thews, C. E. Max, K. G. Tirsell, and other colleagues in the Livermore Laser Fusion program.

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## Surface Diffusion by an Atomic Exchange Mechanism

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On Ir(110), tungsten adatoms move preferentially across  $[1\overline{10}]$  channels rather than along them, as observed on other channeled planes. The jump mechanism underlying this anomalous behavior has been tested in an atom probe. It is found that after crosschannel motion, an iridium adatom is left on the iridium surface rather than the tungsten originally deposited. These observations provide the first experimental evidence for atomic diffusion on a metal by exchange of the adatom with an atom from the substrate.

In the past, it has generally been assumed that atomic diffusion on metals occurs by hopping over the surface. Observations of single metal atoms on channeled planes such as (211) of tungsten and (110) of rhodium<sup>1</sup> fit in well with this view. Migration on these surfaces is one dimensional (1D); as expected from the atomic arrangement of such planes, atoms move only in the direction of the surface channels. Recently, however, quite a different behavior has been found on some face-centered-cubic metals. On Pt(110), modeled in Fig. 1, Bassett and Webber<sup>2</sup> observed 2D diffusion of platinum and iridium atoms, with frequent movements across as well as along the  $[1\overline{10}]$  rows. On iridium (110), diffusion of iridium and tungsten atoms occurs preferentially across the channels.<sup>3</sup> Cross-channel motion of atoms may be visualized<sup>2, 4</sup> in two different ways: (1) via atomic jumps *over* the rows of lattice atoms forming the ridges, which *a priori* appear more unlikely than jumps along the channels; or (2) by an exchange process. In the latter, the adatom takes the place of a lattice atom in the side of the channel; it is the displaced lattice atom that continues the diffusion. We report here the first experimental evidence that cross-channel diffusion does, in fact, occur by an exchange mechanism.

The idea underlying our experiments is simple. Consider what happens when a foreign adatom,



FIG. 1. Hard-sphere model of the (110) plane of an fcc lattice. Outermost lattice layer in grey, second layer in white, adatom in dark grey. (a) Ideal  $1 \times 1$  surface. (b) A reconstructed  $1 \times 2$  surface, obtained by removal of alternate [110] rows.

such as tungsten, is deposited on a (110) plane of iridium. In cross-channel motion by a hopping process, the tungsten atom just jumps from one channel to the next. However, if exchange is the dominant mechanism, then after diffusion an iridium atom rather than the original tungsten should appear in the adjacent channel. To distinguish between the two mechanisms it is sufficient to determine the chemical identity of the adatom after cross-channel motion. This is now possible in the atom probe.<sup>5</sup> We have carried out such measurements for the diffusion of tungsten atoms on the (110) plane of iridium.

In our instrument,<sup>6</sup> the iridium surface formed by field evaporation below 50 °K can be photographed with atomic resolution, as in a simple field-ion microscope. From the overall dimensions of the (110) plane, and the number of  $[1\overline{10}]$ rows on it, we conclude that the distance from one row to the next is roughly twice as large as in the bulk. An atomic arrangement conforming to such a channel spacing is indicated in the model in Fig. 1(b). It is worth noting that the structure of Ir(110) produced by field evaporation appears to agree with that proposed for macroscopic (110) planes of iridium on the basis of low-energy electron-diffraction (LEED) measurements.<sup>7</sup>

To use the atom probe for determining the chemical identity of atoms diffusing on Ir(110), it is first necessary to calibrate the mass scale. In our experiments, all under ultrahigh-vacuum conditions, this was done by determinations on the iridium substrate (Fig. 2), and on adatoms. For the latter, iridium and subsequently tungsten atoms were evaporated in separate experiments onto an iridium sample at T < 50 °K. Without ever warming the surface, the atoms were pulse desorbed, and their time of flight was determined. It should be noted that in each experiment only a



