data by the function $p_{66}{}^{E=0} = A + B/(T - T_0)$. Adjustment of the parameters A, B, and T_0 to produce a best "least-squares" fit produced $p_{66}{}^{E=0} = -0.0345 - 2.132/(T - 117.45^\circ)$ with an rms error of 0.013 which is comparable with the experimental uncertainty indicated by the error bar in Fig. 1. The value of $T_0 = 117.45^\circ\mathrm{K}$ is in excellent agreement with the more accurate value of 117.7 °K determined from the elastic constant measurements,[2] and the value $B = -2.132$ is in satisfactory agreement with the value of 1.78 predicted by Eq. (7), in view of the various experimental errors.[8]

Conclusion. – The observed anomaly in the Pockels' coefficient $p_{66}{}^{E=0}$ is due to the Curie-Weiss behavior of the clamped dielectric constant $\epsilon_{33}{}^{x=0}$. The expression for $p_{66}{}^{E=0}$ which we have found is of the same form as the expression for the anomalous elastic constant $C_{66}{}^{E=0} = C_{66}{}^{P=0} - (a_{36}{}^2 \epsilon_{33}{}^{x=0})/4\pi$ since both anomalies result from the divergence of $\epsilon_{33}{}^{x=0}$. The fact that $p_{66}{}^{E=0}$ approaches a finite upper bound as $T \to T_c{}^+$ (as does $\epsilon_{33}{}^{x=0}$)[1] emphasizes again that any discussion of the ferroelectric transition in KDP must include the piezoelectric coupling of polarization and strain which leads to a 4.3° difference in the Curie temperatures for the "clamped" and "free" crystals.[1,2]

---

†Work supported by the Advanced Research Projects Agency and by the U. S. Army Research Office (Durham).

*Present address: The University of Rochester, College of Engineering and Applied Science, Space Science Center, River Campus Station, Rochester, N. Y. 14627.

‡Alfred P. Sloan Research Fellow.

[1] F. Jona and G. Shirane, Ferroelectric Crystals (Pergamon Press, New York, 1962).

[2] The experimental arrangement was the same as that discussed by E. M. Brody and H. Z. Cummins, Phys. Rev. Letters 21, 1263 (1968).

[3] The total intensity of light scattered from the X_y shear acoustic mode in KDP was measured by I. M. Aref'ev et al., Fiz. Tverd. Tela 8, 272 (1966) [translation: Soviet Phys. – Solid State 8, 222 (1966)], and shown to be in qualitative agreement with the temperature dependence of $(C_{66}{}^{E=0})^{-1}$.

[4] n_x was obtained from data of R. A. Phillips, J. Opt. Soc. Am. 56, 629 (1966).

[5] J. F. Nye, Physical Properties of Crystals (Oxford University Press, London, England, 1957).

[6] H. H. Landolt and R. Börnstein, Elastic, Piezoelectric, Piezo-optic, and Electro-optic Constants of Crystals (Springer-Verlag, Berlin, Germany, 1966), Group III, Vol. 1.

[7] The value of $p_{66}{}^{E=0}$ quoted in Ref. 6 is −0.0685. The value −0.0552 was obtained by direct comparison in the Brillouin spectrometer using a single crystal of quartz as a calibrating standard. V_V scattering from the $\vec{q} = \vec{y}$ phonon in quartz was used. In essence the $p_{66}{}^{E=0}$ coefficient in KDP is normalized against the p_{13} coefficient in quartz. $p_{13} = 0.259$ (from Ref. 6) is used. [Recently $p_{13} = 0.27$ has been found by T. S. Narasimhamurty, J. Opt. Soc. Am. 59, 682 (1969)].

[8] The electro-optic coefficient r_{63} of KDP has recently been reported to be 10.6×10^{-10} cm/V [R. S. Adhav, J. Opt. Soc. Am. 59, 414 (1969)]. If this value is used in place of that of Ref. 6, then Eq. (7) becomes

$$p_{66}{}^{E=0}(T) = p_{66}{}^{P=0} - 2.1(T - 117.7^\circ\mathrm{K})$$

indicating much better agreement between theory and experiment.

