 3. For each sample irradiated we have measured the total number of fissions which occurred and the total number of U^{237} atoms formed. These are tabulated in the third and fourth columns of Table I as the number of the corresponding events per mole of uranium. For those runs for which we have an accurate measure of the x-ray exposure we have also tabulated the number of events per mole of uranium per 100 r of x-rays. A number of the runs were made with the sample 25 cm from the betratron target (the usual distance was 55 cm) and for these, an accurate determination of the x-ray exposure was not made. This close distance was used in order to get more intensity.

In calculating the number of photofissions which occurred during the betatron runs, it was assumed that the yield of barium and molybdenum photofission products was 0.9 that from slow neutron fission of U^{235}. There are no experimental data on this point, but the above figure seems reasonable since it is known that fission becomes more nearly symmetric as the energy of excitation is increased. All the absolute fission yields which appear in this paper depend on this assumption of course and if it is incorrect, the fission yields will be incorrect accordingly.

In Fig. 2 are shown the yields (events per mole of U^{238} per 100 r of x-rays) as a function of the betatron energy, the two solid curves representing the experimental data. The U^{237} yield curve has quite a different shape as well as absolute value from the fission yield curve. The other curves in this figure are discussed below.

In Fig. 3 the ratio of the number of fissions to the number of U^{237} atoms formed is plotted as a function of betatron energy. As can be seen, this number is very roughly constant at 0.4 except for a valley in the neighborhood of 12 Mev. This last feature is discussed below.

These yield curves have been analyzed by what are now standard procedures[10,11] to give cross sections as functions of discrete gamma-ray energy. In this procedure the bremsstrahlung spectrum is taken to be that calculated by Schiff.[12] The cross sections so calculated, particularly at the higher energies, are very sensitive to the experimental fluctuations so that the detailed shapes of the cross sections should not be taken too literally.

The cross-section curves calculated from the experimental data are shown in Fig. 4. The cross section for the reaction $U^{238}(\gamma,n)U^{237}$ appears to have a strong maximum at the low energy of 11 Mev, decreasing sharply above this. The reaction $U^{238}(\gamma,$ fission) has a broad maximum at approximately 14 Mev. The latter result is in agreement with previously reported measurements of the U^{238} photofission cross-section shape.[13,14]

The range of uncertainty in these cross sections is indicated in Fig. 5. The solid line is the cross section deduced for the reaction $U^{238}(\gamma,n)U^{237}$ from the experimental data. The broken lines represent other arbitrarily chosen curves which resemble in some features the solid curve. We have used these cross-section curves and the Schiff bremsstrahlung spectrum to calculate back to the hypothetical yield curve to which each of these would

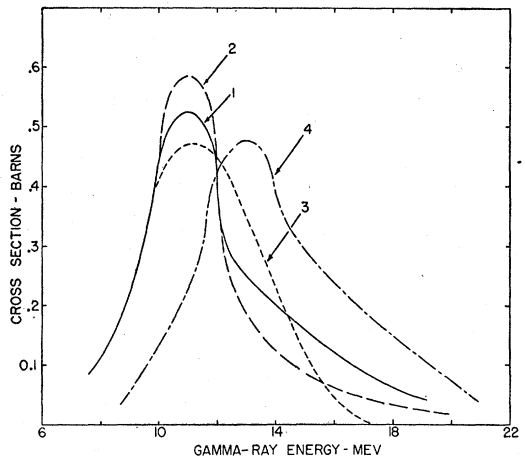

FIG. 5. Cross section for the reaction $U^{238}(\gamma,n)U^{237}$ *vs* gamma-ray energy. Curve 1 is the experimental one. Curves 2, 3, and 4 are arbitrarily chosen to resemble Curve 1 in some features; they give the yield curves numbered correspondingly in Fig. 2.

[10] B. C. Diven and G. M. Almy, Phys. Rev. **80**, 407 (1950).
[11] L. Katz and A. G. W. Cameron, Can. J. Phys. **29**, 518 (1951).
[12] For a discussion, see G. D. Adams, Phys. Rev. **74**, 1707 (1948).
[13] W. E. Ogle and J. McElhinney, Phys. Rev. **81**, 344 (1951).
[14] R. E. Anderson and R. B. Duffield, Phys. Rev. **85**, 728 (1952).

correspond.[15] These yield curves appear in Fig. 2 with numbers indicating the cross-section curve to which they correspond.

It is evident from curves 2 and 3 of Fig. 2 that changing the heights or detailed shape, particularly on the high energy side, of the cross-section curve makes only a small difference in the yield curve. However, curve 4 shows that shifting the position of the maximum by 2 Mev makes a great difference in the yield curve. We conclude from these that the cross section has a maximum near 11 Mev but that its exact shape is uncertain.

