Measurements with a magnetic spectrometer⁵ of the reaction $B^{11}(d,p)B^{12}$ showed that the cross section for the 2.72-Mev level was 5% of the cross section for the 2.62-Mev state. Recent measurements' of protons from $B^{10}(t,p)B^{12}$ confirm that the 2.72-Mev state is weakly excited. With a different mode of excitation one cannot rule out the possibility of a greater population of the 2.72-Mev level; however, the total cross section for this group of α particles is not significantly larger than for the groups corresponding to single levels.

The 7.0-Mev level which we observe in B^{12} is unambiguously present, and our 8.05-Mev level is quite

⁵ M. M. Elkind, Phys. Rev. 92, 127 (1953).

A. A. Jaffe, et. al., Proc. Phys. Soc. (London) A76, 914 (1960)

strongly excited. Neither of these levels was observed in the total neutron cross-section measurements' on B¹¹. If these levels were due to target contaminants we should have observed additional sharp α -particle peaks of higher energy due to these same contaminants. There are several reasons⁴ why peaks might not have been seen are several reasons why peaks inight not have been seen
in the neutron work; e.g., the B^{12} level could have $J=0$.

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Angular Distribution of Alpha Particles Emitted by Oriented Np^{237} Nuclei

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Np²³⁷ nuclei were aligned through the electric quadrupole and magnetic hyperfine couplings in $NpO_2Rb(NO_3)$, cooled to 0.2–4.2°K. A complete experiment, with rotatable monocrystalline sample, solidstate counter, thermometer, and goniometer, was enclosed in a copper container filled with He' gas and thermally attached to $\frac{1}{2}$ mole of paramagnetic salt which could be cooled magnetically. The measured temperature dependence of the α -particle anisotropy gives $A < 0$, $P > 0$ for the signs of the hyperfine coupling constants in $NpO₂⁺⁺$. The α particles were observed to be emitted preferentially along the direction of the nuclear angular momentum vector. The results are consistent with $P-\pi$ bonding in the NpO₂⁺⁺ ion and with Hill and Wheeler's prediction of the role of barrier penetration in α emission from nonspherical nuclei.

I. INTRODUCTION

LPHA —PARTICLE emission from oriented nuclei was first considered by Spiers.¹ In a formal way he pointed out that if there were angular momentum changes in the alpha emission process, a spatial anisotropy of emission would in general be expected when the parent nuclei were oriented. No specific nuclear model was considered in this work, however, and no detailed prediction of the character of this anisotropy was given.

A general theory of angular momentum effects in alpha emission has been given by Rose' in which the angular distribution of the alpha particles is completely specified in terms of nuclear matrix elements.

It was suggested by Hill and Wheeler³ that a nonspherical shape for an alpha-emitting nucleus should have an effect upon the angular distribution of the alpha particles emitted. For example, they found that for a nucleus of prolate spheroidal shape the potential barrier against alpha-particle emission should be both lower and thinner near the nuclear "poles" than near the nuclear

"equator." Assuming a uniform probability of alphaparticle formation over the nuclear surface, they pointed out that a strong preferential alpha emission near the nuclear "poles" would be expected. Using a simplified WKB treatment they estimated this preferential emission to be sixteen times more intense at the polar than at the equatorial region for a moderate spheroidal deformation of the nucleus of $c/a \approx 1.1$. They further suggested that these effects could be investigated by nuclear orientation experiments, but angular momentum effects were not included in their discussion. Barrier effects in alpha emission have also been studied by Christy,⁴ whose conclusions are in general accord with those of Hill and Wheeler.

Recently, more complete theoretical discussions of alpha emission including angular momentum effects and considering specific nuclear models have been given, for example, by Brussaard and Tolhoek,⁵ Rasmussen and Segall,⁶ Steenberg and Sharma,⁷ and Fröman.⁸ It is

^{*}Operated by Union Carbide Corporation for the U. S.Atomic Energy Commission. '

J. A. Spiers, Nature 161, ⁸⁰⁷ (1948).

² M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957), pp. 176–186.

