STUDY OF ELASTIC AND INELASTIC INTERACTIONS OF ATOMS AND MOLECULES BY SMALL ANGLE FAST MOLECULAR BEAM METHOD

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It is more than fifty years since the method of fast (energy ~1 keV) molecular beam scattering at small angles (10^{-4} – 10^{-2} rad) is used to study elastic scattering of atomic particles (atoms, molecules and ions). On the base of such experiments the repulsive interaction potentials were found. To extend the possibilities of this method to study inelastic processes we have designed and constructed the experimental setup at the Institute for Problems in Mechanics of Russian Academy of Sciences which enabled us not only to measure the fact of particle scattering (differential cross section measurements) but simultaneously to define the energy of a scattered particles (energy loss spectra measurements). Thereby one experiment combines both angular and energy measurements and allowing to obtain doubly differential cross sections. This was achieved by using the position-sensitive microchannel-plate detector and the energy loss spectra time-of-flight measuring technique. Measurements of doubly differential cross sections for various systems including molecules proved good possibilities for determination of not only elastic and inelastic differential cross sections, but also the studies of low-lying electronic states of molecule excitation for collision of fast projectile and target particles.

Introduction

Collisions of atoms, molecules and ions are of great importance for understanding the macroscopic properties of substance. Molecular beam method is the most straight one to study elementary processes during the collisions. Usually the experiments used thermal (energy E~0.01-0.1 eV) or fast (E~1 keV) beams for these purposes. The thermal beams are used in studies of collision processes governed by the potential well region and the attractive part of potential, while fast molecular beams imply collisions of particles with interaction energies of the short-range repulsive potential region. The fast molecular beam method first appeared in the USA at the beginning of the 40s when the supersonic aviation demanded the knowledge of substance properties at extreme conditions. The real experiment for such conditions is very difficult or next to impossible hence the mathematical simulations are often the only way to estimate these properties. Professor I.Amdur from Massachussetts Institute of Technology began measurements of the integral cross sections with the fast molecular beams [1]. The measurements yielded the short-range repulsive interaction potentials. He used them to calculate the high temperature gas properties. The fast molecular beams experiments in the USSR were linked with the space explorations and they had been started by professor V.B.Leonas back in the 60s in Moscow State University [2] to be continued in the Space Research Institute in Moscow. Nowadays this laboratory is situated at the Institute for Problems in Mechanics of Russian Academy of Sciences. For more than fifty years the fast molecular beam method is used to study the elastic collisions of atomic particles (atoms, molecules and ions). We decided to expand possibilities of this technique for studies of both elastic and inelastic collisions. This became possible by simultaneous angular and energy measurement of the scattered particle.

The particle energy is gained by the time-of-flight technique. Therewith today we have doubly differential cross sections measurements. Such measurements of doubly differential cross sections for various systems including molecules proved good possibilities not only to define elastic and inelastic differential cross sections, but also to study the excitation of low-lying electronic states of molecules for collision of fast projectile and target particles. This paper describes the experimental apparatus and some results on doubly differential cross section measurements and provides their interpretation.

Experimental facilities

Here is a brief description of the experimental apparatus for measuring both angular distribution and energy-loss spectra. The device enables us to study doubly differential cross sections for atoms, molecules and ions. A detailed description of the experimental apparatus can be found in papers [3], [4]. The time-of-flight technique was used to measure the collision energy losses [5], while the position sensitive detector measured the scattered particles angular distribution. Figure 1 shows the block diagram of the experimental apparatus.

