ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E7-87-50

Yu.A.Lazarev, Yu.Ts.Oganessian,
I.V.Shirokovsky, S.P.Tretyakova, V.K.Utyonkov,
G.V.Buklanov

OBSERVATION OF DELAYED NUCLEAR FISSION IN THE REGION OF ^{180}Hg

Submitted to "Europhysics Letters"

1987

It is an indubitable fact that the most precise and detailed experimental information about the probability and mechanism of nuclear fission is that gained at low excitation energies comparable to, say, the nucleon binding energy. However, low-energy fission, such as neutron-induced and spontaneous fission, remained until recently the "privilege" of the actinide nuclei. Nowadays fission properties of the actinide nuclei are explored thoroughly enough, so that experimental results obtained just in the region of Ra to Fm form a general basis for understanding the fission mechanism, a source material for testing fission theories and models, and a starting ground for estimating fission properties of nuclei outside the well explored region. At the same time, the variations of Z and N are small within the traditional region of low-energy fission and thus the ground-state properties of the fissioning nuclei are fairly similar; the fission barriers here, although showing certain variations of the double-humped shape, have a nearly unchanged amplitude of about 5 to 6 MeV; and virtually the same set of the most probable fragment nuclei is formed in the exit channel of fission. Hence it is not surprising that in progressing from Ra to Fm low-energy fission properties of nuclei only rarely display appreciable qualitative changes. In broad outline, fission of the ordinary actinide nuclei seems quite monotonous. Therefore, along with increasingly precise multiparameter correlative measurements in the traditional region, of great urgency are experiments beyond its limits, i.e., with nuclei having a more unusual nucleon composition. Considerable variations in Z and N of the initial nucleus would make it possible to appreciably change the alignment of forces between collective and single-particle factors in fission, to elucidate in a more distinct way the role of intrinsic structure of the initial nucleus and that of nascent fragment nuclei, to alter the dynamical conditions under which fission proceeds, etc. -- eventually, to gain critically important material for a deeper understanding of the properties and mechanism of the fission process. Indeed, interesting results have been obtained in attempts to study spontaneous fission properties of the heaviest Fm isotopes and of nuclides beyond Fm (see, e.g., ref. /1/). However, in progressing into the transfermium region one encounters increasingly serious difficulties because of low yields and short lifetimes of the nuclei under study as well as of rather complicated background conditions. So far these studies have been extended only up to kurchatovium -- element 104. On the other hand, the extension of low-energy fission studies to the region of preactinide elements -- from Ra toward Pb -- is blocked by the absence of suitable targets with Z = 84-87 and, what is more important, by drastic losses in the fissility of nuclei resulting from a rapid augmentation of their fission barriers with decreasing Z.

The situation changes considerably in the case of nuclei far from β stability. The fissility of these nuclei may turn out to be sufficiently high in a much wider Z range since the macroscopic component of the fission barriers is expected to decrease substantially in moving farther from the line of β stability (see, e.g., ref. /2/). At the same time, the β-decay energies grow rapidly as one goes off the β-stability line. Then valuable possibilities for large-scale research into low-energy fission involving nuclei of many elements from the second half of the Mendeleev Periodic Table are offered by β-delayed fission, i.e., fission from excited states populated in $\beta^{\pm}$ decay or electron capture (EC). The phenomenon of β-delayed fission was discovered in 1965 by Flerov and coworkers /3/ at Dubna and subsequently has been studied in a number of other laboratories too (for a recent review, see ref. /1/). Although until now β-delayed fission has been observed only for the actinide nuclei, there are good reasons to believe /4/ that this phenomenon should also take place for nuclei of considerably lighter elements.

Beta-delayed fission is expected to occur with a detectable probability when the total β-decay energy of a precursor, $Q_{\beta(EC)}$, is comparable to the fission barrier amplitude of the daughter nucleus, B_f^{max}. Then a certain branch of β(EC) decay can lead to the population of rather high-lying states of the daughter nucleus, which possess large fission widths. Although fission from the states near the barrier top occurs "instantaneously" -- on a time scale of $\tau_f \sim 10^{-14}$-10^{-15} s, the population of the fissioning states takes place according to the half-life of the precursor, $T_{1/2} \gg \tau_f$, which just determines the time distribution of fission events (if they, of course, are recorded without coincidences with any marks of the preceding β(EC) decay). Thus, there appears a "delay" -- the factor