D. L. Hill and J. A. Wheeler, Phys. Rev. 89, 1133 (1933).

⁴ R. F. Christy, Bull. Am. Phys. Soc. 30, 66 (1955).
⁵ P. J. Brussaard and H. A. Tolhoek, Physica 24, 233 (1958).
⁶ J. O. Rasmussen and B. Segall, Phys. Rev. 103, 1298 (1956).
⁷ N. R. Steenberg and R. C. Sharma, Ca (1960).

P. O. Froman, Kgl. Danske Videnskab. Selskab, Mat. -fys. Skrifter 1, No. 3 (1957).

generally expected that barrier effects will be the dominating influence in the anisotropy of alpha emission.

The first measurements $^{9-11}$ of the anisotrophy of alpha emission from oriented nuclei were for Np^{237} . In these experiments, the nuclei were oriented through the unusually large electric quadrupole and magnetic hyperfine couplings in the $NpO₂^{++}$ group of the salt $NpO₂Rb(NO₃)₃$. In this rhombohedral salt, all of the linear NpO_2^{++} groups lie parallel to the crystalline c axis.^{12,13} Thus, the c axis is the asymmetry axis (z direction) for the nuclear orientation, and alpha emission from the oriented nuclei was measured relative to this direction. It was found in these first experiments that as the temperature was lowered and the nuclei became oriented, the preferential alpha emission was perpendicular to this axis.

The hfs coupling constants for this salt have been measured by Llewellyn'4 and by Bleaney, Llewellyn, measured by Llewellyn¹⁴ and by Bleaney, Llewellyn
Pryce and Hall,¹⁵ and discussed theoretically by Eisen-Pryce and Hall,¹⁵ and discussed theoretically by Eisenstein and Pryce,¹⁶ and more recently by Pryce.¹⁷ They expressed their measured results at zero applied magnetic field in terms of a spin Hamiltonian,

$$
H = A I_s S_z + B (I_x S_x + I_y S_y) + P [I_z^2 - \frac{1}{3} I (I+1)], \quad (1)
$$

with the experimentally determined parameters

$$
A = \pm 0.16547 \pm 0.00005
$$
 cm⁻¹,

$$
|B| = 0.01782 \pm 0.00003
$$
 cm⁻¹,

$$
P = \mp 0.03015 \pm 0.00005
$$
 cm⁻¹.

The absolute signs for A and P were not determined experimentally, but only their relative sign. In early experimentally, but only their relative sign. In early theoretical discussions,¹⁶ Λ was given as positive and P as negative. When this assignment was used for the interpretation of our measurements on alpha emission from oriented Np nuclei mentioned above, the unexpected result of predominant alpha emission from the nuclear equatorial region was obtained. It was thus desirable to determine the signs of A and P experimentally. We have done this by extending our earlier nuclear alignment measurements to a lower temperature region.

A qualitative understanding of the nature of these recent experiments may be gained from a consideration

¹⁴ P. M. Llewellyn, thesis, St. John's College, England, 1957 (unpublished), Chap. 4.

- ¹⁵ B. Bleaney, P. M. Llewellyn, M. H. L. Pryce, and G. R. Hall, Phil. Mag. 45, 992 (1954).
- ¹⁶ J. C. Eisenstein and M. H. L. Pryce, Proc. Roy. Soc. (London) $A229$, 20 (1955).

FIG. 1. Energy-level diagram of Np in $NpO_2Rb(NO_3)$, for different choices of the signs of the hyperfine coupling constant A and P. There are small $(\leq 1\%)$ admixtures of states of different m_I and m_S in most of the levels.

of Fig. 1, where the two possible energy-level diagrams are shown which correspond to the alternative sign choices for A and P . Due to the "interference" of the magnetic and electric coupling effects in H , there is a clustering of the levels. For case I of Fig. 1 with $A>0$, $P<0$, the lowest level is in a degree isolated, so that when the temperature is lowered the system will tend to condense into this lowest level, leading to a relatively large degree of nuclear alignment. For case II, however, the lowest group of three doublets is especially closely spaced and contains a complete set or shell of the magspaced and contains a complete set or shell of the magnetic substates of $I=\frac{5}{2}$. In our temperature region this group would contribute very little to the nuclear alignment, which arises for this case primarily from the next higher doublet for which $m_I = \pm \frac{1}{2}$. The alignment would therefore be substantially smaller than for case I. It is also evident from Fig. 1 that the relative populations of the different substates would vary in an entirely different way for the two cases as the temperature was lowered. Thus a measurement of the temperature dependence of the nuclear alignment will serve to distinguish between the two cases.

