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Abstract: The structure of adatom nuclei on metal crystal planes (1)

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The geometrical structure of adatom nuclei involved in the nucleation of a subsequent crystal plane is of fundamental interest as it reflects the energetics of adatom-adatom and adatom-surface interactions. Investigations of adatom nucleus structure by field ion microscopy have yielded some very interesting results. Bassett(1) and Fink and Ehrlich(2) have observed that on W(110) the heterogeneous nucleation of Ni, Pd, Ir, and Pt adatoms initially proceeds by the formation of stable linear chain nuclei oriented in one of the close-packed directions of the substrate. Bassett found that only beyond a critical chain length of roughly 4 Ni and 8 Pd atoms will the nucleus assume a two-dimensional island configuration. In addition, Fink and Ehrlich found that a two-dimensional island configuration of 12 Ir adatoms on W(110) is metastable and reconfigures into linear chain nuclei with heating.

In the course of our research we have observed chain forming nuclei in other heterogeneous systems. For example, with Pd on Ta(110), a linear chain oriented in the close-packed direction on the surface is the most stable structure for nuclei of 8 or fewer adatoms; only for 9 or more adatoms is a two-dimensional island configuration stable. Similarly, mixed metal clusters of Pt and Pd on W(110) were also found to initially nucleate in the form of stable linear chains.

Further experiments produced the interesting result that the formation of stable linear chain nuclei is not limited to heterogeneous systems(3). In particular, for the self-adsorption of Ir on Ir(100), linear chain nuclei oriented in a close-packed surface direction are the most stable configuration for 5 or fewer adatoms. Two-dimensional islands are only the most stable configuration for 6 or more Ir atoms. These facts are evidenced by the direct observation of transformations of metastable two-dimensional islands of 5 adatoms to stable linear chains, and metastable 6 adatom linear chains to stable two-dimensional islands. In contrast, with Ir on Ir(111), two-dimensional island formation begins immediately at the tri-atomic nucleus for which a triangular shape is the most stable.

To interpret such chain to island transformations in homogeneous systems, we have modeled the Ir on Ir(100) system as a lattice gas(4). The critical chain length of 5 Ir atoms, together with the observed stable island geometries for 6 and 7 adatoms are found to imply relatively tight bounds on the adatom-adatom interaction energetics once reasonable assumptions are made regarding falloff with distance. Specifically, the effective second near-neighbor interaction must be repulsive and of a magnitude lying between approximately 1/4 and 1/3 that of the nearest-neighbor attraction. In addition, the effective third near-neighbor interaction must be attractive and have a magnitude between roughly 1/6 and 2/7 that of the nearest-neighbor attraction. The fact that the third near-neighbor interaction cannot be considered to be negligible if the experimental results are to be explained, provides rather direct evidence that relatively long-range interactions exist between the Ir adatoms on Ir(100). Such considerations of adatom cluster geometries are suggestive of further experiments which may provide substantially more detail regarding the sign and magnitude of adatom-adatom interactions on surfaces.

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