for the analyzing powers of the first excited states of ^{26}Mg and ^{22}Ne. On this basis the experimental data in the region 10°–35° strongly suggest transfers which are predominantly $d_{5/2}$. In fact the data points in this region lie close to those measured for the ^{26}Mg ground state. Since the DWBA predictions for a pure $d_{5/2}$ transfer do not alter much in moving from the ground state to the first excited states, this also suggests that the amount of $d_{3/2}$ admixture is small for these transitions. Finally it should be noted that for any $d_{3/2}$ admixture greater than 40% the predicted analyzing power is opposite in sign to the measured powers.

In conclusion, the results presented here strongly suggest that the admixture of $d_{3/2}$ into $d_{5/2}$ transfer for proton pickup to the 1.81-MeV (2^+) state of ^{26}Mg and 1.27-MeV (2^+) state of ^{22}Ne is small. This result is in contrast with the possibility raised by Clement, Georgiev, and Perez[3] that in the case of ^{22}Ne the admixture might be $\gtrsim 40\%$.

One of us (N.E.S.) would like to acknowledge valuable discussions with Dr. C. F. Clement. The support of Professor G. C. Morrison for this work is also acknowledged.

---

[1] C. F. Clement, Nucl. Phys. A213, 469, 493 (1973).

[2] C. F. Clement and S. M. Perez, J. Phys. A 7, L93 (1975).

[3] C. F. Clement, B. Georgiev, and S. M. Perez, J. Phys. G 2, L77 (1976).

[4] M. Arditi, L. Bimbot, H. Doubre, N. Frascaria, J. P. Garron, M. Riov, and D. Royer, Nucl. Phys. A165, 129 (1971).

[5] A. K. Basak, J. A. R. Griffith, M. Irshad, O. Karban, E. J. Ludwig, J. M. Nelson, and S. Roman, Nucl. Phys. A229, 219 (1974).

[6] J. M. Nelson and B. E. F. Macefield, Birmingham University Report No. 74-9 (unpublished).

[7] A. A. Debenham, J. A. R. Griffith, M. Irshad, O. Karban, and S. Roman, Nucl. Phys. A167, 289 (1971).

[8] J. E. McQueen, J. M. Joyce, and E. J. Ludwig, Nucl. Phys. A151, 295 (1970).

[9] P. M. Endt and C. van der Leun, Nucl. Phys. A214, 144 (1973).

[10] B. Mayer, H. E. Conzett, W. Dahme, D. G. Kovar, R. M. Larimer, and Ch. Leeman, Phys. Rev. Lett. 32, 1452 (1974).

[11] H. Breuer *et al.*, in Jahresbericht 1975, Max-Planck-Institut für Kernphysick, Heidelberg (unpublished), p. 77.

Strong Molecular Effects in Heavy-Ion–Induced Carbon and Nitrogen *K* Auger Transitions

R. Mann
Gesellschaft für Schwerionenforschung, D-6100 Darmstadt, West Germany, and Institut für Kernphysik der Universität, D-6000 Frankfurt am Main 90, West Germany

and

F. Folkmann
Gesellschaft für Schwerionenforschung, D-6100 Darmstadt, West Germany

and

K.-O. Groeneveld
Institut für Kernphysik der Universität, D-6000 Frankfurt am Main 90, West Germany

(Received 8 October 1976)

K Auger electrons from C bound in CH_4, C_2H_2, and CO_2 molecules, from N bound in NH_3 and N_2, and from Ne, induced by Ar^{12+} (56 MeV) bombardment, were recorded with an electrostatic energy analyzer. The *K* Auger spectra from atoms in different chemical compounds exhibit significantly different line intensities and linewidths. Linewidths of metastable states are explained by the Doppler broadening arising from Coulomb explosion of the target molecule.