The value of the integrated photonuclear cross section up to 20 Mev for the reaction $U^{238}(\gamma,n)U^{237}$ from the above data comes out to be 2.6 Mev-barns and for the reaction $U^{238}(\gamma,\text{fission})$ the corresponding value is 1.2 Mev-barns.

IV. DISCUSSION

Our experiments are in general accord with the results one might expect on the basis of the statistical compound nucleus model. Complete predictions from the model have not been made because of the difficulty of treating the fission process. The questions of the total integrated photodisintegration cross section and the multiplicity of neutron emission have been discussed by Levinger and Bethe[16,17] and by Eyges.[18]

Previous measurements have shown that the threshold for photofission of U^{238} is approximately 5.3 Mev[19,14] and that the threshold for the reaction $U^{238}(\gamma,n)U^{237}$ is 5.97 Mev.[5] The photofission threshold predicted by the liquid drop model calculations for U^{237} is 7 Mev[20] but the indications from other data are that all photofission thresholds for heavy elements are considerably lower, perhaps 5.3 Mev.[19,21] The neutron binding energy in U^{237} is probably about 5.2 Mev.[22] These numbers indicate that a U^{238} nucleus with excitation energy between 6 Mev and approximately 11.3 Mev can either undergo fission or neutron emission. If we let σ_f represent the cross section for the reaction $U^{238}(\gamma,\text{fission})$ and σ_n the cross section for the reaction $U^{238}(\gamma,n)U^{237}$, our experimental results show that the ratio $\sigma_f/(\sigma_f+\sigma_n)$ decreases somewhat between 8 and 11 Mev but has the average value 0.20 to 0.25.

For excitation energies in excess of 11.3 Mev, the U^{237} nucleus left after single neutron emission may either emit a second neutron or may undergo fission. Either process will serve to increase the experimentally observed ratio of the number of fissions to the number of residual U^{237} atoms as a function of betatron energy. This effect is apparent in the data of Fig. 3, and accounts for the rise above a betatron energy of 12 Mev. The cross section for the reaction $U^{238}(\gamma,n)U^{237}$ definitely appears to decrease sharply beyond a gamma-ray energy of 11 to 12 Mev, though as shown by the curves, the exact dependence on energy is rather uncertain.

The product of the $(\gamma,2n)$ reaction, U^{236}, was not measured in these experiments, To do so is very difficult experimentally because of its long half-life. As pointed out above, it should have a threshold of about 11.3 Mev and the cross section may be appreciable in the energy range 15 to 20 Mev.

Levinger and Bethe have shown that a value of 0.23 (independent of excitation energy) for the relative probability of fission of a uranium nucleus would reconcile the neutron yield data at 320 Mev with the value of the integrated photodisintegration cross section predicted by the sum rule formula and neutron yield data for lower Z elements. This formula is

$$\int_0^\infty \sigma dW = 0.06(NZ/A)(1+0.8x),$$

where N, Z, and A are the neutron number, proton number, and mass number of the nucleus in question, and x is the fraction of the neutron-proton force that has an exchange character. These authors show that the 320-Mev data for Cu, I, Ta, and Bi are in accord with this formula written as

$$\int_0^\infty \sigma dW = 0.14NZ/A.$$

This formula predicts an integrated cross section for uranium of 7.9 Mev-barns. This agrees with the observed neutron yield at 320 Mev if the probability of photofission is as given above and if the neutron multiplicity is proportional to the excitation energy.

The coefficient of NZ/A in the above formula required to fit the 320-Mev neutron yield data is higher than expected even for $x=1$. Eyges[18] has shown that the lower energy data for Zn^{64}, Sb and Ta^{181} indicate a smaller value of this coefficient, and he attributes the difference to mesonic interaction at the higher energies. His evaluation indicates an integrated cross section to 20 Mev, for uranium, of 4.7 Mev-barns.

Our data gave integrated cross sections to 20 Mev of 1.2 Mev-barns for photofission and 2.6 Mev-barns for $U^{238}(\gamma,n)U^{237}$. They indicate a total integrated photodisintegration cross section to 20 Mev of at least 3.8 Mev-barns but give no measure of the cross sections for $(\gamma,2n)$ and $(\gamma,3n)$. A value of 1 Mev-barn for the $(\gamma,2n)$ cross section seems not unreasonable and is consistent with the other cross-section data if one assumes that the relative probability of fission as compared to neutron emission is independent of the excitation energy and has the value of about 0.25.

