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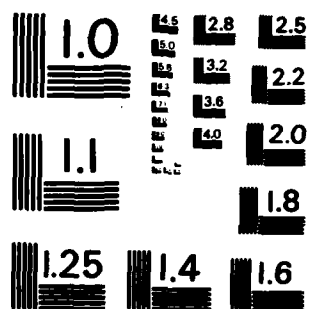END
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Negative-Ion Formation from Surface Scattering
and the Anderson Correlation Energy U

by

Kai-Shue Lam, K. C. Liu and Thomas F. George

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NEGATIVE-ION FORMATION
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CORRELATION ENERGY
H^+ + CESIATED W(100)
GOOD AGREEMENT WITH EXPERIMENT

20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A theoretical investigation of negative-ion formation from positive-ion-surface scattering is presented from a unified point of view. Based on the time-dependent Anderson-Newns model, the correlation energy U is seen to play an important role in the two-electron transfer process. Calculations of the probability of negative-ion formation are in good agreement with experiments on the conversion of $H^+(D^+)$ to $H^-(D^-)$ by scattering from a cesiated W(100) surface.

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NEGATIVE-ION FORMATION FROM SURFACE SCATTERING

AND THE ANDERSON CORRELATION ENERGY U

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Abstract

    A theoretical investigation of negative-ion formation from positive-ion-surface scattering is presented from a unified point of view. Based on the time-dependent Anderson-Newns model, the correlation energy U is seen to play an important role in the two-electron transfer process. Calculations of the probability of negative-ion formation are in good agreement with experiments on the conversion of $H^+(D^+)$ to $H^-(D^-)$ by scattering from a cesiated W(100) surface.

Charge-exchange processes arising from monoenergetic ion beams scattered from solid surfaces have recently been the subject of much experimental[1-5] and theoretical[6-10] interest. Most of these studies focus on the process of ion neutralization, involving the transfer of a single electron from the surface to the singly-charged ion.[1-3,6-10] However, owing to the increase of applications involving the transfer of two electrons in a variety of situations, such as plasma fusion[11,12] and the generation of high-energy neutral beams,[11] attention has also been directed to processes of negative-ion formation.[4,5,13]

For ion neutralization, the majority of theoretical work has been based on the Anderson-Newns model,[14] originally proposed for the explanation of local-ized magnetization in transition metal alloys and subsequently applied to various other problems such as chemisorption on metals[15] and mixed valence in solids.[16,17] In these applications of this model, the correlation energy U, arising from the Coulomb repulsion between the two electrons of opposite spin in the same discrete level, plays a crucial role. However, in studies of ion netrualization (or the reverse process of atom ionization), this important quantity is either implicitly assumed to be infinite[6] or completely ignored.[7-9] Such approaches are thus incapable of accounting for negative ion formation: $U = \infty$ completely supresses the transfer of a second electron, while if only single-electron transfer is considered, U is irrelevant.

Previous theoretical work[13] on negative-ion formation has centered on the time-dependent width and shift of the valence level of the impact ion, where the time dependence is due to the motion of the ion. It was assumed that first neutralization occurs via a nonresonant Auger process, while the

nuclear motion of the ion can subsequently bring the valence level into resonance with a band state and thus effect the transfer of the second electron. The quantity U again does not play an important role in this theory.

In the present work, we use the Anderson-Newns model to examine the effects of U on the two-electron transfer process in relation to the dynamics of the nuclear motion of the impact ion. This represents the first time that charge-exchange processes in surface scattering have been considered from a unified point of view. We shall see that the negative-ion formation depends crucially on the finite value of U, and in fact, recent experimental results[5] can be explained in terms of our theory.

The time-dependent Anderson-Newns Hamiltonian including the correlation energy term is given as

$$H(t) = H_0 + V(t),$$ (1)

where

$$H_0 = \sum_\sigma \varepsilon_d c_{d\sigma}^+ c_{d\sigma} + \sum_{k\sigma} \varepsilon_k c_{k\sigma}^+ c_{k\sigma} + U n_{d\sigma} n_{d,-\sigma}$$ (2)

$$V(t) = \sum_{k\sigma} [V_{kd}(t) c_{d\sigma}^+ c_{k\sigma} + V_{kd}^*(t) c_{k\sigma}^+ c_{d\sigma}].$$ (3)

