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A Study of Ag/Ag(100) Thin Film Growth with Scanning Tunneling
Microscopy

by

Wen, Jianming

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Ames Laboratory, U.S. DOE

Iowa State University

Ames, Iowa 50011

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A study of Ag/Ag(100) thin film growth with scanning tunneling microscopy

Jianming Wen

Major Professor: Patricia Thiel

Iowa State University

We used STM to study metal thin film growth processes. The following are main conclusions:

1. Large metal cluster diffuse on Ag(100) surface. The metal-metal system large adatom cluster diffusion is reported for the first time. The evaporation-condensation of adatoms dominating the vacancy cluster diffusion process is suggested. The diffusion coefficients is on the order of $10^{-18} \text{ cm}^2\text{s}^{-1}$.

2. Metal film evolution behaviors at different coverage regime are examined. At low coverage, cluster diffusion dominates the coarsening process. At high submonolayer coverage, vacancy cluster repining dominates the process. At moderate coverage, edge-running dominates the process.

3. The vacancy cluster diffusion is also observed. The vacancy were formed by controlling the deposition flux and film coverage. The method is simple and offers similar advantages of the sputtering method. The diffusion coefficients is on the order of $10^{-18} \text{ cm}^2\text{s}^{-1}$, is smaller than that of adatom clusters, and has a scaling factor α roughly close to 0.5 in $D \propto N^\alpha$. The evaporation-condensation of vacancy monomers dominating the vacancy

cluster diffusion process is suggested.

4. We have presented a number of observations of dynamical phenomena of Ag cluster on Ag (100) with UHV-STM. Ag cluster on Ag (100) at room temperature are mobile. The interaction between clusters and between clusters and surface steps are mediated via 2D dilute adatom gas on the terrace and different adatom concentrations around clusters and steps. Background gases and impurities on cluster edges can strongly affect film coarsening process. The former can enhance clusters dissolution, but the latter can stabilize the local structure and geometry of the Ag film.

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CHAPTER 1

GENERAL INTRODUCTION

1. The importance of thin film growth kinetics

Thin films are attracting more and more attention in both the industrial and scientific communities. Many applications of thin films have been developed in industry. By using various growth methods, thin films can be used in optics, microelectronic devices, magnetic recording media, and as protective coatings. In order to improve existing applications and to find new ones, it is essential to understand what makes them so useful in applications and what factors affect their properties. Therefore, an understanding of film growth processes is necessary. Scientifically, many fundamental interactions, such as the interaction between the atoms that comprise the film and substrate, or the interaction between film atoms, are of great interest to surface scientists; studies of these interactions can provide dramatic insights into the nature of thin films and therefore, can further drive technology forward. In every application, the film structures, including morphology and microstructure, and adhesion between film and substrate are critical to the film's properties and therefore its performance. Studies of the mechanisms that control film morphology, microstructure and adhesion thus are important. Film growth kinetics can provide important information regarding the film structure and adhesion. Film growth is an atomistic process. The chemistry and physics of the system can be better understood if the information provided is at an atomic level.

The purpose of this chapter is to provide an overview of thin film growth kinetics. In this chapter, first, a general microscopic description of growth process is given. The focus is mainly on metal-on-metal systems, although the principles are applicable to other systems. Then, a short literature review of metal-on-metal growth with emphasis on diffusion and coarsening is introduced. Finally, a general introduction of scanning tunneling microscopy is given since it is the main technique employed in this study.

2. Overview of growth kinetics

a. What will happen during film growth

Thin films can be grown in many different ways. The growth process relevant to this study can be classified as vapor deposition. It is one of the most widely used deposition techniques. As the name implies, the technique consists of vaporization of the solid material by heating it to sufficiently high temperatures and condensing it onto a cooler substrate to form a film. The whole system needs to be placed in a vacuum chamber since in atmosphere evaporator cannot be functional and substrate and vapor source can be contaminated.

Most metal-on-metal systems are expected to have one of three classical growth modes in terms of thermodynamics, as shown in Figure 1: (1) sequential filling of layers, defined as Frank-van der Merwe mode; (2) after initial layer growth under critical thickness, agglomeration into three-dimensional clusters, defined as Stranski-Krastanov mode; and (3) agglomeration into three-dimensional (3D) clusters, defined as Volmer-Weber mode. The growth mode is predicted on the basis of the surface free energy calculations involving

Frank-van der Merwe

Stranski-Krastanov

Volmer-Weber

Figure 1. The three classical growth modes

□ Adatom

⊗ Substrate

substrate, film, and interface (γ_s , γ_f , and γ_i , respectively). For homoepitaxial systems, the system for which the substrate and deposition materials are the same, layer-growth is predicted for all film thickness. For heteropitaxial growth, where the substrate and deposition materials are not identical, 3D clustering is always the case. However, under certain deposition conditions, the growth may be far from equilibrium. Then the kinetic processes involved in film growth can be critical in terms of controlling film structure.

During film growth, adatoms are continuously deposited onto the substrate at a certain rate. The adatoms that land on the substrate will have a certain lifetime to migrate on the surface and interact with the surface atoms as well as with each other. In some cases, these interactions may cause growth of substrate steps, intermixing of deposited atoms and substrate atoms, and alteration of further film growth. These interactions result in nucleation on the surface, two-dimensional (2D) condensation of adatoms into islands and their subsequent growth, and the continuous growth from 2D to 3D clusters.

Growth kinetics depend on the fundamental surface processes of surface adsorption, desorption, and diffusion. Often the thermodynamically favored film states are 2D islands and 3D clusters on the substrate rather than a random distribution of adatoms. Film growth is a non-equilibrium process where surface phase separation occurs and growth kinetics play an essential role. As illustrated in Figure 2, there are two ways to make phase separation occur. In order to enter the two-phase coexistence regime from one phase state, one can quench the system. During quenching, the system is brought to a temperature-concentration regime where the single phase state is no longer stable. This is the isoconcentration

Figure 2. Phase Diagram of Surface with Film and Substrate

Path A and Path B can bring the system into two-phase coexistence regime. For explanation, see text

transition. This process is shown by Path A. The regime now consists of two phases, a dense state phase (2D islands or 3D clusters) and a dilute adatom phase. The other way is to enter the two-phase coexistence regime isothermally, represented by Path B. Continuous deposition of adatoms onto a substrate at a constant temperature drives the system into a supersaturated state, i.e., it makes the 2D vapor pressure of the adatoms higher than that at equilibrium. The system tries to move back to equilibrium by forming a condensed phase, e.g. 2D islands or 3D clusters. The large decrease in molar entropy accompanying the process indicates that condensation is a thermodynamically favored process. The driving force for condensation comes from the lowering of the potential energy of the adatoms when they form bonds to each other. Since the adatoms are continuously being deposited, this relaxation toward equilibrium is taking place continuously.

There are three major stages in the formation of a condensed phase from a 2D vapor: nucleation, early stage growth, and late stage growth. Arriving adatoms make a random walk on the surface and, upon meeting each other, form nuclei. This is the nucleation process. In this stage, the nuclei are generated in large number but with a narrow size distribution. Random nucleation and spinodal decomposition are the dominant processes in nucleation. In random nucleation, the 2D adatoms are in a metastable state. This means that an energy barrier exists to form the two-phase regime, i.e., to form nuclei. The system needs to overcome this barrier to relax. On the other hand, in spinodal decomposition, the 2D adatom adlayer is unstable and forms nuclei spontaneously without a barrier to overcome. While nucleation continues the existing nuclei start to grow by

capturing more adatoms. The nucleation rate slows down when supersaturation is reduced. In the early growth stage, nucleation may still occur. However, the growth of 2D clusters is more pronounced. In this stage, depending on the deposition parameters, coalescence may occur, although this process mainly takes place in later growth process. This stage is a transition stage between nucleation and the later growth stage.