In the present experiment, this temperature dependence has been studied over the temperature range $0.2 - 4.2$ ^oK, by inferring the nuclear alignment from measurements of alpha-particle anisotropy. The angular distribution of alpha particles emitted by oriented nuclei has been calculated by Rose,² whose result is

$$
W(\theta) \propto \sum_{L,L'} A_{LL'} \sum_{\nu} G_{\nu}(T) C(LL'\nu; 00)
$$

$$
\times W(IILL'; \nu I') P_{\nu}(\cos\theta). \quad (2)
$$

L. D. Roberts, J. W. T. Dabbs, G. W. Parker, and R. D.

Ellison, Bull. Am. Phys. Soc. 1, ²⁰⁷ (1956). "J.W. T. Dabbs, L. D. Roberts, and G. W. Parker, Physica 24, S69 (1958)

S69 (1958).

¹¹ L. D. Roberts, J. W. T. Dabbs, and G. W. Parker, *Proceedings*
 of the Second United Nations International Conference on the
 Peaceful Uses of Atomic Energy, Geneva 1958 (United Nations,

Geneva, 195

F. Duncan, Spectroscopic Properties of Uranium Compounds (McGraw-Hill Book Company, New York, 1949), p. 15.
 13 W. H. Zachariasen, Acta Cryst. 7, 795 (1954).

¹⁷ M. H. L. Pryce, Phys. Rev. Letters 3, 375 (1959).

This may be expressed as

$$
W(\theta) \propto \sum a_{\nu} G_{\nu}(T) P_{\nu}(\cos \theta), \tag{3}
$$

where a_r contains all the nuclear properties and G_r measures the nuclear alignment;

$$
G_{\nu}(T) = \frac{\operatorname{Tr}(T_{\nu 0}e^{-H/kT})}{\operatorname{Tr}(e^{-H/kT})}.
$$
 (4)

The quantities $A_{LL'}$ are intensities for $L = L'$ and products of amplitudes for $L \neq L'$; I and I' are initial and final nuclear angular momenta; L is the alpha-particle angular momentum; $C(LL'v; 00)$ and $\overline{W}(II\overline{L}L'; vI')$ are, respectively, Clebsch-Gordan and Racah coefficients. Only terms with ν even are present, and ν must be no greater than $L+L'$, and also no greater than 2I. For Np^{237} , $I=\frac{5}{2}$; only $\nu=0$, 2, or 4 can be present. The theoretically admissible term involving $P_4(\cos\theta)$ was in fact not observed, and in the following only the terms with $\nu=0$ and 2 will be considered.

In Eq. (4), H is the spin Hamiltonian for the alignment forces, given for our case by Eq. (1), and $T_{\nu 0}$ is an element of an irreducible tensor, given by

$$
T_{00} = (2I+1)^{-\frac{1}{2}},\tag{5a}
$$

$$
T_{20} = \left[\frac{180(2I-2)!}{(2I+3)!}\right]^{\frac{1}{2}} [I_z^2 - \frac{1}{3}I(I+1)].
$$
 (5b)

It follows from the theory given above that a measurement of the coefficient of $P_2(\cos\theta)$ as a function of temperature can be compared with $G_2(T)$ within a proportionality constant which depends upon nuclear properties. The two possible sign choices for ^A and P give two different functions $G_2(T)$; these are cases I and II discussed previously.

In the following, after a description of the experimental procedure, the results are compared with the two possible functions $G_2(T)$ which we have calculated dipossible functions $G_2(T)$ which we have calculated directly from the results of the experiment of Llewellyn.^{14,15} The signs of A and P are thus determined experimentally. A calculation of a_2 is then given, based on current nuclear theory, and compared with experiment. Finally, the direction of preferential alpha emission is compared to the prediction of Hill and Wheeler.³