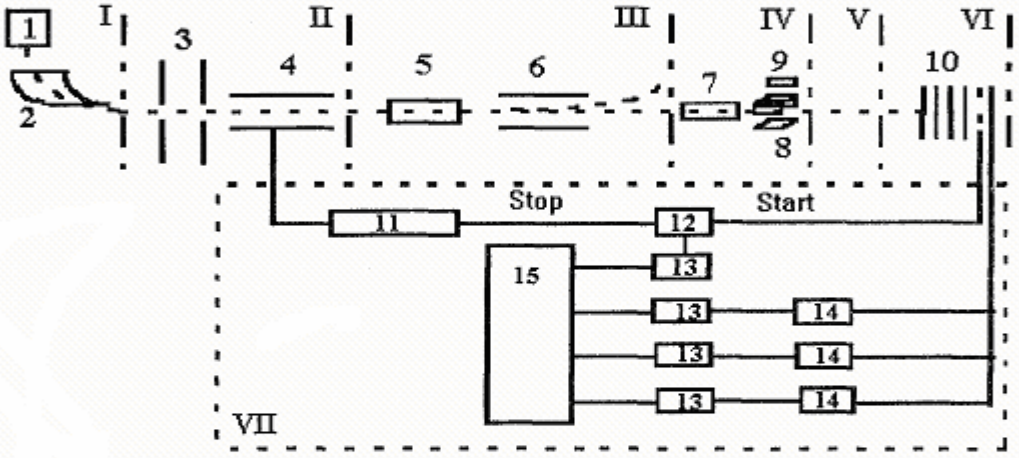

Fig.1. Block diagram of the experimental apparatus: 1-ion source, 2-magnetic mass analyser, 3- shaping slits, 4-modulating capacitor, 5-charge-exchange chamber, 6-deflecting plates, 7-scattering chamber, 8-beam monitor, 9-mutually perpendicular plates, 10-detector, 11-generator, 12-time-to-amplitude converter, 13-amplitude-to-digital converter, 14-charge-sensitive amplifier, 15-personal computer.

Section I forms an ion beam with a wide range of ion masses and the particle energies from 1 to 5 keV. Section II modulates the ion beam to create very short particle pulses necessary for the time-of-flight technique [5]. The modulated ion beam passes through the charge exchange chamber 5 (section III) where ions are neutralized. The survived ions are removed from the beam by the deflecting plates 6. Thereafter the neutral particle beam enters the scattering chamber 7 (section IV). Having travelled through the section V the scattered beam particles are detected by the detector 10. As a detector we use a home made assembly of three microchannel plates of 56 mm in diameter with the wedge and strip collector [4]. Our detector has a grid between the third microchannel plate and the collector. The grid signal is used as the time reference for the particle detection moment. The measuring-control complex VII comprises a personal computer 15 and a set of CAMAC modules (12, 13, 14). The control program makes it possible to process information in real time mode and visualize the results. The time of flight is determined by the time-to-amplitude converter 12 referenced by the signals from the modulating generator 11 (stop signal) and the collector grid (start signal). Three signals from the collector elements are amplified by the charge sensitive amplifiers 14 and digitized by the amplitude-to-

digital converters (13). The coordinates of a detected particle are determined from the collector element charge values [6]. Only those events are selected which satisfy the pre-assigned condition (e.g., rejecting events with the total charge values beyond the fixed limits may effectively bar both the thermal detector noise pulses and double particle events).

Results and discussion

The experimental apparatus tests involved scattering of the He-N_2 and He-CO systems for fast beam of He with the energies of 1.5 and 2.5 keV. The earlier measured differential cross section by a movable detector [7] is known to have the rainbow-like peak. The existence of such a peak was confirmed in [8]. We put forward a hypothesis [7, 9] that this peak is related to electronic or vibrational excitation. The earlier study of the inelastic energy-loss spectra [10] confirmed the electronic origin of the excitation of N_2 molecule in collisions with the fast projectile particles. The new experimental facility permits one to combine the measurements of the differential cross sections and the energy-loss spectra for all angles. Figures 2 and 3 demonstrate the results for He-N_2 systems. Black squares in Fig. 2 represent the experimentally measured differential cross section (reduced coordinates $\tau=\theta\cdot E$ and $\rho=\sigma\cdot\theta^2$ are used, where θ is the scattering angle, σ – differential cross section). In Fig. 2 one can see the rainbow like singularity for reduced angles of 10–14 eV·rad. Peak I in Fig. 3 corresponds to the elastically scattered particles (zero loss energy), peaks II and III show the energy loss due to the inelastic collisions. The ratios of peak I to peaks II and III accordingly reveal the elastic (open circle in Fig.2) and inelastic (triangles in Fig 2) differential cross sections.