In the later growth stage, also called the coarsening process, the islands continue to grow by capturing free adatoms but other processes, i.e., combining two islands into a larger one upon impact, dominate. This process is favored since the surface free energy is reduced during the coalescence. When deposition is stopped and supersaturation is locally eliminated, island growth is usually thought to be dominated by Ostwald ripening. Ostwald ripening is characterized by large clusters growing at the expense of smaller clusters. Small clusters evaporate and their atoms diffuse on terraces and incorporate into larger clusters. The driving force for the ripening process is a concentration gradient of adatoms associated with the difference in vapor pressure over islands of different size. At this time, coalescence is also possible if the clusters are mobile. This stage is characterized by the dramatic reduction in cluster density.

This simple picture is applicable only in a binary system, when there are two different phases and no intermixing of adatoms and substrate atoms occurs. If the coverage is in the multiple layer regime, the process described will be complicated further, since interlayer diffusion and film roughness may occur. Therefore, the study of submonolayer deposition can eliminate some of these complications while providing valuable information

about growth.

Realistically, the other way to reduce and eliminate supersaturation in the 2D lattice gas is the incorporation of adatoms at surface steps. The surface by no means is flat. Steps always exist due to imperfect plane orientation. Steps are generally considered good places to trap adatoms because step atoms have higher coordination. Therefore when an adatom arrives on the surface, it diffuses on a terrace and takes one of the following ways to settle: colliding with another adatom or several other adatoms to form a new island (nucleation), exchanging with a surface atom (surface alloying), diffusing into the bulk of the substrate (interdiffusion), migrating into an existing island (growth), migrating into a substrate step (step incorporation), or leaving the surface and returning to the gas phase (reevaporation).

Figure 3 shows these processes.

Island formation and incorporation at surface steps are two competing processes. The spatial distribution of islands formed after a submonolayer deposition reflects this competition. Near steps the island number density will be lower than that far from the steps because the adatoms that land near the surface steps are able to reach the steps before meeting other adatoms to form islands. From the atomistic point of view, the most important microscopic aspects underlying the above processes are the interaction of adatoms with steps (including substrate steps and island steps) and the surface migration of adatoms. The interaction between adatoms and surface steps includes 2D condensation and desorption from steps, step barriers for adatom to cross steps, and diffusion along steps.

The general expectation is that all surface steps are good adatom traps, i.e., all

adatoms reaching a step from its down terrace (ascending) by surface diffusion will be trapped at the step to lower the total surface potential energy, since the surface steps are conceptually identical to 2D cluster edges. The existence of an energy barrier at a step for adatoms to cross downward is assumed for some systems, corresponding to an adatom being reflected back when it approaches a descending step. The physical picture to explain the existence of this barrier is that at the edge before an adatom is falling down over the step the adatom will have a lower coordination number and hence higher energy than on the flat terrace. In other words, the adatom would rather stay on the terrace than cross the step. This barrier also affects the smoothness of film growth. In the case where the step barrier is high, adatoms are not able to cross downward. Therefore, each adatom will be forced to stay on the terrace where it landed. Consequently, there is a difficulty to fill one layer completely, resulting in 3D growth. A knowledge of the existence of a possible barrier for downward flow is therefore of great importance for the understanding of the growth process. Since not all systems have step barriers, the physical picture mentioned above is not universal.

Once an adatom reaches to an edge, it may diffuse to find the edge site with lowest energy and then stay there. If its binding to the edge is not very strong or the temperature is high enough, the adatom may diffuse along or even desorb from the edge. The ease with which adatoms can move along the edge depends upon the atomic structure of the edge and the interaction between the adatoms and the edge. The diffusion barrier for edge migration is lower than that for laterally desorbing from the edge onto a terrace, but higher than that

for migrating on the flat terrace because of the extra bonding with edge atoms. Due to these possible interactions between adatoms and steps, the growth kinetics can change dramatically. Furthermore, the important role steps play in film growth can be described by a process known as step propagation. In this process the incorporation of adatoms to steps dominates over nucleation and growth on terraces. It can be considered as a heterogeneous nucleation process at steps and the result is step growth. This process is more likely to occur on high step density surfaces.

b. General experimental methods

Physical and chemical properties of thin films are determined to a large extent by their structure and morphology, therefore by the growth mode. Studies on these subjects have revealed a variety of complex atomic surface structures and growth behaviors. Some of these studies have employed averaging techniques, such as scattering methods and diffraction methods, from which local atomic structures and film morphological information can only be obtained indirectly. The different growth modes have been investigated from the coverage dependence of spectroscopic intensities or from the intensities/widths of diffraction beams at different coverages. Other studies have involved high resolution tools to reveal detailed local information on the atomic scale directly. These methods include field emission and transmission electron microscopes, but due to experimental limitations, only limited systems have been studied under extreme conditions. Basically, these methods can track the system for a very long time. Surface structure and morphology changes as a

function of time can provide insight to the kinetics. These are direct measurements. A newer technique, scanning tunneling microscopy (STM) can provide, in real space, direct information on film dynamics with high resolution and large field, because of the local nature of the measurements. In particular the different aspects of the later growth stages can be studied and understood to a large extent from systematic STM observations. Thus, more and more studies have been carried out with this tool. In this chapter, section 4, a discussion of STM's principles of operation will be given.

3. Literature review

The purpose of this section is to provide a brief review on published experimental results regarding surface diffusion and coarsening.

From the aforementioned description of growth processes, the dynamics of adatoms and clusters are very important to film morphology and structure. In fact, surface diffusion is one of the most important fundamental interactions in the metal-on-metal thin film growth. In all stages of film growth, diffusion is the central step involved. As Doll and Voter have said, *"Diffusion plays the role in interfacial phenomena that gas-kinetic collisions do in gas-phase chemistry. An understanding of surface diffusion is a prerequisite for understanding the rate of a variety of interfacial phenomena, including surface chemical reactions, crystallization, annealing, and vapor deposition. Conversely, surface diffusion itself serves as a valuable microscopic probe of both surface topography and adsorbate/surface interactions"*[1]. Only under extreme conditions, such as when the

substrate temperature is low enough, or when the binding between adatom and substrate is very strong, are the adatoms fixed on the sites where they land. In this growth regime, diffusion is not operative and will not be discussed in this study. We will begin with single atom diffusion.

A substrate is not flat. From the microscopic view, there are steps, defects, and contamination. At the atomic level, the arrangements of surface atoms affect surface corrugation. For example, an fcc(111) surface is less corrugated than an fcc(110) surface. The moving of adatoms from one surface site to another is called surface diffusion. Surface diffusion encompasses a broad phenomena. The atomic motion on the surface can include that of individual atoms and clusters, diffusion of vacancies, rearrangement of clusters, and stability of clusters. The direct consequence of these motions is changes in surface morphology and surface features.

The traditional picture of single atom diffusion on low-index surface is the hopping of the atom, i.e., the atom moves from one energy minimum site to another minimum site via a potential minimum pathway, a saddle point. A simple picture of atom hopping on surface can be viewed as a rigid spherical ball rolling on a rigid surface lattice consisting of a matrix of rigid balls. The activation energy for surface diffusion is the energy difference between the saddle point energy and the energy associated with the equilibrium binding position. One expectation would be that increasing the binding energy of the atom to the surface will make the atom more difficulty to migrate, i.e., higher activation energy. Another expectation would be that rougher surfaces will have a higher activation energy