The indicies d and k denote the valence state of the impact ion and the conduction band states of the solid, respectively, σ is the spin index and $n_{d\sigma} = c_{d\sigma}^+ c_{d\sigma}$. The interaction Hamiltonian, $V(t)$, responsible for the electron transfer between the band and valence states, is the only explicitly time-dependent part. The motion of the projectile ion can be phenomenologically

taken into account by using the specific form[6]

$$V_{kd}(t) = V_{kd}e^{-\lambda|t|},$$ (4)

where λ, the sole dynamical parameter in our model, is directly proportional to the normal impact velocity. Hence λ controls the duration of the bound-continuum interaction, while V_{kd} (time-independent) determines its strength.

To lowest order, the perturbative solution for the time-evolution operator $\tilde{T}$ in the interaction picture, which contributes to the S-matrix for negative-ion formation, is given as

$$\tilde{T}^{(2)}(t,-\infty) = (-i)^2 \int_{-\infty}^{t} dt' \tilde{V}(t') \int_{-\infty}^{t} dt'' \tilde{V}(t''),$$ (5)

where

$$\tilde{V}(t) = e^{iH_0 t} \tilde{V}(t) e^{-iH_0 t}$$ (6)

is $V(t)$ expressed in the interaction picture. At $t = -\infty$, the ion is taken to be infinitely far from the surface, and $t = 0$ is the instant of impact. It is apparent from Eq. (3) that, since negative-ion formation involves ultimately the transfer of two electrons, only terms of even powers in $\tilde{V}$ in the expansion of $\tilde{T}$ contribute to the S-matrix.

Figure 1 illustrates schematically the electron-transfer process to lowest, i.e., second, order as contained in Eq. (5). Physically, every factor of $\tilde{V}(t)$ corresponds to the transfer of one electron from the band level ϵ_k to the ϵ_d level of the ion, or the conjugate process of electron transfer from ϵ_d to ϵ_k. Therefore, the contribution to the S-matrix from

4

higher-order terms of $\tilde{T}$ can be diagrammatically generated in a straight-forward manner. Such terms (neglected in this work), however, may involve those final states in which the solid becomes electronically excited, with levels higher than the Fermi level occupied. In general, low-ordered processes are favored by high impact velocities (large λ).

To lowest order, the time-dependent probability for negative-ion formation is then given as

$$P(t) = \int_{\epsilon_L}^{\epsilon_F} d\epsilon \int_{\epsilon_L}^{\epsilon_F} d\epsilon' \rho(\epsilon)\rho(\epsilon') |<k_\uparrow k'_\downarrow|\tilde{T}^{(2)}(t,-\infty)|I>|^2, \tag{7}$$

where ϵ_F and ϵ_L are the Fermi energy and conduction band edge of the solid, respectively, $\rho(\epsilon)$ is the density of states of the band, and the initial and final states, $|I>$ and $|k_\uparrow k'_\downarrow>$ respectively, are described in Fig. 1. The matrix element in Eq. (7) can be evaluated by using Eqs. (2)-(6) to give the result

$$<k_\uparrow k'_\downarrow|\tilde{T}^{(2)}(t,-\infty)|I> = -\frac{v^2}{2}\frac{1}{\lambda+i(U-\epsilon-\epsilon')/2}e^{[2\lambda+i(U-\epsilon-\epsilon')]t}\left(\frac{1}{\lambda-i\epsilon}+\frac{1}{\lambda-i\epsilon'}\right),$$

$$t \leq 0 \tag{8a}$$

$$= -\frac{v^2}{2}\frac{1}{\lambda+i(U-\epsilon-\epsilon')/2}\left(\frac{1}{\lambda-i\epsilon}+\frac{1}{\lambda-i\epsilon'}\right)-\frac{v^2}{2}\frac{1}{\lambda-i(U-\epsilon-\epsilon')/2}$$

$$\times\left(\frac{1}{\lambda+i\epsilon}+\frac{1}{\lambda+i\epsilon'}\right)\left\{e^{-[2\lambda+i(U-\epsilon-\epsilon')]t}-1\right\}$$

$$+2v^2\lambda\left\{\frac{1}{(\lambda^2+\epsilon^2)[\lambda-i(U-\epsilon')]}\left\{e^{-[\lambda-i(U-\epsilon')]t}-1\right\}\right.$$

