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DISCLAIMER

SCATTERING OF DIATOMIC AND POLYATOMIC MOLECULES FROM THE (100) CRYSTAL FACE OF PLATINUM

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ABSTRACT

The angular distribution of diatomic (CO, N_2, O_2, NO, H_2, D_2) and polyatomic (CO_2, N_2O, C_2H_2, NH_3 and methylene cyclobutene) molecules were monitored upon scattering from the clean and carbon covered (100) crystal face of platinum. All of the scattered molecules (except ammonia) exhibit directed scattering with intensity maxima at or near the specular angle, indicating poor energy accommodation during collision. On scatterings CO and C_2H_2 from an adsorbed layer of carbon monoxide or acetylene the angular distributions were cosine-like that indicate significant energy accommodation between the incident gas molecules and the surface. It appears that if the vibrational energy of surface atoms and the translational energy of the incident molecules are of the same magnitude, the incident particles will be reflected. The near complete energy accommodation of the scattered molecules in the presence of adsorbed gases could take place if high frequency surface modes are created on adsorption of these low molecular weight gases. This way, there is matching of vibrational frequencies of the collision pairs (gas molecules - adsorbed molecules) which allow strong vibrational (V-V) energy transfer.

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I. INTRODUCTION

Studies of the scattering of atomic and molecular beams from solid surfaces reveal the nature of energy transfer between an incident atom or molecule and surface atoms. For monatomic gas atoms energy can only be exchanged between the translational modes of the incident atom and the vibrational states of atoms in the surface. The interaction of monatomic gases with solid surfaces has been studied sufficiently to allow the development of the "hard-cube" and "soft-cube" scattering models.[1,2] These models, and more sophisticated models, explain qualitatively many of the scattered beam properties: its surface and beam temperature dependence and its dependence on the mass ratio $(m_{gas}/m_{surface})$. The major assumption of the cube models, that momentum changes only along its perpendicular component while the magnitude of the parallel component is conserved, stands up well under experimental scrutiny.

For diatomic molecules, however, the interaction between the gas and the solid is more complex due to the possibility of additional energy exchange between the internal states of the molecule (rotation and vibration) and the vibrational modes of the surface atoms. That such energy exchange takes place efficiently has been demonstrated by the striking observations of Saltsburg et al.[3] in their studies of the scattering of H_2, D_2 and HD molecules from the Ni(100) surface. H_2, whose rotational energy states were separated by $\Delta E_{rotation} > kT$ at the experimental temperatures, showed a sharp intensity maximum at the specular angle when directed at the solid in the form of a molecular beam, while beams of HD and D_2 for which $\Delta E_{rotation}$ is of

the order of kT, produced broad angular distributions, indicating efficient energy exchange. Further evidence that surface scattering involves the excitation of rotational energy states was obtained by increasing the beam temperature and observing changes in the angular distribution of scattered H_2. An energy accommodation model, developed by Feuer[4] for the case of diatomic molecules scattered from surface atoms, does indeed predict efficient energy exchange between surface vibrations and the rotational modes of diatomic molecules. Experimental studies have indicated that energy transfer takes place efficiently when the magnitude of the surface vibrational energy quantum is comparable to the energy to be transferred to or from the internal states of the gas molecule. At temperatures above the Debye temperature, the thermal energy, 3RT, may be used to estimate the maximum quanta of vibrational energy that is transferred by a surface atom.

In order to investigate further the nature of energy transfer between the internal modes of molecules and the vibrational modes of surface atoms, we have studied the angular distribution of several diatomic and polyatomic species (CO, NO, O_2, NO_2, H_2, D_2, CO_2, N_2O, C_2H_2, methylene cyclobutene and NH_3), scattered from the clean (100) face of a platinum single crystal and from the same surface covered by a layer of graphite.[5] We have also monitored the angular distribution of molecules scattered from a layer of like molecules (CO, C_2H_4) adsorbed on the surface.