due to higher saddle point. The diffusion of single metal atoms on metallic surface has been studied mainly by field ion microscope (FIM). FIM is capable of locating the position of an atom. An individual atom on a well defined low-index single crystal surface can increase local field strength, and therefore, enhance field ionization. A bright spot due to the higher emission is an indicator of the location of the atom. The locations of the atom as a function of time can provide diffusion information. Due to the nature of FIM, a limited number of surfaces have been used to study different problems. Wang and Ehrlich [2], and Bassett and Parsley [3] have measured diffusion barriers for various metal atoms (Pd, Ni, Rh, Ir, and Re) on W(211). They found that with increased bulk sublimation energies of the elements, the diffusion barriers were increased. This means that the surface diffusion barriers were higher for higher-melting-point metals. Tung and Graham [4] measured Ni on Ni(110). The row-and-trough surface of fcc(110) is a good choice to examine the anisotropic diffusion behavior. When the activation energies for in-channel and cross-channel diffusion were compared, they found in-channel diffusion had a barrier of 0.23 eV while the other was 0.32 eV. Ayrault and Ehrlich [5] conducted a systematic study of Rh on Rh surfaces with different atomic roughness. They found that the activation energy increased with increased atomic corrugation of the surface. For the more isotropic surfaces (111) and (100), with increased corrugation, the barriers were 0.16 eV and 0.88 eV, respectively. The isotropic surfaces are surfaces which have identical atom arrangement in both the x and y directions on the surface plane. The anisotropic surfaces are surfaces with different atom arrangement in x and y directions on surface plane. For anisotropic surfaces (311), (110), and (331), with

increased corrugation, the barriers were 0.54 eV, 0.60 eV, and 0.64 eV, respectively. Chen and Tsong [6] measured Ir self-diffusion on unreconstructed and reconstructed Ir(110) surfaces. They concluded that the higher diffusion barrier in reconstructed Ir(110) in-channel diffusion than that in unreconstructed was due to a slightly larger corrugation after reconstruction.

In addition to the traditional diffusion pattern, a second diffusion mechanism for single atoms is proposed, i.e., exchange-displacement, to interpret unexpected experimental results. Bassett and Webber [7] studied Pt self-diffusion on Pt(110). They reported that on this row-and-trough surface, instead of seeing the expected anisotropic motion of the atom, the atom had 2-dimensional migration and the diffusion barrier of in-channel diffusion was similar to the barrier of cross-channel motion. To explain this surprising experimental finding, they proposed the exchange-displacement theory, in which diffusion in the cross-channel direction results when an adatom pushes one atom in the channel wall onto the adjacent channel and then itself occupies the channel wall position. Because the atoms in the channel wall have less bonding than atoms on the terrace, these atoms are easier to replace energetically. Wrigley and Ehrlich combined FIM and time-of-flight mass spectrometry [8] to provide more experimental evidence to support the exchange-displacement mechanism by their study of W on Ir(110). This system is a heterogeneous system, and the chemical identity of adatom and wall atom can be detected. Mass spectrometry was used to identify the atoms monitored by FIM. More convincing results came from Pt on Ni(110) [9].

Since the exchange-displacement mechanism was proposed and confirmed

experimentally in 1978, all measurements were done on rough surfaces such as fcc(110). However, the theoretical work about self-diffusion on fcc(100) by Feibelman [10] indicated that the exchange-displacement process can take place on atomically-smooth surfaces since the activation energy of this process is less than that of hopping process. The exchange mechanism on an atomically-smooth surface is as follows: The adatom occupying the four-fold hollow site on a fcc(100) surface, for example, starts pushing down toward one of the four surface atoms forming a four-fold hollow site. The adatom is lowering its height while the surface atom is raising its position. At the saddle position of the process, these two are at same level above the surface, and the bonding configurations are the same. Then, the adatom starts to move into the vacancy left by the surface atom. At the same time, the surface atom moves to the diagonally opposite four-fold hollow site. That the exchange-displacement mechanism is favored over traditional hopping can be explained because the former allows less bond-breaking during the process than the latter. Kellogg and Feibelman [11] and Chen and Tsong [12] confirmed this mechanism by their studies of self-diffusion on Pt(100) and Ir(100), respectively. Ni on Pt(100) [13] and Pt on Ni(100) [14] also follow the same mechanism. It will be very interesting to see whether this mechanism can occur on the atomically-smoothest surfaces, fcc(111) or hcp(001). So far, there are no reports.

Metal cluster diffusion has also been investigated, mainly with FIM. Both the motion of individual atoms in the cluster and the motion of the center of the cluster attract a great deal of attention [15-18], since the relationship between individual atom displacement and the center of the entire cluster can provide information on the cluster diffusion mechanism.

The center of the cluster can diffuse because of the movement of atoms around the cluster periphery, and because of dissociation-reattachment of periphery atoms. The cluster may migrate under simultaneous movement of every atom in the cluster, i.e., the entire cluster gliding on the surface.

Dimers, the smallest possible cluster, have been studied extensively. For row-and-trough surfaces, two types of dimers exist. If the dimer is oriented perpendicular to the surface row, the dimer is called cross-channel dimer. When the two atoms are oriented in the same direction as the rows, the dimer is named an in-channel dimer. There are many studies about cross-channel dimer diffusion. All these studies show that individual atoms undergo a series of motion which causes the dimers' mobility. Studies of Re_2 on $\text{W}(211)$ [15], W_2 on $\text{W}(211)$ [16], W_2 on $\text{W}(110)$ [17], Pt_2 on $\text{W}(110)$ [18] all suggested that the displacement of the center of cluster was due to displacement of each atom, not due to the simultaneous movement of the whole cluster. Furthermore, the activation energies for dimers in many cases were found to be comparable to or smaller than that for single atom, such as W_2 on $\text{W}(110)$ [19], Rh_2 on $\text{W}(110)$ [20], Mo_2 on $\text{W}(211)$, Re_2 on $\text{W}(211)$ [15], Pt_2 on $\text{W}(110)$ [18]. The possible explanation for this observation is that the dimer stays at a surface with less potential roughness than the monomer does [21]. This helps to reduce the diffusion barrier. The energies at saddle points will also be reduced due to interaction between atoms of cluster [15,22,23,24]. These observations show that cluster diffusion is as important as single atom diffusion during thin metal growth.

Experimental studies are also available for clusters larger than dimers, but the

number is limited and the clusters are general smaller than 20 atoms. A study of Pt on Rh(100) [25] indicated hexamers have a higher activation energy than trimers, tetramers, and pentamers which have the same activation energy. The observed cluster shapes may help to explain the results. The trimer, tetramer, and pentamer are most stable when they line up with a one-dimensional configuration. The same shape in clusters ($n=3,4$, and 5 respectively) may be responsible for the same activation energy if they have same rate-limiting step in the diffusion process. For the hexamer, the most stable configuration is a two-dimensional cluster. The different rate-limiting step with a higher barrier gives the hexamer lower mobility. A study of Ir self-diffusion on Ir(111) by Wang and Ehrlich [26] showed Ir atoms migrating on the edge of a 12-atom cluster and consequently the center of the cluster had a short displacement.

The general trend of the relationship of the cluster size with activation energy is that the activation energy increases as the cluster size increases for these small clusters. Pt on W(110) [18], Ir on Ir(111) [27], Rh on W(110) [20] all indicate this general trend. However, Kellogg [28] reported an interesting result for Rh cluster (2-12 atoms) diffusion on Rh (100). He found that clusters with a square or rectangular shape have higher activation energies than clusters with extra atoms at the cluster edges. Thus, the activation energy of diffusion exhibits an oscillatory behavior as a function of cluster size. This behavior rules out the gliding of the entire cluster as the diffusion mechanism. Instead, atom migration around cluster edges is more likely for these small clusters, since extra energy is needed to release an atom from an edge of the cluster.

Thus far cluster diffusion has been shown to include individual atoms in a cluster contributing to the entire cluster's motion via hopping. There is little experimental evidence showing that individual atoms can facilitate the cluster diffusion via exchange [29]. Kellogg and Voter showed that the Pt dimer on Pt(100) diffuses via an exchanging process in which one atom exchanges with a substrate atom in a similar manner as in the monomer case and the other atom in the dimer basically stays stationary. The results of the displacement are that the center of the dimer moves and the dimer changes orientation. The activation energy for this process is lower than that of the monomer. For Pt trimers, the exchange and hopping both contribute to the motion of Pt_3 , and the activation energy is comparable to that of the monomer.

