NEGATIVE MUON SPIN ROTATION EXPERIMENT[†]

T. Yamazaki, K. Nagamine, S. Nagamiya and O. Hashimoto
Department of Physics, University of Tokyo, Bunkyo-Ku, Tokyo, Japan and
Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720, U.S.A.

ABSTRACT

Results of the experimental project using negative muon spin rotation at Lawrence Berkeley Laboratory are reported. The following have been discussed briefly. i) The bound muon g factors from the viewpoints of the relativistic and nuclear polarization effects. ii) Strange depolarization phenomena in terms of the difference between the muonic-atom probe and its equivalent nuclear probe iii) Application of μ^-0 probe to studies of magnetic oxides. iv) Total muon capture rates in the actinide region.

INTRODUCTION

In this paper we will present results of the negative muon spin rotation project which has been running for the past two years at the 184 inch Cyclotron at the Lawrence Berkeley Laboratory.[1] The principle of the muon spin rotation (μSR) is to determine the muonic Larmor frequency and the relaxation time by observing μ - e decay asymmetry from polarized muons time-differential wise. This is quite an interdisplinary project spanning over meson physics, nuclear physics and atom/solid-state physics. There are two kinds of muon spin rotation, namely, μ^+SR and μ^-SR. Table I summarizes the characteristic features of both methods.

TABLE I

Characteristic features of μ^+SR and μ^-SR

	μ^+	μ^-
Spin	1/2	1/2, when bound to I = 0 nucleus
g factor	2.002 in μ_μ	a little smaller
Polarization	100%	~18%
Asymmetry	$1 + 0.33 \cos\theta$	$1 + 0.06 \cos\theta$
Lifetime	2.2 μsec	2.2 μsec ~80 nsec with Z
Electron yield	100%	100% ~4% with Z
Location	interstitial	at nucleus
Size	diffusing	$r_\mu \sim \frac{260}{Z}$ fm
Character	light proton	pseudo nucleus of (Z - 1)e

[†] Supported by Japan Society for the Promotion of Science, the National Science Foundation and the U.S. ERDA.

The positive muon is characterized as a "light proton" and its application to solid state studies is getting popular by now, and we have submitted some results to this conference.[2] In μ^-SR we use the bound muon (namely, the ground state muonic atom) as a probe. There were a series of pioneer work on μ^-SR during the years of 1958-62 at Columbia,[3] Chicago and Dunba,[4] but since then its importance has almost been forgotten. It is rather surprising that even the bound muon g factors in heavy nuclei had not been measured until we started this project. In this paper we will restrict ourselves to μ^-SR.

The negative muon ($1s_{1/2}$) bound to a spinless nucleus of charge Ze looks like a pseudo nucleus of charge (Z - 1)e, if seen from outer atomic electrons, but the charge and magnetization distributions are different (see Fig. 1). Three questions may be raised. i) How different is the bound-muon g factor from the free muon g factor? ii) How different are the muonic atom and its equivalent nucleus? iii) Are applications to solid state studies promising?

The experimental apparatus we employed is shown in Fig. 2. The time difference between the stopped muon and the decay electron was measured by a digital clock counter (HP5360A) to a precision of 0.1 nsec and recorded by a PDP-15 computer. A detailed discription is given in ref. 1) and ref. 5).

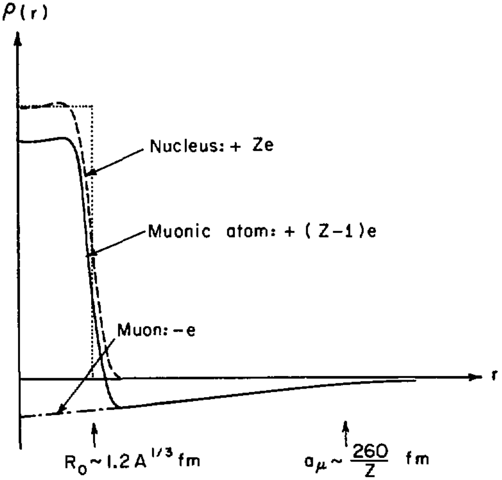

Fig. 1. Charge distribution of the muonic atom μ^-Z, as compared with that of an equivalent nucleus of charge (Z - 1)e.

