PHYSICAL REVIEW C VOLUME 38, NUMBER 1 JULY 1988

Unexpected recoil systematics of intermediate energy spallation products

Michael J. Tobin* and Paul J. Karol
Department of Chemistry, Carnegie-Mellon University, Pittsburgh, Pennsylvania 15213
(Received 21 March 1988)

Integral recoil properties of 25 nuclides produced in the interaction of 800 MeV $p+{}^{89}Y$ were determined using activation techniques. The forward-to-backward recoil ratios are a factor of 3 larger for *n*-deficient products compared to near-stable and *n*-excess products of similar mass, a behavior not discovered in previous studies. The results are interpreted as an indication that the near-stable and *n*-excess products, previously assumed to be the result of conventional *N*-*N* intranuclear cascades, actually reflect a significant contribution from "target fragmentation" channels.

I. INTRODUCTION

Interest in intermediate- and high-energy nuclear reactions remains undiminished in the approximately 40 years since Serber's first pronouncement.[1] The surge in activity in recent years is at least partially motivated by two issues. First, the production of light fragments in association with high-energy processes is not very well understood.[2] Second, there is the suspicion that new phenomena and new states of matter might become evident in heavy-ion reactions at ultrarelativistic energies. Critical to the planning or understanding of any such venture is confidence in the interpretation of preexisting results. However, there is currently a tension between two prevailing concepts. One maintains that nucleon-nucleon (*NN*) two-body interactions provide an adequate explanation of results, as argued most recently from emulsion experiments.[3] The other point of view, supported by a recent thick-target thick-catcher activation study,[4] emphasizes the prevalence of a *multifragmentation* process that is incompatible with the *NN* viewpoint. We have performed a new activation study that strongly suggests *both* of these perspectives are operative, even for reactions induced by 800-MeV protons.

The thick-target thick-catcher integral recoil experiment remains one of the few available techniques that provide information on the kinematic histories of heavy spallation products produced in intermediate- and high-energy nuclear reactions. In the experiment, a target foil of thickness W mg/cm^2 is sandwiched between two low-Z catcher foils, each of which provides 2π geometry and sufficient thickness to stop all relevant recoiling nuclei. The target stack is oriented perpendicular to the projectile beam. The fraction of product nuclei recoiling in the forward, "F," and backward, "B," hemispheres relative to the direction of the beam is measured for the radioisotopes of interest.

Thick-target integral recoil experiments are interpreted in the framework of the Serber cascade-evaporation model.[1] It is assumed that a forward directed velocity v is imparted to the residual nucleus as a result of the nucleon-nucleon intranuclear cascade, and an isotropically distributed velocity V is imparted during the evaporative relaxation step. Using a reasonable range-velocity relationship, v and V may be inferred from the experimental recoil fractions for each product according to equations derived from geometrical arguments.[5,6] From the values of v and V, two kinematic quantities are calculated: E^* and $\langle T \rangle$.

The excitation energy E^* deposited in the cascade residue is estimated by recourse to Monte Carlo intranuclear cascade (INC) simulations. An observed correlation between cascade momentum and E^* is expressible as[7]

$$\frac{E^*}{E_{\text{cn}}}=S\frac{v}{v_{\text{cn}}} \quad (\text{slope } S=0.8)\,, \tag{1}$$

where the subscript "cn" denotes the value imparted for complete momentum transfer to a hypothetical compound nucleus. $\langle T \rangle$, the kinetic energy imparted in the relaxation step of the reaction, is given by

$$\langle T \rangle=\tfrac{1}{2}A_R V^2\,, \tag{2}$$

where A_R is the product mass number.

The Serber two-step model of the spallation process has been extensively and successfully applied.[8] Nevertheless, measurements of the recoil properties of spallation products have yielded results that appear to lie outside the conventional interpretation. For example, Kaufman *et al.* found that the derived excitation energies of spallation products from the reaction of 1 GeV $p+$Au first increased with increasing mass loss from the target as expected, but then leveled off and even *decreased* with further mass loss.[9] Several studies using protons or heavy ions have now confirmed the sideways- and backward-enhanced recoil behavior of reaction products.[4,10] Also of relevance are counter studies of the reaction (480 MeV $p+$Ag) which showed that nearly 90% of the integrated cross section of stable light fragments ($4\le A\le 24$) are formed with high kinetic energies and forward-peaked angular distributions inconsistent with two step production systematics.[11] Lynch has recently reviewed the various experiments and their discordance with the two-step model.[2]