ROLE OF DOORWAY STATES IN NEUTRON CAPTURE IN ^{93}Nb RESONANCE*

Karim Rimawi,† R. E. Chrien, J. B. Garg,† M. R. Bhat, D. I. Garber, and O. A. Wasson
Brookhaven National Laboratory, Upton, New York 11973

(Received 16 September 1969)

A comparison between the γ-ray spectra following the capture of $l = 0$ and $l = 1$ neutrons in niobium shows characteristic differences which are not expected with a purely statistical model of compound nucleus decay. The data suggest that the strong $E1$ γ rays are due to particle-hole annihilations across the $Z = 50$ proton shell, and demonstrate the influence of two-particle, one-hole "doorway" states in the neutron capture process.

Although most features of slow-neutron reactions are characteristic of the formation of a compound nucleus and its subsequent statistical decay, there are several notable exceptions. These include the presence of size resonances in the $l = 1$ and $l = 0$ neutron average cross sections (or strength functions),[1] and the effects of intermediate structure, or doorway states.[2] Departures from the usual statistical-model predictions also occur in the radiative decay of the unbound states formed by slow-neutron capture. The strengths of transitions to final states of single-particle character[3] are observed in some cases to be enhanced over the statistical-model predictions, and partial-width correlations are also found to occur.[4]

From the present work, evidence for the effects of two-particle, one-hole "doorway states"[5] is presented in the γ spectrum following p-wave capture in resonances of ^{93}Nb.

Differences in the spectra from s- and p-wave capture are related to the decay of two-particle, one-hole states to one-particle final states by a particle-hole annihilation process.

Neutron resonance γ rays from a 798-g sample of niobium powder were examined with the Brookhaven National Laboratory high-flux beam reactor fast-chopper time-of-flight facility. P-wave resonances at 35.8, 42.2, 94.3, and 243.7 eV, and s-wave resonances at 105.8, 119.2, and 193.8 eV were investigated and the energies and intensities for transitions to 136 final states in ^{94}Nb were determined. The s- and p-wave resonances have characteristically different spectra. These are indicated in Fig. 1, where the interval-averaged reduced γ-ray widths are plotted as functions of excitation energy in ^{94}Nb, with a 300-keV averaging interval. The widths have been summed over the four p-wave and three s-wave resonances. The s-wave spectra are clearly consistent with a statistical decay of the compound nucleus, but the p-wave spectra exhibit enhanced transitions to states near the ground state of ^{94}Nb. These are $E1$ transitions and several of them exceed the modified single-particle estimate[6] of the partial width by an order of magnitude.

The properties of low-lying states in ^{94}Nb have been exhaustively studied by Jurney et al.[7] In the simplest approximation the ground-state configuration may be regarded as a ^{90}Zr core with one proton and three neutrons outside it. The configurations arising from the coupling of the three neutrons in the $2d_{5/2}$ shell and the proton in the $1g_{9/2}$ shell lead to low-lying positive-parity states. Neighboring orbitals above the $2d_{5/2}$ are the $3s_{1/2}$, $2d_{3/2}$, and the $1g_{7/2}$. Thus, the low-lying states are primarily of positive parity and many exhibit $l=2$ and $l=0$ stripping patterns. Negative-parity states can be constructed by lifting a proton from the $2p_{1/2}$, $1f_{5/2}$, or $2p_{3/2}$ into higher even-l orbitals. These proton-excited states are not expected to be seen in the (d,p) reaction, as is confirmed by the results of Sheline et al.[8] The purely statistical decay of a highly excited neutron-resonant state would be expected to populate excited neutron and proton configurations equally.

The evidence of Fig. 1 indicates that this is not the case. Furthermore, in capture in the $l=1$ resonances, there is a marked tendency preferentially to populate states strongly excited in the (d,p) reaction. The detailed comparison between the resonance-averaged (n,γ) and the (d,p) intensities is shown in Fig. 2, and the data are listed in Table I. The correlation coefficients between the (n,γ) and (d,p) strengths, defined by the relation

$$\operatorname{corr}(x,y)=\frac{\Sigma(x-\bar{x})(y-\bar{y})}{\sigma_x\sigma_y},$$