BOUND-MUON g FACTOR

Already in 1928 Breit[6] predicted that the electron in a K shell orbit must possess a g factor different from the free-electron value ($g_{free} = -2$) due to its relativistic motion. Breit gave a correction $\delta g/g \cong -(\alpha Z)^2/3$. While this effect was not examined in ordinary atoms because of the lack of techniques to measure the g factors of deeply

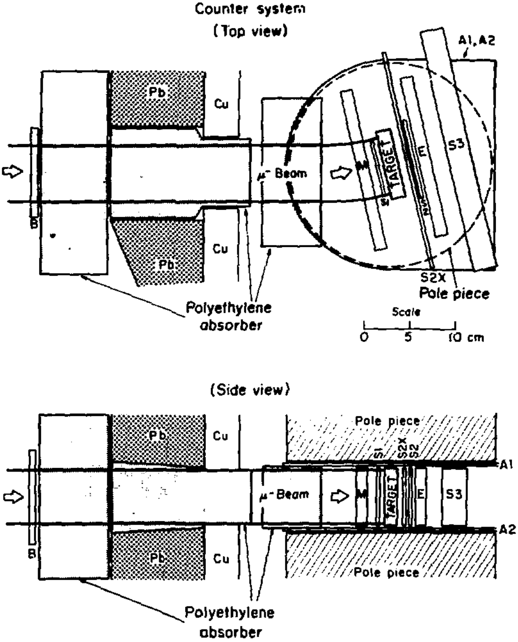

Fig. 2. Experimental setup for μ^-SR at the 184" cyclotron of Lawrence Berkeley Laboratory. All the counters were plastic scintillators connected to RCA 8575 phototubes through photoguides. These counters were placed between two pole pieces of the 9-inch Varian magnet with its gap of 7.6 cm. The A1 and A2 counters (3 mm thick) were fixed to the surfaces of the pole pieces in order to eliminate the background from the iron cores. The fast logic was as follows:

$$\text{“stopped } \mu\text{”} = B\cdot M\cdot S1\cdot\overline{(S2X + A1 + A2)},$$

$$\text{“decay e”} = S2X\cdot S2\cdot E\cdot S3\cdot\overline{(B + M + S1 + A1 + A2)}$$

bound electrons, it is straightforward to study this effect in the ground state of muonic atom which consists of one muon in the $1s_{1/2}$ state and a zero-spin nucleus. In the muonic case the finite nuclear size has to be taken into account, as calculated by Ford et al.[7] The first observation of the Breit effect was reported by Hutchinson et al.[3] who determined the g factors of the bound muons in light elements up to Z = 16. We extended such measurements to heavier nuclei, where greater effects are expected, but it became exceedingly difficult to determine the g factors precisely because of the shorter lifetimes and the smaller yields of decay electrons. The results reported by

Yamazaki et al.[5] are summarized in Fig. 3, which shows presence of quite large correction in the g factors of muons bound to heavy nuclei in accordance with the finite size calculation of Ford et al., but the precision was not good enough to discuss any indication of nuclear polarization.

In principle the bound muon can polarize the nucleus from its ground state ($I = 0^+$) to magnetic dipole states ($I = 1^+$) through the electromagnetic interaction (muon-nucleus hyperfine coupling), which gives rise to a small correction to the g factor. The essentially same effect is seen in nuclear magnetic moments in a much more pronounced way (so called core polarization), but this effect has never been separated from other possible effects. A large inherent problem is that the bare M1 operator for one nucleon in the nucleus is by no means the same as the one for the free nucleon because of the mesonic exchange current, as shown experimentally by the Tokyo group.[8] Therefore, the bound-muon g factor will be a quite unique probe with no ambiguity in deducing

