already partly occupied shell of ^{94}Mo and ^{96}Mo. This is in agreement with results from muonic atoms[4,9,10] and measurements of isotope shifts of electronic atoms.[11,12]

This effect may also be seen from the differences of charge distributions in Fig. 4. Here the charge distribution differences $\Delta\rho(r)$ times r^2 of neighboring isotopes are compared to that of ^{94}Mo and ^{92}Mo (shaded areas). The addition of the two neutrons to the empty $2d_{5/2}$ shell of ^{92}Mo results in a polarization of the protons such that *more* charge from the inner region of the nucleus is moved *further* outward than by adding two neutrons to the already partially filled shell. In going from ^{98}Mo to ^{100}Mo another shell ($3s_{1/2}$) is filled and results in a noticeably different shape for $r^2\Delta\rho(r)$; it is somewhat broader than that of ^{92}Mo and ^{94}Mo, but coincides with it at large radii.

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Anomalous Vibrational-State Distribution in N_2^+ ($B^2\Sigma_u^+$) after Charge Exchange of He_2^+ with N_2†

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We measure cross sections for N_2^+ ($B^2\Sigma_u^+$) production in 5-eV He_2^+-N_2 and He^+-N_2 collisions by observation of emission in the first negative system of N_2^+. For He_2^+ the cross section is unusually large (~ 10 Å^2) and the vibrational-state distribution shows primarily $v'=0$ and 1, consistent with vertical ionization of N_2. He^+ produces a much wider distribution, as do most other simple ions. Explanations for the large cross section and observed state distributions are proposed.

Production of excited N_2^+ in interactions of N_2 with helium and its ions (He^+ and He_2^+) has been studied extensively, primarily by observation of luminescence from a discharge in a mixture of the gases.[1] Recently a nitrogen-ion laser, operating at 3914, 4278, and 4709 Å in the first negative (1N) system of N_2^+ ($B^2\Sigma_u^+ \to X^2\Sigma_g^+$), has been reported[2] and, since this laser is pumped by the reaction

$$He_2^+ + N_2 \to N_2^+(B^2\Sigma_u^+) + 2He, \qquad (1)$$

these charge-transfer processes take on additional importance. This paper reports the results of experiments in which spectra (1900–8500 Å) from the products of He^+-N_2 and He_2^+-N_2 collisions at 5 eV were separately obtained. Since these experiments were performed with a mass-selected primary ion beam impinging on N_2 gas at low pressure, microscopic details of the collision processes are obtained. Absolute cross sections were measured for the most prominent features of the spectra, and the cross section for He_2^+ charge exchange yielding product N_2^+ (B) is roughly two orders of magnitude greater than the analogous He^+-N_2 process. Higher-resolution studies of the $\Delta v = 0$ and $\Delta v = -1$ sequences of N_2^+ $(1N)$ show that for He_2^+-N_2 collisions N_2^+ $[B^2\Sigma_u^+(v'=0)]$ is the predominant product while He^+-N_2 collisions yield a distribution extending beyond $v' = 10$. For He_2^+-N_2, the distribution is approximately that predicted from Franck-Condon factors[3] between the target N_2 $[X^1\Sigma_g^+(v=0)]$ and vibrational levels of N_2^+ (B); this observation is contrary to the results of most studies of charge exchange in ion-neutral collisions[4] below ~10 keV.

The apparatus[5] provides an electron-impact-produced, magnetically mass-selected ion beam directed into a collision cell containing N_2 gas at 1.5 mTorr. For He^+ the electron impact energy was maintained below ~50 eV ensuring ground-state ions; He_2^+ ions were formed under conditions[6] which yield primarily $X^2\Sigma_u^+$ $(v=0)$. Photons from the collision cell enter a Jarrell-Ash 0.25-m scanning monochromator with interchangeable 1180-line/mm gratings blazed at 3000 and 5000 Å; filters were used, where appropriate, to suppress second-order lines. The detector was an EMI 9659 QAM photomultiplier tube and single-photon counting was employed. Complete spectra were assembled by computer normalization of overlapping portions of adjacent scans; data were corrected for the efficiency of the detector and for grating, window, and filter transmissions.