The energy transfer appears to be poor (except for ammonia) when scattering takes place from clean or carbon covered platinum surfaces. On the other hand, efficient energy transfer takes place on scattering

CO or C_2H_2 in the presence of adsorbed (CO or C_2H_2 molecules. These results can be explained assuming the presence of high frequency vibrational modes associated with the adsorbed molecules. This way, there is a matching of vibrational frequencies of the collision pairs (surface atoms--gas molecules) which produces the efficient vibrational-vibrational (V-V) energy transfer. These vibrational frequencies may be estimated from the simple models[6] that were proposed.

II. EXPERIMENTAL

The apparatus used for these experiments has been described in detail elsewhere.[7] In brief, the molecular beam emanates from a source consisting of a multi-channel array and a reservoir maintained at a pressure of several torr. Differential pumping is used between the source and the scattering chamber to reduce the ambient gas pressure to UHV levels. At the single crystal surface, the incident flux is approximately 10^{13} molecules/cm^2/sec, modulated at 150 Hz to allow phase sensitive detection of the scattered signal. During the experiments a high speed ion pump maintains the background pressure in the scattering chamber in the low 10^{-9} torr range. The scattered beam in detected by a quadrupole mass spectrometer with an ionizer acceptance angle of $7°$. Due to the open construction of the ionizer, the ion intensity is proportional to the number density of molecules in the range of the acceptance angle. Scattered signals are reported in the plane of incidence; no attempt was made to analyze the out-of-plane scattering intensity.

Surface structures of the clean surface or of those resulting from adsorbed gases were monitored by low energy electron diffraction (LEED) optics built into the scattering chamber. The Pt(100) single crystal surface was characterized by a LEED diffraction pattern indicating a (5×1) surface structure when clean, and a (1×1) structure following the adsorption of carbon monoxide, carbon or hydrocarbons.[8] Its cleaning required heating in an oxygen pressure of 10^{-5} torr at 800°C for an extended period (10 minutes-100 minutes depending on the amount of carbon on the surface). A layer of surface carbon was the

the major impurity encountered in these experiments. Surface cleanliness
was determined by Auger electron spectroscopy (AES), carried out using
the LEED electron optics. Typical Auger spectra of a clean and of a
CO covered platinum surface are shown in Fig. 1. An auxilliary titanium
sublimation pump was used to reduce the partial pressure of CO in the
ambient when desired. In each experiment it was possible to monitor
not only the scattered beam intensity as a function of scattering angle,
but also to detect the incident beam intensity. This way the data could
be presented in terms of the mormalized beam intensity $I(\theta)/I(\text{beam})$ that
allows a direct comparison of all of the scattering curves for different
gases.

III. RESULTS

A. Scattering from the Clean Pt(100)-(5×1) Surface

Figures 2-4 show the angular distributions of CO, N_2 and O_2 scattered from the clean platinum surface. The cleanliness of this surface during an experiment could easily be checked since the diffraction beam intensities due to the (5×1) surface structure are known to deteriorate rapidly upon the adsorption of ambient gases (mostly CO). Scattered beam intensities have been normalized to the incident beam intensity so that all of the beam intensities in these figures can be compared directly. The angular distribution of the scattered beam is peaked at or near the specular angle for all of these diatomic gases. It should be noted that for these experiments the source temperature (which defines the beam temperature T_{beam} for a Maxwellian velocity distribution) was 300°K while the surface temperature was somewhat hotter (475°K). It was necessary to maintain this slightly elevated temperature during the scattering experiments with the clean platinum surface because the surface temperature must be above the point at which CO in the α binding state desorbs. Otherwise, the surface would soon be covered with a layer of α-CO.[8]

Figure 5 shows the angular distribution of nitric oxide, NO, scattered from the (100) face of platinum. The surface temperature for this experiment was 1200°K because the thermal dissociation of N_2O was also being investigated.[9] Due to the higher surface temperaturs, the angular distribution is broader than those previously shown and the peak intensity appears at the position nearer to the surface normal, than the specular angle, as it should according to various models for

momentum transfer from the hot surface to the colder gas. The angular

distribution is still peaked however, and not cosine in nature, thus

indicating the lack of complete energy accommodation as in the case

of all the other incident diatomic molecules.