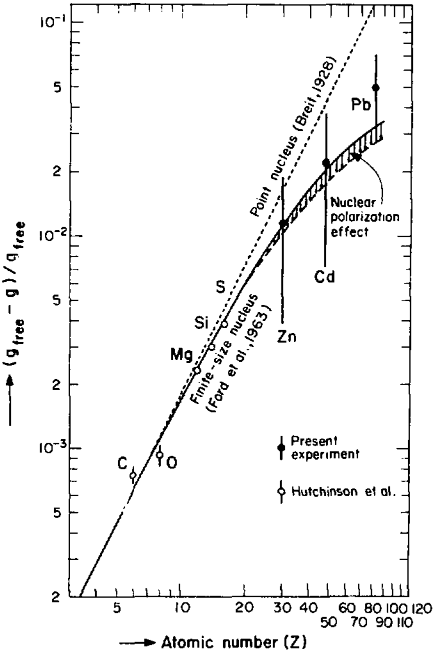

Fig. 3. Relativistic effect (or binding effect) on the g factor of bound muons. The dotted curve shows the relativistic correction for a point nucleus.[6] The solid curve shows the calculated values by Ford et al.[7] where the finite size effect of the nucleus is taken into account. The difference between the solid and broken curves is the nuclear polarization effect calculated by Ford et al. The experimental data after the corrections for the Knight shift and diamagnetism are plotted.

the magnetic core polarizability of the nucleus. At this moment we can only say that high precision measurements will be possible in the near future and that precise determination should depend on further understanding of the difference between the bound muon probe and the equivalent nucleus in terms of the solid-state effect.

STRANGE DEPOLARIZATION OF NEGATIVE MUONS

The ground-state muonic atom in comparison with the impurity nucleus of charge Z - 1 is expected to show a substantial hyperfine anomaly because its charge and magnetization distributions are entirely different from those for the corresponding nucleus.[9] We can define the following anomaly factors both for the static hf frequency $\Delta\omega$ and for the dynamic relaxation time T_1,

$$F_1 \equiv (\Delta\omega)_\mu/(\Delta\omega)_{\text{nucleus}}, \quad F_2 \equiv (g^2T_1)_{\text{nucleus}}/(g^2T_1)_\mu \qquad (1)$$

The Dubna group[4] reported depolarization of μ^- in Mo, Pd and other paramagnetic metals but this is rather surprising in view of the known NMR data. We found that the Mo metal shows no depolarization (see Fig. 4) in contrast to the Dubna result. We

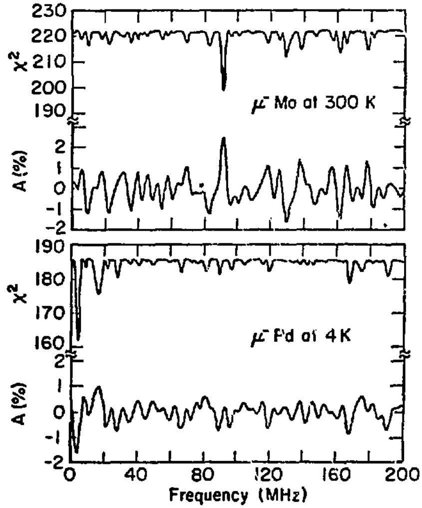

Fig. 4. Chi-squares and precession amplitudes versus Larmor frequency which are obtained by fitting an experimental time distribution to

$$N(t) = N_0 \exp(-t/\tau)\,[1 - A\cos(2\pi f t + \phi)]$$

after the subtraction of the long-lived component and background. Upper: μ^-Mo at 300 K. Lower: μ^-Pd at 4 K. In μ^-Mo a free precession signal is observed, while in μ^-Pd no distinct signal is seen.

studied the Pd case very carefully at various temperatures, using a very pure metal including only 5 ppm Fe.[10] Such a study was hoped to be promising in obtaining F_1 and F_2, as large negative Knight shifts are observed in the corresponding nuclear case, Rh in Pd.[11,12] We expect T_1 for μ^-Pd at 4K to be 16 μsec, which is much longer than the μ^- lifetime of 0.1 μsec, and the Knight shift at 4K to be -0.16[11,12] under the assumption of no hyperfine anomaly ($F_1 = F_2 = 1$).

