tions of diffractive and refractive optical processes permit aplanatic or chromatic correction in lenses, spectral dispersion, imaging from a single optical element, and other manipulations of light (7). The capability to pattern LCs on curved surfaces also provides routes for the fabrication of displays with wide viewing angles.

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30 January 1997; accepted 15 April 1997

Perturbed Equilibria and Statistical Energy Redistribution in a Gas-Phase S_N2 Reaction

Stephen L. Craig and John I. Brauman*

The relative translational energy of one set of reactants in the gas-phase equilibrium between chloride ion and chloroacetonitrile $^{35}Cl^- + {}^{37}ClCH_2CN \rightleftharpoons {}^{37}Cl^- + {}^{35}ClCH_2CN$ is varied without affecting the energy distribution of the other set. The steady-state ratio of ionic isotopes changes so that there is an increase in the concentration of energized reactants. The ratio provides a measure of the translational energy dependence of the rate constant for the bimolecular nucleophilic substitution (S_N2) reaction. The observed energy dependence is indistinguishable from that predicted by Rice-Ramsperger-Kassel-Marcus theory, suggesting that the increased translational energy redistributes statistically in the collision complex.

We report here two interesting results obtained in a study of the translational energy dependence of the gas-phase, chloride-exchange reaction of chloroacetonitrile

$$^{35}Cl^- + {}^{37}ClCH_2CN \underset{k_b}{\overset{k_f}{\rightleftharpoons}} {}^{37}Cl^- + {}^{35}ClCH_2CN \qquad (1)$$

where k_f and k_b are the rate constants for the forward and back reactions, respectively. First, we show that selectively increasing the center-of-mass collision energy between one set of reactants in Eq. 1 without changing the energy of the other set causes the concentration of the energized reactants to increase. Although this behavior may seem to contradict LeChatelier's principle, it is not exceptional and is qualitatively consistent with a well-known kinetic model. Second, we examine the energy dependence of the rate constant and compare our results to those of earlier studies of gas-phase S_N2 reaction dynamics, which have focused on halide–methyl halide reactions. Although the methyl halide systems exhibit nonstatistical behavior, the dynamics of the larger S_N2 system in Eq. 1 appear to be statistical, in that the reaction rates are consistent with a

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statistical reaction rate theory analysis.

The experiments were conducted in a Fourier-transform ion cyclotron resonance spectrometer (ICR). The kinetic energy of ions of a particular mass-to-charge ratio (m/z) is manipulated by a radio-frequency (rf) potential that is resonant with the cyclotron frequency of the ions. The rf signal is used in a reversible mode (*1*), so that it alternately accelerates and then decelerates the resonant ions, with no net effect on the nonresonant ions (*2*, *3*). Thus, two sets of ions with different velocity distributions may be simultaneously trapped in the ICR: the nonresonant ions, whose velocity distribution is thermal, and the resonant ions, whose distribution is shifted on average toward higher velocities.

When energy is added to only one set of reactants in a system at equilibrium, LeChatelier's principle would seem to suggest that the steady-state concentrations would shift to lower the concentration of energized reactants. For many systems, this expectation is correct, because an increase in reactant energy leads to an increased rate of product formation and a simultaneous reduction in the concentration of energized reactants. Gas-phase S_N2 rate constants, however, provide a contradiction to this expectation, because they are well known to exhibit negative energy dependencies (*4–7*). Equation 1 is such a reaction, and we observe that, when we increase the reactant energies, the concentration of energized reactants increases.

We have established that the observed changes in the isotope ratio are not artifacts and that they reflect changes in the reaction rate constant of the translationally excited reactants (*8*). At the steady state

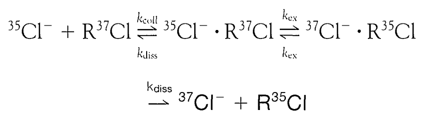
$$k_f[^{35}Cl^-][^{37}ClCH_2CN] = k_b[^{37}Cl^-][^{35}ClCH_2CN] \qquad (2)$$

and so

$$\frac{[^{35}Cl^-]}{[^{37}Cl^-]} = \frac{k_b[^{35}ClCH_2CN]}{k_f[^{37}ClCH_2CN]} \qquad (3)$$

The pressure of the neutrals remains constant in the ICR. Under thermal conditions, k_b and k_f are equal (isotope effects are negligible), and the ratio of the Cl^- isotopes is the naturally occurring ratio of 3:1 (Fig. 1A). Selective acceleration of $^{35}Cl^-$ does not affect k_b, and so the increase in the isotope ratio reflects a decrease in k_f with additional kinetic energy (Fig. 1B). Similarly, acceleration of $^{37}Cl^-$ reduces k_b and results in a decreased ratio of $^{35}Cl^-$ to $^{37}Cl^-$ (Fig. 1C).