The scattering of molecular beams of hydrogen and deuterium was

also studied. The results are shown in Figs. 6 and 7 for a clean

platinum surface maintained at 1175°K. Compared to other diatomic

gases scattered from hot surfaces, these curves are intense and the

maxima fall at the specular position as noted in previous studies.[10,11]

While the angular distributions are surprisingly broad for both molecules,

it appears that the angular distribution of scattered deuterium beam

may be somewhat narrower than that of hydrogen and that the overall

intensity may be slightly higher, although the experimental uncertainty

does not permit a definitive answer at this time. Neither the presence

of oxygen nor hydrogen in the ambient of the scattering chamber seemed

to effect the D_2 scattering, but there was a large amount of scatter

in the data and a high background noise level.

Several polyatomic gases were also scattered from the clean (100)

face of platinum. Figures 8 and 9 show the angular distribution of

the scattered beams of two tri-atomic molecules, CO_2 and N_2O. Again,

the angular distributions are peaked at or near the specular angle.

The higher surface temperature in the N_2O study[9] has shifted

the peak intensity somewhat toward the surface normal. Figures 10

and 11 show the angular distributions of scattered beams of two

hydrocarbons, acetylene and methylenecyclobutane. As in the

previous cases the intensity maximum occurs near the specular position

for low surface temperature (acetylene) and is backscattered at high

surface temperatures (methylenecyclobutane). In summary: the

scattered diatomic and polyatomic molecules we have investigated have

angular distributions with intensity maxima at or near the specular

angle.

The only exception to this type of directed scattering from the

Pt(100) is that of ammonia. Ammonia molecular beams had a cosine-like

scattered angular distribution as can be seen in Fig. 12. The ammonia-

platinum interaction appears to be strong that leads to near thermal

equilibrium between the gas molecules and the surface during the collision

time.

B. Scattering from Molecules Adsorbed on
the (100) Face of Platinum

Carbon monoxide and acetylene both chemisorb readily on platinum

at 300°K. Carbon monoxide forms several ordered surface structures

depending on the surface concentration and surface temperature.[8]

Acetylene forms a $(\sqrt{2} \times \sqrt{2})$-R45° surface structure.[12] Although the

surface order is easily discernible by low energy electron diffraction

the high background intensities in the diffraction pattern indicate a

fair amount of disorder in the adsorbed layer and this has been sub-

stantiated by molecular beam studies.

The angular distribution of carbon monoxide scattered from an

adsorbed layer of carbon monoxide on platinum is shown in Fig. 13.

The scattered beam has a very broad angular distribution that does not

change with differing angle of incidence, thus indicating a change in

the nature of gas-solid interaction. This result should be contrasted

with the scattering of CO from the clean platinum surface (Fig. 2) that
had an intensity maximum at the specular angle. The angular distribution
of acetylene scattered from a layer of adsorbed acetylene is shown in
Fig. 14. The scattered beam again has a broad, cosine-like distribution.
These observations seem to indicate that complete energy accommodation
occurs between the surface and the incident molecules within the collision
time.

Carbon forms a graphitic overlayer on the (100) face of platinum.
The resultant ring-like diffraction pattern indicates the presence of
domains of graphite that lie with their basal plane parallel to the
surface and yet show rotational disorder. We have scattered the molecular
beams of several diatomic molecules from this carbon layer. Figures
15a and b show examples that in every case the angular distribution of
the scattered beam was peaked near or at the specular angle, indicating
incomplete energy accommodation between the incident molecules and the
carbon covered surface.

IV. DISCUSSION

One of the general conclusions one may draw from the results

obtained by monitoring the angular distribution of the scattered beams

of diatomic and polyatomic molecules from the clean platinum (100)

surface is that in every case (with the exception of ammonia) the

angular distribution is non-cosine, it has an intensity maximum at

or near the specular angle. The peak intensities and the widths of

the angular distributions may vary depending on the surface temperature.

The kinetic energy of the scattered molecules at the various angles

has not been determined in these experiments. Thus, any conclusion as

to the nature of energy transfer between the surface atoms and the

incident molecules has to be arrived at from the inspection of the

angular distribution. The absence of a cosine-like scattering distri-

bution and the large intensity maximum near the specular angle does

indicate, however, that there is incomplete energy accommodation between

the platinum surface atoms in the single crystal surface and the incident

gas molecules. It is likely that some fraction of the molecules that

appear at the specular angle have velocities identical to the incident

velocity, while others also appear at this angle after undergoing

inelastic scattering.