We observed no significant precession signal at any temperatures (4-300 K), as shown in Fig. 4. This surprising fact indicates either of the following two cases.

i) If this depolarization takes place at the ground-state muonic atom, the relaxation time T_1 should be less than 50 nsec, corresponding to a surprisingly large hyperfine anomaly: $F_2 \gtrsim 300$! This would mean that the hyperfine field felt by the muonic atom is 20 times larger than that felt by the nucleus.

ii) This depolarization may take place before the muon reaches the ground state, i.e., during the slowing down, capture state or muonic atom cascade, where the muon traverses through electrons of high spin densities. This is entriely unknown region both in space and time.

One of the adequate approaches to this problem is to see whether this phenomena is associated with the macroscopic observables such as atomic magnetic moments. The Dubna group observed recovery of the polarization in $Pd_{0.6}H_{0.4}$ metal where the paramagnetism of Pd is completely supressed by hydrogen.[4] Similarly, we can artificially reduce the magnetic moment of Ni by adding the impurity like Cr. We have studied the concentration dependence of the residual muon polarization for paramagnetic Ni and NiCr alloy. No precession signal was observed in paramagnetic Ni at 700 K and 900 K with an external field of 9.8 kG. However, we observed appreciable precession signal in NiCr alloy at room temperature as the Cr concentration increased toward the critical concentration (12%) where the magnetic moment of the alloy ultimately becomes zero. The disappearance of the precession signal takes place for NiCr alloy of 10% Cr concentration, which has an atomic magnetic moment of only 0.1 μ_B.

Further experiments to pin down these questions are being planned.

APPLICATION TO STUDIES OF MAGNETIC OXIDES

The negative-muon spin rotation method has been applied to observe the hyperfine field at μ^-O in paramagnetic MnO at room temperature.[13] This is the first case, where the μ^-SR method is applied to studies of magnetic oxides. The oxygen atom in this magnetic oxide plays an important role in the superexchange interaction which gives rise to antiferromagnetic ordering of Mn^{2+} d-electrons below $T_N = 116$ K. To study local fields at oxygen sites in such an oxide, however, conventional NMR method is hardly applicable, not only because of the quite low natural abundance of ^{17}O (0.04%) but also because of a large quadrupole broadening at non-cubic site, while the μ^-SR method is promising and powerful. Since μ^-O has a lifetime of 1.8 μsec, which is much longer than the lifetimes (100-200 nsec) of μ^- trapped by heavier elements, the muon signal from oxygen can be selectively separated. Furthermore, the μ^-SR method may reveal new phenomena, since the μ^-O probe behaves as a nitrogen-like impurity but with rather broader magnetization distribution as compared with the nitrogen nucleus.

The time spectrum of decay electrons from stopped muons in MnO, as shown in Fig. 5, involves two decay components with mean lifetimes of 232 nsec (μ^-Mn) and 1.84 μsec (μ^-O). The frequency spectrum of the latter component at 6.830 kOe external field is shown in Fig. 5(b) together with that for a carbon target, which reveals a paramagnetic shift of μ^-O in MnO. The relaxation time T_2 has also been obtained. All these results are presented in Table II and compared with the ^{17}O NMR data.[14]

The observed shift $\Delta(\mu^-O)$ is about 1/3 of $\Delta(^{17}O)$, which cannot be accounted for solely by the fact that the 2s electron density at μ^-O is decreased by a factor of 0.6. The present result indicates that i) the presence of μ^- at O increases the local covalency

