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Angular distributions of electrons elastically scattered via K-shell resonances in N_2

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Angular distributions of two K-shell resonances in e-N_2 elastic scattering in the angular range $50° \leq \theta \leq 150°$ have been measured in a crossed-beam apparatus. The dominant partial wave has $l=3$ character, in conformity with recent theoretical predictions and photoionization measurements.

The prominent role played by shape resonances in the continuum dynamics of diatomic molecules has recently been highlighted by inner-shell photoionization[1] and vibrationally-selective photoelectron experiments[2] using synchrotron radiation as well as by theoretical calculations[3] using the continuum multiple scattering model (CMSM). In the ionization of inner-shell electrons the interaction between the ejected electron and the anisotropic molecular field consists of Coulomb (attractive) and centrifugal (repulsive) forces which can combine to form a potential barrier acting on high-angular-momentum components of the final-state wave function giving rise to such resonances. Barrier effects[4] have also been invoked to explain the nonhydrogenic behavior observed within a few rydbergs above the K edges of diatomic molecules in x-ray and pseudophoton absorption spectra. In the case of K-shell photoionization in N_2, the electric dipole interaction which creates a hole in the (σ_u $1s$) orbital results in a photoelectron with angular momentum $l=1$. This ejected p-wave electron experiences the field of the two nitrogen atoms which scatters it into a variety of angular momentum states contributing to the dipole-allowed σ and π ionization channels. The CMSM calculations of Dehmer and Dill[3] have shown that the resonant enhancement of photoelectric current at ~ 1 Ry kinetic energy occurs due to the potential barrier in the f-wave component of the σ continuum wave function even though the $l=3$ component is not the dominant one. This is in contrast to the usual situation in atomic collision physics where high-angular-momentum components are seldom seen. In comparing photon and electron interactions with molecules it is known that, although the long-range part of the scattering potential is dramatically different in the two cases, the inner part is significantly the same since it is dominated by the interactions between nuclei and those electrons in inner orbitals which are the same in both systems. Thus shape resonances which are localized in the molecular core should basically maintain their identity in going from the $h\nu + N_2$ to $e + N_2$ system, apart from an energy shift owing to the difference caused by the addition of an electron in the electron impact case.

We report here for the first time measurements of the angular distributions of K-shell resonances in e-N_2 elastic scattering. Our data provide unambiguous experimental evidence for the $l=3$ character of the dominant partial wave involved in electron scattering at these resonances. Though a K-shell resonance has been detected earlier in different inelastic electron scattering experiments[5] the effect has manifested itself as an extremely small (typically $\sim 0.1\%$) effect superimposed on an enormous continuum; this had made the accurate determination of resonance energy and width (lifetime) difficult and angular distribution studies to define the symmetry and electron configuration of the resonant state impractical. In elastic scattering the contribution from direct (potential) scattering becomes extremely small at large scattering angles ($50° < \theta < 150°$); as a consequence, the resonance effect in this channel is found[6] to be typically 2 orders of magnitude larger than in inelastic decay channels. The relatively large magnitude of the resonance effect enables us to clearly identify two separate N_2^{-*} components in our experiment corresponding to singlet-triplet splitting of the excited N_2^* parent state.

Measurements of elastic scattering intensities as a function of electron energy and scattering angle were carried out in a crossed beam apparatus which has been described elsewhere.[6] Each of the crossed beams has a circular cross section of 1 mm diameter; they intersect at 90° in a field-free region where the background pressure is $\sim 5\times 10^{-7}$ Torr. Electron energy analysis is performed by means of an electrostatic cylindrical mirror analyzer and electrons are detected by a channel electron multiplier operating in the single particle counting mode. Background counts are typically a factor of 4000 smaller than the count rate at the elastic peak, and are nonfluctuating over the entire range of energies and scattering angles investigated. The entire apparatus is interfaced to an on-line microprocessor controlled multichannel analyzer. No attempts were made to determine absolute values of the measured differential cross-section functions.