Results also indicate that energy transfer is inefficient when

the incident molecules are scattered from carbon-covered platinum

surfaces. It appears that the vibrational modes of platinum atoms at

the surface, or of carbon atoms in the graphite layer on the platinum

surface, impart enough momentum to reflect the diatomic or polyatomic

species with high efficiency before adsorption can take place and permit

complete energy accommodation. It should be noted that directed scattering occurs from both of these surfaces regardless of the large difference in atomic weights between platinum and carbon.

Once a layer of adsorbed CO is present on the surface, however, energy accommodation of the incident CO molecules is entirely different from that on clean platinum or on carbon covered surfaces. The incident molecule collides with the carbon monoxide layer and is then reflected with a near-cosine angular distribution indicating significant energy accommodation. Thus, the carbon-oxygen bond of the adsorbed CO molecule has suitable vibrational modes that permit efficient energy transfer between the CO(gas) and CO(adsorbed) while no such modes exist between CO(gas) and C(adsorbed) or CO(gas) and Pt(surface). In the same way, C_2H_2(gas) shows complete energy accommodation with an adsorbed C_2H_2 layer within the collision time as indicated by the distribution of scattered beam that again exhibits cosine character. It appears that these low molecular weight adsorbed gases have vibrational modes that allow rapid energy transfer on collision, while adsorbed carbon, with an even smaller atomic weight, and platinum itself, do not.

Recently Beebe and Dobryzhnsky[13] have suggested that if vibrational modes that are closely similar in energy to the thermal translational energy of the incident particle exist for surface atoms, the incident particles will be reflected. In the absence of such modes, the particle may be adsorbed and must wait for desorption until the surface modes come into phase. Thus translational(T)-vibrational(V) energy transfer between incident molecules and the surface is not very likely if the two are matched in energy, but such an energy transfer

results in a reflection of the molecule from the surface. For these

relatively high temperature experiments (T > Debye temperature), the

thermal energy of surface atoms that is due to the various modes of

lattice vibrations is of the same magnitude as the kinetic energy

of the incident molecule. The energy of excitation of diatomic

molecules to their first excited vibrational state, however, is so

high ($\approx$0.1 eV) that the surface phonon energies ($\approx$0.02 eV) are in-

sufficient to cause vibrational excitation in the diatomic molecule at

the experimental temperatures.

Armand et al.[6] have suggested that adsorption of a low molecular

weight gas on a surface creates high frequency surface modes, the

frequencies being similar to the vibrational frequency of diatomic

gas molecules. On the other hand, the adsorption of high molecular

weight gas molecules with respect to the atomic weight of the adsorbing

surface atoms creates low frequency surface modes whose frequencies

are lower than the vibrational modes of the adsorbing solid. This

theory is yet to be tested rigorously, but it would predict the thermal

accommodation of CO molecules by a layer of adsorbed CO or the accommo-

dation of C_2H_2 molecules by adsorbed C_2H_2, as due to very efficient V-V

energy transfer processes between the high frequency vibrational modes

of the adsorbed molecule and the incident molecule. Also, the vibrational

modes of adsorbed molecules of higher energy than the kinetic energy of

the incident molecule would not allow, according to the Beebe model,

reflection of the incident particles.

So far we have not considered the possibility of the exchange of

energy between the rotational modes of the incident diatomic molecule

and vibrational states of the surface atoms. While the ground and first excited vibrational energy states of the gas molecules are 0.1-0.2 eV apart for most of the diatomic and polyatomic molecules considered here, the first two rotational energy states are $\sim 10^{-3}$ eV apart for nitrogen and for all other molecules with the exception of H_2 and D_2. It has been shown by Saltsburg et al.[3,10] that coupling between the vibrations of surface atoms and the rotations of diatomic molecules can yield strong V-R interactions that may rotationally excite diatomic molecules (at least in the case of H_2, D_2 or HD). However, due to the small energy separation of rotational states for all other molecules, many of the higher rotational states are already excited. Feuer's model predicts that whenever the temperature of the solid exceeds that of the gas (or beam), not only can vibrational energy be transferred from the lattice into rotational modes of the gas, but also some of the rotational energy of the gas may be converted into translational energy. The net effect of these V-R and R-T energy transfer processes should be to broaden the angular distribution of the scattered beam and to impart a diffuse component to the scattered gas. This energy transfer mechanism might be especially effective in the case of ammonia due to its large number of rotational and vibrational modes, and may give rise to the observed cosine-like distribution.