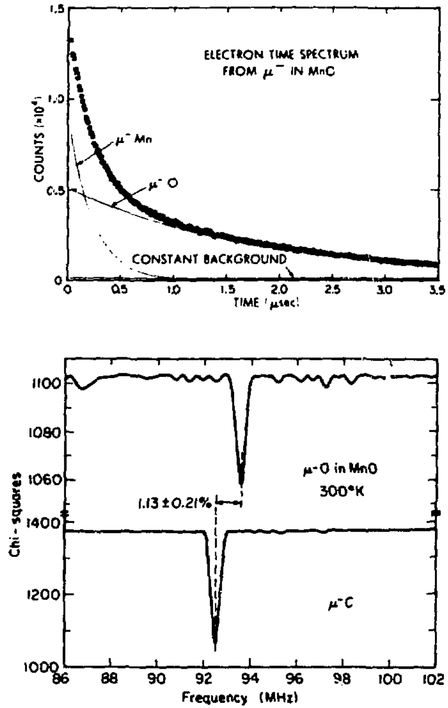

Fig. 5. Upper: Time spectrum of decay electrons from the muons stopped in a MnO target. The long-lived component comes from μ^-O and the short-lived one from μ^-Mn.
Lower: The χ^2 versus the Larmor frequency for the μ^-O component in paramagnetic MnO at room temperature and at H = 6.830 kOe (upper) and for a carbon target under the same experimental conditions (lower), showing a paramagnetic shift of 1.16 ± 0.21% after the correction for the g-factor difference between μ^-O and μ^-C.

and thus enhances the local superexchange interaction, which gives rise to reduction of the polarization of the neighbouring Mn^{2+} d spins and that ii) the local exchange frequency is also increased so as to reduce the relaxation rate.

Let us emphasize that the negative muon is not a passive probe but an active probe in the sense that it changes the local structure of solid states and detects such a change by itself. This is a perfectly dilute solid state system which cannot be accessible by conventional means.

TABLE II

Summary of the paramagnetic shift Δ and relaxation time T_2 observed at room temperature by different methods for paramagnetic MnO. The relaxation times have been obtained from extrapolation of experimental data to zero external magnetic field.

Probe	g-Factor (μ_N)	Δ (%)	T_2 (μsec)	g^2T_2 ($10^2\mu$sec)	$\Delta^2g^2T_2$ ($10^{-2}\mu$sec)	Ref.
μ^-O	17.76	1.16 ± 0.21	$1.5^{+0.8}_{-0.4}$	$4.7^{+2.5}_{-1.3}$	$6.4^{+3.7}_{-1.5}$	present
^{17}O	-0.7575	3.21 ± 0.02	74 ± 26	0.42 ± 0.15	4.4 ± 1.6	14

MUON CAPTURE RATES IN ACTINIDE NUCLEI

As a byproduct of the μSR experiments we studied the total muon capture rates in the actinide nuclei.[15] So far several lifetime measurements of bound muons have been reported.[16-20] They were carried out mostly by observing fission fragments after muon capture, except in ^{238}U where the lifetime was also determined through the detection of electrons emitted from muons. In order to study the systematics of muon lifetimes in this actinide region in more detail we have measured lifetimes of muons bound to ^{232}Th, ^{235}U, and ^{239}Pu through the detection of electrons. Another goal of this experiment is to look at any difference between τ_e (lifetime for the electron decay mode) and τ_f (lifetime for the fission mode), since recently Bloom[21] pointed out that there could be a difference between τ_e and τ_f for ^{238}U due to the presence of a fission isomer ($\tau \sim 200$ nsec) in ^{238}U which might be fed by the radiationless muonic 2p-1s transition.

The experimental results are shown in Table III and compared with measurements through fission. The present value (81.5 ± 3.0 nsec) of $\tau_e(^{238}\text{U})$ is shorter than that reported by Sens,[20] but still a little longer than τ_f (= 76.8 ± 0.8 nsec). The values of τ_f for ^{232}Th, ^{235}U and ^{239}Pu are scattered too much to be compared with the present τ_e values.

The muon capture rate, Λ_c, derived from muon lifetime ($\Lambda_c = 1/\tau - 1/\tau_{free}$ where τ_{free} is the lifetime of a free muon), is expressed within the framework of Primakoff's gross theory[22] by

$$\Lambda_c(A,Z) = \gamma K^2 \times <\rho> [1 - \delta(A - Z)/2A] \qquad (2)$$

where δ is the nuclear correlation parameter, $<\rho>$ is the overlap of the muon and nuclear charge, and γK^2 is a constant. Reduced capture rates, $\Lambda_c/<\rho>$, versus $(A - Z)/2A$ are plotted in Fig. 6. The straight line corresponds to $\gamma K^2 = 83.29 \times 10^{-31}$ cm^{3}s^{-1} and $\delta = 3.11$ which have been obtained by Fillippas et al.[23] from the best fits for relatively high-Z elements from Cu to Pb [$(A - Z)/2A$ ranged from 0.28 to 0.30]. Roughly speaking, our data fall on this straight line. However, on the other hand, it seems that, in actinide nuclei, the reduced capture rates for the electronic decay mode are less dependent on $(A - Z)/2A$ than in lighter nuclei. It is obvious that the above formula (2) may not hold well at around $(A - Z)/2A = 1/\delta \sim 0.32$ at