Similar results for He-CO system are shown in Figs.4 and 5.

The elastic and inelastic differential cross sections shown in Figs.2 and 4 can be used for evaluation of the repulsive interaction potentials. The procedure of such an evaluation and results for the He-N_2 and N_2-N_2 systems are discussed in detail in [11].

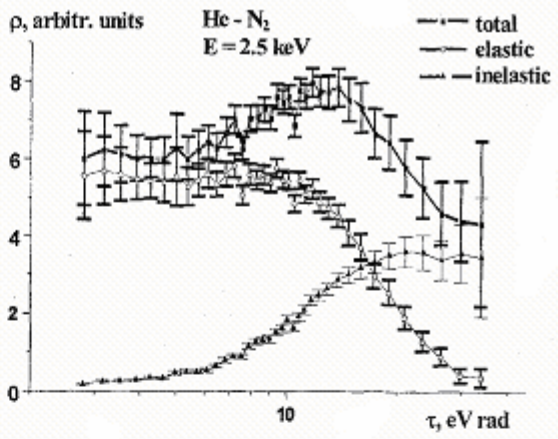

Fig.2. Differential cross sections of He-N_2 system.

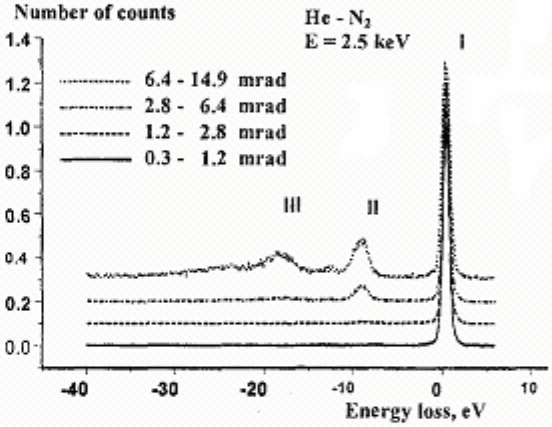

Fig.3. Energy loss spectra of He-N_2 system.

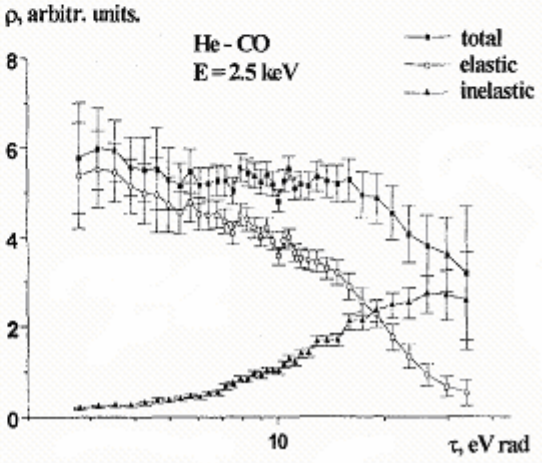

Fig.4. Differential cross sections for He-CO system.

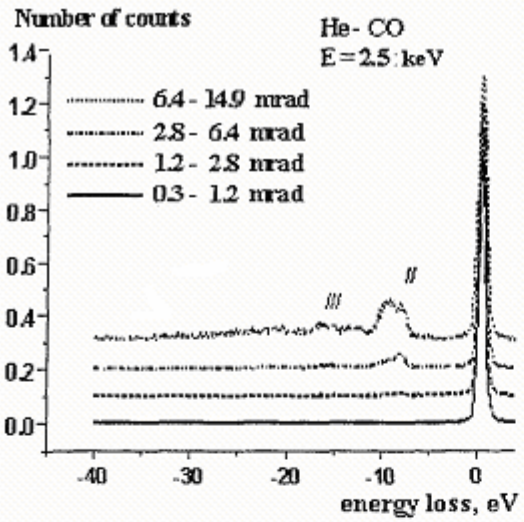

Fig. 5. Energy-loss spectra for He-CO system.