that essentially facilitates the performance of experimental studies. As regards the probability of β-delayed fission, $P_{\beta\text{-}df}$*), it is a very sensitive function of the difference ($B_f^{max} - Q_{\beta(EC)}$): the systematics of empirical $P_{\beta\text{-}df}$ values for subbarrier β-delayed fission of the actinide nuclei (see table 2 and fig.19 in ref. /1/) demonstrates that a 1 MeV increase in ($B_f^{max} - Q_{\beta(EC)}$) results in a factor of at least 10^2 decrease in $P_{\beta\text{-}df}$. This produces a due basis for obtaining information about the fission barrier from empirical data on the probability of β-delayed fission.

As follows from our previous examinations /4,1/, the appropriate conditions for β-delayed fission to occur can be realized in a wide range of neutron-deficient nuclei achievable with high yields in heavy ion reactions -- not only in the interesting region of near-magic precursors with Z = 87-91 and N < 126, such as, e.g., ^{208}Ac, ^{212}Pa or the lighter isotopes of Ac and Pa, but also in the region of <u>preactinide</u> elements, in particular, for the ultraneutron-deficient nuclei of Tl to Bi. Figure 1 shows an example of the case of Tl $\xrightarrow{\beta^+(EC)}$ Hg. We see that β-delayed fission of Hg isotopes becomes possible if one moves by 15-20 neutrons far off the β-stability line. Yet the actual A value at which β-delayed fission will come into play depends critically on the pattern of change in the macroscopic part $\tilde{B}_f$ of the fission barrier height with respect to the number of neutrons in the Hg nuclei. Therefore, as we have stressed previously /4,1/, β-delayed fission provides a unique probe for studying the Z,N variations of the fission-associated macroscopic properties of nuclei as well as for determining the fundamental parameters characterizing a drop of nuclear liquid, such as the fissility parameter x or the surface-asymmetry parameter k_s /6/ which governs the isospin dependence of the surface energy of a nuclear drop. Moreover, on condition of sufficiently high yields, β-delayed fission could furnish a unique chance also for more detailed studies of low-energy fission properties of exotic nuclei -- for measuring, e.g., the fragment mass and kinetic energy distributions, prompt neutron multiplicities, etc. It is noteworthy that, say, the ^{180}Hg nucleus

*) By definition, $P_{\beta\text{-}df}$ is the probability of fission of the daughter nucleus per one event of β(EC) decay of the precursor; $P_{\beta\text{-}df} = \sigma_{\beta\text{-}df}/(\sigma_\beta \cdot b_\beta)$, where $\sigma_{\beta\text{-}df}$ is the cross section corresponding to the measured yield of β-delayed fission events, whereas σ_β and b_β are, respectively, the production cross section and the β(EC) branch for the precursor.

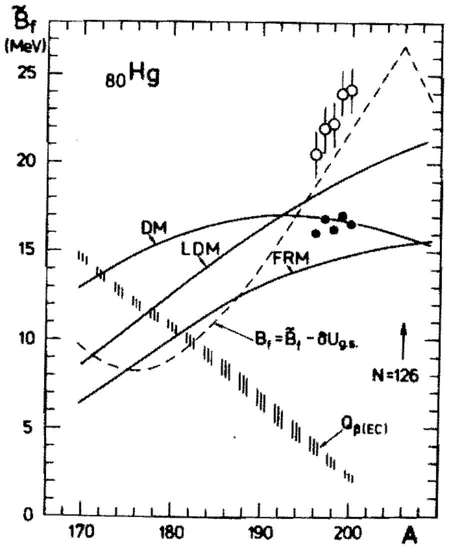

Fig. 1. The isospin dependence of the macroscopic part $\tilde{B}_f$ of the fission barrier heights for Hg isotopes, as predicted by the droplet model of Myers (DM) /5/, the liquid drop model of Myers and Swiatecki (LDM) /6/, and the finite range model of Sierk (FRM) /2/. The dashed curve shows the fission barriers $B_f = \tilde{B}_f - \delta U_{g.s.}$ composed of the $\tilde{B}_f$ values by Sierk /2/ and of the ground-state microscopic corrections $\delta U_{g.s.}$ by Möller and Nix /7/; note that around ^{180}Hg the $\delta U_{g.s.}$ corrections are small. The open circles represent the empirical fission barrier heights obtained in ref. /9/ from statistical-model analyses of the measured excitation functions for fission induced by light charged particles with $A \leqslant 4$; the closed circles show the macroscopic barrier heights $\tilde{B}_f$ obtained by removing the calculated $\delta U_{g.s.}$ corrections of Möller and Nix /7/ from the empirical B_f values. The triplets of vertical bars drawn for each even A outline, according to the mass tables /6-8/, the "corridor" of the $Q_{\beta^+(EC)}$ values for the odd-odd Tl isotopes; the bar heights thus reflect a scatter of the $Q_{\beta^+(EC)}$ predictions.