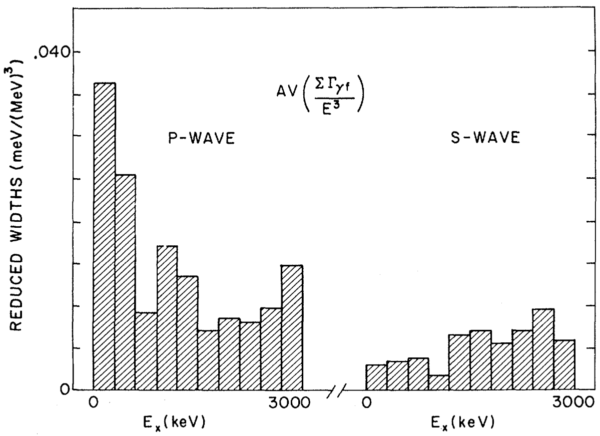

FIG. 1. A plot of the reduced γ-ray intensities from s- and p-wave resonances of niobium. The intensities are given by $\langle\Gamma_\gamma\rangle/E_\gamma^3$, averaged over 300-keV energy intervals, and summed over resonances.

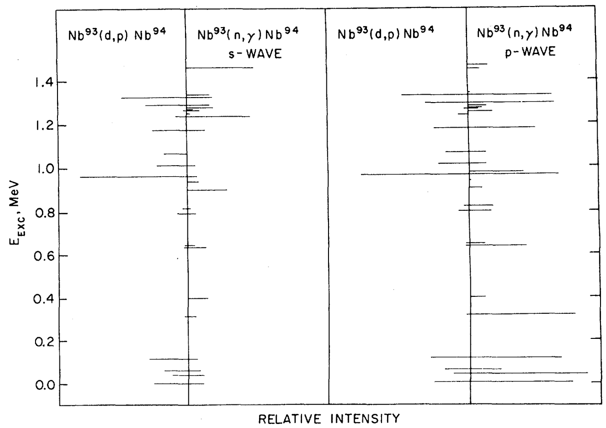

FIG. 2. A comparison of (d,p) and (n,γ) strengths for s- and p-wave resonances. The (n,γ) strengths are averaged over resonances.

Table I. The observed radiative widths for ^{93}Nb$(n,\gamma)^{94}$Nb. The differential (d,p) cross sections are shown for comparison. The widths are expressed in MeV. Where no entry is shown, the intensity is below the sensitivity of the experiment. Only transitions to states below $E_{ex} = 1.5$ MeV are tabulated here.

		p-wave Resonances				s-wave Resonances			(d,p)
E_γ (keV)	E_{exc} (keV)	E_n=35.8 eV	E_n=42.2 eV	E_n=94.3 eV	E_n=243.7 eV	E_n=105.8 eV	E_n=119.2 eV	E_n=193.8 eV	$d\sigma/d\Omega$ mb/sr
7229.3	0	16.29 ± 0.96			0.85 ± 0.14	0.26 ± 0.12	0.46 ± 0.09	0.63 ± 0.07	0.33
7188.5	40.8		0.56 ± 0.13	17.76 ± 0.86		1.43 ± 0.17			0.16
7171.0	58.4	2.13 ± 0.28	1.31 ± 0.28	0.75 ± 0.42	0.45 ± 0.22			1.06 ± 0.09	0.24
7116.3	113.0		12.51 ± 0.68		1.11 ± 0.31			0.74 ± 0.08	0.37
6917.3	312.0	11.98 ± 0.45	0.13 ± 0.22	0.70 ± 0.46	1.99 ± 0.30		0.55 ± 0.05		0.04
6832.8	396.6	0.23 ± 0.31	0.56 ± 0.19	1.19 ± 0.48		1.65 ± 0.20			0.00
6597.1	632.3		0.41 ± 0.24		4.53 ± 0.30	0.38 ± 0.15		1.02 ± 0.07	0.04
6587.7	641.6	0.43 ± 0.22	0.48 ± 0.25		0.65 ± 0.26			0.43 ± 0.06	0.04
6434.8	794.5		0.92 ± 0.23	0.79 ± 0.42	0.60 ± 0.26	0.53 ± 0.25			0.10
6412.8	816.6		0.01 ± 0.30	2.65 ± 0.53		0.06 ± 0.17			0.05
6332.5	896.8		0.56 ± 0.21		0.59 ± 0.37		1.09 ± 0.11	1.50 ± 0.13	0.00
6297.0	932.3				0.12 ± 0.22	0.50 ± 0.19	0.20 ± 0.11		0.00
6271.5	957.8	6.07 ± 0.54	3.18 ± 0.27		0.51 ± 0.29			0.61 ± 0.11	1.00
6258.6	970.7		2.83 ± 0.26	3.04 ± 0.55	0.53 ± 0.29	0.20 ± 0.21			0.00
6252.5	976.9			2.45 ± 0.54					0.00
6218.1	1011.3		0.84 ± 0.36	0.93 ± 0.44			0.43 ± 0.11		0.39
6170.3	1059.0	0.52 ± 0.25	0.38 ± 0.32	0.79 ± 0.43					0.22
6059.2	1170.1	1.41 ± 0.28	1.29 ± 0.51	1.71 ± 0.76	1.47 ± 0.24	0.29 ± 0.17	0.43 ± 0.10	0.29 ± 0.10	0.32
6049.8	1179.5		2.28 ± 0.51	0.95 ± 0.44	1.10 ± 0.17	0.33 ± 0.17			0.00
5995.5	1233.8					0.59 ± 0.20	1.12 ± 0.07	1.85 ± 0.17	0.10
5981.0	1248.3		2.20 ± 0.43				0.05 ± 0.09		0.00
5972.4	1256.9	1.01 ± 0.28					0.28 ± 0.09	0.35 ± 0.11	0.04
5966.1	1263.3	1.29 ± 0.25				0.31 ± 0.20			0.00
5955.9	1273.4	0.55 ± 0.26		0.37 ± 0.52	0.56 ± 0.25	0.66 ± 0.22	0.41 ± 0.16	0.62 ± 0.11	0.00
5948.1	1281.2	0.97 ± 0.24	5.40 ± 0.46		1.11 ± 0.25	0.33 ± 0.26	0.42 ± 0.16	0.42 ± 0.14	0.39
5907.4	1321.9		2.94 ± 0.26	3.38 ± 0.49	0.81 ± 0.17	0.02 ± 0.19	0.42 ± 0.20	0.89 ± 0.17	0.61
5896.0	1333.3				0.07 ± 0.12	1.24 ± 0.19			0.00
5881.5									0.00
5781.2	1448.2			0.94 ± 0.54	0.06 ± 0.26		0.08 ± 0.13		0.00

