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Oscillatory Ion Yields of He^+ Scattered from Atomic and Solid Pb Targets

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Ion yields of He^+ scattered from atomic and solid Pb in the energy range from 200 to 2000 eV into a large scattering angle are reported. The beam results show an oscillatory structure of the ion yield as function of the energy which is comparable to the same effects observed for a solid target. Thus the atomic nature of the quasiresonant charge exchange is confirmed. The data also give evidence for solid-state effects influencing the oscillations of the ion yield.

Oscillations in the yields of ions scattered from solid targets have been reported first by Erickson and Smith.[1] It has been shown[2] that the effect is most pronounced for nine elements of the third, fourth, and fifth groups of the periodic table, i.e., from Ga to Bi as targets, and He^+ as the scattered particle. The experimental observation is that as a function of primary energy (or velocity) the yield of backscattered ions into a given scattering angle oscillates with intensity ratios up to a factor of 5. Measurements as a function of scattering and impact angle to the surface[3] suggest that the charge-exchange process occurs in a binary collision, i.e., it can be understood as an atom-atom collision. The influence of the solid state seems to be marginal as far as the oscillations are concerned. The overall distribution of backscattered ions is, however, governed by the (elastic) differential scattering cross section and by the possible neutralization processes.[4] The empirical interpretation[1,2] of the oscillations as being due to quasiresonant charge-exchange processes between the He 1*s* level and the *d* levels of the above-mentioned elements has been supported by the first theoretical approaches.[5,6]

In this Letter we report results from the scat-

tering of He^+ ions from solid Pb targets, Pb on Ni in submonolayer coverage, and—for the first time—from an atomic Pb beam. The experimental setup has been described earlier.[7] It is an UHV system containing a low-energy ion source (50 eV to 2 keV) and a 90° spherical energy analyzer (resolution $\Delta E/E = 2\%$) positioned at a laboratory scattering angle of 90°. The setup allows the quick exchange of a solid target mounted on a manipulator with an oven which produces an atomic beam of Pb. A second smaller oven is used to prepare thin layers of Pb on different targets *in situ* under control with the ion beam. Further, absolute coverage determinations are obtained from Rutherford backscattering measurements in a 2-MeV accelerator system.[8] Details of the experimental procedures will be published elsewhere.[9]

Figure 1 shows the ion yield of He^+ as a function of the ion energy for four targets: a sputter-cleaned solid Pb target, thin layers of Pb on a clean Ni surface ($\frac{1}{10}$ and $\frac{1}{3}$ of a monolayer), and an atomic beam of Pb. The change in overall yield reflects first of all the different target densities. For the solid targets the yield is to a good approximation proportional to the surface density of Pb atoms and, within the experimental errors, to the density of the beam; i.e., we do not have to invoke large changes of the neutralization effect as a function of coverage in the energy range around 1 keV. We can then write the ion yield $Y \propto NP(v, \vartheta)d\sigma(v, \vartheta)$, where N is the atomic density and $d\sigma$ the differential scattering cross section, which is a function of the ion velocity and the scattering angle. P is the probability that an ion escapes neutralization, which for a given atom-ion combination is assumed to be a function of velocity and scattering angle only. Hence changes in the yield curves (Fig. 1) for the different targets reflect changes of the electronic state of the Pb atoms. These changes are seen as more pronounced, "high-frequency" oscillations above 1 keV for the thin layer and as a peak splitting and slight shift of position for both the thin layer and the atomic beam compared to the bulk target. Furthermore, for the beam target the "amplitude" of the oscillations is smaller compared to those from solid targets. For the solid targets this value is nearly independent of the Pb surface density. These results give clear evidence for the atomic character of the charge-exchange interaction, but they present also the first experimental evidence for solid-state surface effects in this interaction process.