$$\left.+\frac{1}{(\lambda^2+\epsilon'^2)[\lambda-i(U-\epsilon)]}\left\{e^{-[\lambda-i(U-\epsilon)t}-1\right\}\right\} \qquad t > 0 \tag{8b}$$

where $\epsilon = \epsilon_k - \epsilon_d$ and $\epsilon' = \epsilon_{k'} - \epsilon_d$ with ϵ_d set as the zero of energy, and $V = V_{kd}$ is assumed to be independent of energy. As a first approximation, we have also assumed that U and ϵ_d are constant within the collision region. This latter assumption is not expected to affect the results qualitatively.

We now turn to calculations of the probability P(t) of negative-ion formation and its limiting value at $t \to \infty$, P(∞), for various choices of the parameters U and λ. P(∞) represents the experimental observation of outgoing negative ions after the scattering event is complete, and P(t) reflects the behavior of the transient states. The following fixed numerical values were adopted: ϵ_L = -10 eV, ϵ_F = -0.06 eV and $\lambda = \pi\rho V^2$ = 0.43 eV, where we have assumed the band to have no important structure so that the density of states ρ can be taken as constant in the integration over the band in Eq. (7). Although the last two numbers have been used previously in a study of charge transfer in the Na/W(100) system,[8] they have no special significance in our present work, which is to investigate in general the effects of the variation of λ and U, especially the latter.

Intuitively, we expect the probability P(∞) to be small when the repulsive correlation energy U is large. However, P(∞) also depends on λ, i.e., on the velocity of the impact ion. As shown in Fig. 2, there is a peak for P(∞) at a small value of λ for each U. The explanation for this is that since small λ implies a long duration of interaction whereby from the uncertainty principle the resonance requirement is stringent, it is impossible for the second electron in the solid to overcome the barrier U. On the other hand, large λ can ease the resonance requirement -- energy conservation can be violated for short-duration processes -- but it also limits the actual time available for electron transfer, resulting in a small P(∞). As a consequence, for each U there exists an optimal value of

λ, λ_m, for which the probability attains a maximum. Moreover, λ_m increases as U is increased, due to the fact that a shorter inter-action time is favorable for the second electron to transfer non-resonantly as U becomes larger.

The close relationship between resonant electron transfer and the ion velocity becomes obvious when we look at the time evolution of the probability of negative-ion formation. Figure 3 displays P(t) for various values of U with fixed λ. Each probability curve has a peak at very short time t_m ($\sim$ 0.2 femtoseconds). We see that t_m is smaller for larger U, in accordance with the arguments given above. In addition, the degree of transient negative-ion formation, measured by the ratio $P(t_m)/P(\infty)$, is more pronounced for larger U. Figure 4 provides yet another manifestation of the striking transient behavior; namely, although there is an optimal λ_m at each U for the limiting value of the probability, $P(\infty)$, this is not necessarily the case in the transient region.

To test our theory, we shall compare it with measurements on the conversion of $H^+(D^+)$ to $H^-(D^-)$ by scattering from a cesiated W(100) surface at different grazing angles θ.[5] For this purpose, we identify λ as $v\cos\theta$, where v is the magnitude of the velocity, and phenomeno-logically introduce a velocity-dependent interaction, $\Delta = 0.43 \exp(-0.01v)$ (in the unit of eV), to account for the loss of particles due to penetra-tion into the surface. The variation of θ is thus equivalent to the variation of the normal impact velocity λ. Our results, given in Fig. 5, are in qualitative agreement with the experimental ones,[5] where for all incident energ $\sim$ the $\sim$onversion probability goes

through a maximum. Quantitative comparisons have not been attempted since
precise information on critical parameters, especially Δ, is still lacking.
For the cesiated W(100) surface, among other complications leading to
unreliable data for parameters is the theoretical evidence of a lowering
of the work function by multiple dipole formation.[18,19]

In this work we have demonstrated, through varying the dynamical
conditions of the impact ion, the significance of the correlation energy U
in negative-ion formation from positive-ion-surface scattering. Though U
in general decreases the probability for negative-ion formation, one can
always exploit the experimentally controllable dynamical conditions (varying
v and θ) to achieve an optimal result for a given system. Moreover, there
may even be the possibility of exploiting the characteristic transient behavior
of P(t), since for finite U, $P(t_m)$ is always larger than (∞) except for very
large λ. For very small λ, on the other hand, our perturbation approach may
not yield correct results, since the long interaction times then allowed may
require higher-order processes than the second-order one be considered. Our
results have been shown to be in good qualitative agreement with experiments.
A more elaborate calculation is needed which takes into account the lowering of the
valence level of the ion near the surface[18,19] is needed for quantitative
comparision with experiments.