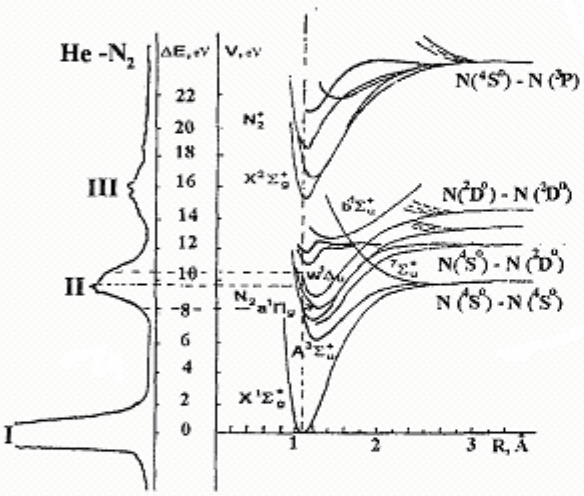

Fig. 6. Comparison of energy loss spectra of He-N_2 system with electronic terms of N_2 molecule.

On the base of Figs.3 and 5 a conclusion can be drawn that only excitation of molecules during the collisions with He atoms takes place (electronic excitation levels of He lie approximately near 20 eV and higher). Fig.6 helps to understand the origin of the peaks at the energy loss spectra for He-N_2 system. Here we can see the energy shift of these peaks (the left part of Fig.6) in comparison with the energy of vertical transition from zero vibration level of the nitrogen molecule ground state to the excited states (dot line in the right part of Fig.6). So peak II may be linked with excitation of nitrogen molecule on different electronically excited states.

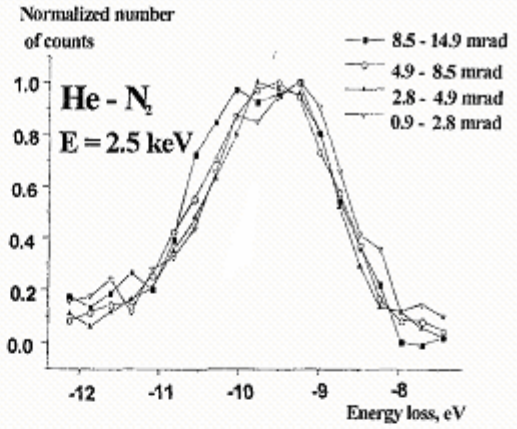

Fig.7. Dependence of peak II for He-N_2 system on scattering angle.

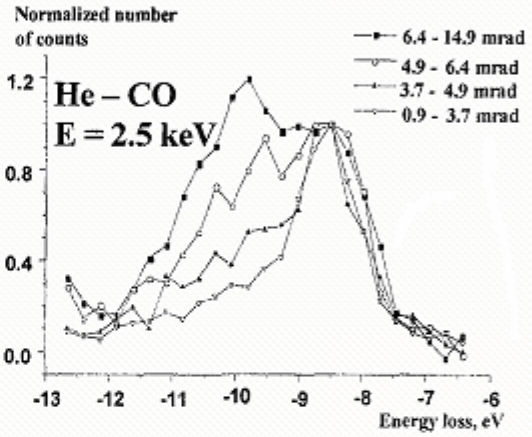

Fig.8. Dependence of peak II for He-CO system on the scattering angle.

The electron selection rules can be used to choose necessary transitions. Similar consideration can be applied to He-CO system. Consider the behavior and origin of peaks II for He-N_2 and He-CO systems. Figures 7 and 8 show the behavior of peak II for these systems for different scattering angles (all curves were normalized to 1). From these figures one can see the different behavior of these peaks. Dealing with the He-N_2 system all these normalized peaks almost coincide for different scattering angles. As for the He-CO system they have different shapes for different angles. This means that the origin of the peaks is connected with transitions to several excited states.