FIM has been proven to be a successful and mature method to measure adatom and cluster diffusion on surfaces. During the last three decades, FIM has been the main technique to provide understanding of the fundamental phenomena of diffusion of atoms and small clusters at the atomic-scale. Very detailed diffusion information is available only for atom/small cluster. However, this method has severe drawbacks, including that the substrate is limited to a very small size, generally around $100\text{\AA}$. In an FIM apparatus, the substrate is in the form of a tip. Therefore, the substrate materials are limited to those from which an emission tip can be made. The tip has surface structure other than an ordinary crystal surface. An FIM tip has a near circular shape with descending steps at the edge of the terrace in all directions. Coexistence of planes with different Miller indices may be the case. Under these conditions, therefore, the size of the cluster for a specific surface plane is

limited.

Compared with FIM, STM is a relatively new technique. It can view a larger area with atomic resolution than FIM. The intermediate length-scale of the observation it provides can be very useful for understanding thin film growth, since this is the area range between FIM and averaging techniques such as diffraction and scattering methods. One example of an averaging method was the characterization of diffusion of Cu on Cu(100) indirectly. Ernst [30] used He scattering to investigate the system over a wide range of coverage and temperature. They concluded that the diffusion of Cu adatoms on Cu(100) is via exchange.

STM has been used in studies of nucleation [31,32,33,34], island shape and size [35], growth mode [36,37], and interactions between adatoms and steps [32, 35]. A few measurements of diffusion with STM at an atomic level are available. Gane and coworkers[38], using STM, observed that isolated Pd adatoms can diffuse on Ge(111) at elevated temperatures via the exchange pathway. The same group also described concerted motion of an adatom row on Ge(111). Mo [39] and Stroscio [40] used STM to measure the average island density to obtain diffusion information with the modeling of the nucleation and growth processes. In another STM study, Sb on Si(001)[41], Mo usedSTM to study the Sb dimer diffusion. For metal-on-metal systems, no experimental literature is available on the mobility of large clusters.

Vacancy cluster diffusion can play an important role in film ordering kinetics. 3D vacancy clusters have been studied extensively in crystal growth processes and in metallic

alloys [42,43,44,45,46]. Recently the study of two-dimensional vacancies emerges. A two-dimensional vacancy cluster can be viewed as the inverse of a two-dimensional cluster.

Thus, STM is an ideal tool to study this phenomenon also. One recent STM experiment by de la Figuera *et al.* [47] showed that artificially created (with STM tip) two-dimensional Cu vacancy clusters on clean Cu(111) are immobile, but on Co-modified Cu(111), Cu vacancy clusters undergo surface diffusion. The presence of Co on the Cu surface is needed to lower the surface diffusion barriers. Another example of 2D metal vacancy diffusion was shown by Comsa [48]. Ag vacancies were created by ion sputtering on a Ag(111) surface. Brownian motion described the diffusion of the vacancies. The relationship between cluster size and diffusion coefficient is also addressed. All these studies were carried out on a triangular substrate.

Once a film has been deposited, two processes are considered as main contributors to the evolution of the films: Ostwald ripening and dynamic cluster coalescence. The concept of Ostwald ripening was first developed by Ostwald [49], then later advanced by Lifshitz, Slyozov and Wagner [50,51]. This phenomenon has been studied extensively [52]. The idea of dynamic cluster coalescence was first used in thin film growth by Zinsmeister [53]. This term refers to the coalescence of clusters due to random motion. It has been found that cluster mobility can affect film growth in many aspects, such as the cluster size distribution [54]. Both Ostwald ripening and dynamic cluster coalescence will result in a decrease both in the number of clusters, and the size distribution as a function of time for these mechanisms. It is difficult to distinguish these two mechanisms by just looking at the

final results [55]. In experimental studies of later stage growth, the cluster size and shape are characterized with scattering methods [56], scanning electron microscopy (SEM) [57], scanning transmission electron microscopy (STEM) [58], low energy electron microscopy (LEEM) [59], and STM [60,61]. Futamoto, *et al* [57], by using SEM, showed Ga clusters on GaAs(001) underwent different coarsening processes under different conditions, i.e., at 833K cluster coalescence dominated, while at 758K Ostwald ripening dominated. SEM was also used to investigate systems involving metals: Ag on Mo(100) [62], Ag on W(110) [63], Ag on Si(111) [64]. However, the studies were limited by the low resolution of SEM. LEEM, with higher resolution ($\sim 20\text{\AA}$), was used by Bauer [59] to observe the cluster dynamics of Au on Si(111). STM was used on Si, Ge on Si(100) [60,65] to study cluster shape and size. On Fe/Fe(100), STM was used to analyze the cluster density in order to characterize the growth kinetics.

In this thesis, STM is used to investigate a metal-on-metal system, Ag on Ag(100). Both large adatom cluster diffusion and vacancy cluster diffusion are studied. The mechanisms involving diffusion are discussed. Diffusion coefficients are measured. Growth processes in the late stage are also investigated in detail with STM. Other dynamic processes, such as biased cluster motion, are studied as well.

4. Scanning tunneling microscopy

a. Introduction

Since its first successful operation with atomic resolution in 1982 [67], scanning tunneling microscopy (STM) has been widely used to study a great variety of problems, including many traditional surface science problems, as well as many new ones. This technique redefines the concept of microscopy because it allows us to see atoms and other small features on surfaces in a wide range of environments such as in air, liquid, and ultrahigh vacuum [68]. In this section, the general concepts of STM are discussed.

Because of its atomic resolution and the ability to scan large areas, STM is an ideal tool for studying surface kinetic processes at the atomistic level. Using STM, we are capable of achieving a more accurate and comprehensive understanding of growth processes microscopically. This technique is based on quantum mechanical tunneling, in which electrons penetrate with nonzero probability through an energy barrier higher than the electrons' total energy. In STM, an atomically sharp metal tip is brought within a few Å to a sample surface such that the wavefunctions of the tip electron states overlap with those of the sample. A tunneling current will flow between the tip and the sample when a bias voltage is applied between them. The tunneling current has exponential dependence on the distance between the tip and the sample, i.e., the thickness of the barrier. At a typical tunneling distance, a roughly one Å change in the gap spacing will cause the tunneling current to change by a factor of ten. In the operation of an STM, piezoelectric elements (orthogonal piezoelectric triple or piezoelectric tube) force the tip to move in three

orthogonal directions: X and Y parallel to the sample and Z perpendicular to the surface. As the tip scans across the surface, the width of the microscopic tunneling gap changes due to the surface corrugation in the electron density contours, resulting in a change in the tunneling current. If this changing tunneling current is plotted vs. the X and Y positions, it directly gives an image of the surface corrugation. Another way to scan is to control the tip in the z direction with a feedback voltage to maintain a constant tunneling current, then to plot current vs. the X and Y positions to reflect the surface corrugation. This latter mode is called the constant-current mode. This constant-current contour mapping is most widely used because the feedback circuit prevents the tip from crashing into the surface, which is particularly important for rough surface. Because of the sensitive dependence of the current on the gap spacing together with an atomically sharp tip, an STM has extremely high resolution: $\sim 1\text{\AA}$ laterally and $\sim 0.1\text{\AA}$ vertically, thus enabling us to resolve atomic features on the surface. Furthermore, there is no fundamental limit on the maximum scanning range of STM, and views of large areas with atomic resolution can be obtained.

b. Theory

It is a great challenge to interpret STM images theoretically, since a theoretical treatment needs to relate the constant-current contours to the variation of surface properties of the samples. Early analyses of the tunneling current and STM image were based on analogy with the one-dimensional tunneling problem [69]. The relationship between current I and bias V is obtained from the following equation at low voltage and temperature [69]:

$$I \propto \exp(-2kd)$$

where k is the decay constant of the wave function in the tunneling barrier, and d is the separation between sample and tip. The decay constant, k , is related to the effective local work function ϕ by $k=(\hbar/2\pi)^{-1}(2m\phi)^{-0.5}$. The exponential dependence of the tunneling current on distance gives STM its high sensitivity.