Acknowledgments

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Figure Captions

Fig. 1. Schematic diagram of electron transfer in negative-ion formation.
ε_U is the upper and ε_L the lower edge of the band. (a) Initial
state $|I>$: valence state ε_d empty, band filled up to the Fermi
level ε_F. (b) Intermediate states $|k_\uparrow>$ and $|k'_\downarrow>$: states cor-
responding to the neutralized atom; one electron transferred
from the ε_k or $\varepsilon_{k'}$ level to the ε_d level. The arrows denote
the spin states of the electrons, and the solid and hollow
circles represent electrons and holes, respectively. (c) Final
states $|k_\uparrow k'_\downarrow>$: negative-ion states; two electrons transferred
to the ε_d level.

Fig. 2. $P(\infty)$ vs λ for various values of U. Energy is in the unit of eV.

Fig. 3. $P(t)$ vs t for various values of U with fixed λ. As U increases,
the characteristic short-time behavior becomes more pronounced.
Energy is in the unit of eV, and time is in the unit of 6.59 x
10^{-16} sec $(\frac{\hbar}{eV})$

Fig. 4. $P(t)$ vs t for various values of λ with fixed U. The units are the
same as in Fig. 3.

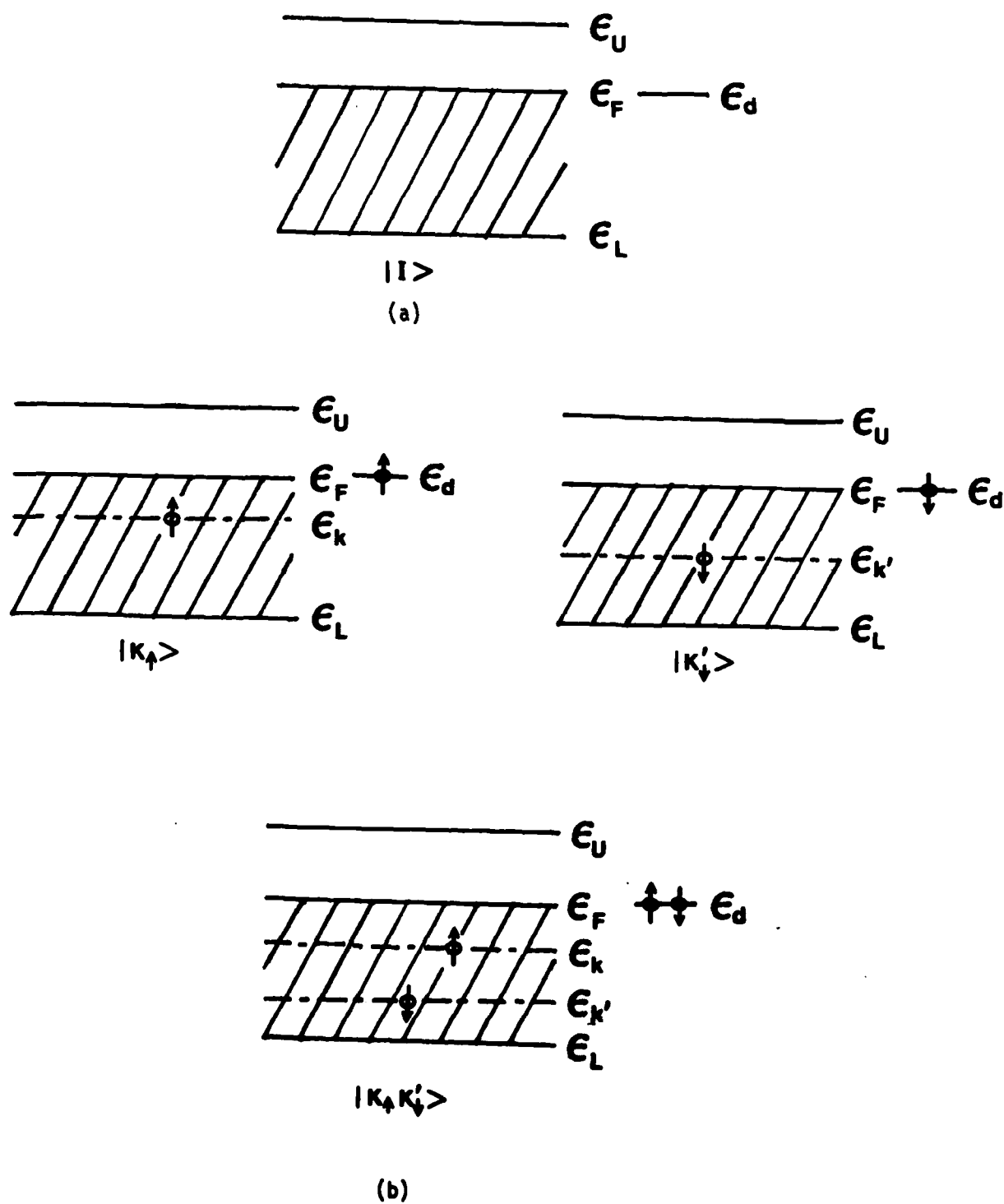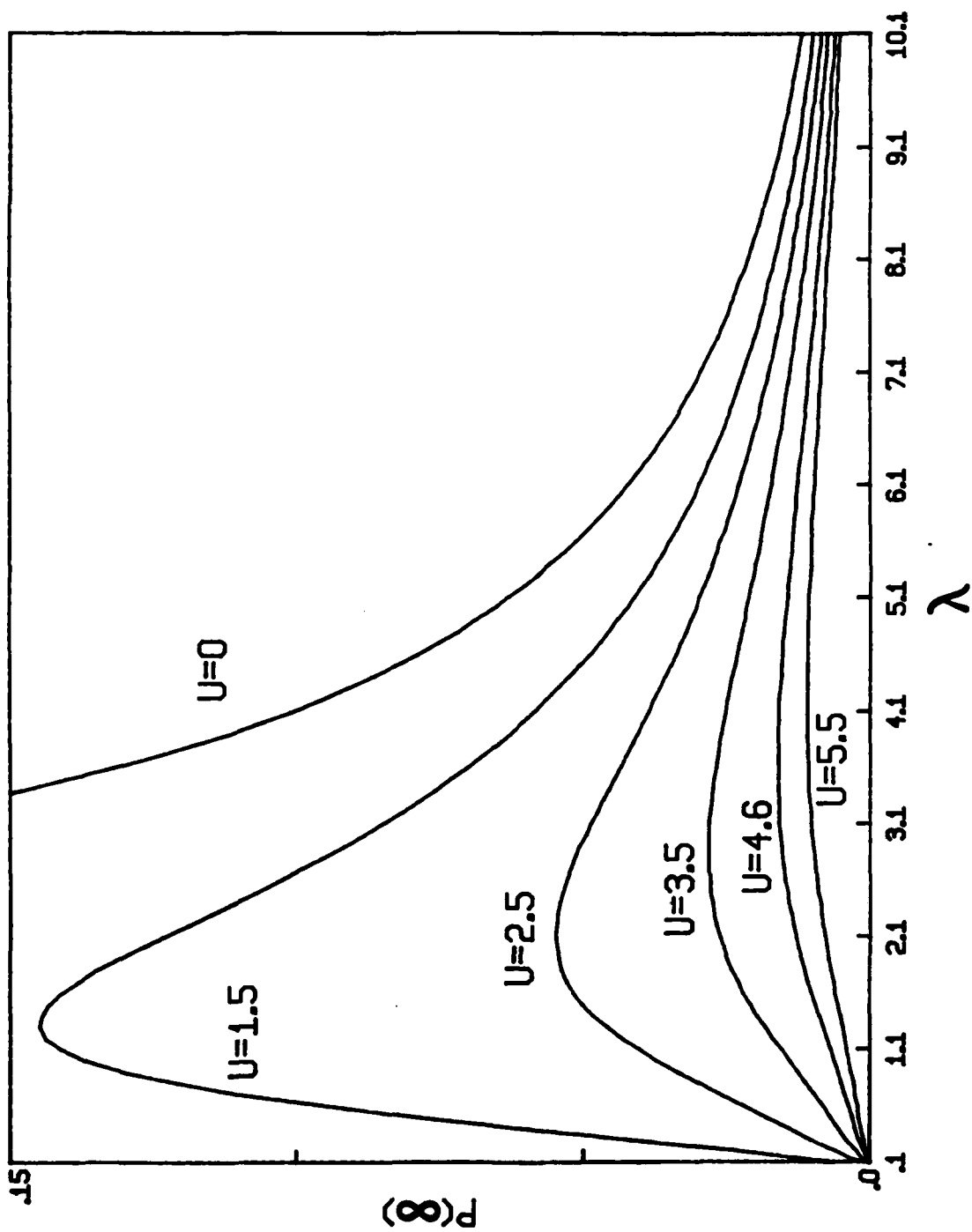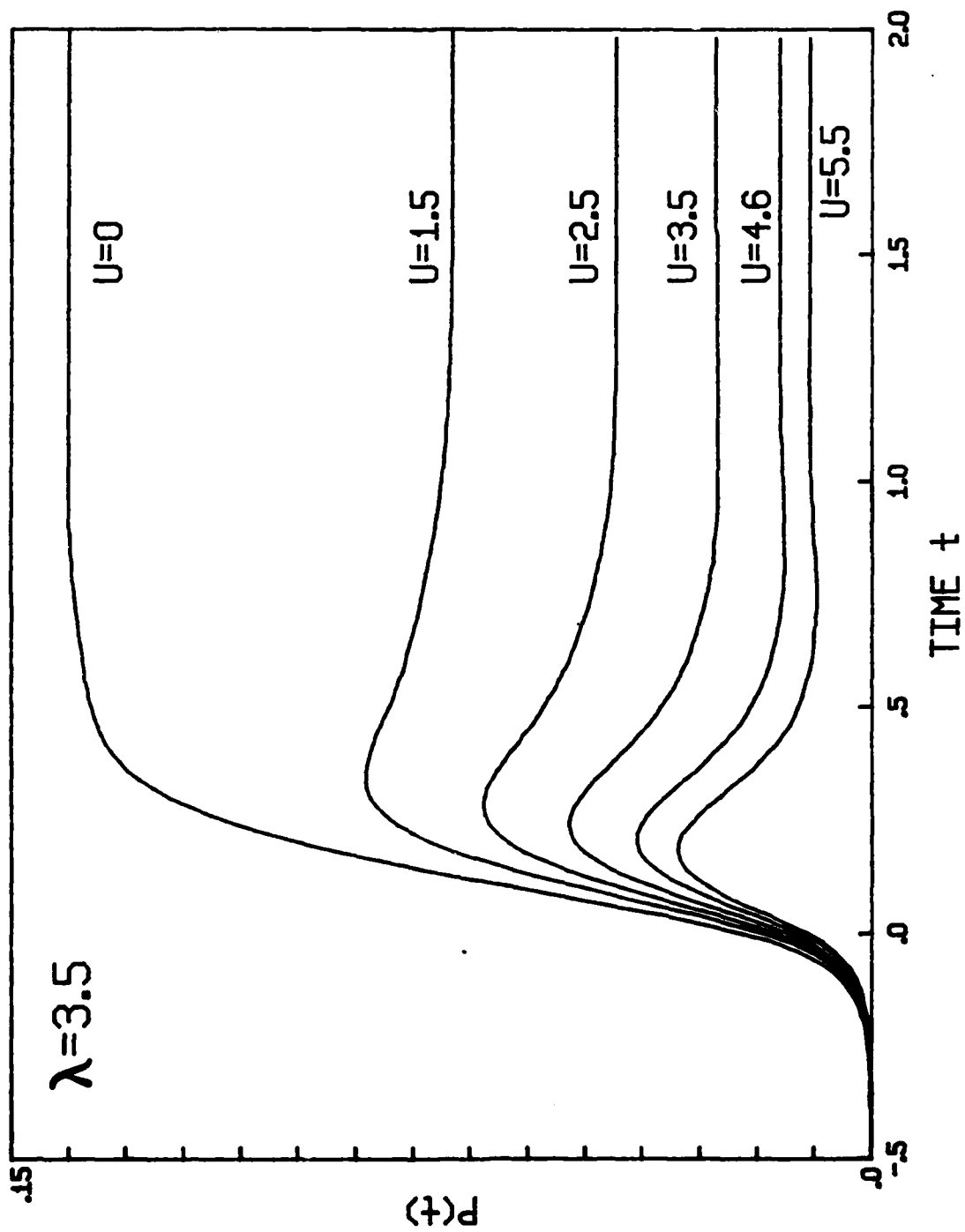Fig. 5. $P(\infty)$ vs θ, the incident angle of impact. $v_1 < v_2 < v_3 < v_4$.