Using the energy-loss spectra and diagrams of terms of N_2 and CO molecules in the frameworks of Frank-Condon model we have tried to

interpret the peaks II to be a result of transition from ground to excited electronic states in N_2 and CO molecules and to determine the contribution of these electronic states. To do this we assumed that each transition on separate vibrational level fits the Gauss function with the center of this transition and the dispersion equals to dispersion of the elastic scattered beam. In this case the Gauss functions sum whose relative height is determined by Frank-Condon coefficients stipulates the electronic transition. Varying the heights of this way constructed functions per different electron transition we fit the calculated results to the experimental curve. According to the state selection rules $a^1\Pi g$, $a'^1\Sigma_u^+$, $w^1\Delta_u$, the terms were chosen for the He-N_2 system. Figure 9 demonstrates the compliance of the calculated and experimental pattern for the He –N_2 system (angle range is 1.6-14.9 mrad). The transition on term $a'^1\Sigma_u$ is seen to be suppressed and peak II for the He-N_2 system can be very well reproduced by transition on $a^1\Pi g$ and $w^1\Delta_u$ levels. The contribution of these transitions is determined by the ratio of the areas of peaks per each transition (see Fig. 9) and for this system does not depend on the scattering angle.

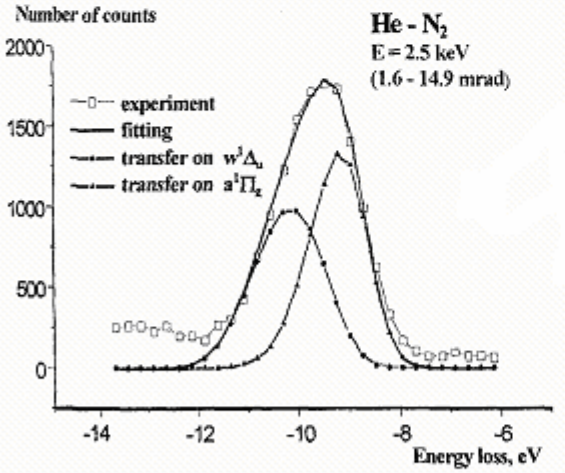

Fig.9. Interpretation of peak II for He-N_2 system.

For the He-CO system we choose $A^1\Pi$, $I^1\Sigma^-$ and $D^1\Delta$ as possible candidate terms. Figure 10 demonstrates the experimental curve fitting (the angular range is 6.4-14.9 mrad). The experimental curve is seen to be very well reproduced by only these two terms $A^1\Pi$ and $D^1\Delta$.