II. EXPERIMENTAL PROCEDURE AND RESULTS

As in our earlier work, $9-11$ the experiment was performed by cooling a monocrystalline sample of $NpO_2Rb(NO_3)$ to the desired temperature and counting the emitted alpha particles at known angles to the crystalline c axis. In all, twenty-three low-temperature runs were made using a single sample, the earlier work having shown that the results are reproducible from sample to sample. In order to reduce alpha-particle heating, the sample was prepared by growing a thin

layer (0.75 mg/cm²) of $NpO_2Rb(NO_3)$ onto a single crystal of the isomorphous compound $UO_2Rb(NO_3)_2$. The base crystal and the surface layer were grown from saturated solutions in $16M$ HNO₃ at approximately 50'C. Numerous x-ray diffraction studies" have confirmed that the uranyl crystals are indeed single crystals, and that the neptunyl salt grows in the same lattice, oriented in the same direction as the underlying material. The amount of Np^{237} in the sample was determined by alpha counting in a known geometry. Separate determinations were made for prism faces and cap faces, because of differing growth rates. The uranyl and neptunyl salts were both checked for radioactive contamination, and none was observed.

A schematic diagram of the experimental arrangement is shown in Fig. 2. An isothermal copper enclosure, filled with He³ gas for cooling the sample surface, contained the sample in a holder which could be rotated on small ball bearings, a germanium surface-barrier alpha small ball bearings, a germanium surface-barrier alph
counter,¹⁹ rotation indicators, and a carbon resistanc counter,¹⁹ rotation indicators, and a carbon resistance.
thermometer.²⁰ This sample enclosure was in therma contact with $\frac{1}{2}$ mole of small manganous ammonium sulphate (MAS) crystals. The whole assembly was suspended on cotton threads in a vacuum so it could be cooled by adiabatic demagnetization of the MAS, using a large Weiss-type electromagnet. The vacuum chamber was mounted within Dewar vessels for liquid He and liquid N_2 . Thermal contact was made—and brokenbetween the low-temperature assembly and the liquid helium bath by means of a mechanical heat switch.^{21,21}

FIG. 2. Schematic diagram of experimental arrangement.

¹⁸ R. D. Ellison (private communication).

F.J. Walter, J. W. T. Dabbs, and L. D. Roberts, Rev. Sci. Instr. 31, 756 (196O).

²⁰ J. R. Clement and E. H. Quinnell, Rev. Sci. Instr. 23, 213 (1952).

²¹ F. J. Webb and J. Wilks, Proc. Roy. Soc. (London) A230, 549

 $(1955$ ²² R. Berman, J. Appl. Phys. 27, 318 (1956).

FIG. 3. Typical angular distributions at $1.92\textdegree K$ and $0.19\textdegree K$. The solid lines are least-squares fits to Eq. (6).

After the sample enclosure had been cooled to the desired temperature, the large Weiss magnet was removed, and the sample was rotated by means of the interaction between external, rotatable Helmholtz coils and a small permanent magnet attached to the sample holder. At each temperature, a run consisted of at least two independent counts at each angle. For thirteen of the runs, counting was done only at $\theta = 0^{\circ}$ and $\theta = 90^{\circ}$; for the other ten runs, counts were taken at 15° intervals. In all cases the counting sequence was arranged to minimize the effect of any slow drifts that might have been present; no drifts were in fact observed. For each run, the temperature was determined by measuring the susceptibility of the MAS with a set of mutual inductance coils and a ballistic galvanometer. The temperature readings were corrected for the effects of shape and readings were corrected for the effects of shape and
filling fraction of the MAS,²⁸ and for the departures of its susceptibility from Curie's law.²⁴ In every case, the temperature variation during a run was small because of the large heat capacity of the MAS; the average temperature was used when the variation was measurable. The carbon resistance thermometer was cemented into a copper block whose size and shape approximated that of the sample crystal. This block was mounted on plastic supports so that it could be cooled essentially only by the He' gas. Measurements using this thermometer confirmed that the temperature rise of the sample above its surroundings was negligible.

The pulses from the solid-state counter were amplified in a linear amplifier, whose output signal was applied to two discriminators set at different discrimination levels to study the variation, if any, of angular distribution with pulse height. During two runs, the pulse-height

distribution was recorded on a multichannel pulseheight analyzer. The results were determined to be independent of the discrimination level.