However, when considering STM scanning on a surface it is necessary to solve the three-dimensional tunneling problem. This involves the electrodes' Fermi level states which have complex spatial structures, leading to complex electronic interactions between electrodes. Moreover, it is uncertain what the distance, d , actually represents under this 3D scanning condition.

One of the most successful attempts to interpret STM images has been made by Tersoff and Hamann [69]. They considered that the tip-surface interaction was weak when the separation was about a few Å. These two weakly coupled electrodes allowed them to use first-order perturbation theory based on Bardeen's transfer Hamiltonian approach. This method requires explicit expressions for the wavefunctions of the tip and the sample surface. Since the actual atomic structure of the tip is generally not known, they started with the simplest possible model, a spherical symmetric wavefunction, for the tip. The size of the tip is not important as long as the spherical symmetric wavefunction is maintained. Their results state that the tunneling conductance is proportional to the local density of states of the sample, at tip position and at Fermi energy. The tip path maps out a contour of constant Fermi-level local density of states (LDOS) of the surface. This theory has clarified

many important questions such as the resolution of the microscope. This theory is used to explain the STM images of the reconstructed Au(111) surface, which has a larger surface unit cell than a normal metal surface lattice [69]. However, the theory is somewhat simplistic, since both geometric and electronic structures of the tip must play an important role in the formation of the image. Thus this theory can be considered as a "macroscopic" approach. The theory cannot provide an explanation of the STM atomic resolution on a close-packed metal surface.

Taking into account the atomic structure of the tip, Chen [70] further developed the theoretical treatment of STM theory. His theory enabled one to explain the atomic resolution STM image of Al(111), a close-packed metal surface without reconstruction. He used a d_z^2 -tip. The microscopic view of STM introduced in detail by him is based on symmetry considerations of the electronic states of the tip and the sample surface and therefore emphasizes the atomic-level imaging of STM.

Lang [71] introduced a model incorporating the microscopic and macroscopic pictures of STM. He described the tip and sample by an atom adsorbed on a jellium model of the surface. This theory considers the total current due to real atoms on the sample and tip. In his work, a sodium tip was scanned over a surface with adsorbed sodium, sulfur, and helium atoms. Tip displacement versus lateral separation for constant current was plotted. This plot was compared to contour plots of constant Fermi level local density of states and constant total density for each of the three surfaces versus the sodium tip. This method successfully explained the STM images of single-atom adsorbates on metals.

A perturbative treatment of tunneling will break down if the tip-surface separation becomes extremely small, i.e., if the wavefunction overlap of tip and sample surface becomes significant. In this case, the perturbative method is no longer valid. Non-perturbative treatments of tunneling based on the Green's function method [72, 73], which goes beyond the perturbation approximation, is used to improve the approximation. These more intensive STM theories will not be reviewed here.

c. Instrumentation

(1) Scanning Mechanism

A very precise and very stable driver for controlling the tunneling tip in three orthogonal directions is necessary in STM, since the tip rasters in the X-Y plane while responding to the surface corrugation in the Z direction with a feed-back loop. Piezoelectric materials can generate mechanical strain under the influence of an electric field across them. The deformation of material can provide linear motion at the sub-angstrom level and up to a hundred micrometers [74]. This capability, as well as the simple relationship between the strength of applied field and mechanical deformation results in this type of material being used almost exclusively in STM scanners. High piezoelectric-constant piezoelectric materials are used for small scanners in order to have enough scan area.

Currently, scanners with two different configurations are widely used, tripod and single tube. In a tripod scanner, three piezoelectric material bars are mounted orthogonally to each other. Two electrodes are placed across the two ends of each other. A polarization

process is needed before the bar can be used. A high electric field is employed across the two electrodes to force the randomly oriented dipoles in the material to orient in the direction of field, and then the field is removed. The polarization of the material will remain unless it is annealed to its Curie temperature resulting in the loss of polarization. The Curie temperature is the temperature at which the material's dipoles will reorient randomly. Under the influence of an applied field with the same direction as the polarization direction, the polarized material will expand in the direction of applied field and contract in the direction perpendicular to the field direction.

The second type is a single tube scanner. The outside of the tube is sectioned into four electrically separated parts along the circumference. Electric fields with the same strength but opposite signs are applied to the two opposite quadrants. Therefore, one quadrant will expand and the other will contract. This bending will cause the lateral movement. The tunneling tip is mounted at one end of the tube. The tube is polarized radially with a field applied between the inner and outer walls of the tube.

(2) Approach Mechanism

In order to bring the tip to within a tunneling range, one must move the tip over a macroscopic distance (hundreds of microns) with a precision of several tens of nm without crashing the tip into sample. An approach device is designed to do this. Several schemes are possible. Some designs are based on purely mechanical means [74], using fine-thread screws or a level system that transforms a coarse motion into a very fine linear tip approaching

motion until the tunneling current is detected. The other type involves the expansion and contraction generated by large size piezoelectric materials with a variety of clamping mechanisms to provide step motion of the scanner.

(3) Vibration Isolation

Effective vibration isolation is one of the critical elements in achieving atomic resolution by STM. Because the tunneling current is very sensitive to the separation between the tip and the sample, any disturbance due to external vibration can cause large noise in STM signals. The typical atomic corrugation amplitude is about a few tenth of an Å. Thus, the fluctuation of the separation must be reduced to much less than 0.1 Å in order to achieve a good atomic image.

To overcome the external vibration influence on the tip-sample junction, two approaches have been employed: (1) high rigidity of STM assembly, (2) a vibration isolation system [74]. In general the trend has been toward compactness and simplicity to improve rigidity. It has become clear that increasing the rigidity of STM can be more conveniently achieved than perfecting a vibration isolation system. If the gap spacing between tip and sample can be held constant against all external vibration by using a rigid STM design then no isolation system is needed. But in practice, there are some limitations in improving the rigidity of STM, for example, the approach device and sample itself need to be able to move. In addition, other factors, such as operational convenience and construction cost, should be considered. Therefore, a combination of both is widely used. To date, the suspension spring with eddy-current damping is the most efficient vibration isolation system

for a very rigid STM. However, this system is large. An elastomer system is also in use.

This system uses a stack of metal plates which are separated by rubber pieces. By increasing the number of metal plates, reasonably good vibration isolation can be achieved in a modest space. If the STM is not rigid enough, oif it is maintained in an Ultrahigh Vacuum (UHV) or cryogenic environments more sophisticated vibration isolation systems are needed.

Pneumatic systems, as a secondary vibration isolation stage, are often used to float the whole STM or the entire UHV-STM.

5. Dissertation organization

This thesis includes four papers. Paper 1: ^{removed} Diffusion of Large Two-Dimensional Ag Clusters on Ag(100), appears in Physical Review Letters Volume 73 on page 2591, 1994.

Paper 2: ^{removed} Coarsening Mechanisms in a Metal Film: From Cluster Diffusion to Vacancy Ripening, has been submitted to Physical Review Letters. Paper 3: Large ^{removed} Ag Vacancy

Cluster Diffusion on Ag(100), will be submitted to Surface Science. Paper 4: ^{removed} Surface

Dynamical Phenomena, will be submitted to Surface Science. Following the last paper are general conclusions, references cited in Chapter 1, and appendices that describe the apparatus and data summary.

CHAPTER 6

CONCLUSION

During film growth, adatoms are continuously deposited onto the substrate at a certain rate. The adatoms that land on the substrate will have a certain lifetime to migrate on the surface and interact with the surface atoms as well as with each other. In some cases, these interactions may cause growth of substrate steps, intermixing of deposited atoms and substrate atoms, and alteration of further film growth. These interactions result in nucleation on the surface, two-dimensional (2D) condensation of adatoms into islands and their subsequent growth, and the continuous growth from 2D to 3D cluster.

Growth kinetics depend on the fundamental surface processes of surface adsorption, desorption, and diffusion. Often the thermodynamically favored film states are 2D islands and 3D clusters on the substrate rather than a random distribution of adatoms. Film growth is a non-equilibrium process where surface phase separation occurs and growth kinetics plays an essential role. In this work, the cluster mobility are studied extensively.

