ENERGY-RELATED ATOMIC AND MOLECULAR STRUCTURE AND SCATTERING STUDIES

Annual Progress Report for Period July 1, 1978 - June 30, 1979

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ABSTRACT

We have initiated a program to measure the electric dipole polarizabilities, higher energy moments and cross sections for the scattering of low energy electrons by alkali halide molecules, their dimers and larger clusters. Preparatory work is also being carried out to perform similar measurements on the important atomic species, N, O, C and S. This report describes work performed during the first year of the program (July 1, 1978 - June 30, 1979), and presents some preliminary data of the measurements of the polarizabilities of (CsCl)_2, (RbCl)_2 and (KCl)_2. A description is contained of a new scattering apparatus now under construction that will enable us to perform scattering experiments with high energy (<0.80 eV) and angular (<1°) resolution, with state preparation and analysis.
During the first year of our DOE project, we have been engaged primarily in design and construction work, although we have already performed some preliminary experiments, and have obtained some data for several alkali halide molecules. Our activities to date can be divided into the following parts:

1) atomic polarizabilities of heavy atoms
2) molecular polarizabilities of alkali halides
3) electron collisions with alkali halide molecules.

Both polarizability programs are being planned for our existing "polarizability" apparatus, but modifications of the apparatus are being made to accommodate these new programs. Because of the desirability to exploit our existing beams system as fully as possible, and to maximize our resource utilization as much as is feasible, we have undertaken to design these experiments so that they can be performed on a time-shared basis on the same machine. This is not a particularly trivial operation, since (as will be seen below) these programs are not completely compatible.

Similarly, our scattering experiments are being planned for a new machine, which is now under construction (primarily with the help of an NSF capital equipment grant), and which will be time-shared with an ongoing NSF sponsored electron-atom scattering program. Details are presented herewith.

I.1 Polarizabilities

For practical reasons as discussed below, we have reversed the time schedule as described in our original proposal, and have actually started the molecular polarizability experiments during the first year, while going through a design and construction phase of the atomic experiments, rather than vice-versa.

I.1.1 Atoms

We are preparing the heavy atom experiment, as described in Section 9, p. 29 of our original proposal. For this purpose we require a new refractory atomic beams source. Such a source,
using electron bombardment, has been designed, and it is now under construction. So that this experiment could be conveniently time-shared with the polar molecule experiment, we are constructing an entire new source assembly, mounted on its own flange, with all required feed-throughs, terminals and motions. This assembly is presently in the Physics Department machine shop.

Because we are anticipating signal-to-noise problems with some of the more recalcitrant heavy atoms (e.g., uranium), and also in order to supply additional state selection when needed, we are now in the process of designing a magnetic quadrupole neutral beam focussing magnet. This neutral beam focuser will be tunable by means of a variable magnetic field, i.e., will possess a variable focal length, to enable atoms possessing different temperatures and magnetic moments to be focussed. It will lie directly downstream of the atomic beam, so as to subtend as large a solid angle as possible. We estimate that the beam intensity will be increased using the focuser, over the straight inverse square beam, by a factor of about 100. The focuser will work for essentially all elements in the periodic table except the rare gases and the alkaline earths (which possess \(^1S_0\) ground-state configurations).

On the other hand, the molecular experiments generally require an electric six-pole focussing configuration, for molecules possessing electric dipole moments. Accordingly, both multipole assemblies (magnetic quadrupole and electric hexapole) are being designed to slip inside a permanent magnetic yoke assembly (which is too heavy and bulky to disassemble) so that either one or the other, but not both, could be used as needed. The electric six-pole configuration is described below.

In some cases the E-H balance technique will be used. This method is particularly suitable for use with atoms which possess atomic magnetic moments, with either simple (one-electron) or no hyperfine structure, e.g., indium \((5s^25p^2P_\frac32)\) and uranium-238 (no hfs). Strontium and barium \((^1S_0)\) cannot be balanced, since they do not possess magnetic moments. For these elements, and also to obtain better resolution for any element, a high-resolution
mechanical velocity selector is very desirable. Using such a selector, the beam experiences little dispersion as it is deflected by the inhomogeneous electric field. We have been fortunate in acquiring a six-component, slotted disc velocity selector on extended loan from Dr. Stockdale at Oak Ridge National Laboratory. An additional chamber to contain this selector has been built and inserted in our apparatus, between the source and interaction chambers.

I.1.2 Molecules

We have already undertaken some preliminary electric field deflection experiments using alkali halide beams. These preliminary experiments, performed without velocity or state-selection, have now been completed. While analysis of the results is complicated, because of the large velocity dispersion, and the enormous quantity of rotational states (not to mention the $J^2$ total number of $M_J$ states), our results have already yielded some interesting information. We briefly describe the experiment here.