The observed angular distribution may be expected to have the form

$$
V(\theta) = V(\theta) \left[1 + b_2 F(T) P_2(\cos \theta) \right],\tag{6}
$$

where b_2 is a geometrical correction²⁵ for finite source and counter size; for this experiment, $b_2=0.93$. The alpha-particle anisotropy is measured by $F(T)$, the experimentally determined coefficient of $P_2(\cos\theta)$, which is to be compared with $a_2G_2(T)$, the theoretically predicted anisotropy in Eq. (3). The function $V(\theta)$ is the angular distribution at high temperature (no nuclear alignment) and depends on the shape of the sample crystal. The counter characteristics made it impossible to measure $V(\theta)$ directly at high temperature in this experiment. This function was therefore deduced by comparing the data of the present experiment in the comparing the data of the present experiment in the temperature range $1.2 - 4.2^{\circ}$ K with the earlier results^{10,11} taken in good geometry in this temperature region.

For each run, the counting data were fitted by least squares to Eq. (6). Typical data taken at 1.92 and 0.19° K are shown in Fig. 3, together with the leastsquares fits to Eq. (6) for these two runs. The slopes

FIG. 4. Size of anisotropy as a function of temperature, com-
pared with normalized $G_2(T)$ functions for the two possible
choices of the signs of A and P.

 23 H. B. G. Casimir, Magnetism and Very Low Temperature

⁽Cambridge Vniversity Press, New York, 1940), pp. 9—13. 24A. H. Cooke, H. Meyer, and %. P. Wolf, Proc. Roy. Soc. ('London) A233, 536 (1956).

²⁵ M. E. Rose, Atomic Energy Commission Report ORNL-2050 (1956) (unpublished).

give $b_2F(T)$. It should be noted that, as in the earlier experiments, the alpha emission was enhanced near $\theta = 90^{\circ}$, with $F(T) < 0$.

The data for all the runs are presented in Fig. 4, which shows $F(T)$ as a function of temperature. The errors shown in Fig. 4 are standard deviations based on counting statistics only; the scatter of the points for each run is consistent with these deviations. Also shown on Fig. 4 are the two curves for $G_2(T)$, calculated from Eqs. (1) and (4) and normalized at $1.13\textdegree K$ on the basis of the earlier experimental results.

It is clear from Fig. 4 that the data admit only the sign choice $A<0$, $P>0$. This conclusion follows directly from the two experimental results, the microwave resonance¹⁴ and the present one, and from angular-momentum theory.² No nuclear or electronic structure model is involved. This experimentally determined sign model is involved. This experimentally determined sign
choice is opposite to the earlier expectation,¹⁶ but is consistent with a more recent theoretical result of Pryce.¹⁷ Pryce.¹⁷

III. DISCUSSION OF RESULTS

A. Size of Anisotroyy

The spins and parities of the levels in Pa²³³ populated by the alpha decay of Np^{237} have recently been compiled by Asaro²⁶ and are given in Table I. Using this information, it has been possible to calculate the constant a_2 in Eq. (3), and thus to make a quantitative comparison between the theoretical and experimental anisotropies. The calculation contains two adjustable parameters S and 5' which give the relative intensities of 5-wave alpha particles (as against D -wave intensities D and D^7 ; $S+D=S'+D'=1$) for the two transitions involving $L=0$ alpha particles. For this calculation, we set $S'=S$ based on alpha-particle decay systematics. $6, 8, 27$ In any

^a From F. Asaro (private communication).
^b These levels have an admixture of $K = 3/2$.

case, the value chosen for S' will affect the result only slightly because of the low intensity of this transition. For other transitions, only the lowest possible value of angular momentum was considered.²⁷ Figure 5 shows the angular momentum was considered. Figure 5 shows the calculated value of a_2/a_0 as a function of S; a_0 is the amplitude of isotropic alpha emission. The S and D waves can be either in phase or out of phase; these are plotted to the right and left, respectively, of the origin in Fig. 5. Also shown is the least-squares value of a_2/a_0 redetermined from the data of the earlier experiments in good geometry; see Fig. 6. The comparison of Fig. 5 suggests that the S and D waves are in phase in such a way as to give the largest possible anisotropy. The value of S for maximum anisotropy is in the range predicted by alpha-particle decay systematics. $8,27$

In Fig. 6 the data from the earlier experiment are plotted as a function of temperature, along with $a_2G_2(T)$, using for a_2 its maximum theoretical value. The data are not inconsistent with this value of a_2 .

