are emitted isotropically within a cone of ± 45° opening around the beam axis, a rough estimate for the splitting probability was 4%.

The interesting question is whether the splitting of the Kr-like products is a fast nonequilibrium decay or a sequential decay of an equilibrated nucleus. From the data of Fig. 2, the first hypothesis seems more likely. The fact that for comparable excitation energies (same TKEL* bins) the presence of the splitting products is observed at 12.1 MeV/u but does not appear in the distributions at 8.2 MeV/u is hard to understand in a pure statistical picture. An explanation based on the sequential fission of the Kr-like nuclei would require a drastic decrease of the fission barrier due to an increased angular momentum dissipation at the higher bombarding energy. This however is not predicted by model calculations[15] based on the γ-multiplicity experiments at lower bombarding energies.[7]

In conclusion the new feature emerging from the reaction ^{86}Kr-^{166}Er at a bombarding energy of 12.1 MeV/u is the enhanced production of elements lighter than the projectile, which at low-energy losses appears as a skewness of the element distributions and at high-energy losses develops into a pronounced shift of the maximum. The experimental results can be understood in terms of a dynamical splitting of the projectile-like fragments which prevails over the emission of light charged particles. This suggests that in DIC between heavy nuclei at high incident energies a considerable fraction of the dissipated energy is at first concentrated in few collective degrees of freedom, rather than in a small local region (hot spot).

We thank G. Augustinski, H. Daues, H. J. Beeskow, and M. Ludwig for their assistance in preparing the detection system, H. Folger for the target preparation, and the operators of the Unilac accelerator for the production of an excellent Kr beam of 12.1 MeV/u.

[(a)] On leave from Texas A & M University, College Station, Tex. 77840.

[(b)] On leave from the Institute of Nuclear Research, Swierk, Poland.

[1] Y. Eyal *et al.*, Phys. Rev. Lett. 41, 625 (1978).
[2] B. Tamain *et al.*, Nucl. Phys. A330, 253 (1979).
[3] W. U. Schröder *et al.*, Ann. Rev. Nucl. Sci. 27, 465 (1977).
[4] H. Sann *et al.*, in *Proceedings of the Topical Conference on Heavy-Ion Collisions, Fall Creek Falls State Park, Tennessee, 1977*, CONF-770602 (National Technical Information Services, Springfield, Va., 1977).
[5] G. Rudolf *et al.*, Nucl. Phys. A330, 245 (1979).
[6] C. K. Gelbke *et al.*, Phys. Lett. 70B, 415 (1977).
[7] A. Olmi *et al.*, Phys. Rev. Lett. 41, 688 (1978).
[8] M. Dakowski *et al.*, to be published.
[9] H. Sann *et al.*, Nucl. Instrum. Methods 124, 509 (1975).
[10] U. Lynen *et al.*, Nucl. Instrum. Methods 162, 657 (1979).
[11] P. Doll *et al.*, Gesellschaft für Schwerionenforschung Annual Report No. 79-11, 1978 (unpublished).
[12] D. E. Greiner *et al.*, Phys. Rev. Lett. 35, 152 (1975).
[13] Y. P. Viyogi *et al.*, Phys. Rev. Lett. 42, 33 (1979).
[14] U. Lynen, in Proceedings of the Symposium on Heavy Ion Physics from 10 to 200 MeV, Upton, New York, 1979 (to be published), and to be published.
[15] G. Wolschin *et al.*, Phys. Rev. Lett. 41, 691 (1979), and private communication.

Observation of Selected Molecular-Orbital X Rays in Coincidence with Separated-Atom *K* X Rays

J. J. O'Brien, E. Liarokapis, and J. S. Greenberg
A. W. Wright Nuclear Structure Laboratory, Yale University, New Haven, Connecticut 06520
(Received 20 June 1979; revised manuscript received 7 December 1979)

A cascade relationship between molecular-orbital (MO) and separated-atom x rays has been utilized to study selected MO transitions. Depending on the mechanism of vacancy production, the observed MO spectra are interpreted as reflecting $2p\sigma \rightarrow 1s\sigma$ transitions or a mixture of $2p\sigma \rightarrow 1s\sigma$ and $2p\pi \rightarrow 1s\sigma$ transitions. The coincidence selection also directly corroborates a previous assignment of MO transitions to the $2p\sigma$ state.