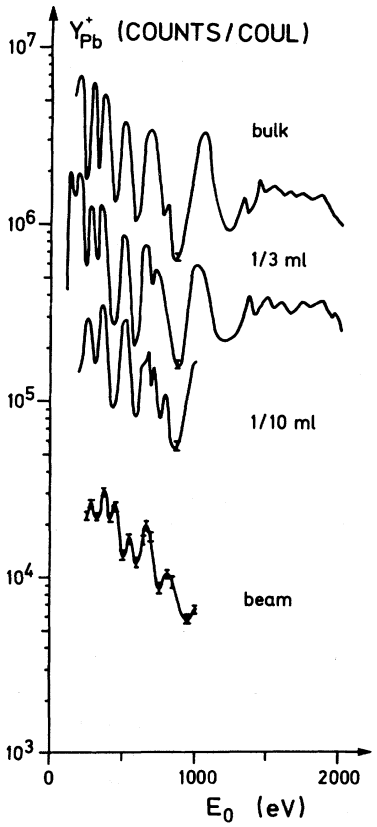

FIG. 1. Ion yield as function of energy for solid Pb, $\frac{1}{3}$ and $\frac{1}{10}$ of a monolayer of Pb on Ni, and atomic Pb for the scattering of He^+ into a laboratory scattering angle of 90°, angle of incidence 45° (solid targets). The yields are normalized to incident charge and energy window of the analyzer.

From the theory[5,6] and from earlier work on ion-atom scattering[10,11] the ion yield is expected to follow approximately $Y^+ \propto \exp(-v_0/v)\sin^2\alpha$ where $\alpha \approx \langle Ea\rangle/\hbar v - \beta$. Here v_0 is a characteristic velocity, v the ion velocity, β a phase factor, and

$$\langle Ea\rangle = \int_{a_m}^{a_c}(E_2 - E_1)dR,$$

where $E_2 - E_1$ is the distance between the two electronic energy levels involved during the collision along the internuclear separation R between the beginning of the interaction a_m and the distance of closest approach a_c (for details see Ref. 10). Essentially the "frequency" and the "amplitude" of the oscillation depend on the evolution of the molecular orbital formed during the collision. If this is not known, even qualitative discussion of the results is difficult. In Fig. 2 we have assembled the energy levels in question for He, solid Pb, and atomic Pb. The difference

of the latter two arises from the difference in work function[12] of 4.25 eV and ionization potential[13] of 7.41 eV, and the fact that the $6s6p$ electrons form bands in the solid.[14,15] Quasiresonant charge exchange can be expected between the Pb $5d$ and the He $1s$ levels and from Pb $6p6s$ to He $2p2s$. The band structure of the $6p6s$ electrons in the case of the solid makes oscillatory resonance effects unlikely, even though exchange to the He $2p2s$ levels is likely and essentially resonant because of the match of the energies. Perhaps the small additional oscillations which show up with the thin Pb layers on Ni are due to this exchange, since in that case (submonolayer coverage, Fig. 1) the Pb band will not form, and not necessarily all four $6s6p$ Pb electrons will be involved in the (probably also energetically broadened) surface bond. Results with the atomic beam in this energy range (>1 keV) could not be obtained with sufficient accuracy at present. We expect to see this fine structure in that case as well.

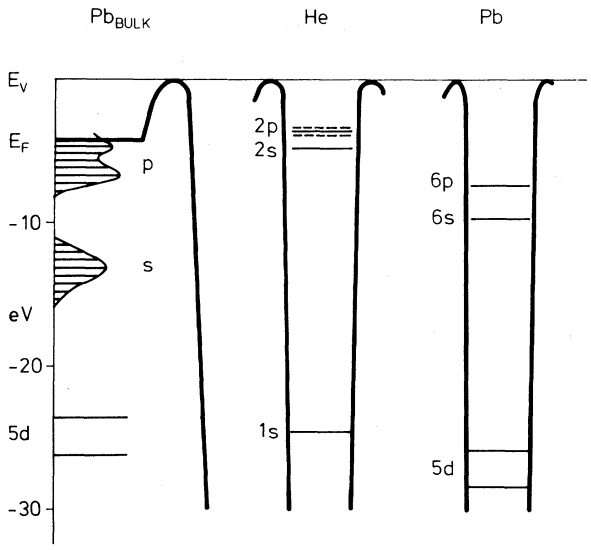

FIG. 2. Energy levels of the outer-shell electrons of He and atomic and solid Pb. Density of states of solid Pb are from Refs. 14 and 15.

