AD-A254 482

ATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (*Leave blank*)

2. REPORT DATE
4 August 1992

3. REPORT TYPE AND DATES COVERED
Reprint

4. TITLE AND SUBTITLE
A Proposal to Produce Velocity-selected and State-selected Molecular Beams Using the Ballistic Effect

5. FUNDING NUMBERS
PE 63220C
PR S321
TA 13
WU 01

①

6. AUTHOR(S)
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7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Phillips Lab/GPOS
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Massachusetts 01731-5000

8. PERFORMING ORGANIZATION REPORT NUMBER
PL-TR-92-2195

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES *Yap Analytics Inc., Lexington, Massachusetts 02173
Reprinted from The Journal of Chemical Physics, Volume 97, Number 1, 1 July 1992

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; Distribution unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT (*Maximum 200 words*)

A method for producing molecular beams containing molecules in the desired internal (rotational, vibrational, electronic, etc.) state and traveling with the desired velocity is proposed. The method is illistrated by describing the circumstances that would lead to a state-selected and velocity-selected N_2 beam resulting from collisions of Li+ with N_2.

92-22898

92 8 18 081

14. SUBJECT TERMS
Molecular beams, State-selected, Velocity-selected, Ballistic collisions, Energy transfer, Chemical reactions

15. NUMBER OF PAGES
2

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	SAR

PL-TR-92-2195

Reprinted from

THE JOURNAL
OF
CHEMICAL PHYSICS

VOLUME 97 NUMBER 1

1 JULY 1992

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pp. 737-738

Published by the

AMERICAN INSTITUTE OF PHYSICS

AIP

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(Received 3 February 1992; accepted 12 March 1992)

The study of chemical reactions and energy transfer processes using molecular beams has revolutionized our understanding of the mechanisms underlying these processes.[1] Still, except for a few fortunate instances,[2] most of the processes studied involve reactants with low internal excitation[3] or with Boltzmann distributions at the oven temperature.[4–6] In this letter we propose a method for producing molecule beams in a desired internal state and traveling with a desired velocity. The rates of chemical reactions are known to depend, sometimes strongly, upon the initial state of the reactants. One should expect greatly improved understanding of the chemical reactions and the possibility of studying many more reactions using the ideas presented here. We illustrate the method by describing the circumstances leading to the production of a desired N_2 beam resulting from collisions of N_2 with Li^+ in crossed beams—a system studied extensively by Toennies and co-workers.[3]

The method is based upon the ballistic effect discovered about 19 years ago.[4–6] Herschbach and co-workers studied the collisions between an alkali halide molecule and an Ar atom at about 1 eV relative translational energy by crossing two beams and measuring the laboratory recoil velocity of the alkali halide molecule as a function of the laboratory scattering angle. In addition to a peak centered at the recoil velocity of elastically scattered alkali halide molecules (pseudoelastic peak, formerly called elastic peak[7]), another equally strong peak centered at the recoil velocity corresponding to the motion of the center of mass (ballistic peak) was observed. Obviously, the ballistic peak is contributed to only by those alkali halide molecules which are almost stationary in the center-of-mass (c.m.) frame and have nearly all of the collision (relative translational) energy converted into the internal (vibrational and/or rotational) energy.

A theory of ballistic collisions[8] gives excellent agreement with the measured results for the CsF–Ar system at the laboratory scattering angles of 30° and 60°. According to this theory, when the differential cross section for the scattering of alkali halide molecules is measured along the direction of the c.m. velocity, only a few transitions contribute to the ballistic peak. This is in contrast to several hundred inelastic transitions making up the pseudoelastic peak. Further, when the relative translational energy equals the energy of one of the rotation–vibration transitions [resonant T-(V,R) process], this transition may have a differential cross section which is larger than that of any other neighboring transition by as much as an order of magnitude. This gives us a state-selected beam traveling at the speed of the c.m. It should be pointed out that the differential cross section of the resonant transitions in the c.m. frame is nearly equal to the nearby nonresonant transitions. The resonant transition has a much larger differential cross section in the laboratory frame because of a greater Jacobian of transformation from the c.m. to the laboratory coordinates. Physically it means that, for the correct viewing geometry, the signal from the entire 4π steradian solid angle in the c.m. frame may be collected by a small solid angle in the laboratory frame.

The differential cross sections are calculated using an exact formulation of a three-dimensional quantum-mechanical impulse approach (IA) calculation described earlier.[9] Physically, one may use the impulse calculation when the time duration of the collision is much shorter than the periods of vibrational and rotational motion of the diatom. The nuclei constituting the diatom may then be considered stationary during the collision and the intramolecular potential is the generator of the momentum distribution of the constituent nuclei. The atom–molecule potential, in the IA, is written as the sum of the atom–atom potentials and the atom–atom scattering amplitudes are added to obtain the atom–molecule scattering amplitudes. The atom–atom potential, in the present study, is described by a hard core interaction. Following the previous study,[9] the hard core radius for the Li^+–N interaction is taken to be 1.62 Å.