Although the study of molecular-orbital (MO) x rays is expected to provide one of the more direct sources of information on quasimolecular states formed in heavy-ion collisions, the acquisition of such information has been severely limited by the lack of distinctive features in MO x-ray spectra.[1] Moreover, since previous experiments have been confined to the observation of

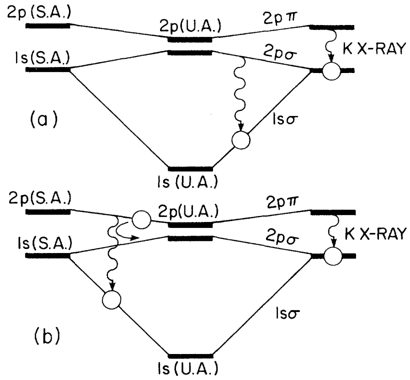

FIG. 1. Schematic correlation diagrams showing (a) the cascade relationship between $2p\sigma \to 1s\sigma$ MO transitions and SA K x rays. (b) Also shown are the effects of rotational coupling which may lead to ($2p\pi \to 1s\sigma$, K x ray) cascades if $1s\sigma$ vacancies are present in the incoming part of the collision, as in two-collision MO x-ray production.

the total spectrum, any conventional one-to-one spectroscopic correlation between energy levels in the quasimolecule and specific radiative transitions has yet to be established. This Letter reports on a method which isolates selected MO transitions. To illustrate the method, we present results from measurements on the Nb-Nb collision system which provide the first direct association of a selected part of the MO spectral profile with specific transitions in the quasimolecule.

The principle of the method is illustrated in Fig. 1(a). In the MO description of heavy-ion-atom collisions, the $2p\sigma$ orbital correlates to the 1s level of the separated atom (SA). Excluding, for the time being, a consideration of dynamic couplings which may rearrange vacancies after the MO transition, a $2p\sigma \to 1s\sigma$ MO transition will lead to a 1s SA vacancy. Thus a cascade relationship is anticipated between $2p\sigma \to 1s\sigma$ MO transitions and SA K x rays. Transitions from other levels to the $1s\sigma$ level as well as transitions filling vacancies in the $2p\sigma$ or lesser bound states do not lead to a 1s SA vacancy. A coincidence requirement between MO and K x rays, therefore, separates out $2p\sigma \to 1s\sigma$ transitions from the total MO x ray spectrum. In principle this method can be extended to higher-lying shells.

Several important considerations can modify this straightforward interpretation. As noted above, dynamic couplings between MO states may alter the cascade sequence. For example, in the event that the MO transition occurs before the nuclei reach their minimum separation, as in two-collision MO x-ray production where a K-shell vacancy is brought into the second collision,[2] rotational coupling of the $2p\sigma$ and $2p\pi$ orbitals at small internuclear separations will lead to a ($2p\pi \to 1s\sigma$, K x ray) cascade sequence [Fig. 1(b)]. Similarly, radial coupling involving the $2p\sigma$ state can contribute to a mixing of transitions in the coincidence spectrum. It is also clear that the strict exclusion of transitions to the $2p\sigma$ and higher-lying MO states in the coincidence spectrum can be modified by a dynamic rearrangement of vacancies. These considerations as well as other background coincidence events will be discussed further in connection with the data presented below.

A series of (x ray, x ray) coincidence experiments were carried out using 100-, 160-, and 200-MeV ^{93}Nb beams on ~500-μg/cm^2 ^{93}Nb target. The continuum x rays were detected with 1000-mm^2 planar hyperpure germanium detector with a suitable absorber to reduce the characteristic x-ray intensity, and the K x rays were monitored with a NaI detector. The principal difficulty encountered in these measurements originates with the very small (10^{-6} to 10^{-8}) continuum x-ray yields per keV per K x ray emitted. However, even with large counting rates of ~10^5 counts/sec in the K x-ray counter, necessitated by acceptable data-accumulation rates, most of the measurements were carried out with true coincidence counting rates exceeding the chance rates by an order of magnitude in the interesting regions of the continuum spectrum.

Figure 2(a) displays the x-ray spectrum observed in coincidence with K x rays and compares it with a singles spectrum. A high-energy continuum is observed in coincidence with K x rays. Such a continuum, indeed, is anticipated because of the cascade relationship between transitions to the $1s\sigma$ MO and K x rays. Furthermore, by normalizing the coincidence and singles spectra at the high-energy region where they possess very similar shapes, it becomes evident that the rapid increase observed in the continuum of the singles spectrum below ~35 keV is largely missing in the coincidence measurement. Again, the exclusion of these radiations in the coincidence spectrum is expected if they are linked with MO transitions to higher-lying molecular orbitals such as to the $2p\sigma$ MO.[3]