Many studies of atomic Auger spectra produced by bombarding noble gases with ions of various masses and charge states have been reported.[1,2] The center of gravity of the spectrum shifts to lower energies as the projectile charge state and energy increase, as a result of multiple ionization and excitation of the colliding partners in a single collision.[3-5] Energy shifts and line broadening of Auger transitions from simple molecules like CH_4, N_2, and CO_2 from electron and proton impact have been reported.[6-8] In this Letter we present the first measurements of Auger lines

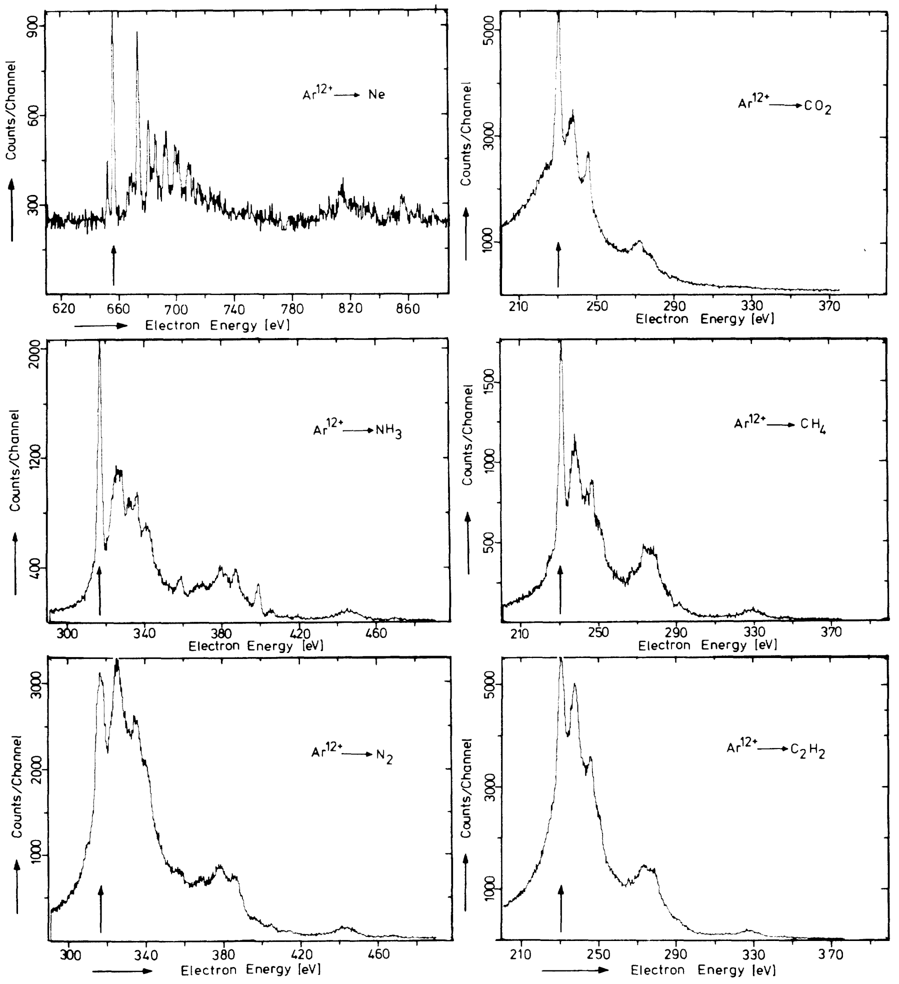

FIG. 1. Auger transitions in Ne and in different C- and N-containing molecules from Ar^{12+} (56 MeV) impact. The arrow indicates the $1s2s2p \rightarrow 1s^2$ transition.

from atoms in different molecules excited by heavy-ion impact, where the molecular binding is expected to break up in the collision process.

An Ar^{12+} beam of 56-MeV energy from the UNILAC accelerator at Gesellschaft für Schwerionenforschung, Darmstadt, passed through a gas jet target. Electrons from the target atoms ejected at 155° with respect to the incident beam direction were analyzed with an electrostatic spectrometer consisting of a retarding field followed by a spherical mirror analyzer.[9] The overall resolution amounts to less than 2.5 eV full width at half-maximum (FWHM), independent of electron energy. The gas pressure in the collision region was constant within 1% during the experiment and is estimated to be 10^{-2} Torr.

Figure 1 presents K Auger spectra from the target gases Ne, CH_4, NH_3, CO_2, C_2H_2, and N_2. The Ne spectrum (with an energy resolution of 1.7-eV FWHM) in Fig. 1 exhibits well-separated lines, most of which have been identified as transitions in lithium- and beryllium-like ions.[5,10] The lowest-energy line at 652 eV in the Ne spectrum is identified as the $1s2s^2 \rightarrow 1s^2$ Auger transition; the most intense lines are seen at 656 eV ($1s2s2p \rightarrow 1s^2$) and 672 eV ($1s2p^2 \rightarrow 1s^2$).