In summary, the absence of high frequency vibrational modes for surface atoms that would match the vibrational excitation energies of incident diatomic molecules is the likely cause of the poor energy accommodation on platinum or carbon covered platinum single crystal surfaces. Adsorbed carbon monoxide or actylene facilitate efficient

energy transfer between the surface and the incident molecules. This
can be explained by assuming the creation of high frequency surface modes
on adsorption of these gases that allow strong V-V energy transfer
between the collision pairs.

V. COMPARISON WITH OTHER SCATTERING RESULTS

It is possible to compare the scattering results above, with data previously obtained for scattering various diatomic and polyatomic molecules from the (111) surface of several other fcc metals, at the (110) face of tungsten and different polycrystalline solids. In general, there is good agreement with our observations of poor accommodation of the incident beam on clean surfaces and efficient accommodation on gas covered surfaces. Coltharp, Scott and Muschlitz[14,15] have studied the scattering of nitrous oxide from a tungsten ribbon composed primarily of crystallites having the (112) orientation. At room temperature they detected a cosine angular distribution for the reflected gas. The ambient pressure in their scattering chamber was 10^{-6} torr, high enough to insure that the surface was covered by adsorbed gases. They also observed, however, that high surface temperatures yielded pronounced lobular distribution with broad intensity maxima in the vicinity of the specular position. These same authors report that both nitrogen and nitric oxide are backscattered from a tungsten surface heated to 2500°K. Although our platinum surface temperature was considerably lower, we have also observed that nitric oxide scattered back toward the surface normal from the hot surface in agreement with their observation. The scattering of nitrogen molecules has been studied by several authors.[15,16,17] Hurlbut[16] found that nitrogen was scattered diffusely (i.e. with a cosine angular distribution) from polished steel, polished aluminum, and unpolished glass except at near-grazing incident angles ($\theta_{incident} \geqslant 80°$) where some peaking was observed. In view of the poor residual vacua achieved, (10^{-6} torr) and the presence in the

work chamber of hydrocarbon vapors from the diffusion pump oil, the

specimen surfaces were undoubtedly contaminated and we would thus expect

them to yield distribution of scattered gas molecules close to the

cosine pattern. Stickney and Hurlbut[18] and Stickney[19] have measured

the momentum transfer between several gases (He, Ne, Ar, N_2, H_2, CO_2)

and the surfaces of tungsten, aluminum and platinum, through the use

of a sensitive torsion balance. Angular distributions are not determined

and thus, direct comparison of their data with those reported in

this paper cannot be carried out. In the only other systematic inves-

tigation of the scattering of polyatomic gases besides the studies

reported here, Saltsburg et al.[17] have reported the scattering of several

molecules, H_2, HD, D_2, O_2, CO and CO_2 from an epitaxially grown (111)

silver single crystal. Their results for nitrogen show a slight back-

scatter but the surface was held at 560°K in their experiment, as

compared to T_S = 475°K in our work where not backscatter was observed.

A better comparison between their work and our work may be achieved

by looking at the scattering of nitrogen and oxygen. Their data indicated

that both curves have maxima at the same location and are similar in

intensity and peak shape. From our figures (Figs. 3 and 4) we can see

that the scattering of nitrogen and oxygen from the Pt(100) surface

yields the same results. Additionally, CO_2 scattering in their system

produces a lower intensity maximum than the scattering of either N_2 or

O_2, although all three gases have peak intensities at the same angle.

Comparing our data with theirs shows that similar trends exist for

the platinum surface.

There are no data in the literature for hydrocarbon scattering
except for methane, which shows backscatter from a (111) silver surface
at T_S = 560°K.[20] Smith and Merrill[21] have studied the irrerversible
chemisorption of ehtylene on a Pt(111) single crystal by combined LEED
and helium beam techniques, but evidently did not attempt to scatter
the gas. Thus, our results for acetylene and methylenecyclobutane
must stand alone. The likelihood of electronic or vibrational excitation
of C_2H_2 by the clean platinum surface is small, although we already
know from our work that this gas readily adsorbs and decomposes on
platinum. Methylenecyclobutane seems to be the largest projectile
(13 atoms) used to date in gas-solid scattering experiments.