These qualitative observations seem to confirm the anticipated selective properties of the coincidence requirement. However, before a direct association with MO transitions can be made,

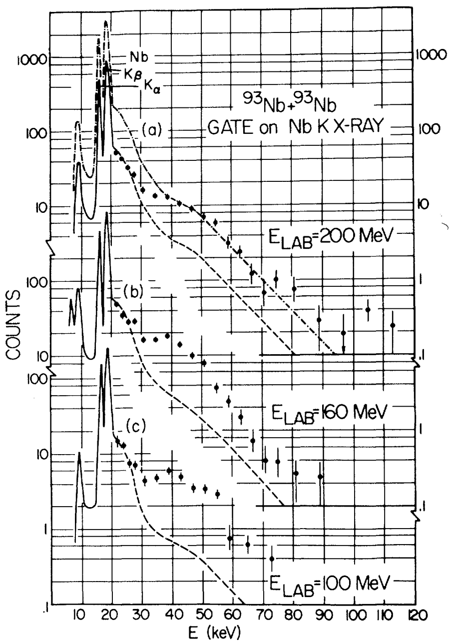

FIG. 2. Solid lines and data points show spectra of x rays in coincidence with SA K x rays. Solid lines and their continuation into the dashed curves represent singles spectra and reflect contributions to the coincidence spectra from multiple-vacancy processes. Above 30 keV the data have been averaged over twenty channels. The dot-dashed curve in (a) shows the singles spectrum normalized to the coincidence data in the region 40–70 keV.

other mechanisms leading to coincidences between continuum x rays and K x rays have to be examined. Continuum x-ray sources such as nucleus-nucleus bremsstrahlung[4] and radiative ionization[5] were found not to produce any significant coincidence background. On the other hand, a more significant background is implied by the presence of K-K x ray coincidence events in the data. These reflect both sequential K vacancy production by a single projectile in different independent collisions in the solid target and multiple-vacancy production in a single collision. Multiple vacancies can also generate coincidences between MO and K x rays through one of the vacancies ($1s\sigma$ or $2p\sigma$) decaying via a MO transition, K or L, while another emits a separated-atom K x ray. For these events the two x rays do not necessarily form a cascade sequence and may include MO transitions from higher-lying states. As demonstrated below, however, the magnitude of this background depends sensitively on the bombarding energy so that it can be reduced to a small fraction of the coincidence spectrum at the lowest bombarding energy used in these measurements.

A measure of this MO x-ray background is obtained by noting that with the singles spectrum normalized to the K-K x-ray coincidence yield, both the spectral distribution and the intensity of the singles continuum spectrum closely reflect the MO x-ray coincidence background. This simple correspondence is exact for the fraction of coincidences originating with multiple $2p\sigma$ and $1s\sigma$ vacancies formed in sequential collisions, since the vacancies each decay with the characteristic singles spectral distribution. Measurements of the K-K x-ray coincidence yield as a function of target thickness have shown that for a target thickness of 500 μg/cm^2 sequential collisions contribute approximately 50% of the double-vacancy production at the bombarding energies utilized.[6] For the background due to multiple-vacancy production in a single collision, consideration has to be given to the MO x-ray production mechanism and to the influence of the impact-parameter dependences for single- and multiple-vacancy production on the shapes of the singles and coincidence MO spectra and on the MO to K x-ray intensity ratios. Although the experimental information is not available to assess these questions in detail, only small differences are expected between the normalized singles and coincidence photon distributions in this case also. Differences in the spectral shape due to these effects are largely suppressed by the large dynamical broadening, particularly at high photon energies. Differences in the ratio of MO to K x-ray intensities in the two cases were explored through calculations using impact-parameter dependences scaled from measurements[7] to evaluate single- and double-vacancy probabilities and also through further coincidence measurements[6] which examined double-vacancy production, $(2p\sigma^{-2})$ and $(1s\sigma^{-1}, 2p\sigma^{-1})$, in a number of symmetric and asymmetric collision systems formed with Ni and Nb projectiles. These considerations indicate that the normalized singles spectrum closely reflects the intensity of coincident MO transitions to the $2p\sigma$ level due to $(2p\sigma^{-2})$, while also accounting for the background from transitions filling $1s\sigma$ vacancies within a factor <2. Therefore, with the background from all sources of multiple-vacancy production comprising only a small fraction of the total coincidence intensity at the low bombarding energies where this coincidence technique is most useful, uncertainties in utilizing the normalized singles spectrum to represent this background have only a minor effect on the background-subtracted MO x-ray yield.