Within the current models[5,6,10] the amplitudes are characterized by a velocity term v_0 and the frequency of the oscillations by $\langle Ea\rangle/\hbar$ which has the dimension of a velocity as well and a is the quantity which contains the information about the dynamics of the electron states during the collision. The exponential damping term arises from a Landau-Zener charge-exchange model,[10] or it is described by the effect of the conduction electrons in the charge-exchange process,[6] i.e., the interaction of the conduction electrons with the excited states of the ion. Experimentally we observe no change of the amplitudes with target thickness and type of substrate, but

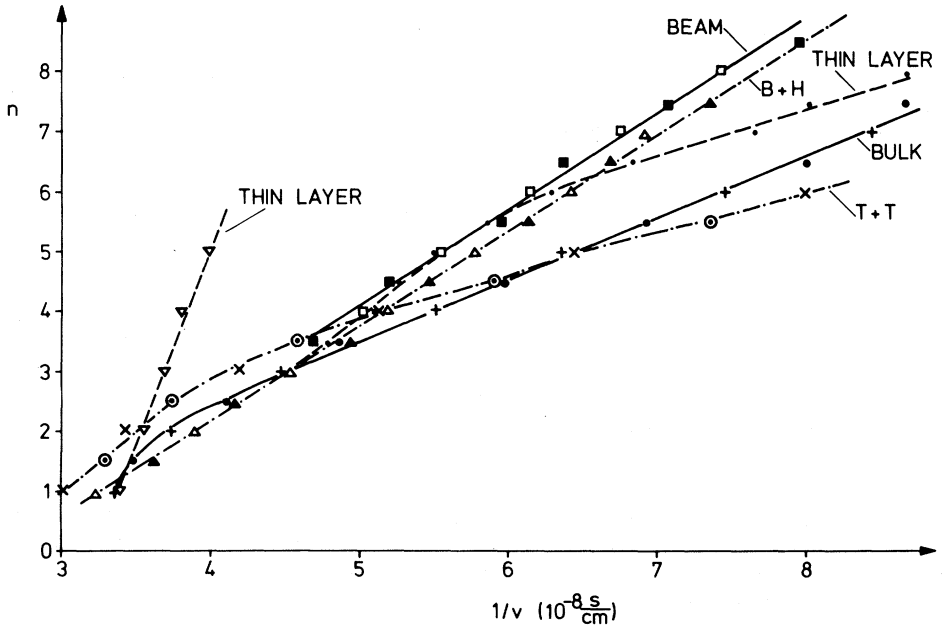

FIG. 3. Number of phase of the oscillations (Fig. 1) as a function of the inverse velocity of the scattered He^+ in comparison with theoretical results (T+T, Ref. 5; B+H, Ref. 6). The triangles are for the small oscillations from the thin layer above 1 keV (Fig. 1) (maxima only).

lower amplitudes with the Pb beam. These findings seem to contradict the assumption that the conduction electrons play a major role in the damping.

The frequency term can be evaluated from a plot of the phase number of the oscillations versus the reciprocal ion velocity (Fig. 3). Approximately linear dependencies are found, the slope, which is proportional to $\langle Ea \rangle$, being steeper for the beam data, and for the additional oscillations observed in case of thin Pb films on Ni. The theoretical results seem to agree with the bulk results[5] or with the beam results.[6] The thin-layer results are intermediate between the theories. This difference of the theoretical values may be accidental in view of the simplifications and the arbitrary choice of some parameters entering into the calculations. Our results may help to define these parameters. Deviations from linearity (Fig. 3) are surprisingly small since this is only expected for $E \sin\vartheta = \text{const}$ (E the energy, ϑ the scattering angle). Furthermore, it is an approximation for small ϑ only.[10,11] The data suggest a larger difference of the energy levels in case of the $5d$-$1s$ exchange for the beam than for the bulk. An even larger separation for the assumed $6s6p$-$2s2p$ exchange is indicated. Comparable data were obtained for Pb layers on Al, Si, and Cu substrates which will allow a more detailed discussion of the oscillatory fine structure.[9] Our main concern here is the direct comparison of the beam and the solid-state targets.

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Electron-Photon Coincidence Measurements in Electron Scattering from Atomic Hydrogen

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Angular correlations have been measured between electrons exciting the $2p$ state of hydrogen and the Lyman-α decay photons at incident energies of 70 and 100 eV and electron scattering angles of 5°, 10°, and 15°. The information obtained on the scattering amplitudes to the magnetic substates is compared with recent calculations, none of which are in good agreement with all of the data.

Electron-impact excitation of atoms in the low- to intermediate-energy range has been the subject of much theoretical work. Besides being of interest as a fundamental quantum mechanical scattering problem, electron-impact excitation has important applications in other fields of