is far from the traditional region of low-energy fission by 10 units in Z and 40 units in N, whereas in terms of information about the fission probability and mechanism the study of β-delayed fission of this nucleus would be quite analogous to that of thermal-neutron-induced fission of ^{179}Hg.

In this letter we report on experiments which have led to the detection of β-delayed fission of nuclei in the region of ^{180}Hg.

For producing ultraneutron-deficient Pb, Tl, and Hg nuclei, isotopically enriched targets of ^{144}Sm (88.8%), ^{147}Sm (96.4%) and ^{150}Sm (95.0%) were irradiated by 230-MeV ^{40}Ca and ^{40}Ar beams provided by the U-400 cyclotron at JINR, Dubna. The U or Th contamination in the targets did not exceed $2 \cdot 10^{-8}$ g/g. The experiments were carried out by using the technique described in ref. /10/. The ^{40}Ca (or ^{40}Ar) beam with an average intensity of about $5 \cdot 10^{12}$ particles/s struck tangentially the lateral surface of a cooled copper cylinder onto which surface an about 2 mg/cm^2 layer of the target substance was deposited. This cylindrical target (serving simultaneously as a recoil catcher) rotated with a set constant velocity relative to the mica fission fragment detectors arranged around it. Earlier, this technique was widely used in experiments aimed at synthesizing transfermium elements (see, e.g., refs. /10,11/) and permitted the detection of spontaneously fissioning nuclei produced with cross sections as low as the tenth fractions of a picobarn (1 pb = 10^{-36} cm^2).

In the present exploratory experiments, searches were carried out for delayed fission activities with half-lives $T_{1/2} \gtrsim 0.1$ s. The relevant experimental data are presented in table 1 and fig. 2 from which it follows that the effect of a delayed fission has been detected in all the four reactions studied. The most striking fission activity, with $T_{1/2} = 0.70^{+0.12}_{-0.09}$ s, was observed in the ^{144}Sm + ^{40}Ca reaction (fig. 2). The measured yield of the 0.7-s activity conforms to a cross-section of some 50 pb (with an accuracy within a factor of 2-3). In the reactions on targets of ^{147}Sm and ^{150}Sm as well as in the ^{40}Ar-induced reaction the yields of delayed fission fragments decrease substantially, whereas the time distributions of the fission events recorded indicate a considerable increase in half-lives.

Evidently, the origin of any delayed fission activities should primarily be associated with nuclear decay processes occurring at the lowest excitation energies. Although at present we, strictly speaking, <u>cannot</u> exclude completely a possible manifestation of more exotic processes, the most probable source of the fission activities

Table 1

Delayed fission activities detected in the reactions of ^{40}Ca and ^{40}Ar projectiles with targets of ^{144}Sm, ^{147}Sm and ^{150}Sm

Reaction	I [a)]	t [b)]	N_f [c)]	$T_{1/2}$ [d)]	Y [e)]
$^{144}Sm + ^{40}Ca$	0.42	1.4	87	$0.9^{+\infty}_{-0.3}$	4.0
	0.46	4.9	93	$0.70^{+0.12}_{-0.09}$	4.7
$^{147}Sm + ^{40}Ca$	0.6	4.8	26	{ $0.3^{+0.7}_{-0.2}$	0.4
				{ >5	0.5
	0.7	19	26	-	0.7
	0.8	200	16	28^{+14}_{-8}	0.5
$^{150}Sm + ^{40}Ca$	0.7	5.1	23	-	0.6
	0.2	210	5	≳30	0.5
$^{144}Sm + ^{40}Ar$	0.5	2.2	6	≳1.4	0.3 [f)]

a) Beam dose, in units of 10^{18}.

b) Period of revolution of the target, in seconds.