Figure 2 displays data obtained at three bombarding energies together with the background determined using the singles spectral distribution normalized to the K-K x-ray coincidence yield. The ratio of the measured coincidence events to the background increases from 4 to 12 as the bombarding energy decreases from 200 to 100 MeV. This trend reflects the more rapid increase with projectile energy observed for double-vacancy production compared with single-vacancy production. It is evident that such a background subtraction can account for essentially all the events in the photon energy region between 20–30 keV. Even the small fraction of the prominent intensity increase observed below ~ 35 keV in the singles spectrum which remains in the coincidence spectrum is attributable to multiple-vacancy processes. As previously noted this is expected if this region of the singles spectrum is dominantly associated with the transitions to the $2p\sigma$ level.[3] The region between 30 and 70 keV, however, cannot be accounted for by a plausible multiple-vacancy-related background which would not reflect unreasonably large deviations from the singles spectral distribution or would not simultaneously greatly overestimate the events in the lower-energy region. Therefore it is concluded that an excess of coincidence events between 30 and 70 keV remains above this background, particularly at lower bombarding energies, corresponding to cascade emission following the production of a single $1s\sigma$ vacancy. From a comparison of the MO singles and coincidence spectra we find that the cascade MO x rays constitute, respectively, 20% to 25% of the total MO spectrum in the range 30 to 70 keV as the projectile energy increases from 100 to 200 MeV.

Beyond the identification of the high-energy continuum in the coincidence spectra with transitions to the $1s\sigma$ level, its unique association with the particular $2p\sigma \rightarrow 1s\sigma$ transition requires that there is no dynamic rearrangement of electrons from the $2p\sigma$ MO into the level vacated by the MO transition. The absence of transitions to the $2p\sigma$ state in the cascade spectrum suggests that the rearrangement of electrons in a one-collision MO x-ray emission process is not an important effect to consider in this connection. To the extent the two-collision mechanism also contributes to K MO x-ray emission in the Nb-Nb system, vacancy transfer by $2p\sigma - 2p\pi$ rotational coupling can admix $2p\pi \rightarrow 1s\sigma$ transitions into the coincidence spectrum [see Fig. 1(b)]. This aspect of the spectrum's composition is yet to be resolved in detail, since the relative contributions from one- and two-collision processes to the spectral profile are not known quantitatively, presently, for our collision conditions. A measure of this admixture can be obtained by estimating the two-collision fraction of the total measured MO spectrum from a scaling law,[8] and using scaled $2p\sigma - 2p\pi$ coupling probabilities[7] in a quasistatic-type calculation[2] of the two-collision MO x-ray emission process. Such considerations indicate that $2p\pi \rightarrow 1s\sigma$ transitions constitute the major fraction of the cascade events detected, with the $2p\pi \rightarrow 1s\sigma$ component attaining its maximum value of ~ 25% near the midpoint of the spectrum and decreasing to a few percent at the MO spectrum limits, essentially independent of the bombarding energies studied.

In summary, these experiments have demonstrated that MO transitions to the $1s\sigma$ state can be selected from the total MO spectrum in a coincidence measurement with K x rays. With only one or two specific transitions to consider, meaningful tests of many aspects of the theory then become possible. For example, the isolation of MO transitions clearly would be advantageous in studies of the emission anisotropy for MO radiation,[9] which in turn could provide a direct measurement of the $2p\pi \rightarrow 1s\sigma$ contribution to the coincidence spectrum.

We wish to thank T. Zouros, J. Schweppe, J. Thomas, and P. Vincent for their participation in this work. We also acknowledge informative communications with R. Anholt, B. Müller, and J. Kirsch regarding the calculation of MO spectra and dynamic coupling effects. This work was supported by the U. S. Department of Energy, Contract No. EY-76-C-02-3074.

[1] W. E. Meyerhof, T. K. Saylor, S. M. Lazarus, W. A. Little, B. B. Triplett, and L. F. Chase, Jr., Phys. Rev. Lett. 30, 1279 (1973); C. K. Davis and J. S. Greenberg, Phys. Rev. Lett. 32, 1215 (1974).

[2] W. E. Meyerhof, T. K. Saylor, S. M. Lazarus, A. Little, B. B. Triplett, L. F. Chase, Jr., and R. Anholt, Phys. Rev. Lett. 32, 1279 (1974).

[3] K. H. Heinig, H. U. Jager, H. Richter, and H. Woittennek, Phys. Lett. 60B, 249 (1976).