In their investigation of the scattering behavior of polyatomic
gases on (111) silver, Saltsburg et al.[17] found that the CO and CO_2
scattering distributions are quite similar . This is in agreement with
our observations of CO and CO_2 scattering from the (100) platinum surface
if we take into account the adsorption of CO on Pt surfaces and that
CO does not adsorb on silver at 300°K. The surface of silver was also
employed in a study of the angular distribution of scattered ammonia.[20]
The pattern was found to be very broad and almost completely diffuse
in nature, which is similar to the data shown in Fig. 12 for ammonia
scattered from Pt(100).

Much attention has been directed toward the scattering of hydrogen
and deuterium from metal surfaces, and data now exist for experiments
conducted on Ni,[3,22] Au,[23] Ag,[10,24] Pt[11,25,26] and W.[27] For all of
these solids, provided the surface is clean, the maximum intensity
occurs at the specular positions, although the dispersion or half-width

of the scattered beam varied somewhat. In order to explain the broad

peak that was observed in many experiments between the surface normal

and the specular angle in the angular distribution of H_2, HD, and D_2,

Palmer et al[3,10,24] invoked energy transfer between the rotational

energy states of the incident hydrogenic species and the vibrational

modes of the surface atoms in the silver and nickel lattices. Their

model for R-V energy transfer is further supported by the beam tempera-

ture dependence of the deuterium scattering distribution. There can be

no doubt about the markedly different scattering distributions observed

by Palmer et al. when hydrogen isotopes were impinged on the epitaxially

grown thin film. Our studies of H_2 and D_2 scattering from the Pt(100)

surface and those of Smith[28] for D_2 from the Pt(111) surface show no

peaks other than the specularly directed one, and we find no evidence

for marked differences between the angular distributions of scattered

H_2 and D_2 molecular beams. Smith and Merrill,[11] however, have found

a spatial distribution reminiscent of those observed by Palmer et al.

when D_2 is scattered from an unetched Pt(111) crystal surface. This

result indicates that perhaps surface roughness is important in effecting

R-V energy transfer between incident diatomic molecules and the solid.

It appears that the epitaxial silver and nickel surfaces used in the

experiments by Palmer et al. could have been considerably rougher on

an atomic scale than the well-characterized platinum single crystals

used in our work and the work cited above. In addition, the surface

temperature in our studies was higher (1175°K) than the surface tempera-

ture used in the scattering studies with silver (570°K). If microscopic

disorder enhances rotational coupling to the solid, even a surface "disordered" through heating (due to the large mean square displacement of surface atoms) could possibly cause significant internal excitation of hydrogenic molecules. Unfortunately, systematic variation of the surface temperature during the scattering of H_2 and D_2 beams have not been carried out mostly because of the likely contamination of the platinum surface at lower temperatures by the adsorption of carbon monoxide from the ambient. Such study will be carried out in the near future to uncover the causes of the discrepancy between the deuterium beam scattering characteristics reported by the various investigators.

We have already pointed out that the hydrogen and deuterium data presented here (Figs. 6 and 7) show rather broad peaks. At T_S = 775°K Smith[28] reported a half-width of 29.5° for D_2 scattering from Pt(111) and a maximum intensity of 1.5%, down from 3.6% at T_S = 375°K. These values may be compared to those we find at 1175°K for D_2 scattering from the Pt(100): the maximum intensity 0.9% and the half-width 50°. One would expect a lower intensity and perhaps broader dispersion due to the higher surface temperature. Feuer's analysis[4] of the energy exchange between rigid rotors of diatomic molecules and a solid surface also helps to explain the observations as discussed earlier.

