Scattering Angle Resolution in Measurements of Velocity-Changing Collision Kernels

Recently, McCaffery, Reid, and Whitaker [1] proposed that differential cross sections can be directly determined from collision experiments in cells. Here we examine aspects of Ref. [1] and conclude that their technique, as described, can at best produce $\approx 30°$ resolution.

In Ref. [1], a narrow-band pump-laser velocity selectively excites an active molecule with a well defined v_z' using the Doppler shift. This molecule then undergoes a velocity-changing collision (VCC) with a perturber atom which changes v_z' to v_z. A frequency-scanned probe laser detects the population at each final velocity v_z. The VCC kernel $W(v_z' \to v_z)$ describes this redistribution and is defined as the probability density per unit time for changing from v_z' to v_z due to a single VCC. The kernel can be calculated given the velocity distributions and the differential cross section $d\sigma(\theta)/d\Omega$, where θ is the center-of-mass scattering angle [2].

They noted that for infinite perturber mass and infinite initial velocity v_z', the collision axis and energy are well defined [1], since now the active molecule's velocity is parallel to the laser propagation axis and the center-of-mass frame is also the laboratory frame. Then $W(v_z' \to v_z) = 2\pi d\sigma(\theta = \cos^{-1}(v_z/v_z'))/d\Omega$, producing a one-to-one correspondence between v_z and θ.

In practice it is very difficult to select molecules (or atoms) with $v_z' \gg u_a$ ($u_a^2 = 2kT/m_a$). Since free molecules normally absorb with essentially Lorentzian profiles which often have widths $\approx 0.01 u_a$, for pump-laser detunings corresponding to $v_z' \gtrsim 3u_a$, the nonresonant Lorentzian wing excitation of molecules with $v_z'/u_a \approx 0$ is greater than the resonant excitation of atoms with $v_z' \gtrsim 3u_a$. The velocity vector is no longer along the laser axis when $v_z' \lesssim 3u_a$ due to the thermal transverse velocity and, while their idea relied on $v_z' \gg u_a$, their results [1] were obtained for $v_z' = 371$ ms^{-1} ($u_a = 1050$ ms^{-1}). In Fig. 1(a) we reproduce their Fig. 2 and show our calculations of the VCC kernel using the hard-sphere expression given in Ref. [2] and a Monte Carlo result using their 40° model cross section.

To understand the differences between their results and ours, in Fig. 1(b) we show calculated $W(v_z' \to v_z)$ using $d\sigma/d\Omega(\theta) = \delta(\theta - \theta_0)$ for several θ_0. For $v_z' = 0$, symmetry requires $W(v_z' \to v_z) = W(v_z' \to -v_z)$, and thus, with $d\sigma/d\Omega(\theta) = \delta(\theta - 45°)$, the kernel has two maxima near $v_z' \approx \pm u_a \sin(45°)$. For $0 < v_z' \lesssim u_a$, the kernel acquires an asymmetry but retains some vestiges of the two maxima as seen for $\theta_0 = 10°$ and 45° in Fig. 1(b). They stated that the signal was a convolution of a Lorentzian and a scattering angle-dependent function [1], which is, however, in general the kernel $W(v_z' \to v_z)$ [3] and not $d\sigma/d\Omega$. Here, since the kernels for $\theta_0 = 135°$ and 180° are completely overlapped by the kernel for 90° in Fig. 1(b), the scattering angle-dependent function is not $d\sigma/d\Omega$ and consequently $d\sigma/d\Omega$ could not have been uniquely measured.

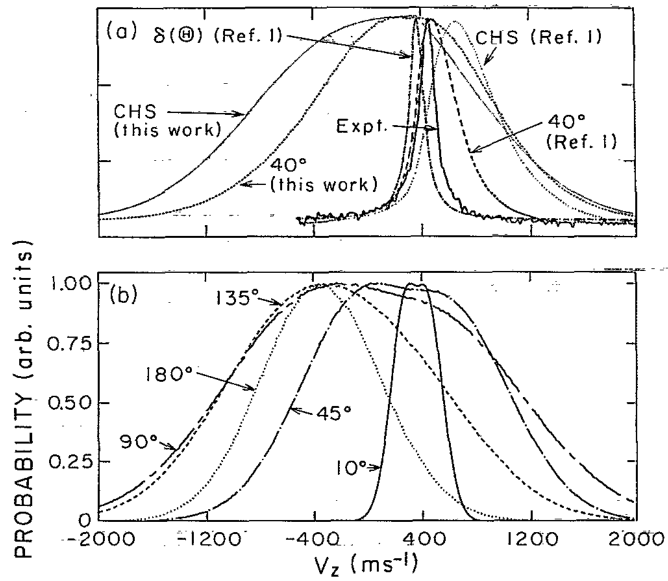

FIG. 1. (a) Experimental data and predicted line shapes of Ref. [1] and VCC kernels for classical hard spheres (CHS) and the 40° model cross section of Ref. [1]. (b) VCC kernels for $v_z' = 371$ m/s, $m_p/m_a = 9.4$, and $d\sigma(\theta)/d\Omega = \delta(\theta - \theta_0)$, where $\theta_0 = 10°$, 45°, 90°, 135°, and 180°.

They [1] also indicated that the pump laser was tuned to 371 ± 50 ms^{-1} whereas the peak of the experimental data is at 460 ms^{-1}. Their results imply that the peak corresponds to $\approx 35°$ scattering. However, the kernels for $\theta_0 = 10°$ and 45° in Fig. 1(b) suggest that there must be a larger maxima near 280 ms^{-1}. If the pump laser was actually tuned near 460 ms^{-1}, their signal may have been primarily from molecules which have undergone very small or no velocity changes [2,3].

Even when $v_z' = 10u_a$, a mass ratio of $m_p/m_a = 9.4$ causes no loss of angular resolution. Thus, the thermal transverse velocity distribution and the constraint of $v_z' < 3u_a$ limits the resolution of this technique to $\approx 30°$.

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