Figure 1 shows spectra obtained with 20-Å resolution (half width at half-maximum). Ordinates, U, are in the same units, photons per ampere per second. Cross sections, σ, for processes which yield intense emission in the He_2^+ system are indicated. These cross sections were calibrated by observation of $H\alpha$ emission from 100-eV He^+-H_2 collisions and normalization to the value reported by Isler and Nathan.[7] For comparison, emission which appears to result from the $\Delta v = +1$ sequence in the He^+ system is shaded; the cross section was calculated after subtrac-

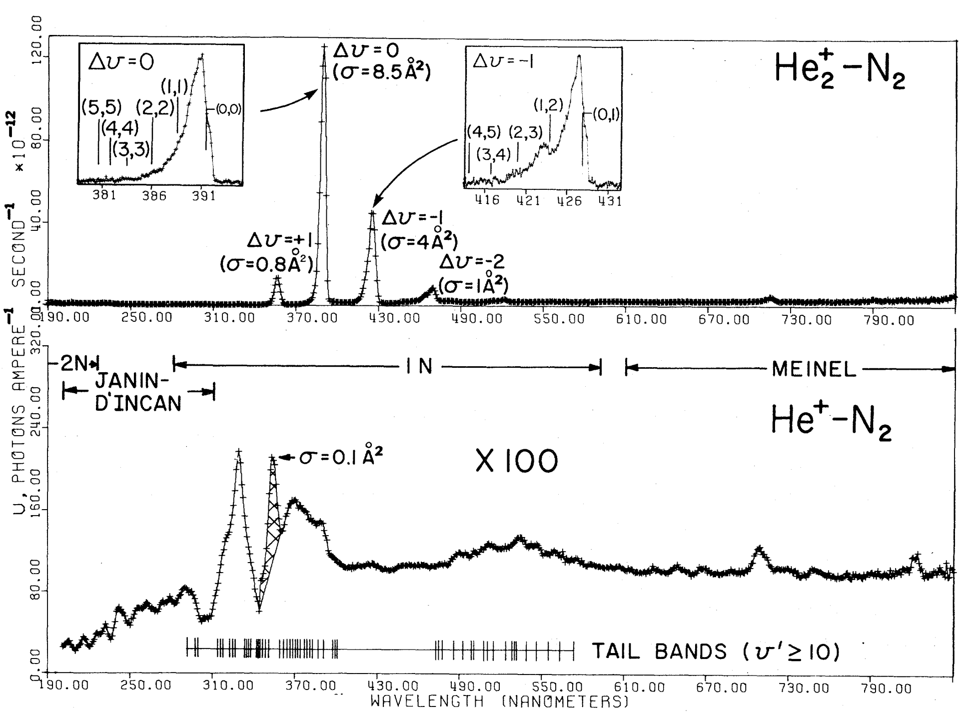

FIG. 1. Emission spectra from He_2^+-N_2 and He^+-N_2 collisions at 5 eV taken with 20-Å resolution. The insets in the He_2^+-produced spectrum are higher resolution scans of the $\Delta v = 0$ and -1 sequences of N_2^+ $(1N)$. Band heads are from Ref. 1c. The known emission systems of N_2^+ are indicated in the lower spectrum.

tion of the intensity which appears to be from other transitions. The pertinent lifetimes[8] are short enough to permit the assumption that all decays occur in the detection region. Aside from the large difference in the luminescence cross sections, the He_2^+-N_2 spectrum shows few prominent features other than the indicated sequences of N_2^+ (1*N*), while He^+ impact produces the 2*N* system as well as a number of previously reported, unresolved N_2^+ emissions.[9,10] Also present in the He^+-N_2 spectrum are "tail bands" of N_2^+ (1*N*). These bands, which originate in high vibrational levels of the N_2^+ *B* state ($v' > 10$), have been observed by Brandt, Ottinger, and Simonis[4a] in low-energy Ne^+-N_2 collisions but not from Ar^+-N_2 collisions. They attributed the tail bands in the Ne^+-N_2 spectrum to the high recombination energy of Ne^+ (21.6 eV), which permits population of the necessary v' levels; observation of tail bands in the present work is consistent with this interpretation since He^+ has a comparably high recombination energy (24.6 eV).

There is a significant difference between the two processes studied here. The repulsive He-He potential causes a three-body final state for the He_2^+-N_2 reaction; thus the products have available more phase space than the two-body final state of the He^+-N_2 process. The energetics are favorable for selective population of N_2^+ (*B*) by He_2^+; Fig. 2 shows pertinent states[11] of N_2, N_2^+, and He_2 ($a^3\Sigma_u^+$),[1c] the repulsive He-He potential,[12] and a Morse curve[13] for He_2^+. Vertical acceptance of an electron by He_2^+ to the He-He repulsive curve has an effective recombination energy in the range 18.3–20.3 eV, with maximum probability at about 19 eV, resonant with low vibrational levels of N_2^+ (*B*). The He^+ recombination energy on the other hand is fixed at 24.6 eV, not having the latitude provided by the repulsive He-He potential. (Note that the ground state of He lies so deep that production of excited He or He_2 is prohibited at 5-eV translational energy.) The variable recombination energy covering the resonant range probably accounts for the unusually high cross section and may play a crucial role in the nitrogen-ion laser.