A typical elastic scattering function at a scattering angle of 110° is shown in Fig. 1(a). The two prominent dips are attributed to shape resonances associated with excitation of a nitrogen $1s$ electron to the π_g $2p$ orbital and the simultaneous capture of the incident electron in a potential barrier, resulting in the formation of a highly-excited, temporary N_2^{-*} state. The centroids of the two resonances occur at 398.95 and 400.96 eV, respectively. A Breit-Wigner type of analysis[6] of the line shapes yields resonance energies of 399.30 and 401.95 eV, and widths of 0.53 and 1.4 eV, for the lower- and higher-energy features, respectively. Promotion of a $1s$ electron from the σ_u $1s$ orbital to the lowest empty orbital π_g $2p$, will yield a $^1\Pi_u$ state as well as a lower-energy dipole-forbidden $^3\Pi_u$ state. By measuring the energy difference between the vibrational ground state of the parent configuration and the respective resonance energies we determine electron affinities of 0.79 and 0.84 eV for the lower- and higher-energy resonances, respectively. Pre-

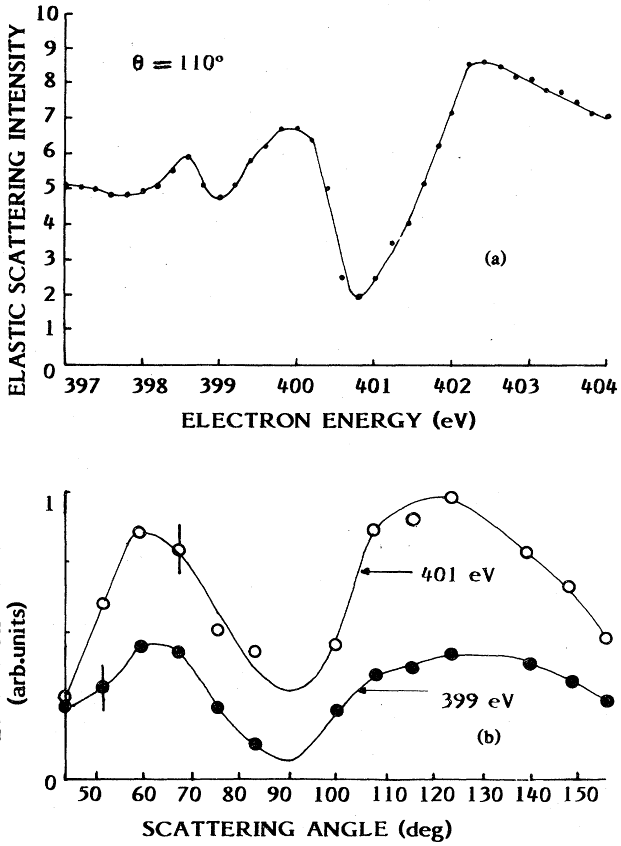

FIG. 1. (a) Elastic scattering cross section for e-N_2 collisions at a scattering angle of 110°. (b) Angular distributions at electron energies of 401 and 399 eV.

cisely which orbital is occupied by the extra electron that results in negative ion formation can be deduced from the resonance angular distribution function shown in Fig. 1(b).

The angular distribution of electrons scattered via resonant states is governed by Legendre polynomials $P_l(\cos\theta)$ of order l. Although elastic scattering angular distribution data are generally difficult to interpret since they always contain some contribution from nonresonant scattering, the present data [Fig. 1(b)] show prominent peaks at 60° and 120°, ruling out p-wave scattering. These peaks, in conjunction with the dip at 90°, indicate that the scattering is predominantly due to an f wave at both the resonance energies. The parity of the resonance state must therefore be odd (u symmetry). Since the core excitation is from σ_u $1s$ to π_g $2p$, this leads to an overall u symmetry for the core excited state $(\sigma_u 1s^{-1}\pi_g 2p)$. Therefore, the orbital in which the incoming electron is trapped must have g symmetry. The lowest normally unoccupied orbital fulfilling this criterion is π_g $2p$. Thus the electronic configuration for the resonant state is $\sigma_u 1s^{-1}\pi_g 2p^2$. The dominant partial wave in the angular distribution is $l=3$; this indicates that the quantum number $\Lambda \geq 2$ for the N_2^{-*} state. This results in a unique overall symmetry $^2\Delta_u$. This contradicts the usual atomic physics picture where high-l partial waves are excluded from atomic cores and lends credence to the view that centrifugal barrier phenomenon involving high-angular-momentum species is likely to be widespread in molecules. The origin of the double structure seen in the elastic scattering cross section remains open as the spin forbidden $^4\Delta_u$ state cannot result from two electrons in π_g $2p$ orbital due to Pauli principle.

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