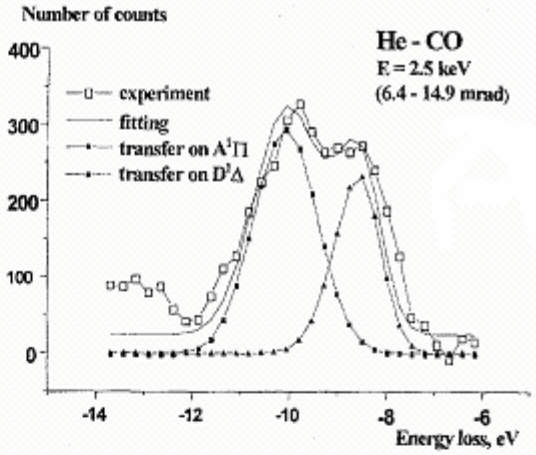

Fig.10. Interpretation of peak II for He-CO system

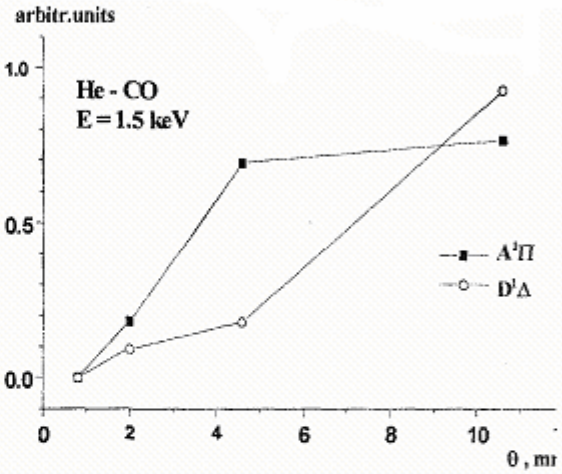

Fig.11. Contribution of excitation $A^1\Pi$ and $D^1\Delta$ terms versus scattering angle for He-CO system (beam energy 2.5 keV)

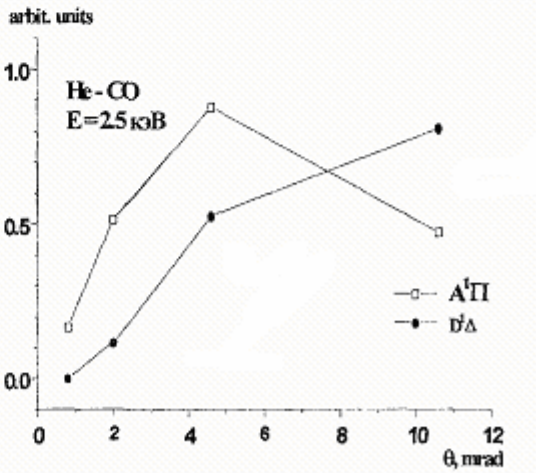

Fig.12. Contribution of excitation $A^1\Pi$ and $D^1\Delta$ terms versus scattering angle for He-CO system (beam energy 1.5 keV)

Such compliance gives a possibility to determine the dependence of transition cross sections to separate excited states. Figures 11 and 12 demonstrate such dependencies for the collisional excitation of the $A^1\Pi$ and $D^1\Delta$ terms for the He-CO system on the scattering angle for two beam energies.

Conclusions

The constructed experimental apparatus for fast beam small angle scattering allows one to measure the doubly (with respect to angle and energy) differential cross sections. The position-sensitive detector and time-of-flight technique to measure the scattered particles energy made possible to fulfill such experimental ambitions. The doubly differential cross section measurements for He-N_2 and He-CO systems demonstrate good possibilities to study elastic and inelastic collisions of atoms and molecules including collisional excitation of the low-lying electronic states. Such experiments permit to separate elastic and inelastic collisions measurements, and to identify transitions contributing to the singularities in the energy-loss spectra.

References

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ДОСЛІДЖЕННЯ ПРУЖНИХ І НЕПРУЖНИХ ВЗАЄМОДІЙ АТОМІВ І МОЛЕКУЛ МЕТОДОМ СКОЛІМОВАНОГО МОЛЕКУЛЯРНОГО ПУЧКА

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Метод малокутового (10^{-4} – 10^{-2} рад) розсіювання швидких (енергія ~1 кеВ) молекулярних пучків понад 50 років використовується для вивчення пружного розсіювання атомних частинок (атомів, молекул, іонів). На основі таких експериментів знайдено потенціали відштовхувальних взаємодій. З метою розширення можливостей цього методу на вивчення непружних процесів нами спроектовано і створено експериментальну установку в Інституті проблем механіки Російської академії наук, яка дозволила не тільки встановити факт розсіювання частинки (вимірювання диференціального перерізу), а й одночасно визначити енергію розсіяних частинок (вимірювання спектрів енергетичних втрат). Таким чином один експеримент поєднує кутові та енергетичні вимірювання і дозволяє отримати подвійні диференціальні перерізи. Вимірювання подвійних диференціальних перерізів для різних систем включно з молекулами підтвердили хороші можливості для визначення не тільки диференціальних перерізів пружного і непружного розсіювання, а й досліджень збудження нижчих електронних станів молекул для зіткнень швидких налітаючих частинок і мішеней.