c) Number of fission tracks recorded. It corresponds to the time interval of 0.06 t to 0.85 t, with t being the period of revolution of the target (note that there are no mica detectors around the zone in which the beam hits the target; see fig. 5 in ref. /10/).

d) Half-life conforming to fission tracks recorded. A dash in this column means that no decay is observed at the given speed of revolution of the target. For all reactions except for $^{144}Sm + ^{40}Ca$, the indicated $T_{1/2}$ values should be considered to be effective ones since the recorded tracks may originate from complex rather than single fission activities.

e) Relative yield (number of delayed fission events per beam particle), in units of 10^{-16}. The total detection efficiency is taken into account in the determination of Y.

f) In this reaction, the relative yield Y of the 0.7-s fission activity does not exceed 0.1.

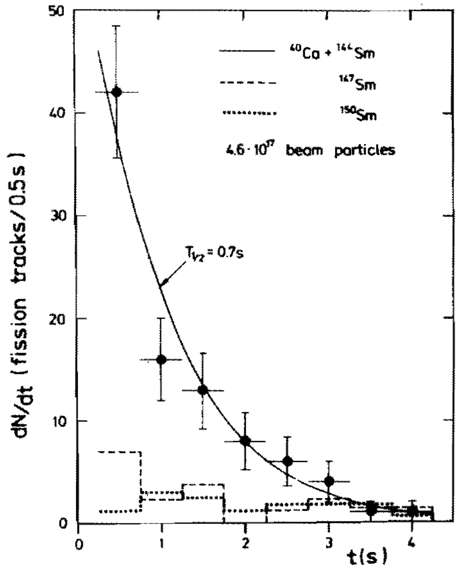

Fig. 2. The time distributions of the delayed fission events detected in the reactions of ^{40}Ca projectiles with targets of ^{144}Sm, ^{147}Sm and ^{150}Sm. For the reactions on ^{147}Sm and ^{150}Sm, the distributions (histograms) are shown after renormalization to the beam dose of $4.6\cdot10^{17}$ for which the $^{144}Sm + ^{40}Ca$ data are presented. Note the linear scale of ordinates.

observed seems to be the β-delayed fission. It is this phenomenon that has been predicted in refs. /4,1/ for the ultraneutron-deficient nuclei of Bi-Tl. If that is the case, an examination of the data of table 1 in the light of the radioactive properties of the residual nuclei formed after particle emission from the compound system ^{184}Pb with the initial excitation energy $E^* \approx 40-75$ MeV leads to the assumption that fission with $T_{1/2}$ = 0.7 s occurs in the decay chain $^{180}Tl \xrightarrow{\beta^+(EC)} {}^{180}Hg^*$. In fact, the radioactive properties of the majority of nuclei from the region of $A \sim 174-184$ and $Z \leq 82$ are known well, whereas the properties of unknown nuclides can be estimated reliably by a short extrapolation of the empirical data. Again, the value of $T_{1/2} = 0.70^{+0.12}_{-0.09}$ s determined rather accurately restricts sharply the range of possible precursors of β-delayed fission. Thus Pb isotopes with $A < 182$ should undergo predominantly α decay with half-lives that are expected to be noticeably shorter than 0.7 s. All the neutron-deficient Hg isotopes with $A \geq 175$ as well as Au isotopes with $A \geq 173$ are presently known well (see ref. /12/ and references therein); none of them can serve as a precursor of β-delayed fission with $T_{1/2}$ = 0.7 s. As regards Tl, the lightest known thallium isotope is ^{179}Tl with $T_{1/2} = 0.16^{+0.09}_{-0.04}$ s, which has been observed by detecting its α decay /12/, whereas the odd-odd isotopes ^{184}Tl and ^{186}Tl are known to undergo β^+(EC) decay with $T_{1/2}$ = 11 s and 26 s, respectively. The Tl isotopes with A = 180-183 have not so far been observed (only short-lived isomeric states in ^{181}Tl and ^{183}Tl are known /12/). At the same time, from the above-mentioned data it is clear that the half-life of ^{180}Tl should be close to 1 s, with comparable branches for α and β^+(EC) decay; Takahashi et al. /13/ predict $T_{1/2\beta^+(EC)} \approx 2$ s for this nucleus. Therefore ^{180}Tl is expected to be the most probable precursor of the β-delayed fission with $T_{1/2}$ = 0.7 s. Finally, the fission activities detected in the $^{147}Sm + {}^{40}Ca$ and $^{150}Sm + {}^{40}Ca$ reactions seem to be due to β-delayed fission in the isobaric chains $Tl \xrightarrow{\beta^+(EC)} Hg$ and/or $Pb \xrightarrow{\beta^+(EC)} Tl$ with larger A values (more probably even ones); appropriate chains can also be indicated in the case of the $^{144}Sm + {}^{40}Ar$ reaction. In fact, as A increases, the $Q_{\beta^+(EC)}$ values decrease, the half-lives become longer, and the difference $(B_f^{max} - Q_{\beta^+(EC)})$ grows steeply, see fig. 1. On the other hand, a move toward less neutron-deficient nuclei is accompanied by a considerable increase in their production cross sections. In addition, branching ratios for ground-state α decay as well as those for β-delayed proton and α-particle emission decrease sharply in shifting toward the valley of β-stability. As a net result of concurrent but opposite actions of the two above-mentioned groups of factors, the effect yielded on targets of ^{147}Sm and ^{150}Sm or with an ^{40}Ar beam diminishes, compared to the $^{144}Sm + {}^{40}Ca$ case, by one order of magnitude or more, but still it remains at the level of cross sections of the order of several pb.