II. EXPERIMENT

In the current work, attention was direct to short-lived products. All irradiations were performed at Los Alamos

National Laboratory using the LAMPF 800-MeV external proton beam. Target stacks consisting of 10-mg/cm^2 ^{89}Y foil sandwiched between 7-mg/cm^2 ^{27}Al catchers were irradiated for several minutes in each experiment. Following the short exposures, rapid radiochemical separations were employed to isolate gallium, germanium, arsenic, and bromine from the individual target and catcher foils for assay by Ge(Li) spectroscopy. Nuclides were identified on the basis of γ-ray energy and half-life. Photopeak count rates and chemical yields were used to obtain the F and B fractions for each isotope.

III. RESULTS

The experimental results are presented in Table I as the "forward-to-backward ratio" (F/B) and $2W(F+B)$, the "effective recoil range."[6] The values of $2W(F+B)$ have been corrected for scattering effects[12] at the target-catcher interface. The tabulated results are the arithmetic mean of three replicate determinations. Since the recoil data are expressed as fractions, the reported uncertainties reflect only random experimental errors. The principal sources of random error arise from target-catcher alignment (1%), chemical yields (3%), counting statistics per decay curve fits (1–30%), and parent-daughter separation times (3%).

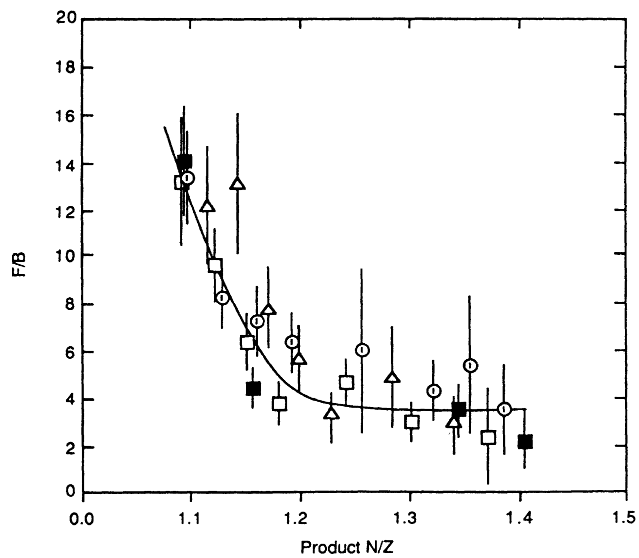

FIG. 1. Forward-to-backward ratios from the reaction (800-MeV $p+{}^{89}$Y). Error bars reflect uncertainties from decay curves, detector efficiencies, and chemical yields. Open circles correspond to Ga isotopes ($\Delta A = 15$–24); solid squares to Ge ($\Delta A = 12$–22); open squares to As ($\Delta A = 13$–20); and open triangles to Br ($\Delta A = 7$–15).

TABLE I. Integral recoil data and calculated parameters.

Nuclide	F/B	$2W(F+B)$ (mg/cm^2)	E^* (MeV)	$\langle T \rangle$ (MeV)
^{65}Ga	13.3±1.9	1.8±0.3	249	5.1
^{66}Ga	8.3±1.3	1.3±0.1	185	4.2
^{67}Ge	14.2±2.2	1.1±0.1	187	2.9
^{67}Ga	7.4±1.3	1.2±0.1	171	3.9
^{68}Ga	6.4±1.2	1.2±0.1	63	4.2
^{69}As	13.4±2.6	1.1±0.1	180	2.9
^{69}Ge	4.6±0.8	1.2±0.1	130	4.2
^{70}As	9.6±1.57	0.9±0.1	152	2.7
^{70}Ga	6.0±3.4	1.1±0.4	158	4.3
^{71}As	6.4±1.17	1.0±0.1	138	3.3
^{72}As	3.9±0.9	1.1±0.1	118	4.6
^{72}Ga	4.5±1.3	1.2±0.2	137	4.7
^{73}Ga	5.6±2.9	1.2±0.4	155	5.0
^{74}Br	12.2±2.5	1.1±0.2	177	3.2
^{74}As	4.8±0.9	1.0±0.1	128	4.0
^{74}Ga	3.6±1.9	1.1±0.3	119	5.1
^{75}Br	13.1±3.0	1.0±0.2	171	2.9
^{75}Ge	3.2±0.7	1.2±0.1	98	4.6
^{76}Br	7.8±1.69	1.2±0.1	165	4.1
^{76}As	3.0±0.5	1.2±0.1	99	5.1
^{77}Br	5.8±1.3	0.8±0.1	167	2.9
^{77}Ge	2.5±1.22	0.9±0.2	70	5.1
^{78}Br	3.6±0.95	0.5±0.1	63	1.7
^{80}Br	4.9±2.1	0.4±0.1	76	1.5
^{82}Br	3.1±1.03	0.4±0.1	56	1.6