Except for an overall energy shift, Auger spectra from CH_4 and NH_3 are very similar in their main features. As in Ne we can therefore classify the intense feature line in the low-energy range as arising from three- and four-electron states having one K hole.[5,10] Satellite and hypersatellite transitions show up in the higher-energy structures.

The linewidths are in a first approximation due to the spectrometer resolution ΔE_{SP} (1.7 eV FWHM for Ne and 2.5 eV FWHM for the other spectra) and due to Doppler broadening by the momentum transfer from the projectile to the target nucleus during the collision. A comparison between the measured linewidth of the dominant $1s2s2p \rightarrow 1s^2$ transition and the calculated kinematic broadening,

$$\Delta E_{\text{coll}} = \left| 8\cos\theta \left[\frac{m_e m_p}{(m_p + m_t)^2} \cdot E E_p \sin^2 \arctan\left(\frac{Z_t^2 Z_p (m_p + m_t) e^2}{2a_0 m_t E_p} \right) \right] \right|, \tag{1}$$

is given in Table I. Equation (1) is derived from the difference between maximum and minimum Doppler shift from the recoiling, Auger-electron-emitting, target atoms. It is assumed that the collison impact parameter is equal to the target K-shell radius $r_K = Z_t / a_0$, with a_0 being the Bohr radius. In contrast to measurements[6-8] with electron and proton bombardment on CH_4, the present CH_4 and NH_3 spectra show no additional line broadening by molecular bonding. The better resolution in Ne compared to that for CH_4 and NH_3 is caused by a larger energy separation between Auger lines from atoms with higher atomic numbers and by the better resolution of the spectrometer used in the Ne measurement.

A dramatic increase in the linewidth and a change of some relative line intensities, however, appear in the N_2 and C_2H_2 spectra. We explain this by an additional kinematic broadening ΔE_{bond} (Table I) due to the nuclear repulsion between the neighboring atoms of similar masses in the molecule which is highly ionized after the Ar^{12+} collision. A straightforward calculation of this repulsion during the "Coulomb explosion," taking into account[11] the molecular bond lengths R, leads to a line broadening

$$\Delta E_{\text{bond}} = 4 \left(\frac{Z_1^* Z_2^* e^2}{R} \frac{m_1 E}{m_2 + m_1} \right)^{1/2}. \tag{2}$$

Here, Z_1^* and Z_2^* are the effective charges of the molecular fragments (estimated to be $Z_1 - 1$, $Z_2 - 1$ to account for the screening by one K-shell electron in the electron configuration in question), and m_1 and m_2 are the fragment masses, where atom 2 ejects the Auger electron. The resulting linewidths

$$\Delta E_{\text{result}} = (\Delta E_{\text{coll}}^2 + \Delta E_{\text{bond}} + \Delta E_{\text{SP}}^2)^{1/2}$$

are in good agreement with the experimental values (see Table I).

Some line intensities depend strongly on the type of molecular environment. In the NH_3 spectrum, e.g., we observe a line at ~398 eV which is barely seen in N_2. The corresponding transition in the carbon spectrum at ~288 eV is much weaker than in NH_3. An anomalous behavior occurs in the CO_2 spectrum, where a broad tail arises below 228 eV.

It should be emphasized that the normal lifetime of the Auger decay is less than 10^{-14} sec whereas the dissociating molecular compounds reach a distance of 10 Å in 5×10^{-14} sec after the collision under the kinematic conditions in this experiment. Thus, transitions from metastable states ($\tau > 10^{-13}$ sec) such as $1s2s2p \rightarrow 1s^2$ (Ref. 1), intensely excited by heavy-ion collision,[12] give rise to lines like those from single atoms where the linewidth is Doppler-broadened by the full dissociation velocity [Eq. (2)]. On the other hand, transitions with smaller lifetimes have widths which are determined by a lower velocity corresponding to the interatomic distance. Such lines may be affected by molecular components giving a shift in energy and a change of line intensities.

Similar measurements made with Kr^{25+} (1.4

TABLE I. Measured and calculated linewidths ΔE in Auger transitions in atoms with different molecular environment.