TABLE III

Lifetimes and reduced capture rates of muons bound to actinide nuclei

Element	(A - Z)/2A	Mode[a)]	Lifetime (nsec)	$\Lambda_{cap}/<\rho>$[b)] ($sec^{-1} fm^3 10^7$)	Reference
^{232}Th	0.3060	f	74.2 ± 5.6	4.60 ± 0.37	16
		f	87 ± 4	3.90 ± 0.18	17
		e	80.4 ± 2.5	4.23 ± 0.13	present result
^{233}U	0.3026	f	61.7 ± 3.8	5.56 ± 0.34	17
^{235}U	0.3043	f	65.3 ± 2.8	5.25 ± 0.22	17
		f	66.5 ± 4.2	5.15 ± 0.33	16
		f	84 ± 6	4.04 ± 0.29	18
		e	78 ± 5	4.36 ± 0.28	present result
^{238}U	0.3067	f	75.6 ± 2.9	4.51 ± 0.17	16
		f	74.1 ± 2.8	4.59 ± 0.17	17
		f	76.0 ± 1.0	4.48 ± 0.06	18
		e	88 ± 4	3.85 ± 0.18	20
		e	81.5 ± 3.0	4.17 ± 0.15	present result
^{239}Pu	0.3033	f	74 ± 14	4.61 ± 0.87	19
		e	77.5 ± 2.0	4.39 ± 0.12	present result

[a)] The detection of fission fragments and decay electrons is denoted by f and e respectively.

[b)] Λ_{cap} defined by $\Lambda_{cap} \equiv 1/\tau - 1/\tau_{free}$.

which the theoretical capture rate given by Eq. (2) becomes zero. Therefore, as the value of (A - Z)/2A approaches closer to 0.32, the effective value of δ should become smaller. Our observations suggest that $\Lambda_c(A,Z)$ may well be expressed by including an additional higher-order term $[(A - Z)/2A]^2$.

If we ascribe the difference of capture rate, $\omega \equiv 1/\tau_f - 1/\tau_e = (0.9 \pm 0.5) \times 10^6$ sec^{-1} for ^{238}U, to Bloom's mechanism, it would indicate that the radiationless excitation, by the 2p → 1s transition feeds the fission isomer with a branch of almost 100%, which, however, seems to be unrealistic.

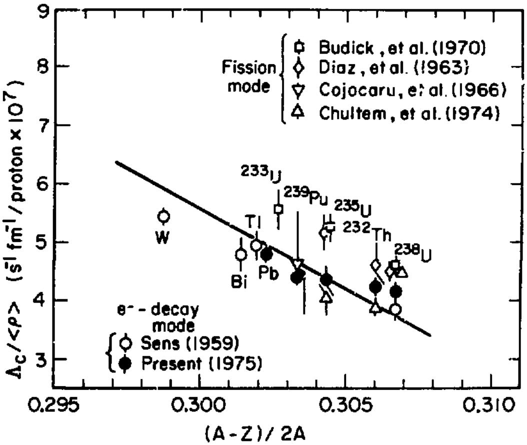

Fig. 6. Primakoff plot of muon capture rates in actinide region. The black circles are the present results. The lifetime of μ^-Pb has also been determined to be 77.5 ± 2.0 ns, and it is involved in this figure.

CONCLUDING REMARKS

We have described briefly what we have done and what we can learn from μSR experiments. We have found that there are many interesting problems, which still remain unsolved. We believe that both μ^+SR and μ^-SR will create new fields of physics in the coming meson factory era.