Although there is the possibility of V-T energy transfer for diatomic molecules from the clean surface and our date indicates that there is evidence for efficient V-V energy transfer between the incident diatomic molecules and diatomic molecules adsorbed on the platinum surface, it is difficult to obtain accurate information on the details of the energy

transfer without an energy analysis of the scattered beam. It appears that to obtain reliable V-V, V-T and T-R energy transfer information one must measure the kinetic energy of the scattered beam by time-of-flight techniques that would modulate the scattered beam and measure the time-of-flight after scattering. Experiments are in progress in our laboratory on a modified apparatus that allows us to measure the translational energy (velocity) of the scattered beam to obtain more detailed information about energy transfer between surface atom vibration and internal energy states of diatomic and polyatomic molecules.

VI. CONCLUSION

Unlike monatomic gases, diatomic molecules may exchange energy with the vibrational modes of surface atoms (i.e. surface phonons) through their vibrational and rotational energy states. From clean surfaces, we infer from the angular distributions that many diatomic and polyatomic molecules scatter without complete energy accommodation. This can be explained by the strong recoil effect due to matching of the translational energy of the incident molecule with the vibrational energy of the surface atoms. The energy accommodation of the incident diatomic molecule appears to be complete when scattered by an adsorbed layer of the same diatomic molecule. Armand's model that postulates, upon the adsorption of low molecular weight gases, the creation of high frequency vibrational modes that have the same magnitude as the vibrational frequencies of the incident diatomic molecules and much higher frequencies than the lattice phonons, can explain these findings. The complete energy accommodation in this case is due to V-V energy transfer that, apparently, is very efficient. V-R and R-T transitions are also possible and when they take place angular distribution of the scattered beam is broadened.

ACKNOWLEDGEMENT

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FIGURE CAPTIONS

Fig. 1   Auger spectra of clean and carbon and carbon monoxide covered
        platinum surfaces.

Fig. 2   Carbon monoxide scattering distribution taken by flashing the
        Pt(100) sample between data points (I_{Beam} = incident beam
        intensity).

Fig. 3   Angular distribution of a nitrogen beam scattered from a
        clean Pt(100) surface (I_{Beam} = incident beam intensity).

Fig. 4   Angular distribution of an oxygen beam scattered from the
        clean Pt(100) surface.

Fig. 5   Nitric Oxide scattering distribution from a clean (100) platinum
        surface.

Fig. 6   Angular distribution of hydrogen beam scattered from a clean
        Pt(100) surface.

Fig. 7   Angular distribution of a deuterium beam scattering from a
        clean Pt(100) surface.

Fig. 8   Angular distribution of a CO_2 beam scattered from a clean
        Pt(100) surface.

Fig. 9   Angular distribution of a nitrous oxide beam scattered from a
        clean Pt(100) surface.

Fig. 10 Angular distribution of an acetylene beam scattered from a
        Pt(100) surface.

Fig. 11 Angular distribution of a beam of methylene cyclobutane scattered
        from a Pt(100) surface.

Fig. 12 Angular distribution of an ammonia beam scattered from a clean
        (100) crystal face of platinum.

Fig. 1

XBL 719-7306

Fig. 2

Fig. 3

Fig. 4

XBL 708-1966

Fig. 5

XBL 7110 - 7347

H₂/Pt (100)-(5X1)
θᵢ =45°
T_B = 300°K
T_S =1175°K

Fig. 6

XBL 7110-7343

D_2/Pt (100) $-(5 \times I)$
$\theta_i = 45°$
$T_B = 300°K$
$T_S = 1175°K$

Scattered % of Incident Beam

θ_r, Angle from Surface Normal (Degrees)

XBL 7110-7342

Fig. 7

CO_2/Pt (100)
Surface: (5x1)
$T_S = 475°K$
$T_B = 300°K$
$\theta_i = 45°$

Fig. 8

XBL 708-1969

N_2O/Pt (100)
Surface: (5x1)
$T_S = 1125°K$
$T_B = 300°K$
$\theta_i = 45°$

scattered % of incident beam

θ_r, angle from surface normal (degrees)

XBL 7012-7439

Fig. 9

Fig. 10

XBL 708-1968

Fig. 11

XBL 7110 - 7344

-38-

XBL7110-7345

Fig. 12

Fig. 13

XBL715-6731

Fig. 14

Fig. 15a

XBL 7110-7348

Fig. 15b

Fig. 15c

XBL 7110-7349

Fig. 15d