An alkali halide molecular beam is formed using an effusive oven source. It is narrowly collimated, and passes through a region containing an inhomogeneous electric field, whose properties are well-known from earlier work. The deflected beam is analyzed by a surface-ionization ("hot wire") detector. Data is generally taken by applying a fixed voltage to the inhomogeneous electric field electrodes and obtaining a deflection profile by measuring beam signal as a function of detector position. Computed and experimental deflection curves are presented in Figs. 1 and 2.

A detailed explanation of these deflection patterns is contained below. First we present a qualitative explanation. Briefly, we believe that the deflection patterns on the right side of Fig. 2 are attributable to the permanent electric dipole moment, and in fact the position of the peak marked A in Fig. 2 yields a rough measurement of the permanent moment which is in agreement with the known values for the alkali halide molecules studied (RbCl, CsCl, KCl). The deflection on the left side is attributable to a superposition of a monotonically decreasing
Fig. 1. Calculated molecular beam deflection profile as a function of displacement from beam axis, for a given electric field, integrated over \( J, M_J \) and the beam velocity. Strong field is to the left. Undeflected beam profile is dashed line.

Fig. 2. Two typical experimental deflection profiles for CsCl at 4500 and 3800 V. Note bumps on strong field (left) side, attributed to the polarizability of the dimer.
background caused by the electric dipole moment, and a peak due to the polarizability of the alkali halide dimer. The dimer consists of two alkali halide molecules oppositely aligned. Accordingly, the dimer will not possess a dipole moment. Its polarizability will be partly attributable to the binding energy of the dimer \( \alpha_d \), and to the polarizability of the monomer \( \alpha_m \).

A polar molecule in the presence of an electric field \( E \), applied in the z direction is characterized by its rotational quantum number \( J \) (angular momentum \( \hbar J (J+1) \)) and its projection along the electric field axis \( M_J \). In general the Stark energy \( E \) is a complicated function of \( E \), for a given \( J, M_J \). In "weak field" the leading term in \( E \) is

\[
E = \frac{\mu_0 E^2}{\hbar^2/2A} \left\{ \frac{J(J+1) - 3M_J^2}{2J(J+1)(2J-1)(2J+3)} \right\}
\] (1)

where \( \mu_0 \) is the (body-centered) electric dipole moment and \( A \) is the molecular moment of inertia. The transverse force \( F_\perp \) exerted by an inhomogeneous electric field is

\[
F_\perp = -\frac{\delta E}{\delta z} = -\frac{\delta E}{\delta E} \frac{\delta E}{\delta z} = \mu_{\text{eff}} \frac{\delta E}{\delta z}
\] (2)

i.e., it is proportional to the derivative of Eq. 1 with respect to \( E \). This quantity is the "effective moment" \( \mu_{\text{eff}} \) of the rotating molecule in an electric field. Fig. 3 shows some relative Stark energies (solid lines), and the effective moments (dashed lines) for a polar molecule for \( J = 3, |M_J| = 3,0 \) and \( J = 7, |M_J| = 7,0 \). Note from Eq. 1 that \( E \) and \( \mu \) depend on \( |M_J| \). All Stark Energies start quadratic in \( E \). At higher fields, for all states for which \( |M_J|/J(J+1) < \cos^{-1}(1/3) \), the Stark energies go through maxima, and the effective moments \( \mu_{\text{eff}} \) through zero. At the position of these "zero moments" it should be possible to observe the polarizability deflections directly, without competition from the permanent dipoles.

In the present experiments we are operating almost exclusively in the "weak field" limit, except for the very small fraction of the non-state-selected beam which is in very low \( J \)-states. This is because the weak field region obtains for the
Fig. 3. Stark energy vs electric field (—) "effective" electric dipole moment vs electric field (----).
potential energy \ll \text{rotational kinetic energy, or}
\[ \mu_0 E << \hbar^2 J^2 / 2A = kT, \]
where T is the molecular beam oven temperature. For \( \mu_0 = 10 \text{ debye} \) (10\(^{-18}\)c.g.s.) and \( T = 1000^\circ\text{K} \), one requires \( E << 10^6 \text{ volt/cm} \), a condition easily satisfied in our present experiments. Typical \( J_{\text{max}}' \), where \( J_{\text{max}} \) corresponds to the most probable J for oven temperature T, range from 50-150 and accordingly, the total number of states we are dealing with here, of the order of \( |J_{\text{max}}^2| \), range from 2500 and higher.