Figure 1 is a plot of the calculated laboratory differential cross section for the excitation of N_2 ($v=0, j=0$) during a collision with Li^+, with a relative translational energy of 0.8825 eV, as a function of the laboratory recoil velocity at the laboratory scattering angle of 48.0° (±0.125°). A beam of N_2 molecules in the state $v=3$, $j=10$ traveling at 1440 m/s may be obtained by filtering out velocities lower than 1350 and higher than 1550 m/s. The character of ballistic scattering in this calculation is qualitatively different from that in the previous work[8] where, when the scattering is viewed along the c.m. velocity, several ballistic transitions are calculated although one transition is still dominant. For the Li^+–N_2 system there is only one ballistic transition. Because of the small rotational constant and low frequency of the vibration of the diatomic molecules studied in the earlier work,[8] several rotation–vibration transitions could be nearly resonant ($\Delta E/E \sim 1$) and appear ballistic. Because of the large rotational con-

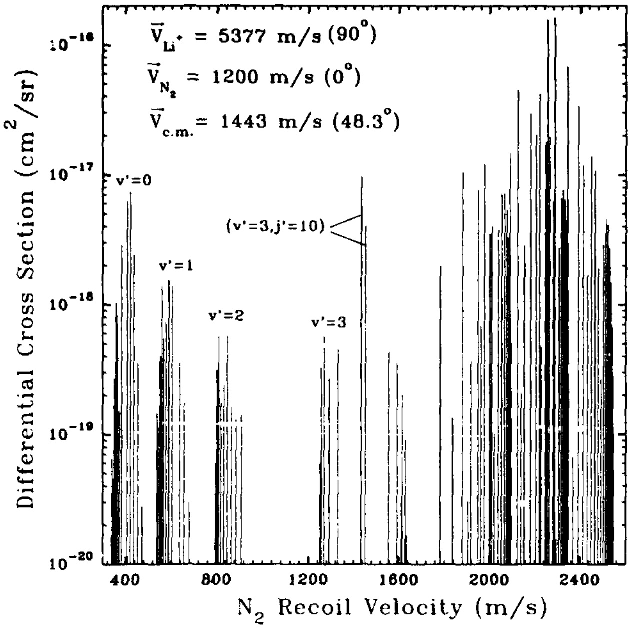

FIG. 1. Differential cross section (cm^2/sr) in the laboratory frame for the scattering of N_2 (initial state $v=0, j=0$) upon colliding with Li^+ with a relative translational energy of 0.8825 eV as a function of the laboratory recoil velocity of N_2 at the laboratory scattering angle (48°±0.125°) coincident with the direction of the c.m. velocity vector (48.3°). The lone ballistic transition to the final state of N_2 ($v'=3$, $j'=10$), $\Delta E/E$ =0.999 87, can be clearly seen.

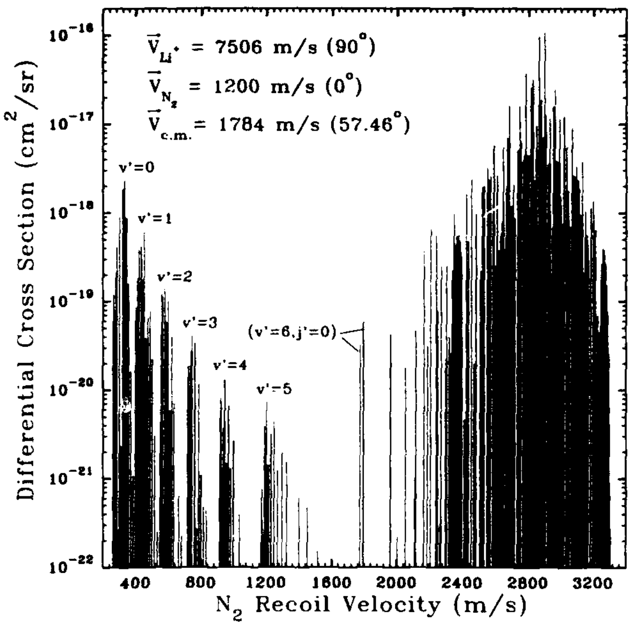

FIG. 2. Differential cross section (cm^2/sr) in the laboratory frame for the scattering of N_2 (initial state $v=0, j=0$) upon colliding with Li^+ with a relative translational energy of 1.68 eV as a function of the laboratory recoil velocity of N_2 at the laboratory scattering angle (57.5°±0.125°) coincident with the direction of the c.m. velocity vector (57.46°). The lone ballistic transition to the final state of N_2 ($v'=6$, $j'=0$), $\Delta E/E$ =0.999 94, can be clearly seen.

stant and high vibrational frequency of N_2 only one transition can be nearly resonant. It is seen from Fig. 1 that if one is interested in defining only the vibrational level, one may obtain a beam of N_2 molecules in $v=3$ by filtering out molecules with recoil velocities below 920 and higher than 1750 m/s. It should be noted, however, that defining only the vibrational level leads to an increase of signal by only a factor of about 2. Figure 2 demonstrates the feasibility of producing velocity-selected N_2 beams in the $v=6$, $j=0$ state. Lower values of the recoil velocities in the figures correspond to small c.m. scattering angles, resulting in transitions to only a few rotational levels having small quantum numbers. The transitions to different vibrational levels are well separated. Large values of recoil velocities, on the other hand, correspond to large c.m. scattering angles leading to large changes in the rotational quantum number during the collision. The rotational transitions are therefore not as neatly bunched within each vibrational level.

A beam containing molecules in nearly any final state may be prepared by giving the appropriate energy to the collision partners provided the c.m. cross section for the particular transition is not too small ($<10^{-22}$ cm^2/sr). The velocity of the beam is equal to the c.m. velocity of the collision partners. The possibilities are limitless.

Part of this research was done while R.D.S. was visiting the Theoretical Institute for Atomic and Molecular Physics in Cambridge, Massachusetts. The research was in part funded by the Air Force Office of Scientific Research.

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[7] Elastic peak was so named because it has the maximum intensity where one would detect the elastically scattered alkali halide molecules. This peak was later shown (Ref. 8) to consist mainly of inelastically scattered alkali halide molecules. This was causing some confusion. At the suggestion of Dudley Herschbach the elastic peak was renamed pseudoelastic peak. The new name is very appropriate because it describes the location of the peak and carries the warning label.
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