the relative GOS sums for $L + M$ electrons and for $K + L + M$ electrons are then, respectively, $S_{LM} = S_1 + S_2$ and $S = S_1 + S_2 + S_3$. From the Bethe sum rule, the absolute value of the latter is the total number $N = 18$ of Kr electrons, so that an effective number of electrons $N_{LM}$ of the $L + M$ shells can be defined as $N_{LM} = N \times S_{LM} / S$, and effective number of electrons of the $K$ shell is then $N_K = N - N_{LM}$. From our 20 measurements (ten angles and their asymmetries) the mean value of $N_K$ obtained together with its standard deviation is $N_K = 1.54 \pm 0.06$. A plot of $N_K(K)$ versus $K$ has shown no significant variation in $N_K$ versus $K$.

This value, obtained for the generalized oscillator strengths of Kr, is similar to the values previously determined from X-ray absorption experiments by Woornk€"u (N$_K = 1.61$) and by Wuilleumier and Bearden (N$_K = 1.46 \pm 0.05$) and with the linear interpolation to the calculations by Wheeler and Bearden (N$_K = 1.465$).

It thus follows that, for the normalization procedure of relative energy loss spectra obtained by electron impact, the Bethe sum rule can be replaced by the partial sum involving the effective number of $L + M$ shell electrons $N_{LM}$, as long as the scattering angle is small enough to allow the asymptotic behavior of the GOS to be reached before the energy loss $E_k$. Using $N = 2$ rather than $N_{LM}$ as it was previously reported introduces a 3% error for the normalization in the case of Ar.

Our results are important for experiments where the investigated energy loss range is not large enough to make use of the Bethe sum rule.

Such a separation into partial contributions to the sum rule is not possible for Ne because the $K$ shell ionization energy is substantially lower than for Ar, and the valence shell GOS does not reach its asymptotic form at this energy loss.

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A simple model for the influence of fluctuations on explosive reactions\(^{a})\)

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Explosions are an important part of combustion,\(^1,2\) The simplest model for an explosive reaction is based upon the chain reaction\(^3\)

\[x' + \text{A} \rightarrow \text{B},\]
\[A + 2x' \rightarrow \text{?},\]

\[A + 2x' \rightarrow 3x' .\]  \hspace{1cm} (1)

This deterministic reaction mechanism has a discrete transition to explosive behavior. Our purpose here is to investigate the influence of fluctuations on this transition. We employ a simple, approximate method that illustrates how composition fluctuations blur the transition between stable and unstable behavior.

In dimensionless form the kinetic equation is

\[\frac{dx}{dt} = -x^2 + x = [x^2A/4],\]  \hspace{1cm} (2)

and the system is open to an infinite reservoir of A.

The exact solution of Eq. (2), for $x(0) = x_0$

\[x(t) = x_0 \exp [-x t] + x_0 \exp (-x t) - 1]^{-1} \hspace{1cm} (3)

predicts that for $x_0 > 1$, $x$ diverges as $t$ approaches the finite time $t = (4)^{-1} \ln [x_0(x_0 - 1)^{-1}]$. This deterministic behavior is lost when fluctuations are present; for $x_0 < 1$, there is a chance that a fluctuation can drive the system into the region $x > 1$.

Fluctuations in concentration may be included through the Langevin equation\(^4,5\)

\[\frac{dx}{dt} = -x^2 + x = A(x)g(t),\]  \hspace{1cm} (4)

where $g(t)$ is chosen to be a Gaussian stochastic variable.

If $A(x)$, the amplitude of the fluctuations, is constant $[A(x) = (x)^{1/2}]$ then Eq. (4) can be transformed\(^6,7\) into a Fokker–Planck equation of the form

\[\frac{\partial P(x, t)}{\partial t} = \frac{\partial}{\partial x} \left[ \frac{\partial}{\partial x} \left( x^2 \frac{\partial P(x, t)}{\partial x} + \beta \phi'(x) P(x, t) \right) \right] \]

\[= - \frac{\partial}{\partial x} J(x, t),\]  \hspace{1cm} (5)

where $D = \beta$ is the relevant diffusion constant and $\phi(x) = \left( \frac{4}{x} \frac{2}{x} (\frac{x}{2} - x/3)^2 \right)$ is the "potential."
An analytic solution of this Fokker–Planck equation is not possible.

There is a simple method\textsuperscript{8} for determining the mean passage time $\tau$ which may be used to calculate $U$, the total flux across the maximum of potential $\beta_0(x)$. Assuming absorbing boundary conditions at $x = 0$ and $x = 1$, and a $\delta$-function initial condition $P(x, 0) = \delta(x - x_0)$, we obtain the set of coupled equations for the total flux $U = \int_0^1 dt j(1, t)$ disappearing across the maximum and $L = \int_0^1 dt j(0, t)$ the total flux disappearing into the origin; clearly $U - L = 1$:

\begin{align}
\tau &= -U \int_0^1 dx p_0(x) \int_0^x \frac{dy}{k p_0(y)} + \int_0^1 dx \frac{dy}{k p_0(x)}, \\
\tau &= L \int_0^1 dx p_0(x) \int_0^x \frac{dy}{k p_0(y)} + \int_0^1 dx \frac{dy}{k p_0(x)}.
\end{align}

The above equations are solved by

\begin{align}
U(x_0) &= \int_0^1 dx \frac{x}{p_0(x)} \int_0^1 dx \frac{x}{p_0(x)}, \\
L(x_0) &= \int_0^1 dx \frac{x}{p_0(x)} \int_0^1 dx \frac{x}{p_0(x)}.
\end{align}

In these equations $p_0(x) = \exp[-\beta_0(x)]/[\int_0^1 dx \exp[-\beta_0(x)]]$. Thus, we arrive at a simple approximate expression for $U(x_0)$ that describes the probability that fluctuations will drive the system from a region of stability $x_0 < 1$ into explosive behavior. The description is approximate since recrossing across the barrier is not permitted.\textsuperscript{3,10}

In order to illustrate this effect, Fig. 1 plots $U(x_0)$ versus $x_0$ for various values of $A_1/k$. As expected, one finds that the transition region of appreciable $U$ is quite narrow for $A_1/k$ large, when the magnitude of the fluctuations $k^{1/2}$ is small compared to the height of the potential $A_1$.

It is amusing to note that the point $x_{1/2}$ where $U(x_{1/2}) = \frac{1}{2}$ may be approximated with little error by assuming $U(x_0)$ is linear near $x_0 = 1$:

\begin{align}
x_{1/2} &= 1 - \frac{1}{2} \frac{\int_0^1 dx \exp[\beta_0(x)]}{2 \exp[A_1/(6k)]}.
\end{align}

In summary we have shown how a simple mean first passage time calculation may be employed to define approximately the influence of fluctuations in a model chemical reaction that exhibits explosive behavior. We shall report a more exact treatment of more complex models elsewhere.

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