Accordingly, one can assume continuous distributions of J, \( M_J \) in a deflection analysis. Thus, the transverse force \( F_\perp \) is
\[
F_\perp = \frac{a \mu_0 E^2 A}{2 \hbar^2 J^2} \left( 1 - 3 \cos^2 \alpha \right)
\]
where \( \cos \alpha = M_J / J \), and \( \alpha \) is the ratio of field-gradient to field. The beam displacement, \( \chi^- \), in the plane of the detector, for a molecule speed \( v \), is
\[
\chi^- = \frac{F_\perp \times \text{geometry factors}}{v^2} = \frac{b}{v^2}
\]
where \( b \) is a constant determined by \( E \), T and molecular and apparatus parameters. When integrated over a Maxwellian velocity distribution Eq. 4 can be used to obtain an expression for \( I(\chi) \), the beam intensity per unit displacement,
\[
I(\chi) = \frac{\text{const} \times I_0}{v^2} \exp \left( -m^2 b / kT \chi \right).
\]
Finally, integrating over \( J \) and \( M_J \), one obtains a distribution such as that sketched in Fig. 1.

In Fig. 2 we show several typical experimental deflection patterns. These are for CsCl for 4500 and 3800 V applied to the inhomogeneous electrodes. The observed deflection patterns are in good agreement with the theoretically derived curves such as those of Fig. 1, except for the bump on the left (strong field) side. We attribute this bump to the polarizability of the alkali halide dimer, which does not possess a dipole moment, and which would necessarily appear on the strong field side, since the polarizability force is always toward strong field.
Fig. 4 shows the position of the peak in KCl as a function of \((\text{applied voltage})^2\). The deflection is quadratic in \(V\), as expected. Using appropriate apparatus parameters, the average polarizability \(\bar{\alpha}\) is given by

\[
\bar{\alpha} = \frac{0.0315 T \times d(\text{mils})}{v^2}
\]

where \(d\) is the displacement of the strong-field peak from the beam axis in mils (0.001"").

Preliminary values for \(\bar{\alpha} = \frac{\alpha + 2\alpha}{3}\), obtained thus far are:

- CsCl: \(59.6 \times 10^{-24} \text{ cm}^3\)
- RbCl: \(47.3 \times 10^{-24} \text{ cm}^3\)
- KCl: \(43.4 \times 10^{-24} \text{ cm}^3\)

I.2 Scattering Experiments; new apparatus

As described in our original proposal, we have been constructing a new apparatus for the purpose of performing electron-atom and electron-molecule experiments. This apparatus will possess higher energy and angular resolution than has hitherto been attainable. It will be capable of studying both ground and laser-excited states, with polarized and state-selected beams. The apparatus is being built with the assistance of a capital equipment grant from the NSF.

The first DOE experiments planned for this machine are measurements of the total and differential cross sections for the scattering of slow electrons by alkali halide molecules, as described in the original proposal. We briefly discuss the present status of this program.

Fig. 5 is a block diagram of the machine, indicating the principal elements necessary for the molecular scattering experiments. Fig. 6 shows the physical setup, including some of the principal components. Referring to that figure, the status of these is as follows:

The vacuum chambers are completed, except for some blank flanges which are being machined now. Most of the vacuum components (pumps, gauges, circuits, etc.) have been already purchased or renovated from existing equipment.
Fig. 4. Strong-field peak displacement vs (electrode-voltage)$^2$ [(Kcl)$_2$ polarizability]
Fig. 5 Block diagram of new scattering apparatus.
Caption Fig. 6: A horizontal view of new apparatus

A. Source chamber, containing supersonic beam oven and cryogenic skimmer.

B. Housing for electrostatic beam focuser.

C. Buffer chamber, containing mechanical velocity selector.

D. Interaction chamber, containing cylindrical electron monochromator, retarding potential electron spectrometer and electron collector.

E. Horizontal and vertical bearings, centered at the interaction region, to guide the motion of the drift tube and detector chamber when measuring molecular angular distributions.

F. Drift tube.

G. Counterweight system to balance drift tube and detector chamber.

H. Detector chamber, to contain surface ionization, impact ionization, photoionization or fluorescence detector.

I. Motor drives for vertical and horizontal motions of the detector.

J. Scanning arm, bolted to bearings "E". The drift tube and detector chamber are mounted on it.

The two I-beams over which everything is mounted, are supported by four vibrational isolators.
The supporting plates and motions for each chamber have been fabricated. The angular scanning motion is finished, except for the motor drive units. The commercial motor drive units have been ordered.

The counterweight system (for the two-dimensional detector system motion) is completed, except for the lead weights, which have been ordered. A tentative time schedule for completion of the apparatus is given in the Renewal Proposal.

Other Activities

Professor Bederson visited both the Lawrence Livermore Laboratory (Isotope Separation Division) and Oak Ridge National Laboratory (Health Sciences Division), primarily to discuss aspects of our DOE research with members of these divisions. He gave seminars at both places, "Electron-Scattering by Laser-Excited Atoms", at LLL on October 18, 1978, and "Highly Polar Molecules - their Polarizabilities and Other Properties", at ORNL, November 13, 1978.
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