The forward-to-backward (F/B) ratio reflects the magnitude of the forward momentum transferred in the initial excitation step. Table I and Fig. 1 show that F/B exhibits an unexpected dependence on product (N/Z) composition. For the near-stable and neutron-excess products ($1.15 \leq N/Z \leq 1.4$), F/B straddles a value of about 4, consistent with earlier work.[11] In contrast, F/B for the neutron-deficient ($N/Z \leq 1.15$) products are a factor of ~ 3 larger. While there have been a few isolated indications of anomalously large F/B for moderately neutron-deficient products,[9,13,14] no particular significance was attached to these. The unequivocal systematic variation of F/B with product N/Z evident in Table I and Fig. 1 has not been previously demonstrated in a nonfissioning system.

Table I also shows that $2W(F+B)$, as a measure of the mean kinetic energy of the product nucleus, increases with ΔA, the mass loss from the target. This trend reflects the greater energy required for increasingly long cascade-evaporation chains.[9,13] For a given product mass, $2W(F+B)$ is constant across stability, in contrast to the marked variation in F/B.

The calculated values of E^* and $\langle T \rangle$ are presented in Table I and Figs. 2(a) and 2(b). Figure 2(a) shows the variation of E^* with ΔA and product N/Z. The expected E^* dependence upon ΔA is clearly evident. However, paralleling the unprocessed F/B ratios, E^* rises sharply for the neutron-deficient products, independent of ΔA. In accordance with established spallation systematics, $\langle T \rangle$ also increases with ΔA, shown in Fig. 2(b). Surprisingly, the variation of $\langle T \rangle$ with product N/Z is inversely correlated with E^*.

IV. DISCUSSION

The observed variation of recoil properties with stability is both unusual and troublesome. In the NN cascade model, higher F/B quantitatively translates into higher E^*: ~ 200 MeV for $N/Z \leq 1.1$ products compared to ~ 125 MeV for near-stable and neutron-excess products, as seen in Fig. 1(a). In the conventional intranuclear cascade calculations, E^* and $\langle T \rangle$ turn out to be correlated. Additional excitation energy results in a longer evaporation chain. Moreover, evaporation calculations show that the probability of excited nuclei far from stability maintaining their "stressed composition" is reduced as excitation energy is increased. Two reasonable explanations come to mind. Since Eq. (1), from which E^* is calculated, is representative of pooled INC results, the biased recoil data in this work might be indicating an unrecognized correlation between momentum deposition and cascade pathway. Alternately, Eq. (1) may be unjustified if the projectile-target interaction does not proceed solely by nucleon-nucleon collisions.

Pursuant to the first argument, that the neutron-deficient products might arise from a nonrepresentative subset of the cascade residual spectrum, the results of a VEGAS (Refs. 15 and 16) INC simulation for (800-MeV $p + {}^{89}$Y) were binned in the following manner. First, the excitation energy and momentum output was grouped according to three N/Z regions mapping "neutron-deficient," "stable," and "neutron-excess" residual composition. If the resulting slope S in Eq. (1) for the first group were significantly smaller than for the latter two, the implication would be that a large amount of forward momentum transfer simply corresponds to less E^* in these products. This would be a conventional explanation for the surprising experimental results. Second, the VEGAS cascade output was pooled in $\Delta A = 2$ increments to look for a ΔA dependence in the energy-momentum relationship Eq. (1). In both explorations, however, linear regression of the individual E^*-momentum bins indicated that S is independent of residue N/Z and cascade depth ΔA, shown in Figs. 3(a) and 3(b). We therefore conclude that the current model of excitation and momentum deposition embodied in the VEGAS simulation does not reproduce the clearly observed variation of forward momentum transfer with product composition.