From the results summarized below a better understanding of thin film growth process is achieved. The information of the metal cluster mobility on metal surface can be used to get information of surface diffusion energy barriers which in turn can help people to understand the fundamental interaction involved in the film growth for other systems.

In summary, we used STM to study metal thin film growth processes. The main conclusions are:

1. Large metal cluster diffuse on Ag(100) surface. The metal-metal system large adatom cluster diffusion is reported for the first time. The evaporation-condensation of adatoms dominating the vacancy cluster diffusion process is suggested. The diffusion coefficients is on the order of $10^{-18} \text{ cm}^2 \text{ s}^{-1}$.
2. Metal film evolution behaviors at different coverage regime are examined. At low coverage, cluster diffusion dominates the coarsening process. At high submonolayer coverage, vacancy cluster repining dominates the process. At moderate coverage, edge-running dominates the process.
3. The vacancy cluster diffusion is also observed. The vacancy were formed by controlling the deposition flux and film coverage. The method is simple and offers similar advantages of the sputtering method. The diffusion coefficients is on the order of $10^{-18} \text{ cm}^2 \text{ s}^{-1}$, is smaller than that of adatom clusters, and has a scaling factor α roughly close to 0.5 in $D \propto N^\alpha$. The evaporation-condensation of vacancy monomers dominating the vacancy cluster diffusion process is suggested.
4. We have presented a number of observations of dynamical phenomena of Ag cluster on Ag (100) with UHV-STM. Ag cluster on Ag (100) at room temperature are mobile. The interaction between clusters and between clusters and surface steps are mediated via 2D dilute adatom gas on the terrace and different adatom concentrations around clusters and steps. Background gases and impurities on cluster edges can strongly

affect film coarsening process. The former can enhance clusters dissolution, but the latter can stabilize the local structure and geometry of the Ag film.

Additional work can be done in the following areas. First, the temperature dependence of clusters' mobility is useful to get information about diffusion barriers. Second, similar measurements for different orientations of Ag substrates are useful to compare the impact of substrates on film growth process.

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APPENDIX I

THE CONSTRUCTION OF THE VACUUM CHAMBER

The vacuum we used to carry out all experiments is composed of two parts. The upper part is used to house all the equipment, including STM, sample transfer arm, evaporation source, etc. It includes two levels. The lower part is used to house all the pumps. The entire chamber (excluding two mechanical pumps) sits on a metal table, which is supported by a servo controlled air suspension unit. The unit provides vibration isolation.

Figure 1. Vacuum chamber upper part, first level ports

Figure 2. The vacuum chamber upper part second level ports

port # 8: view port; port #12: sample transfer arm; port #18: evaporation source
port # 20: STM

Table 1. Port configurations

PORT #	FLANGE	PORT LENGTH INCHES	FOCAL POINT			ELEVATION ANGLE	AZIMUTHAL ANGLE	TUBE DIAM. INCHES	AZIMUTHAL ANGLE OF FOCAL POINT	RADIUS OF FOCAL POINT
			X	Y	Z					
FIRST LEVEL										
1	6" O.D. ROTATABLE CONFLAT	21.15"	0	0	0	90	0	4	0	3
2	2 3/4" O.D. TAPPED NONROTATABLE CF	6.1"	2.598	1.500	13.5	72	30	1.51	30	3
3	2 3/4" O.D. TAPPED NONROTATABLE CF	6.1"	0	3.000	13.5	72	90	1.51	90	3
4	2 3/4" O.D. TAPPED ROTATABLE CF	6.1"	-2.598	1.500	13.5	72	150	1.51	150	3
5	2 3/4" O.D. TAPPED NONROTATABLE CF	6.1"	-2.598	-1.500	13.5	72	210	1.51	210	3
6	2 3/4" O.D. TAPPED ROTATABLE CF	19.62"	0	-4.700	0	90	270	1.51	270	4.7
7	2 3/4" O.D. TAPPED NONROTATABLE CF	6.1"	2.457	-1.725	13.5	72	325	1.51	325	
SECOND LEVEL										
8	8" O.D. NONROTATABLE CONFLAT	8"	0	0.00	13.5	0	20	6		
9	1 1/3" O.D. NONROTATABLE CONFLAT	7"	0	0.00	16.5	0	67	0.75		
10	2 3/4" O.D. ROTATABLE CONFLAT	9"	0	0.00	13.5	0	67	1.51		
11	2 3/4" O.D. TAPPED NONROTATABLE CF	6.5"	0	3.00	13.5	40	90	1.51		
12	2 3/4" O.D. TAPPED ROTATABLE CF	9.5"	0	0.00	13.5	0	90	1.51		
13	1 1/3" O.D. NONROTATABLE CONFLAT	7"	0	0.00	16.5	0	113	0.75	90	3
14	2 3/4" O.D. ROTATABLE CONFLAT	9.3"	0	0.00	13	0	113	1.51		
15	2 3/4" O.D. TAPPED ROTATABLE CF	7"	-1.847	2.36	13.5	42	128	1.51		
16	2 3/4" O.D. ROTATABLE CONFLAT	12.5"	0	0.00	14	0	128	1.51		
17	1 1/3" O.D. NONROTATABLE CONFLAT	7"	0	0.00	16.5	0	143	0.75		
18	2 3/4" O.D. ROTATABLE CONFLAT	9.3"	0	0.00	13	0	143	1.51	128	3
19	8" O.D. ROTATABLE CONFLAT	13"	0	0.00	13.5	0	200	6		
20	8" O.D. ROTATABLE CONFLAT, TAPPED	7"	0	0.00	13.5	0	270	6		
21	6" O.D. ROTATABLE CONFLAT	12"	0	0.00	13.5	0	325	4

Figure 3. The side view of the chamber

APPENDIX II

PREPARATION OF SCANNING TUNNELING MICROSCOPY TIP

In recent years interest in ultrasharp tips has been greatly enhanced by their role in scanning tunneling microscopy (STM). The quality of STM tips is one of the important factors which affect the resolution of the STM images. The procedures to produce sharp tips can be classified into three categories: (1) mechanical method; (2) physical method; and (3) chemical method [1].

Mechanical methods generally involve simple mechanical operations, such as cutting or grading a metal wire made of W, Pt-Ir, Pt-Rh [2,3]. A sharp tip is formed based on the assumption that a single atom or a cluster of atoms must protrude at the cut end and therefore provide the atomic sharpness needed in STM measurements.

Physical methods involve modification of the tips end, such as coating [4], ion sputtering [5], and field evaporation [6].

Chemical method is basically etching a metal wire electrochemically. This method has been used extensively to sharpening metal tips to a very small apex radius [7]. Both ac and dc current are used. Tip preparation setup is simple. The following is a general description of etching process. A metal wire (used as tip material), such as W wire, is placed in an electrolyte (1-2N KOH or NaOH) with only a portion of the wire inside the solution. Another electrode (Pt, Au) is also placed inside the electrolyte. The etching process starts when an ac or dc power supply is connected to the two electrodes. The metal

wire is etched until it breaks into two parts. The broken ends are used as tips.

We use electrochemical etching to make STM tips. The setup is shown in Figure 1. We use W wire (0.25 mm diameter) as tip material. And Au is used as the second electrode. A gold wire about 1mm in diameter is bent as a circular shape with diameter 0.5 inch. The W wire is placed in the center of the Au ring. A 2N KOH solution is used as electrolyte. We use KOH solution to rinse the Au ring to form a thin liquid film on the Au ring after installing the W wire. Then a dc power supply is connected to the two electrodes. We use 5-7V voltage which gives a current of 2-3 mA. The etching process takes about 1 hour. In order to get a smooth etched tip, the position of W wire is adjusted to make sure the etching point constant during the course of etching. The resultant W tips are examined with a scanning electron microscopy (SEM), as shown in Figure 2.

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Figure 1.1. Schematic of tip preparation set-up

Figure 1.2. Schematic of aqueous KOH solution film and gold ring

(1)

(2)

Figure 3. SEM pictures of a W tip with different magnifications

(3)

(4)

Figure 3. cont.