are as follows:

$$\mathrm{corr}\{\bar{\Gamma}_{\gamma i}/E^3, \sigma_{dp}\} = +0.58, \quad p \text{ wave},$$
$$= -0.09, \quad s \text{ wave}.$$

The +0.58 correlation coefficient for p-wave capture falls at the 99.9 percentile of the null-correlation distribution function and is thus inconsistent with zero. The s-wave coefficient, −0.09, falls at the 32 percentile and is consistent with zero correlation.

Such (n,γ), (d,p) correlations have been previously noted in thermal capture,[9] and more recently in resonance capture, as well.[10] It is tempting to attribute this correlation to a single-particle, or valence-neutron, transition from the capturing state, $l=1 \rightarrow l=2$ or $l=0$, leading to enhanced $E1$ radiation. The presence of the $l=1$ strength-function maximum near Nb would seem to support this notion. Nevertheless, our data indicate that this hypothesis is not tenable in this case. The valence-neutron transition concept would require that the (n,γ) widths be correlated with the reduced neutron widths of the initial, resonance states. Such a correlation can be tested from the present data, for the 13 final states fed from p-wave resonances by $E1$ radiation.

The result, for the average correlation coefficient R, is fully consistent with zero:

$$R = \langle \mathrm{corr}\{\Gamma_n^{\;0}, \Gamma_{\gamma i}\}\rangle = -0.09.$$

This result indicates the failure of the valence-transition or single-particle model. An examination of the available single-particle states, however, suggests the alternative hypothesis of a particle-hole annihilation producing the observed strong electric dipole radiation. The approximate position of single-particle states for $A \approx 90$ has been given by Cohen et al.[11] An incoming neutron may interact with protons in the filled $2p_{1/2}$, $1f_{5/2}$, or $2p_{3/2}$ orbits, raising them to states above the $Z=50$ gap. Proton particle-hole states with the requisite parity are plentiful: These are $3s_{1/2}$-$2p_{1/2}$, $3s_{1/2}$-$2p_{3/2}$, $2d_{5/2}$-$1f_{5/2}$, and $2d_{5/2}$-$3p_{3/2}$, and the energy differences for these excitations are all near the observed maximum of 7 MeV. For $l=0$ or s-wave capture, only one particle-hole excitation is available, namely $1g_{7/2}$-$1g_{9/2}$. The particle-hole annihilation would, in this case, lead to $M1$ radiation.