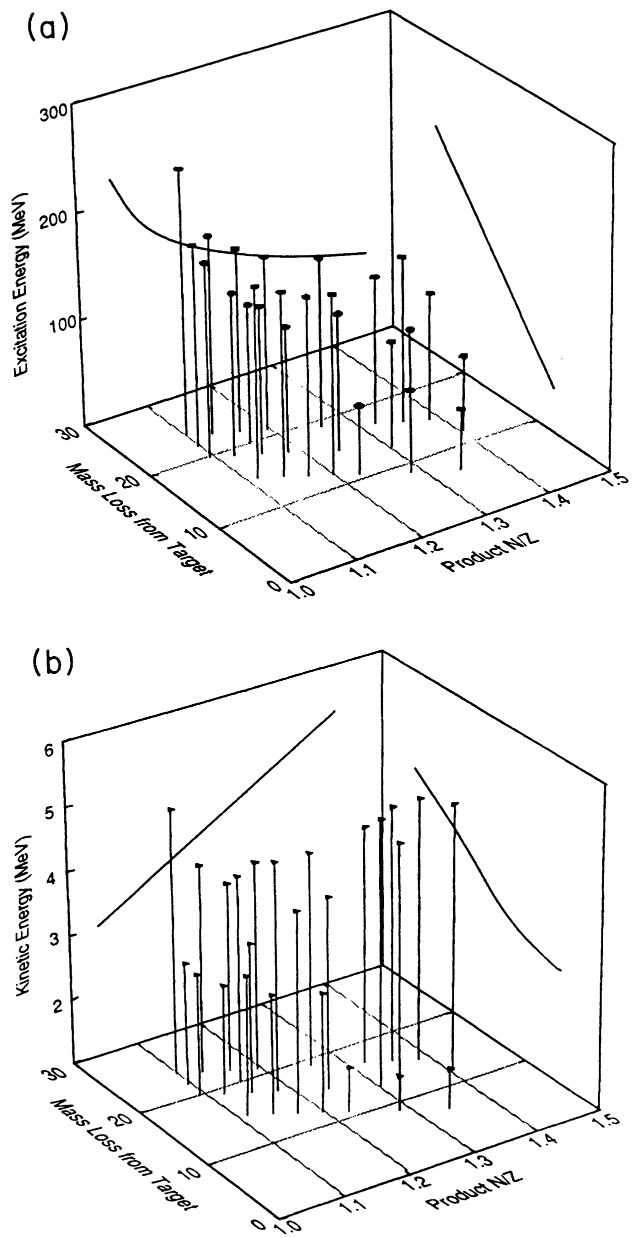

FIG. 2. Kinematic parameters from the reaction (800-MeV $p + {}^{89}$Y) plotted against ΔA and product N/Z. (a) Excitation energy. (b) Kinetic energy. Fitted trend lines are superimposed on the opposite facets.

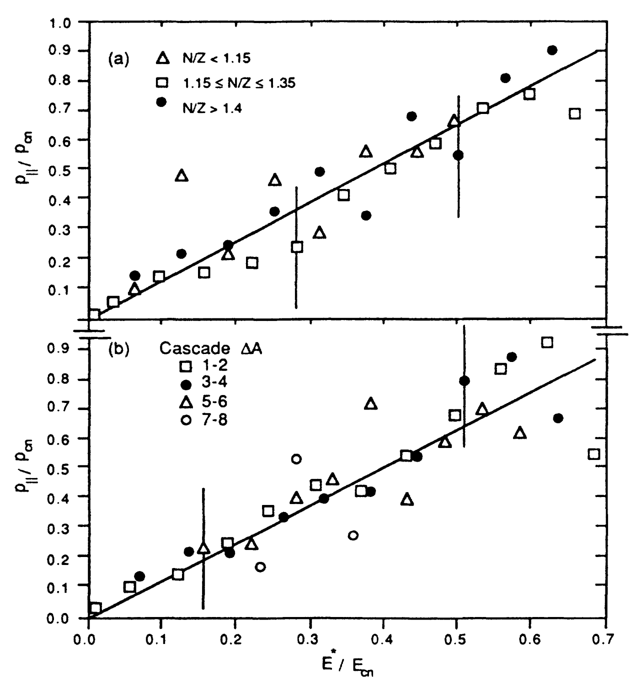

FIG. 3. Mean momentum $p_{\parallel}/p_{\rm cn}$ vs excitation energy $E^*/E_{\rm cn}$. (a) Grouped according to residue N/Z. (b) Grouped according to cascade length (ΔA). Straight lines are within regression error to the expected 0.8 proportionality of Eq. (1).