Fro. 5. Anisotropy coefficient a_2 [cf. Eq. (3)], calculate from Eq. (2) and the data of Table I, compared with experimental result.

B. Direction of Preferential Emission

Referring again to Fig. 1, the experimental result $A < 0, P > 0$ limits consideration to case II. Although the system would condense into a state for which $m_I = \pm \frac{3}{2}$ at a temperature near absolute zero, the situation in the temperature range studied in the present experiment is quite different. The three lowest doublets are almost equally populated, and preferential population of the next lowest states leaves an excess of nuclei in states for which $m_I = \pm \frac{1}{2}$, so that the nuclear angular momentum vector is aligned preferentially perpendicular to the crystalline c axis. The observed enhancement of alpha emission in this direction in all our experiments on \bar{Np}^{237} leads to the conclusion that the alpha particles are emitted preferentially along the nuclear angular momentum vector. This inference follows directly from the microwave resonance data and the results of the present

²⁶ F. Asaro, (private communication).
²⁷ A. Bohr, P. O. Fröman, and B. R. Mottelson, Kgl. Danske
Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 10 (1955).

experiment, and is independent of any assumptions about nuclear shapes or the mechanism of alpha-particle emission.

Under the usual assumption for this region of the periodic table that the nuclear angular momentum vector is oriented along the nuclear symmetry axis, the experimental result means that the alpha particles are emitted preferentially along the nuclear symmetry axis. For a prolate nucleus, this preferential emission is from the "polar" regions, in agreement with the prediction of Hill and Wheeler.³

The question which remains is whether the Np²³⁷ nucleus is in fact prolate, as stated above. We know of no direct experimental evidence available at this time regarding the shape of this nucleus or the sign of the nuclear electric quadrupole moment Q. However, the level diagrams given by Nilsson and others²⁸⁻³⁰ for prolate nuclei in the region $A > 225$ agree very well with experiment.

It is worth noting that for the neighboring nuclei U²³³, U²³⁵, Am²⁴¹, and Am²⁴³, optical hfs spectra have been interpreted³¹ as giving evidence for $Q>0$ in all cases.

Pryce has suggested¹⁷ that $P > 0$ is evidence for $Q < 0$. This inference is based on a theory of Eisenstein and Pryce $16,17$ of the electronic structure and magnetic properties of uranyl-like ions, which assumed that the $O-U-O$ and $O-Np-O$ bonds are of predominantly σ . character. This theory of σ bonding requires P and O to be of opposite sign, so that our result $P>0$ would require $0<0$. An oblate shape for Np²³⁷ would be in conflict with both the expectations of nuclear systematics and the barrier-penetration picture of Hill and Wheeler, δ since the barrier would in this case be weaker at the "equatorial" region.

Fro. 6. Experimental anisotropy as a function of temperature, compared with theory. The data are from the good-geometry experiments described in reference 9.

A somewhat different model of the electronic structure of the uranyl and neptunyl ions³² includes the effects of $P-\pi$ bonding. With this modification, it is possible to reconcile $P > 0$ with $Q > 0$. On the basis of this modification, our experiments on the angular distribution of alpha particles emitted by oriented Np^{237} nuclei are consistent with $Q>0$ and with Hill and Wheeler's prediction of the role of barrier penetration in alpha emission from nonspherical nuclei.

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³² L. D. Roberts and J. W. T. Dabbs, Bull. Am. Phys. Soc. 5, 22 (1960).

<u>. s. – T. Kilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys.</u>
. Medd. 29, No. 16 (1955).
²⁹ B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab

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^{(1959).}

[&]quot;K. L. Vander Sluis and J. R. McNally, Jr., J. Opt. Soc. Am. 44, 87 (1954) and private communication; A. G. Zimin and N. M. Iashin, Soviet Phys. -Doklady I, 419 (1956);N. I.Kaliteevskii and M. P. Chaika, Optika i Spektroskopiya 1, 809 (1956); T. E. Manning, M. Fred, and F. S. Tomkins, Phys. Rev. 102, 1108 (1956); J. Blaise, S. Gerstenhorn, and M. Louvegnies, J. phys. radium 18, 318 (1957).