The particle-hole annihilation pictured above would tend to leave the incident neutron in $2d_{5/2}$ or $3s_{1/2}$ orbits, and therefore tend to favor the population of $l = 2$ and $l = 0$ final states. The positive correlation coefficient in p-wave capture can thus be explained on this basis.

The simple two-particle, one-hole structure envisioned in niobium neutron capture would predict a sizable difference in the ratio of electric dipole to magnetic dipole radiation from p- and s-wave resonances, since it leads to an $E1$ enhancement for p-wave and an $M1$ enhancement for s-wave capture. The experimentally determined ratio of $E1$ to $M1$ strengths can be tabulated for these final states whose parities are known. The results are as follows:

$$\frac{(\langle\Gamma_i\rangle/E_3)E1}{(\langle\Gamma_i\rangle/E_3)M1} = 10.5, \quad p \text{ wave},$$

$$= 3.0, \quad s \text{ wave}.$$

The difference is again inconsistent with a statistical picture of γ-ray de-excitation of the compound nucleus, and fully in accord with the hypothesis of doorway-state decay. The importance of doorway states in determining the structure of the γ-ray spectra from neutron capture has recently been emphasized in the region near $A = 180$ by Earle, Lone, and Bartholomew.[12] The comparison of p-wave and s-wave capture spectra reported here proved additional evidence for this effect.

Previously reported spectra taken with a low-resolution NaI detector by Jackson[13] have been interpreted as indicating that the photon strength function for p- and s-wave resonances is different. The present data indicate a systematic contribution of about 30 meV to the total radiation width for the transitions to the positive-parity states near ground. Thus, the radiation widths of p-wave resonances are expected to exceed those of s-wave resonances by this amount. This 30-meV difference, combined with recent and more accurate determinations of the total radiation widths,[14] suggest that there is no need to invoke a parity-dependent photon strength function for niobium. The difference in total radiation widths can be entirely understood in terms of the nuclear structure of the low-lying ^{94}Nb states.

The authors would like to thank Frederick Paffrath for his tireless work maintaining the rotor system, Vito Manzella for electronic assistance, and Jean Domish for help with the computations.

---

*Work supported by the U. S. Atomic Energy Commission.

†Present address: Physics Department, State University of New York at Albany, Albany, N. Y.

[1] R. S. Carter, J. A. Harvey, D. J. Hughes, and V. E. Pilcher, Phys. Rev. 96, 113 (1954).

[2] J. A. Farrel, G. C. Kyker, E. G. Bilpuch, and H. W. Newson, Phys. Rev. Letters 17, 286 (1965).

[3] B. B. Kinsey and G. A. Bartholomew, Phys. Rev. 93, 1260 (1954).

[4] M. Beer, M. A. Lone, R. E. Chrien, O. A. Wasson, M. R. Bhat, and H. R. Muether, Phys. Rev. Letters 20, 340 (1968).

[5] H. Feshbach, A. K. Kerman, and R. H. Lemmer, Ann. Phys. (N.Y.) 41, 230 (1967).

[6] G. A. Bartholomew, Ann. Rev. Nucl. Sci. 11, 259 (1961).

[7] E. T. Jurney, H. T. Motz, R. K. Sheline, E. B. Shera, and J. Vervier, Nucl. Phys. A111, 105 (1968).

[8] R. K. Sheline, R. T. Jernigan, J. B. Ball, K. H. Bhatt, Y. E. Kim, and J. Vervier, Nucl. Phys. 61, 342 (1965).

[9] B. B. Kinsey, G. A. Bartholomew, and W. H. Walker, Phys. Rev. 83, 519 (1951).

[10] W. V. Prestwich and R. E. Coté, Phys. Rev. 155, 1223 (1967).

[11] B. L. Cohen, R. H. Fulmer, A. L. McCarthy, and P. Mukherjee, Rev. Mod. Phys. 35, 332 (1963); B. L. Cohen, in International Symposium on Nuclear Structure, Dubna, 1968 (International Atomic Energy Agency, Vienna, Austria, 1969).

[12] E. D. Earle, M. A. Lone, and G. A. Bartholomew, Bull. Am. Phys. Soc. 14, 515 (1969).

[13] H. E. Jackson, Phys. Rev. Letters 11, 378 (1963).

[14] W. M. Lopez, E. Haddad, S. J. Friesenhalm, and F. Fröhner, Bull. Am. Phys. Soc. 10, 724 (1965).