As an alternative explanation, we assessed the potential role of a target fragmentation excitation channel. The cross section for production of light fragments from ^{89}Y by 800-MeV protons was estimated in the following manner. From the data of Green *et al.* for light fragment production at forward angles from the interaction of 480-MeV protons and Ag, the integrated yield for $4 \leq A \leq 24$ is 46 mb.[11] The excitation function for this integrated cross section was assumed to be well represented by the excitation function of one of its members, ^{24}Na, which has been extensively investigated and is shown in Fig. 4 for Cu and Ag.[17] The sharply rising parallel excitation functions are characteristic of light fragments, and in going from 480 to 800 MeV amount to factor of 2.7±0.5. Interpolation of the yields between Cu and Ag was used to estimate the "800-MeV $p + {}^{89}\mathrm{Y} \rightarrow (4 \leq A \leq 24) + X$" yield as 150±25 mb.

Several additional observations are relevant. First, the light fragments themselves are known to peak sharply around $N/Z = 1$, with isobaric shapes insensitive to bombarding energy.[11] In the simplest picture, *if* each such light fragment has a complementary target fragment, *then* the latter is necessarily neutron rich. For example, ejection of ^{12}C from ^{89}Y leaves ^{83}As with $N/Z = 1.52$ cf. $(N/Z)_{\mathrm{stable}} = 1.31$. Consequently, light fragment emission is expected to correlate with the production of neutron-rich heavier products and, to a lesser extent, with those near stability as a result of evaporative deexcitation. (The fission channel, which is a serious interference for previous inclusive measurements from heavy targets, is not relevant to the current target-projectile system.) A key corrolary observation is that neutron-deficient spallation products are by and large unreachable by this fragment ejection channel despite earlier surmises to the contrary.[14]

Another observation concerning the significance of the above "rich-poor" dichotomy is that the total yield of ^{89}Y-spallation in the product region studied ($7 \leq \Delta A \leq 24$) is roughly 600 mb. Approximately half of this corresponds to near-stable and neutron-excess products. It follows that a 150±25 mb light fragment production cross section could affect ~50% of the neutron-rich spallation products.

At intermediate projectile energies, observed energetic light fragment emission has been associated by others[8] with inferred slow moving "spectator" or target fragments. That is, the major remnant of the target nucleus (equivalent to what we study) does not undergo significant interaction with the projectile. By implication, the fragmentation spectator would show *relatively* low F/B, particularly for near-stable and neutron-rich products as observed in this work. We repeat, though, that neutron-deficient products would not show this behavior.

The larger values of $\langle T \rangle$ for the neutron-excess products are also consistent with the target fragmentation scenario. Subsequent to the projectile-target collision, the kinetic energy associated with a binary breakup (fragmentation) process should be greater than that arising from a succession of small, randomly directed evaporative recoil kicks.

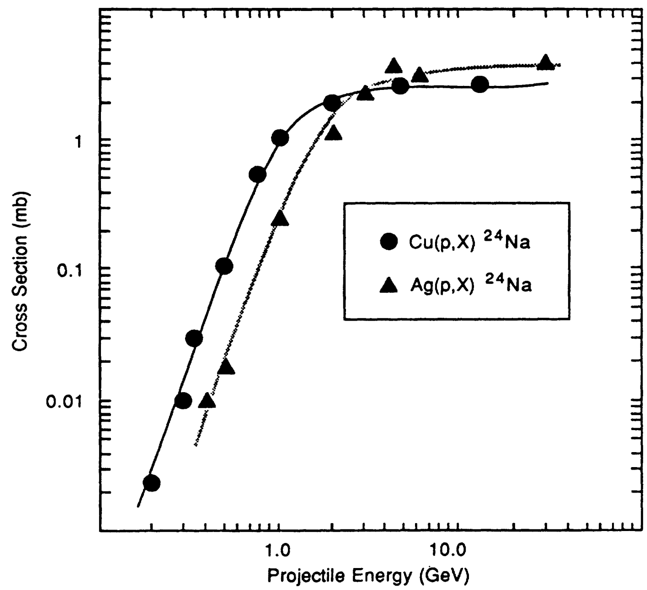

FIG. 4. Excitation functions for ^{24}Na from Cu (solid curve) and Ag (dashed curve), typical of light fragment production.