FIG. 1

Fig. 2

$\lambda = 3.5$

U=0

U=1.5

U=2.5

U=3.5

U=4.6

U=5.5

P(t)

TIME t

FIG. 3

Fig. 4

U=5.5

P(∞)

ANGLE OF INCIDENCE
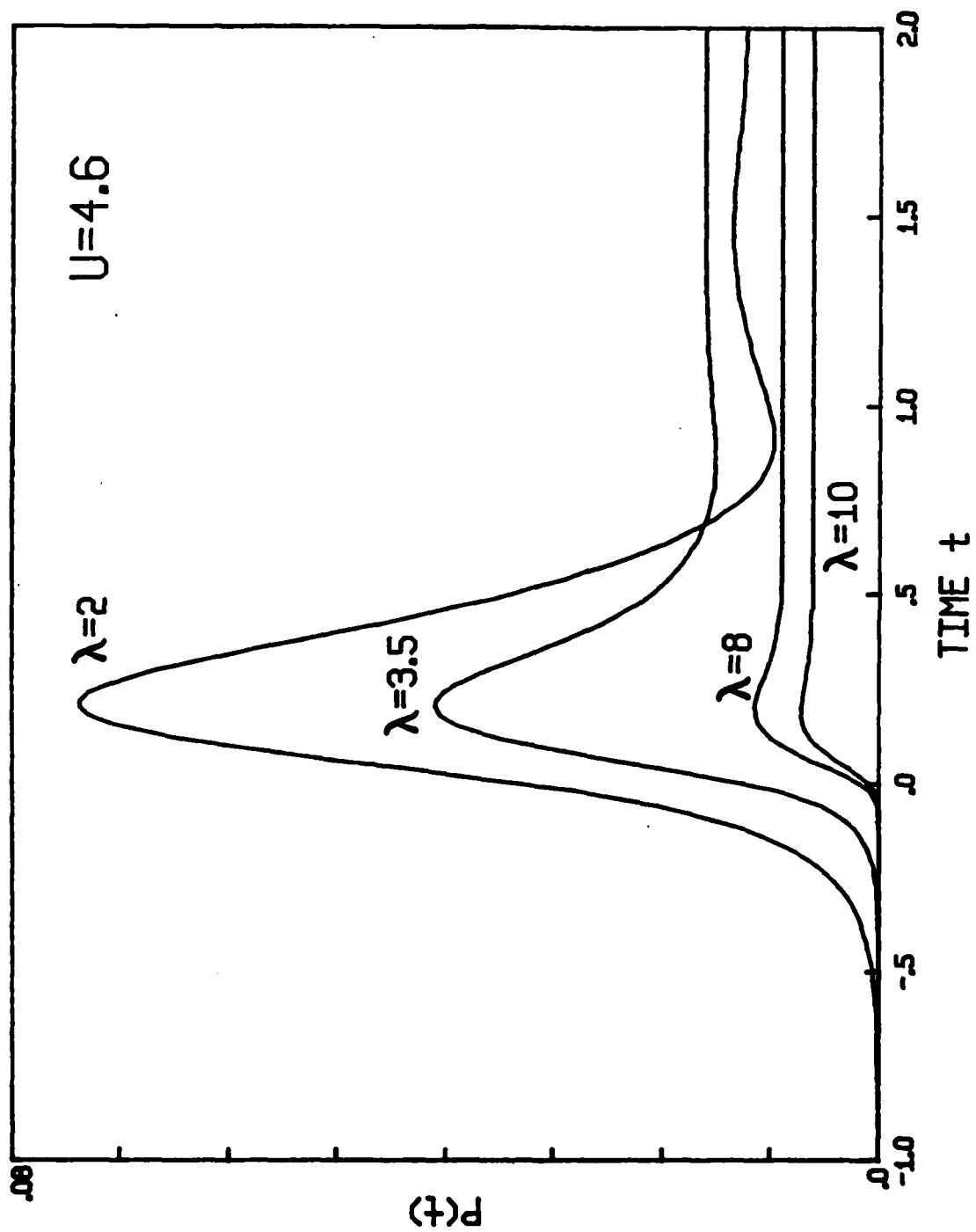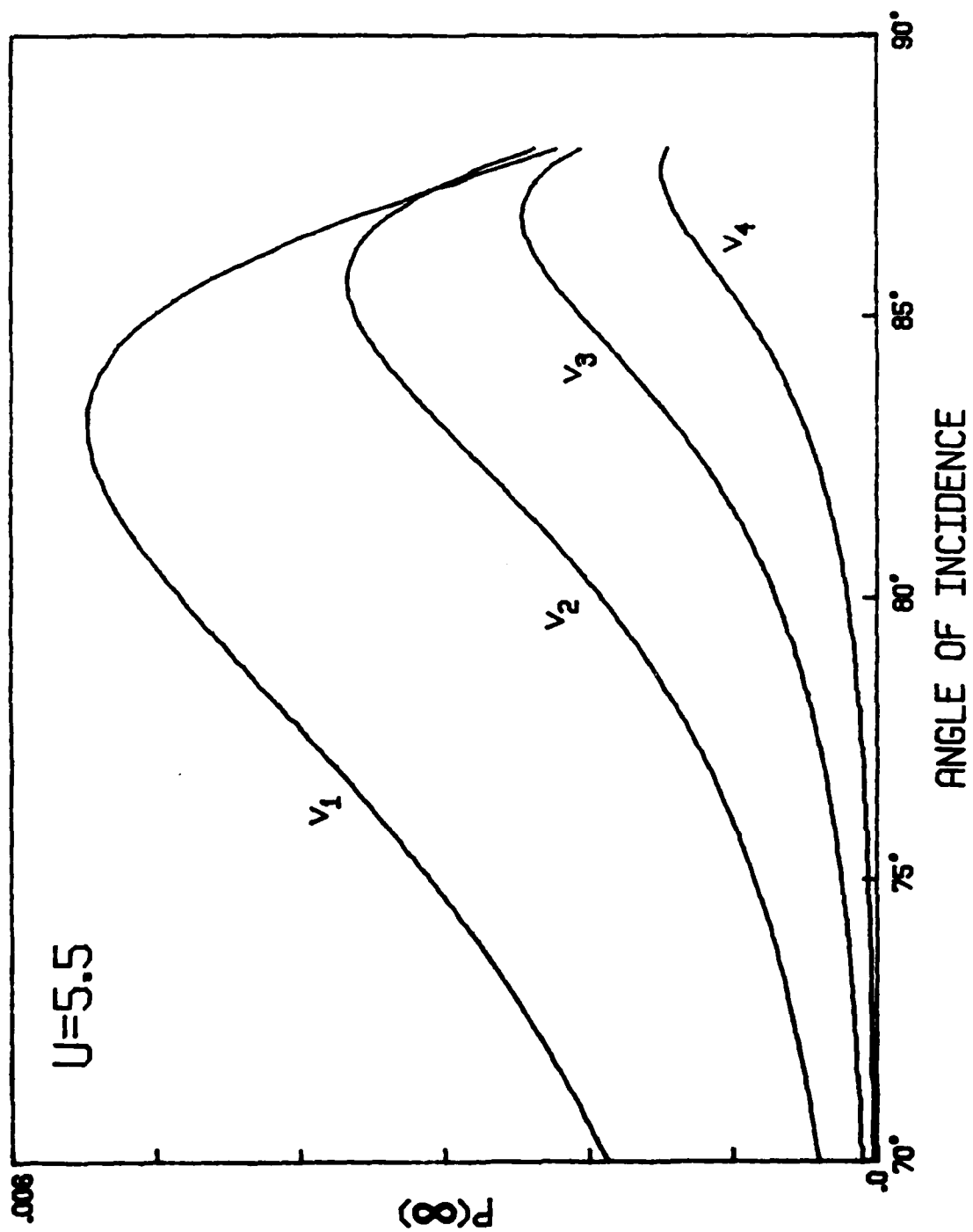
FIG. 5

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