[4] H. P. Trautvetter, J. S. Greenberg, and P. Vincent, Phys. Rev. Lett. 37, 202 (1976); J. Reinhardt, G. Soff, and W. Greiner, Z. Phys. A 276, 285 (1976).

[5] D. H. Jakubassa and M. Kleber, Z. Phys. A 273, 29

(1975).

[6]J. J. O'Brien, E. Liarokapis, and J. S. Greenberg, to be published.

[7]K. Taulbjerg, J. S. Briggs, and J. Vaaben, J. Phys. B 9, 1351 (1976); J. U. Andersen, E. Laegsgaard, M. Lund, and C. D. Moak, Nucl. Instrum. Methods 132, 507 (1976).

[8]R. Anholt, Z. Phys. A 289, 41 (1978), and private communication.

[9]J. S. Greenberg, C. K. Davis, and P. Vincent, Phys. Rev. Lett. 33, 473 (1974); G. Kraft, P. H. Mokler, and H. J. Stein, Phys. Rev. Lett. 33, 476 (1974); W. Wölfli, Ch. Stoller, G. Bonani, M. Stöckli, and M. Suter, Phys. Rev. Lett. 36, 309 (1976).

Diabatic Field Ionization of Highly Excited Sodium Atoms

T. H. Jeys, G. W. Foltz, K. A. Smith, E. J. Beiting,[(a)] F. G. Kellert, F. B. Dunning, and R. F. Stebbings
Department of Space Physics and Astronomy, Rice University, Houston, Texas 77001
(Received 11 September 1979)

Data demonstrating that, contrary to earlier results, laser-excited sodium Rydberg atoms may pass from low-electric field strengths to ionizing electric field strengths along two quite distinct paths is presented. One path involves predominantly adiabatic passage while the other path involves predominantly diabatic passage. Data are presented along with a model that supports this conclusion.

Electric field ionization has become a common technique for the detection and study of highly excited atoms. Proper application of the technique requires an understanding of the possible results of applying an increasing electric field to an atom.

We report the first observation of a completely new feature in the field ionization spectrum of highly excited states of sodium. All previous work on sodium Rydberg states (cf., Gallagher *et al.*[1] in the region $n=15-20$, and Vaille and Duong[2] in the region $n=20-54$) indicates that the path taken to ionization by such atoms in an increasing electric field is primarily adiabatic (i.e., states of the same $|m_l|$ quantum number undergo avoided crossings). We report new observations of sodium atoms excited to high-Rydberg d states ($|m_l|=2$) whose passage, from small electric fields to ionizing electric fields, is primarily diabatic. Data demonstrating both adiabatic and diabatic ionization thresholds are presented along with a consistent model for predicting the field strengths for each.

In the present experiment, excited sodium atoms are produced in zero electric field in a magnetically shielded region. A sodium beam is intersected at right angles by the output of two simultaneously pumped pulsed dye lasers. One laser beam excites the $3^2S_{1/2}-3^2P_{3/2}$ transition ($\lambda=5890$ Å) while the other laser beam excites the $3^2P_{3/2}-n^2S_{1/2}$ or $3^2P_{3/2}-n^2D_{3/2,5/2}$ transition ($\lambda\simeq4100$ Å). The excited atoms are ionized by a pulsed electric field ~3 μsec after excitation. (The laser polarizations are perpendicular to the direction of this field.) The electric field strength, which rises from 0 to 1100 V/cm in ~1 μsec, is generated between two grids parallel to the plane of the sodium and laser beams and centered about their intersection. The electrons liberated at ionization are detected by an electron multiplier whose output goes to a time-to-amplitude converter (TAC). The TAC is started at the beginning of the ionizing voltage ramp and is stopped by the first electron pulse subsequently registered by the detector. The TAC output is fed into a standard multichannel pulse analyzer. For sufficiently low count rates (<0.1 per laser shot) the multichannel analyzer stores data proportional to the probability of a field ionization event per unit time during the 1 μsec ramp. Measurement of the time dependence of the ionizing voltage waveform permits determination of the field strengths at which the ionization events occur. It is necessary to allow for delays associated with electron flight time ($\simeq$6 nsec), transit time in the electron multiplier (~25 nsec), and cables and amplifiers (~30 nsec). The uncertainties in delay measurement and instrument calibrations and in our estimation of field penetration in the interaction region combine to yield a ±5% net uncertainty in the ionization field strength determination.

Figure 1(a) illustrates our field ionization data for sodium d states with $n=30$, 32, 34, and 36. There are two major ionization thresholds in each profile, one at low electric fields and one at