V. CONCLUSION

According to the evidence, we are arguing that the high F/B ratios observed for the neutron-deficient products are representative of the momentum imparted by the conventional nucleon-nucleon cascade. The lower F/B ratios for the neutron-excess and near-stable products, previously thought to be the norm, reflect significant contributions from fragmentation channels associated with the forward ejection of fast, light clusters. In fact, previous comparisons of the near-stable product recoil behavior to INC plus evaporation calculations indicated that the experimental F/B ratios were consistently smaller than the calculated ratios.[9,18,19] Since the existing generations of intranuclear cascade simulations do not incorporate light fragment emission, the continued indiscriminate application of the derived relationship between forward momentum transfer and excitation energy is of questionable validity. This is not entirely unexpected. Discussions in the literature have frequently warned that if light fragment emission were incorporated into the cascade codes, a different momentum-excitation energy relationship than Eq. (1) would be obtained.[2,9,14,20] That concern is deserved.

ACKNOWLEDGMENTS

This study was sponsored by the U.S. Department of Energy. The authors wish to acknowledge the hospitality and assistance of Drs. B. J. Dropesky, L. C. Liu, and G. C. Giesler of Los Alamos National Laboratory. One of us (M.J.T.) submitted this work in partial fulfillment for the Ph.D. degree, Department of Chemistry, Carnegie-Mellon University.

*Present address: Department of Radiation Health, University of Pittsburgh, Pittsburgh, PA 15261.

[1]R. Serber, Phys. Rev. **72**, 1114 (1947).
[2]W. G. Lynch, Ann. Rev. Nucl. Part. Sci. **37**, 493 (1987).
[3]L. M. Barbier, P. S. Freier, R. Holynski, W. V. Jones, A. Jurak, A. Olszewski, O. E. Pruet, C. J. Waddington, J. P. Wefel, B. Wilczynski, W. Wolter, and B. Wosiek, Phys. Rev. Lett. **60**, 405 (1988).
[4]W. Loveland, K. Aleklett, M. Bronikowski, Y. Y. Chu, J. B. Cumming, P. E. Haustein, S. Katcoff, N. T. Porile, and L. Sihver, Phys. Rev. C **37**, 1311 (1988).
[5]N. Sugarman, M. Campos, and K. Wielgoz, Phys. Rev. **101**, 388 (1956); N. T. Porile and N. Sugarman, *ibid.* **107**, 1410 (1957).
[6]L. Winsberg, Nucl. Instrum. Methods **150**, 465 (1978).
[7]N. T. Porile, Phys. Rev. **120**, 572 (1960).
[8]J. Hüfner, Phys. Rep. **125**, 129 (1985).
[9]S. B. Kaufman, E. P. Steinberg, and M. W. Weisfield, Phys. Rev. C **18**, 1349 (1978).
[10]N. T. Porile, D. R. Fortney, S. Pandian, R. A. Johns, T. Kaiser, K. Weilgoz, T. S. K. Chang, N. Sugarman, J. A. Urbon, D. J. Henderson, S. B. Kaufman, and E. P. Steinberg, Phys. Rev. Lett. **43**, 918 (1979); J. B. Cumming, P. E. Haustein, and R. W. Stoenner, Phys. Rev. C **33**, 926 (1986).
[11]R. E. L. Green, R. G. Korteling, and K. P. Jackson, Phys. Rev. C **29**, 1806 (1984), and references therein.
[12]J. A. Panontin and N. Sugarman, J. Inorg. Nucl. Chem. **25**, 1321 (1963).
[13]M. Lagarde-Simonoff and G. N. Simonoff, Phys. Rev. C **20**, 1498 (1979); M. Lagarde-Simonoff, S. Regnier, H. Sauvageon, and G. N. Simonoff, Nucl. Phys. **A260**, 369 (1976).
[14]Y. W. Yu and N. T. Porile, Phys. Rev. C **12**, 938 (1975).
[15]K. Chen, Z. Fraenkel, G. Friedlander, J. R. Grover, J. M. Miller, and Y. Shimamoto, Phys. Rev. **166**, 949 (1968).
[16]G. D. Harp, K. Chen, Z. Fraenkel, G. Friedlander, J. M. Miller, Phys. Rev. C **8**, 581 (1973).
[17]J. Hudis, in *Nuclear Chemistry,* edited by L. Yaffe (Academic, New York, 1968), Vol. I, and references therein.
[18]N. T. Porile and S. Tanaka, Phys. Rev. **135**, B122 (1964).
[19]J. Panontin, N. T. Porile, and A. A. Caretto, Jr., Phys. Rev. **165**, 1281 (1968).
[20]Ø. Scheidemann and N. T. Porile, Phys. Rev. C **14**, 1534 (1976).