[15] The yield curves were calculated by means of a synchro-operated differential analyzer designed by Arnold T. Nordsieck of the University of Illinois.
[16] J. S. Levinger and H. A. Bethe, Phys. Rev. **78**, 115 (1951).
[17] J. S. Levinger and H. A. Bethe, Phys. Rev. **85**, 577 (1952).
[18] L. Eyges, Phys. Rev. **86**, 325 (1952).
[19] Koch, McElhinney, and Gasteiger, Phys. Rev. **77**, 329 (1950).
[20] S. M. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).
[21] J. R. Huizenga and R. B. Duffield, Phys. Rev. **88**, 959 (1952).
[22] Unpublished calculations independently made by K. Way and J. R. Huizenga.

Finally, our experimental cross-section curve for photofission shows a value of 0.1 barn at 17.5 Mev. Charbonnier, Wäffler, and Sherrer reported a value of 0.046±0.015 barn at this energy. This probably constitutes agreement within the large errors of cross-section determination for the bremsstrahlung spectrum.

We are indebted to R. W. Spence of the Los Alamos Laboratory for the calibration of our fission product counter, to A. T. Nordsieck of the University of Illinois for assistance in the operation of the differential analyzer, and to Kevin Flynn of Argonne National Laboratory for the fission product analysis.

APPENDIX

Procedure for separation of neptunium from uranium.

1. A known amount of Np^{239} tracer was added so that the chemical yield could be determined.
2. Potassium permanganate added to oxidize both Np^{237} and Np^{239} to +6 valence state.
3. The neptunium was reduced with sodium nitrite.
4. Neptunium fluoride was precipitated using lanthanum fluoride as carrier.
5. The fluorides were metathesized with sodium hydroxide and dissolved in acid.
6. Steps 2 through 5 were repeated.
7. The neptunium was extracted into an organic solvent as the thenoyltrifluoracetone complex.

PHYSICAL REVIEW VOLUME 89, NUMBER 5 MARCH 1, 1953

Relativity Precession of the Asteroid Icarus

J. J. GILVARRY
Rand Corporation, Santa Monica, California
(Received July 28, 1952)

The relativity precession of the asteroid Icarus is computed to be 10.05″ per century. This value is about one-quarter that of Mercury and exceeds that of any major planet excluding Mercury. Consideration of a figure of merit, which determines the possible precision in measurement of the perihelion advance, indicates that the motion of Icarus can yield a further test of the precessional formula of general relativity.

THE observational evidence[1,2] for the relativity precession of planetary orbits depends primarily on the rate of advance of the perihelion of Mercury. This note considers the possibility of obtaining a further unequivocal check of the precessional formula from the motion of the asteroid Icarus.

Icarus was discovered by Baade in 1949.[3] It is the minor planet of smallest known mean distance from the sun (its perihelion is within the orbit of Mercury) and is one of the highest in eccentricity. The predicted value of the angular advance v per century of the perihelion of Icarus is listed in Table I. For comparison purposes, the corresponding quantities for Mercury, Venus, the earth, and Mars[2] are likewise tabulated. The quantity α of the third column of Table I is a weighted figure of merit of the possible precision in measurement of the corresponding perihelion motion. For the major planets, the parameter α is essentially *kev* as used by Clemence;[2] for an asteroid, the definition of the over-all figure of merit requires modifications which will be explained in a fuller account of this work (to be published elsewhere).

TABLE I. Predicted relativity precessions.

Asteroid or planet	Advance per century, v	Observational figure of merit, α
Icarus	10.05″	>4.4″
Mercury	43.03	3.01
Venus	8.63	0.03
Earth	3.84	0.06
Mars	1.35	0.38

For Icarus, Table I indicates that the predicted relativity precession per century is about a quarter that of Mercury. However, the advance per century of Icarus exceeds that of any major planet excluding Mercury. The observational figure of merit α for Icarus[4] exceeds that of Mercury, and it exceeds by a factor of at least ten that of any major planet excluding Mercury. One notes from the table that only for Icarus and Mercury are the advance v and figure of merit α both relatively high. Thus, these figures indicate clearly that the motion of Icarus can yield a definite observational check of the precessional formula of general relativity, in the course of time, although decades of astronomical observation may be necessary to measure the perihelion advance with sufficient precision. The relativity precessions of other favorable asteroids amount to only one or two seconds of arc per century.

The author acknowledges helpful discussions of this problem with Professor Samuel Herrick of the University of California, Los Angeles (who computed the orbit elements of Icarus employed).

[1] G. M. Clemence, Revs. Modern Phys. **19**, 361 (1947).
[2] G. M. Clemence, Proc. Am. Phil. Soc. **93**, 532 (1949).
[3] R. S. Richardson, Publ. Astron. Soc. Pacific **61**, 162 (1949).

[4] *Note added in proof:*—Further study indicates that, for Icarus, $\alpha \gtrsim 14''$ (this revision is more favorable to the argument of the text).