Previous studies of vibrational-state distributions[4] of N_2^+ (*B*) produced by charge transfer with a variety of positive ions at energies below several keV have shown that in addition to $v' = 0$ and 1, $v' = 2$, 3, and 4 are also significantly populated. This is in contrast to state distributions predicted by a model which assumes vertical ionization of N_2 [$X^1\Sigma_g^+ (v = 0)$] to N_2^+ (*B*). Franck-Condon factors[3] indicate ~90% population of $v' = 0$, ~10% of $v' = 1$, and negligibly small amounts in higher vibrational levels. The ions used in the earlier work, unlike He_2^+, all produced a single, bound neutral after electron transfer. Formation of higher v' levels in slow collisions with ions has been attributed to distortion of the target N_2 molecule in the field of the ion, causing a

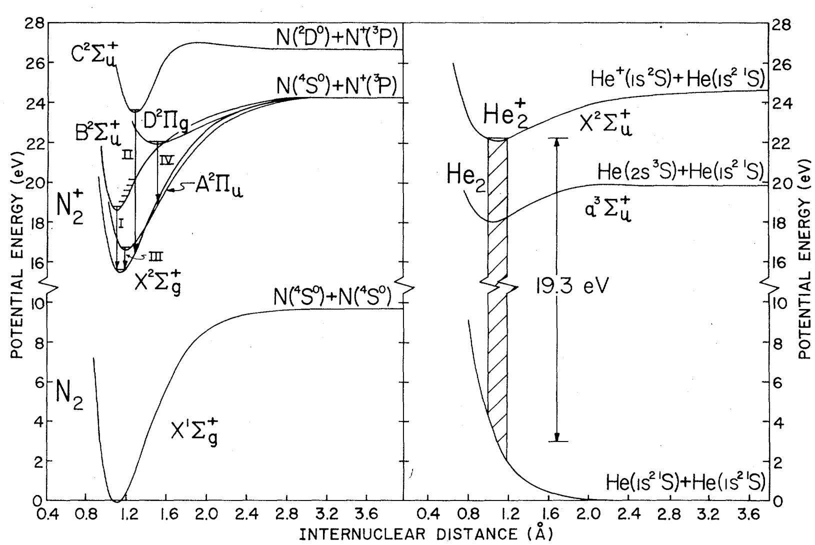

FIG. 2. Potential energy curves for N_2 and N_2^+ (Ref. 11) and He_2 ($a^3\Sigma_u^+$) (Ref. 1c), a Morse potential for He_2^+ (Ref. 13), and the repulsive He-He potential (Ref. 12). The known band systems of N_2^+ are indicated with the Roman numeral designations of Ref. 1c. I, $B \to X$(1*N*); II, $C \to X$(2*N*); III, $A \to X$(Meinel); IV, $D \to A$(Janin-d'Incan).

shift in the equilibrium internuclear separation of N_2 appropriate for calculation of overlap integrals.[4,14] The ratio of cross sections for N_2^+ (B) production in the two systems studied here suggests an effective impact parameter for He_2^+ that is roughly ten times that for He^+, and since the ion-induced dipole potential is proportional to r^{-4}, electron transfer to He_2^+ will occur when the N_2 is essentially unpolarized. Thus the state distribution is expected to be consistent with Franck-Condon transitions from an undistorted nitrogen molecule. The insets in Fig. 1 show the $\Delta v = 0$ and $\Delta v = -1$ sequences of N_2^+ ($1N$) from He_2^+-N_2 collisions under higher resolution (4 Å). Clearly, $v' = 0$ is preferentially populated; a small fraction of $v' = 1$ and an even smaller fraction of $v' = 2$ also occur. This is in sharp contrast to the Ar^+-N_2 experiments of Brandt, Ottinger, and Simonis[4a] who found essentially equal contributions to $\Delta v = 1$ from each of the (0,1), (1,2), (2,3), (3,4) and (4,5) bands at 500 eV. Moore and Doering[4b] observed a ratio of the first four of these bands of approximately 4:3:2:1 in 300-eV H_2^+ collisions. The He^+ experiments reported here show population of even higher vibrational levels. It may be concluded that the differences in the magnitudes of the cross sections satisfactorily account for the differences in the observed state distributions.

The higher-resolution studies shown in Fig. 1 separate the contributions from $v' = 0$ and $v' = 1$. This provides a lower limit (since $\Delta v = -2$ was ignored) of 11.9 Å^2 for the cross section for production of N_2^+ [$B^2\Sigma_u^+(v' = 0)$] in He_2^+ collisions at 5 eV.

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