We would like to thank Professors O. Chamberlain and K. M. Crowe for their hospitality and collaboration which have made it possible for the University of Tokyo group to work at the 184 inch Cyclotron of the Lawrence Berkeley Laboratory. We appreciate excellent operation of the 184 inch Cyclotron by her crew. We are grateful to Professors K. Sugimoto, K. Nakai and S. Kobayashi for their collaborations at the earlier stage.

REFERENCES

[1] The principle and experimental procedures together with preliminary results have been described in the following: T. Yamazaki, K. Nagamine, S. Nagamiya, O. Hashimoto, K. Nakai and S. Kobayashi, Physica Scripta (in press).

[2] K. Nagamine, S. Nagamiya, O. Hashimoto, N. Nishida dn T. Yamazaki, Contributed paper II.18; K. Nagamine, S. Nagamiya, O. Hashimoto, T. Yamazaki and B. D. Patterson, ibid II.19.

[3] D. P. Hutchinson, J. Menes, G. Shapiro, A. M. Patlach and S. Penman, Phys. Rev. Lett. 7, 129 (1961); D. P. Hutchinson, J. Menes, G. Shapio and A. M. Patlach, Phys. Rev. 131, 1362 (1963).

[4] A. E. Ignatenko, Nucl. Phys. 23, 75 (1961).

[5] T. Yamazaki, S. Nagamiya, O. Hashimoto, K. Nagamine, K. Nakai, K. Sugimoto and K. M. Crowe, Phys. Lett. 53B, 117 (1974).

[6] G. Breit, Nature 122, 649 (1928).

[7] K. W. Ford, V. W. Hughes and J. G. Wills, Phys. Rev. Lett. 7, 134 (1961); Phys. Rev. 129, 194 (1963).

[8] T. Yamazaki, T. Nomura, S. Nagamiya and T. Katou, Phys. Rev. Lett. 25, 547 (1970); S. Nagamiya and T. Yamazaki, Phys. Rev. C4, 1961 (1971); T. Yamazaki, J. Phys. Soc. Japan Suppl. 34, 17 (1973).

[9] A. Bohr and V. F. Weisskopf, Phys. Rev. 77, 94 (1950). H. H. Stroke, R. J. Blin-Stoyle and V. Jaccarino, Phys. Rev. 123, 1326 (1961); H. J. Rosenberg and H. H. Stroke, Phys. Rev. A5, 1992 (1972).

[10] K. Nagamine, S. Nagamiya, O. Hashimoto, S. Kobayashi and T. Yamazaki, Contributed paper II.17.

[11] G. N. Rao, E. Matthias and D. A. Shirley, Phys. Rev. 184, 325 (1969).

[12] A. Narath and H. T. Weaver, Phys. Rev. B3, 616 (1971).

[13] S. Nagamiya, K. Nagamine, O. Hashimoto and T. Yamazaki, contributed paper II.15; to be published in Phys. Rev. Lett. (1975).

[14] D. E. O'Reilly and T. Tsang, J. Chem. Phys. 40, 734 (1974).

[15] O. Hashimoto, S. Nagamiya, K. Nagamine and T. Yamazaki, contributed paper II.16.

[16] J. A. Diaz, S. N. Kaplan and R. V. Pyle, Nucl. Phys. 40, 54 (1963).

[17] B. Budick, S. C. Cheng, E. R. Macagno, A. M. Rushton and C. S. Wu, Phys. Rev. Lett. 24, 604 (1970).

[18] D. Chultem, V. Cojocaru, Dz. Ganzorig, Kim Si Chwan, T. Krogulski, W. D. Kuzniecow, H. G. Orthepp, S. M. Polikanov, B. M. Sabirov, U. Schmidt and W. Wagner, Nucl. Phys., to be published.

[19] V. Cojocaru, L. Marinescu, M. Petrascu, G. Voiculescu, A. Ignatenko and M. Omelianenko, Phys. Lett. 20, 53 (1966).

[20] J. C. Sens, Phys. Rev. 113, 679 (1959).

[21] S. D. Bloom, Phys. Lett. 48B, 420 (1974).

[22] H. Primakoff, Rev. Mod. Phys. 31, 802 (1959).

[23] T. A. Filippas, P. Palit, R. T. Siegel and R. E. Weish, Phys. Lett. 6, 118 (1963).