APPENDIX III

THE MAJOR STM DATABASE

Table 1. STM data

File Name	Flux(ML/Min)	Coverage (ML)	Cluster Density x 10 ⁻⁵ (Å ²)	Time Length (min)	
#040793	1.8	0.03	1.5	n/a	
#0412	1.98	0.23	5.6	n/a	
#0412a	2.1	0.35	6	n/a	
#0413	4.8	0.16	6.5	n/a	
#0429	1.6	0.16	5	n/a	
#0503	2.8	0.23	7.4	n/a	
#0823	n/a	n/a		600	steps
#0828	1.68	0.28	1.1	192	high vac
#0831	7.2	0.65		1200	
#0901	9	1.4		1361	2nd evp
#0908	0.18	0.015	0.99	1244	
#0913	0.24	0.007	0.26	1540	
#1221	n/a	n/a		524	steps
#1224	0.56	0.228		300	
#1224a	0.56	1.12		100	
#1227	1.08	0.09	3.1	1705	
#1229	1.08	0.09		396	from 1227
#1230	1.08	0.09		330	from 1227
#010794	0.23	0.015	2	200	
#0110	0.6	0.09	0.52	500	
#0519	n/a	n/a		200	steps
#0528	0.36	0.36	2.2	708	
#0531	0.36	0.9	n/a	1100	
#0602	0.3	0.68	n/a	180	
#0602a	0.3	0.95	n/a	990	
#0603	0.3	1.8	n/a	200	
#0606	0.3	2.2	n/a	30	

Table 1. cont.

#0607	0.3	3.5	n/a	300	
#0608	0.3	0.36	n/a	300	
#0609	0.3	1.1	n/a	1320	
#0610	0.3	8.8	n/a	50	
#0614	1.26	0.57	n/a	110	
#0615	1.26	0.28	n/a	156	
#0617	1.26	1.2	5.9	354	
#0619a	1.26	0.89	n/a	300	
#0620			n/a	n/a	3 times evp
#0621		0.75	n/a	n/a	two layers
#0707	0.28	0.07	2.6	550	
#0711	5.4	0.27	6.9	600	
#0712	0.48	0.05	5.4	1300	
#0715	0.48	0.03	3.8	340	
#032095	0.11	0.69	n/a		
#0322	0.11	0.68	n/a		

APPENDIX IV

SUMMARY OF VIBRATION ISOLATION MEASUREMENTS AND ESTIMATE OF VIBRATIONAL NOISE REACHING THE TIP-SAMPLE JUNCTION

1. Introduction

In order to resolve atoms using scanning tunneling microscopy (STM), it is necessary to minimize vibrational displacements between the scanning tip and the sample surface. Because atomic corrugations on metal surfaces are typically on the order of $0.1 \text{ \AA}$, vibrational amplitude displacements between the tip and the surface should be less than $0.01 \text{ \AA}$. Thus *the vibration goal of the system is on the order of 1 pm or 0.01 \AA* along the tip axis. For our instrument, this corresponds to the horizontal direction. Interatomic spacings are in general larger than the corrugations, therefore the demands upon the vibration isolation in the direction perpendicular to the tip axis (vertical direction for our instrument) are relaxed. Vibration isolation is especially challenging when STM is carried out under ultra-high-vacuum (UHV) conditions.

This report is intended to provide a functional description of methods used for vibration isolation in UHV-STM work. For a rigorous discussion and presentation of the theory of vibration isolation, the reader is referred to refs. 1-6. The vibration findings for the STM laboratory are summarized and presented here. In addition, methods of predicting the

vibration amplitudes transmitted to the STM junction are presented here.

Two major factors contribute to the stability of the tip-sample junction. The first is the rigidity of the STM scanner-sample holder assembly itself; a rigid system transmits smaller vibration amplitudes than does a soft system. (In other words, a greater force is required to deform a rigid assembly.) Such rigidity is built into the STM design and for commercial systems is generally beyond the control of the user. The second factor to be considered is the reduction of environmental vibrations reaching the STM stage. It is this aspect of the vibration isolation system over which the user has considerable control, and is therefore emphasized in this report.

Fig. 1. is a schematic diagram illustrating the principal components of a vibration isolation system for an STM. In Fig. 1a, a single-stage vibration isolation system is employed, while Fig. 1b depicts a 2-stage vibration isolation system. In this figure, a single stage of the vibration isolation system is represented by a damped spring. In practice, the actual vibration isolation stages utilized vary considerably from system to system, but generally consist of one (or more) of the following:

- (a) a series of viton pads
- (b) bungee cord suspension of the STM
- (c) a soft spring suspension of the STM stage with eddy current damping
- (d) pneumatic vibration isolation legs

Each of these vibration isolation methods may be characterized by a resonance frequency, μ_0 , and a damping ratio, ξ .

Figure 1. (a) STM assembly with a single stage of damped vibration isolation. (b) STM assembly with two stages of vibration isolation. Each stage of the vibration isolation system may be characterized by its resonance frequency and its damping coefficient.

2. External Vibrations:

The primary sources of vibration external to the STM stage are:

- (i) Building vibrations
- (ii) Pumps vibrations
- (iii) Chamber vibrations

Buildings generally vibrate at frequencies from 1 - 100 Hz [3-5]. Lower floors exhibit vibrations in the 1-10 Hz range while upper floors have resonance frequencies in the 15-25 Hz range. In addition, ventilation ducts, motors, and transformers give rise to vibrations at the line voltage frequency and its harmonics [6].

The mechanical pumps on a vacuum chamber can introduce vibrations to the STM, typically at the line frequency and its harmonics (30, 60, 120 Hz). Turbomolecular pumps can introduce high frequency vibrations. In order to minimize pump vibrations, the mechanical and turbomolecular pumps are connected to the chamber via soft bellows. (It is not clear at this time whether bellows provide sufficient isolation.)

The chamber does not in and of itself cause vibrations, but may transmit and even amplify external vibrations. For example, the large viewports may be excited acoustically and oscillations in the tunneling current may be observed. Long lever arms, such as the sample transfer arm and the sample holder may also contribute to low frequency vibrations. To minimize the chamber amplification and transmittance of vibrations, the chamber has been constructed of heavyweight stainless steel and a support structure for the transfer arm has been designed. (Note that the approach outlined here has been to *increase* the rigidity of

the chamber.)

In Fig. 2, vibrational amplitude displacements are plotted as a function of frequency for our laboratory. Measurements were carried out on the floor, the support table, and the STM using an accelerometer (from Ames Lab Facilities). These data are for vertical displacements; similar measurements were carried out in the horizontal direction and are presented in the appendix. It may be seen that vibration amplitudes on the order of tens of thousands of Ångstroms occur at low frequencies. The data in Fig. 2a were collected with all of the pumps on, while measurements for Fig. 2b were carried out with all pumps turned off.

3. External Vibration Isolation

As mentioned earlier, spring suspension systems, pneumatic legs, bungee cords, and viton pads are common methods of external vibration isolation. Each of these may be considered in terms of a damped harmonic oscillator. (Such a model is appropriate for these systems in the limit displacements where the spring restoration force is linear with displacement [$F = -kx$]. For coil springs, bungee cords, and pneumatic legs this criteria is met for larger displacements than for viton pad vibrations. Nonetheless, for small displacements, such a model is appropriate for viton pads [5].) The equation of motion for a damped harmonic oscillator is given by [4]:

$$m \frac{d^2x}{dt^2} + b \left(\frac{dx}{dt} - \frac{dx'}{dt} \right) + k(x - x') = 0 \quad (1)$$

where m is the mass supported by the spring (the STM assembly), b is the damping factor, k is the spring constant, and x and x' refer to the displacements from equilibrium positions of the mass and the spring support, respectively. Eqn. 1 reduces to Hook's law in the absence of damping ($b=0$). The damping force in Eqn. 1 is proportional to the relative velocities between the spring support and the supported mass, m . These variables are shown in Fig.