Thus, all the experimental data presented in table 1 and in fig.2 can naturally be explained in terms of β-delayed fission.

In bombarding ^{144}Sm by ^{40}Ca ions, the isotope ^{180}Tl is formed via the p3n evaporation channel which is expected to have a cross section of the order of 0.1 - 1 mb. Then the probability of β-delayed fission in the $^{180}Tl \xrightarrow{\beta^+(EC)} {}^{180}Hg$ chain can be estimated to be $P_{\beta\text{-}df} \sim 10^{-6}$, with an uncertainty of one or one and a half order of magnitude. For a more accurate determination of $P_{\beta\text{-}df}$, experimental data are needed on the excitation function of the $^{144}Sm + {}^{40}Ca$ reaction as well as on the β^+(EC) branch for ^{180}Tl. Postponing a quantitative analysis of $P_{\beta\text{-}df}$ until the necessary data have been obtained, here we will discuss qualitatively what $P_{\beta\text{-}df} \sim 10^{-6}$ means in the context of information about the fission barrier height of the ultraneutron-deficient cold nucleus ^{180}Hg. First of all, the comparatively low value of $P_{\beta\text{-}df}$ indicates that the β-delayed fission observed is a subbarrier process. Yet this process appears to be

only slightly subbarrier one. In fact, using,as a rough guide, the systematics of empirical $P_{\beta\text{-}df}$ values for subbarrier β-delayed fission of the actinide nuclei (see fig. 19 and table 2 in ref. /1/) one can conclude that the fission barrier height for ^{180}Hg is unlikely to exceed by more than 0.5-1 MeV the $Q_{\beta^+(EC)}$ value for ^{180}Tl, which is predicted to be 10.5-11 MeV /6-8/. One should also bear in mind that, in contrast to the actinides, a much steeper dependence of $P_{\beta\text{-}df}$ upon $(B_f^{max} - Q_{\beta^+(EC)})$ is expected for the mercury region. First, for the Hg isotopes fission barriers are anticipated to be appreciably "broader" than, say, for the Cm-Cf isotopes for which, furthermore, the $P_{\beta\text{-}df}$ values seem to be determined by the penetrability of only one (inner) peak of the double-humped barrier. Second, as was noted in ref. /4/, in the vicinity of the proton drip line, where the $Q_{\beta^+(EC)}$ values are rather large, β-delayed proton and/or α-particle emission may compete with β-delayed fission; other things being equal, a contribution from the partial widths Γ_p and/or Γ_α to the total decay width Γ_{tot} can reduce the fission probability Γ_f/Γ_{tot}. Bearing in mind all the above-mentioned points, we believe that a reasonable estimate for the fission barrier amplitude of the cold nucleus ^{180}Hg is given by the value $B_f^{max} \approx 11$-12 MeV.