The kinetic energy distribution of the accelerated ions for these experimental conditions has been determined (*8*), and we can compare the observed change in reaction rate with theory. The double-well potential energy surface for Eq. 1 has been characterized experimentally and theoretically and is shown in Fig. 2 (*9*). We model the reaction as shown in Scheme 1. A two-body collision (k_{coll}) forms an intermediate ion-molecule complex that may either undergo S_N2 chloride exchange (k_{ex}) to form the isomeric complex or dissociate back to reactants (k_{diss}):



$$^{35}Cl^- + R^{37}Cl \underset{k_{diss}}{\overset{k_{coll}}{\rightleftharpoons}} {}^{35}Cl^- \cdot R^{37}Cl \underset{k_{ex}}{\overset{k_{ex}}{\rightleftharpoons}} {}^{37}Cl^- \cdot R^{35}Cl \xrightarrow{k_{diss}} {}^{37}Cl^- + R^{35}Cl$$

Scheme 1.

The overall rate constant (here, k_f) is derived from the steady-state assumption to give Eq. 4:

$$k_f = \frac{k_{coll}k_{ex}}{k_{diss} + 2k_{ex}} = k_{coll}\phi \qquad (4)$$

where $\phi = k_{ex}/(k_{diss} + 2k_{ex})$ is the fraction of collisions that lead to product formation.

The overall energy dependence of k_f reflects the energy dependence of k_{coll} and ϕ. The energy dependence of k_{coll} depends on several factors, including the relative translational energy of the reactants and the dipole, polarizability, and the rotational energy of the neutral. All of the relevant physics may be accounted for implicitly by use of trajectory calculations to calculate k_{coll}. Su has recently reported a parameterized formula that reproduces the results of trajectory calculations accurately, and we use this method in our analysis (*10*). We find that although k_{coll} decreases slightly with added kinetic energy, the observed change in the overall rate constant of Eq. 1 is far too large to be attributed solely to this effect. Most of the energy dependence, therefore, is found within ϕ, which reflects the competition between the dissociation and Cl^- exchange pathways.

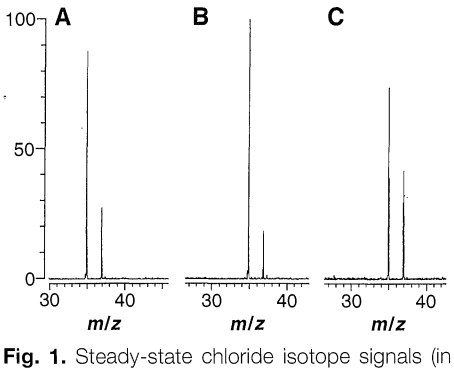


Fig. 1. Steady-state chloride isotope signals (in arbitrary units) for Eq. 1 under (**A**) thermal conditions, (**B**) selective acceleration of $^{35}Cl^-$, and (**C**) selective acceleration of $^{37}Cl^-$. The concentration of the energized reactants increases relative to the thermal values; *m/z*, mass-to-charge ratio.

Both k_{diss} and k_{ex} increase with the energy of the reactant complex. For Eq. 1, k_{diss} increases more rapidly with energy than does k_{ex}, and ϕ decreases (Fig. 2). The energy dependence of ϕ is calculated with Rice-Ramsperger-Kassel-Marcus (RRKM) theory (*11*), in which it is assumed that all of the available energy redistributes statistically across the active degrees of freedom of the system (*12*). Despite concerns about the validity of this assumption (*5*, *13–16*), the energy dependence of the reaction calculated with RRKM theory is indistinguishable from that determined experimentally (Fig. 3). The temperature dependence of Eq. 1 has been shown to agree with that predicted by RRKM theory across a similar range of total energy (*6*, *17*). That k_f varies similarly as a function of total energy for two dramatically different initial energy distributions indicates that most of the energy is randomized in the collision complex.

The agreement between theory and experiment for this reaction stands in contrast to a growing body of literature. Recent theoretical and experimental work on gas-phase S_N2 reactions has questioned the principal assumptions of statistical theories (*13–16*), especially that of com-

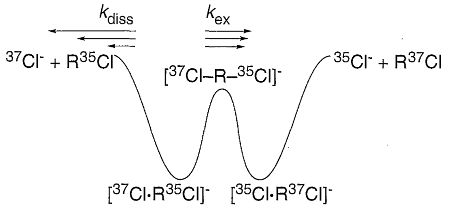


Fig. 2. Double-well potential energy surface for Eq. 1. The S_N2 barrier height is 5.9 kcal mol^{-1} below the separated reactants, so that the S_N2 reaction is favored at total energies below or just above the energy of the entrance channel, whereas the entropically favorable dissociation pathway predominates at higher energies.