FIG. 2. Calibration of atom probe on iridium substrate at T < 50 °K. Background pressure  $< 2 \times 10^{-10}$  mm. Peak at left arises from Ir<sup>+3</sup>, that at right from Ir<sup>+2</sup>.

single atom was placed on the (110) plane; its presence on (110) after deposition was verified by field-ion microscopy at the detector. That the atom had indeed been removed by the field pulse was also checked after each charge-to-mass determination. The results of these measurements are shown in Fig. 3; there data on all charge states observed for a given chemical entity are combined under the same AMU bin. The separation between the iridium and tungsten distributions observed with adatoms is as expected from the calibration on the iridium substrate, and it is clear that iridium adatoms can be readily distinguished from tungsten adatoms in our atom probe.

The results of the crucial experiments are juxtaposed with the calibrations in Fig. 3. In these experiments, a single tungsten atom is deposited on the (110) plane. The surface is then



FIG. 3. Atom-probe measurements on single adatoms pulse desorbed from iridium. Atomic-mass data (AMU) obtained by combing information on all charge states detected on pulsing. Top: Calibration on iridium atoms. Bottom: Calibration on tungsten atoms. In both calibrations, atoms are deposited on surface at 50 °K, and pulsed off without ever warming the substrate. Center: Adatoms detected after depositing tungsten atom on Ir(110), and warming to allow a cross-channel jump.

warmed to  $\approx 270$  °K in order to initiate a crosschannel jump. After cooling to below 50 °K, the atom appearing in the adjacent channel is pulse desorbed and its charge-to-mass ratio is measured. It is apparent from the data in Fig. 3 that after a cross-channel event, an iridium atom rather than the tungsten atom initially deposited on the surface is present in the adjacent channel.

Our measurements thus lead us to conclude that for tungsten atoms on Ir(110), cross-channel motion occurs by exchange of the tungsten adatom with an atom from the substrate, presumably from the protruding rows forming the channels. If this is indeed what happens, then we should also expect to find a tungsten atom incorporated in the substrate on analyzing the first lattice layer. These measurements have been done. After cross-channel motion of a tungsten adatom and subsequent field evaporation of the adatom remaining on the surface we do indeed find tungsten incorporated in the lattice. In a comparable number of blank runs on the substrate, tungsten was never detected.

Several other observations, which will be reported fully elsewhere, also support the conclusion that cross-channel motion of tungsten atoms on the (110) plane of iridium occurs by exchange with lattice atoms. We presume that a similar mechanism accounts for channel crossings in self-diffusion on Ir(110), and also on Pt(110). It is of interest that for both iridium<sup>8</sup> and platinum,<sup>9, 10</sup> LEED measurements on (110) yield a  $1 \times 2$  pattern suggesting a channel spacing twice the normal value. In contrast, for rhodium (110), no rearrangement is indicated by LEED data,<sup>11, 12</sup> and neither are there any indications of crosschannel diffusion.<sup>13</sup> This suggests that the forces responsible for the restructuring of the (110) plane may also favor cross-channel motion by an exchange mechanism.

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## Ripplons, <sup>3</sup>He, and Heat Conduction on the Surface of Superfluid <sup>4</sup>He

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A gradient in the concentration of adsorbed  ${}^{3}$ He, produced by a current of ripplons, has been observed in heat-conduction measurements on the surface of superfluid  ${}^{4}$ He. The  ${}^{3}$ He spreading pressure and  ${}^{3}$ He-ripplon collision rate have been determined from the data.

According to the phenomenological theory of Andreev and Kompaneets<sup>1</sup> the free surface of superfluid <sup>4</sup>He is governed by a two-dimensional (2D) version of the classic Landau theory of superfluidity. "Ripplons" (quantized capillary waves) are the elementary excitations of the sur-

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FIG. 1. Hard-sphere model of the (110) plane of an fcc lattice. Outermost lattice layer in grey, second layer in white, adatom in dark grey. (a) Ideal  $1 \times 1$  surface. (b) A reconstructed  $1 \times 2$  surface, obtained by removal of alternate [110] rows.