Returning now to fig. 1, one can confront this B_f^{max} estimate with theoretical predictions for the macroscopic fission barrier heights $\tilde{B}_f$ of the ultraneutron-deficient Hg isotopes. As is clear from fig.1, the very fact of occurrence, with a detectable probability, of β-delayed fission of ^{180}Hg makes it possible to conclude that the droplet model of Myers /5/ fails to properly predict both the magnitude and the isospin dependence of $\tilde{B}_f$. As regards the liquid drop model of Myers and Swiatecki /6/ and the finite range model of Sierk /2/, their predictions do not contradict our empirical data for ^{180}Hg; however, more definitive conclusions about the degree of adequacy of these versions of the macroscopic theory will become possible only after more quantitative information about the fission barrier height of ^{180}Hg and of nearby ultraneutron-deficient Hg nuclei has been extracted from supplemented data on the probability of β-delayed fission. Note that for extracting such information it is necessary to know the beta strength function $S_\beta(E)$ /14,15/ controlling the population probability of levels of the daughter nucleus in the excitation energy range (E,E + dE) and, in addition, to have certain (a priori) notions about the landscape of the potential energy surface associated with fission of the nuclei under study. The problem of quantitative treatment of empirical data on the probability of β-delayed fission has been discussed at length in papers [4,1] according to which subsequent detailed analyses of our data will be performed.

In conclusion, the detection of β-delayed fission in the region of ^{180}Hg signifies that low-energy fission is not a privilege of the actinide species. Judging from our observations in the Hg region, the ultraneutron-deficient nuclei of many other elements from the second half of the Mendeleev Periodic Table should also undergo β-delayed fission. Proven by the present experiments, this really widespread occurrence of β-delayed fission opens up essentially new and wide prospects for studying the probability and mechanism of low-energy fission at considerable variations of Z, N and N/Z of the fissioning nucleus.

* * *

We take pleasure in thanking Academician G.N.Flerov for his encouraging attitude towards this work. Our debt concerning this work is owed to the late Anatoly A. Pleve, who made a valuable contribution to the development of the highly stable, isotopically enriched samarium targets. We wish to thank also B.N.Gikal, V.B.Kutner and the U-400 cyclotron staff for providing these experiments with the intense beams of calcium and argon ions.

References

1. Oganessian Yu.Ts., Lazarev Yu.A. - In: Treatise on Heavy Ion Science (ed. D.A.Bromley). New York: Plenum Press, 1985, v.4, pp. 1-251.
2. Sierk A.J. - Phys.Rev. 1986, v.C33, p.2039.
3. Kuznetsov V.I., Skobelev N.K., Flerov G.N. - Yad. Fiz. 1966, v.4, p.99, p.279; Yad. Fiz. 1967, v.5, p.271.
4. Lazarev Yu.A., Oganessian Yu.Ts., Kuznetsov V.I. Paper presented at the Int. IUPAC Symp. on the Synthesis and Properties of New Elements (Dubna, September 23-27, 1980); JINR Preprint E7-80--719, Dubna, 1980.
5. Myers W.D. Droplet Model of Atomic Nuclei. IFI/Plenum, New York, 1977.
6. Myers W.D., Swiatecki W.J. - Nucl. Phys. 1966, v.81, p.1; Report UCRL-11980, Berkeley, 1965.
7. Möller P., Nix J.R. - Nucl. Phys. 1981, v.A361, p.117; At. Data Nucl. Data Tables, 1981, v.26, p.165.

8. Liran S., Zeldes N. - At. Data Nucl. Data Tables, 1976, v.17, p.411.
9. Ignatyuk A.V. e. a. - Fiz. Elem. Chastits At. Yadra, 1985, v.16, p.709.
10. Oganessian Yu.Ts. - In: Proc. Int. School-Seminar on Heavy Ion Physics (Alushta, USSR, April 14-23, 1983), D7-83-644. Dubna: JINR, 1983, p.55.
11. Oganessian Yu.Ts. e. a. - Radiochimica Acta, 1984, v.37, p.113.
12. Schneider J.R.H. e.a. - Z. Phys. 1983, v.A312, p.21.
13. Takahashi K., Yamada M., Kondoh T. - At. Data Nucl. Data Tables, 1973, v.12, p.101.
14. Klapdor H.-V. - Prog. Part. Nucl. Phys. 1983, v.10, p.131.
15. Naumov Yu.V., Bykov A.A., Isosimov I.N. - Fiz. Elem. Chastits At. Yadra, 1983, v.14, p.420.

Received by Publishing Department on January 30, 1987.