Target	ΔE_{coll} (eV)	ΔE_{bond} (eV)	ΔE_{result} (eV)	$\Delta E_{\text{measured}}$ (eV)	Molecular bond length R (Å)
Ne	1.39		2.2	2.0 ± 0.2	...
NH_3	1.37	1.0	3.0	2.5 ± 0.3	1.014
CH_4	1.11	0.9	2.9	2.5 ± 0.3	1.093
CO_2	1.11	3.36	4.3	3.7 ± 0.4	1.162
C_2H_2	1.11	4.8	5.5	5.6 ± 0.5	1.21
N_2	1.37	6.85	7.4	7.2 ± 0.5	1.1

MeV/amu) on CH_4, CO_2, NH_3, and N_2 are not shown here. These spectra differ from the Ar^{12+} spectra and show significant increase in the intensity of the $1s^12s^2 \rightarrow 1s^2$ and $1s^12s^12p^1 \rightarrow 1s^2$ transitions and of the double K-vacancy lines (hypersatellites) on the high-energy end of the spectra. The linewidth dependence for different molecules is, however, very similar to that for the Ar^{12+} measurements.

Some valuable properties of this kind of target-molecule spectroscopy should be noted: (1) The energy of the Auger transitions is affected very little by the molecular environment of the Auger-electron-emitting atom (Fig. 1). (2) The linewidths of these transitions, however, reflect kinematic properties of the heavy-ion-molecule reaction mechanism (Coulomb explosion) in cases where levels are involved with decay times longer than the time required for the exploding fragments to acquire sufficient velocity to contribute to a Doppler broadening according to Eq. (2). This opens the possibility of lifetime studies in atomic states of molecules as a new technique. (3) Some intensities are affected strongly; quantitative interpretation of this phenomenon requires involved Hartree-Fock calculations and may yield important and detailed information on the heavy-ion-molecule reaction mechanism.

We conclude that Auger spectra from simple molecules bombarded with heavy ions are more complex than those from noble gases. The general shape of the molecular spectrum is like an atomic one, but different chemical compounds change linewidths and some intensities. The influence of the chemical bonding may depend on the lifetimes of the Auger transition. The linewidth can be explained by an additional kinematic line broadening caused by the molecular dissociation after collision.

Valuable discussions with Dr. Ch. Bhalla, Dr. P. Mokler, and Dr. I. A. Sellin are gratefully acknowledged.

---

[1] B. M. Johnson, D. Schneider, K. S. Roberts, J. E. Bolger, and C. F. Moore, Phys. Lett. 53A, 254 (1975).

[2] M. E. Rudd, Radiat. Res. 64, 153 (1975).

[3] D. Burch, N. Stolterfoht, D. Schneider, H. Wiemann, and J. S. Risley, Phys. Rev. Lett. 32, 1151 (1974).

[4] C. P. Bhalla, D. L. Matthews, and C. F. Moore, Phys. Lett. 46A, 336 (1974).

[5] D. O. Groeneveld, R. Mann, G. Nolte, S. Schumann, R. Spohr, and B. Fricke, Z. Phys. A274, 191 (1975); S. Schumann, K. O. Groeneveld, and K. D. Sevier, Proceedings of the Fifth International Conference on Atomic Physics, Berkeley, California, July, 1976, edited by R. Manus, M. H. Prior, and H. A. Shugart (to be published).

[6] W. E. Moddemann, T. A. Carlson, M. O. Krause, and B. P. Pullen, J. Chem. Phys. 55, 2317 (1971).

[7] N. Stolterfoht, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena and Future Applications, Atlanta, Georgia, 1972,* edited by R. W. Fink, J. T. Manson, I. M. Palms, and R. U. Rao, CONF-720 404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973), p. 1043.

[8] T. A. Carlson, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena and Future Applications, Atlanta, Georgia, 1972,* edited by R. W. Fink, J. T. Manson, I. M. Palms, and R. U. Rao, CONF-720 404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973).

[9] K. O. Groeneveld, W. Kessel, E. Köhler, and R. Spohr, Gesellschaft für Schwerionenforschung, Darmstadt, Report No. GSI-J-2-74, 1974 (unpublished), p. 28.

[10] D. Schneider, C. F. Moore, and B. M. Johnson, J. Phys. B 6, L153 (1976).

[11] H. B. Gray, *Electrons and Chemical Bonding* (Benjamin, New York, 1965).

[12] K. O. Groeneveld, R. Mann, G. Nolte, S. Schumann, and R. Spohr, Phys. Lett. 54A, 335 (1975).