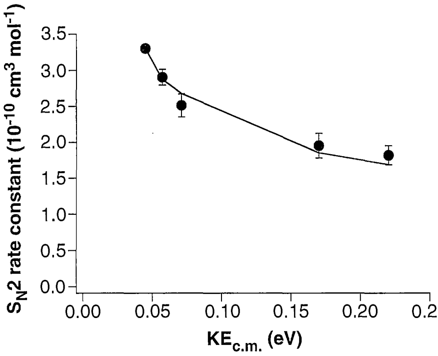


Fig. 3. Observed (●) and expected (RRKM theory, —) S_N2 rate constant for Eq. 1 as a function of average center-of-mass collision energy, $KE_{c.m.}$. Error bars reflect the uncertainty in the relative rate constants. Uncertainty in the absolute rate constants is ±30%.

plete energy randomization. The translational energy dependence of the $Cl^- + CH_3Br$ S_N2 reaction (*5*, *8*), for example, is significantly greater than that predicted by statistical theory (*16*) and differs dramatically from the observed temperature dependence of that reaction (*5*). Graul and Bowers (*14*, *15*) have shown that the kinetic energy release distribution (KERD) of the product ions of exothermic halide–methyl halide S_N2 reactions is significantly lower than that predicted by statistical models, and the product neutral is therefore internally "hot."

Both the translational energy and KERD experiments represent cases in which an ion-molecule complex is formed with energy partitioned nonstatistically between the vibrational and relative translational modes of the two species. Increased translational energy between reactants leads to a reactant complex formed with excited translational modes, whereas the neutral molecule in the product complex is vibrationally hot as a result of the reaction exothermicity. In both cases, poor translation-vibrational coupling in the S_N2 intermediate complex prevents statistical redistribution of energy.

The nonstatistical behavior of the halide–methyl halide systems may reflect the short lifetimes (10 to 100 ps) (*7*) of their intermediate complexes, which Boering and Brauman have shown to be a determining factor of the efficiency of intermolecular vibrational energy transfer (*18*). The short lifetimes are a result of the small size and low association energies (~10 kcal mol^{-1}) (*14*, *19*) of halide–methyl halide complexes, which are not typical of many ion-molecule reactions. The association energy of $[Cl{\cdot}ClCH_2CN]^-$, for example, is ~19 kcal mol^{-1} (*10*). Moreover, the internal vibrations of the neutral molecules have significantly different frequencies (for example, the methyl umbrella mode of CH_3Br differs from similar modes in $ClCH_2CN$) that might influence coupling. Although the limits of poor energy redistribution are yet to be determined, our results for Eq. 1 show that gross nonstatistical dynamics are not general to all gas-phase S_N2 reaction intermediates.

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20. Supported by NSF. S.L.C. was supported by an NSF predoctoral fellowship, an American Chemical Society Division of Organic Chemistry DuPont Merck Pharmaceutical Fellowship, and a John Stauffer Memorial Fellowship.

4 February 1997; accepted 3 April 1997

Accessible Solitons

Allan W. Snyder* and D. John Mitchell

Solitons are ubiquitous. Their description involves abstruse mathematics and is limited to a two-dimensional idealization. A nonlocal model is presented that provides a radical simplification and allows for an elegant description of soliton collisions, interactions, and deformations in two and three dimensions. The model reveals an intimate connection between solitons and the linear harmonic oscillator. It foreshadows a photonic switch in which a bright beam can steer a distant dim beam, and it predicts the existence of noncircularly symmetric solitons.

Solitary waves create their own channel as they travel in a uniform medium, remaining localized and preserving their shape (*1*). But, they can be dramatically altered by colliding with one another (*2*). Solitons are solitary waves that are unaltered by collisions (*1*). The conventional models of soliton propagation are complex; we provide a simple model.

Consider the example of monochromatic light beams in a homogeneous medium whose (squared) refractive index (*3*) is $n^2 = n_L^2 + \delta n^2$. Here, n_L is the constant linear part of the refractive index, whereas δn^2 is the nonlinear induced change caused by the beam. Typical of many materials (*4*–*7*), δn^2 is positive and obeys $\delta n^2 \ll n_L^2$. The conventional model assumes an idealized local response when, by definition, δn^2 at position x is proportional to the beam's intensity at x. This local (Kerr) model admits solitons in two dimensions only. The resulting Schrödinger equation does not have explicit closed-form solutions, except for special initial conditions. Furthermore, the inverse scattering technique that is required for solution is excessively mathematical (*1*). This complexity is not fundamental to solitons, but rather, is a consequence of the specific model.

Consider a model of nonlinearity whose response is highly nonlocal. Then, by definition, a light beam of characteristic radius ρ creates a circularly symmetrical refractive index change δn^2 whose characteristic spatial extent is much larger than ρ and whose axis is set by the beam center and its initial direction. Consequently, δn^2 depends on the integrated intensity of the beam, or power, P. The situation of high nonlocality is analogous to observing distant point sources through a badly blurred lens. When the point sources (light beams) are sufficiently close to the lens axis, the shape of the blur circle (nonlocal response) is indis-

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