3. For a sinusoidal displacement, $x'(t)$, of the spring support, with an amplitude given by x_0' (where $x'(t) = x_0' e^{i2\pi vt}$), the amplitude transmitted by the spring, x_0 , to the supported mass may be referenced to x_0' to yield:

$$\left| \frac{x_0}{x_0'} \right| = \sqrt{\frac{1 + \left(\frac{\xi v}{v_0} \right)^2}{\left(1 - \left(\frac{v}{v_0} \right)^2 \right)^2 + \left(\frac{2\xi v}{v_0} \right)^2}} \quad (2)$$

This function, the amplitude transfer function, is plotted in Fig. 4 for several values of ξ , the damping ratio for the system. The resonance or natural frequency of the spring (also called the eigenfrequency), v_0 , is given by:

$$v_0 = \frac{\sqrt{k/m}}{2\pi} \quad (3)$$

where k is the spring constant and m is the mass. The damping ratio, ξ , is the ratio of the damping coefficient, to the critical damping coefficient:

$$\xi = \frac{\gamma}{\gamma_c} \quad (4)$$

The damping coefficient, γ , and the critical damping coefficient, γ_c , are given by:

$$\gamma = \frac{b}{2m} \quad \text{and} \quad \gamma_c = 4m\pi\nu_0 \quad (5)$$

where, b is the damping factor (from Eqn. 1). (For magnetic damping, $\gamma = 9.69 \times 10^{-9} B l S/\rho$, where B is the magnetic field in Gauss, l is the thickness of the conductor in the direction of B , S is the cross sectional area, and ρ is the resistivity of the conductor.)

In Fig. 4, it may be seen that an undamped spring ($\xi = 0$) offers significant vibration isolation at frequencies above the resonant frequency of the spring. Displacements with characteristic frequencies of greater than 100 Hz are reduced by more than 4 orders of magnitude, while disturbances with frequencies of 10 - 100 Hz result in less than 1% transmission. For disturbances occurring at the spring resonance frequency, amplification of the disturbance occurs. The spring offers no vibration isolation at frequencies below the resonance frequency. The effect of damping the spring is to decrease the amplification of disturbances occurring at or near the natural spring frequency, albeit at the expense of amplitude reduction at higher frequencies. Overdamping of the system considerably reduces the vibration isolation of the spring. It may be seen that at low frequencies (1-100 Hz), vibrational displacements of the laboratory floor of a few thousand Ångströms would result in the transmittance of displacements of more than 10 Å, regardless of damping. From a

Fig. 2. Accelerometer measurements.

Figure 3. Illustration of variables used in Eqn. 1.

design viewpoint, it is desirable to utilize a spring with a resonance frequency far removed from any vibration frequencies and to incorporate only minimal damping. In other words, a very soft spring (small value of k and therefore small resonance frequency, ν_0) with minimal damping should be used. In general, the longer the spring, the lower the resonance frequency [2]:

$$\nu_0 \cong \frac{1}{2\pi} \sqrt{\frac{g}{l}} \quad (6)$$

where l is the relaxed length of the spring and g is the earth acceleration constant. (For example, resonances of 1 and 0.3 Hz would be expected for 25 and 250 cm elongations, respectively. The Omicron STM has 4 springs that are approximately 10 cm in length, with a natural resonance of 1 Hz; the resonance frequency of the Burleigh instrument is 3 Hz while the Park instrument has a double-stage spring system with a resonance frequency of 2 Hz.)

Installation of double-stage vibration isolation systems has been found to provide additional vibration isolation [4-6]. The overall amplitude transfer function for double-stage vibration isolation is given by the product of the two individual suspension systems:

$$\left| \frac{x_0}{x_0'} \right| = \left| \frac{x_1}{x_1'} \right| \times \left| \frac{x_2}{x_2'} \right| = \left| \frac{x_1}{x_1'} \right| \times \left| \frac{x_2}{x_1} \right| = \left| \frac{x_2}{x_1'} \right| \quad (7)$$

where x_1 and x_1' refer to the transmitted displacement and the external displacement, respectively. The displacement passing through the first stage of the vibration isolation system, x_1 , is the source of displacement for the second stage, x_2' , thus $x_1 = x_2'$. This method of determining the overall transmission function of the vibration isolation system is

appropriate for systems using either multiple spring suspension or a vibration isolation system consisting of spring suspension coupled with bungee cords and/or pneumatic legs. (It is also used for a series of stacked viton pads, separated by metal plates [5]) In Fig. 5, the amplitude transfer function for a double-spring suspension system with identical damping and resonance frequencies is shown along with the amplitude transfer function for one of the suspension systems ($\nu_0 = 1$ Hz and $\xi = .10$). Again, it is desirable to minimize the overall resonance frequency of the suspension system and to use minimal damping. Utilization of springs with slightly different resonance frequencies reduces the amplification of the system for disturbances at and near resonances and therefore allows smaller damping ratios to be utilized.

4. The rigidity of the STM scanner assembly

In section 3, the reduction of vibrations reaching the STM stage (isolation of the STM stage from environmental vibrations) was discussed. In this section, the transfer of vibrations between the tip and sample, caused by vibrations which have reached the STM stage, will be considered. It should be pointed out that, if the STM stage were absolutely rigid, that is, if no displacement between the tip and the sample can occur, then there would be no need for the spring isolation discussed in the previous section. Thus, the rigidity of the STM is of primary concern in eliminating vibrations between the tip and the sample. The rigidity of the STM assembly is characterized by its resonance frequency, ν'_0 , and the quality factor, Q' , of the tip-sample junction. The more rigid the structure, the larger the resonance frequency. The quality factor, Q' , is inversely proportional to the damping ratio

for the rigid body; a larger quality factor is associated with a more rigid assembly.

STM piezo scanners typically have resonance frequencies of 10 - 100 KHz. Tripod scanners generally have lower resonant frequencies than do single-tube scanners. (The Omicron STM tripod scanner assembly has a resonance frequency of 20 KHz and that of the Park instruments is 15 KHz. The Burleigh instrument utilizes a single-tube scanner resulting in a resonance frequency of 30 KHz.) The overall resonance frequency of the scanner-tip assembly is generally reduced to 1 - 5 KHz. This decrease in rigidity is caused by joints, junctions, supports, the coarse motion stage, and connectors. We have observed a tip-sample resonance of 5 KHz with the Omicron STM.

The amplitude transfer function (also known as the stiffness or, alternatively, the damping transfer function) for a rigid STM assembly is given by [3,4]:

$$\left| \frac{z_o}{z_o'} \right| = 1 - \sqrt{\frac{1 + \left(\frac{v}{v_o' Q'} \right)^2}{\left(1 - \left(\frac{v}{v_o'} \right)^2 \right)^2 + \left(\frac{v}{v_o' Q'} \right)^2}} \quad (8)$$

where z_o' is the amplitude of the vibration of the STM stage (due to external vibrations), z_o is the resultant amplitude displacement of the tip-sample junction, v is the frequency of the z_o displacement, v_o' is the resonance frequency of the tip-sample assembly, and Q' is the quality factor of the tip-sample junction. (This function is for a sinusoidal displacement, $z'(t)$, of the tip-sample junction, with an amplitude given by z_o' , where $z'(t) = z_o' e^{i2\pi vt}$, analogous to the amplitude transfer function for a soft spring, given by Eqn. 2.) The

amplitude transfer function is plotted in Fig. 6, for resonance frequencies of 1 and 5 KHz (and with a typical Q' value of 30 [3,4]). The rigidity of the STM scanner assembly results in very effective isolation from low frequency vibrations. Fig. 6 shows that increasing the rigidity (higher resonance frequency) drastically improves vibration isolation. The curve associated with the 5 KHz resonance is representative of our instrument. Vibrations of several thousand Ångstroms at frequencies of less than 10 Hz result in tip-sample displacements of less than a few hundredths of an Ångstrom.

5. Combination of soft suspension with the rigid STM scanner assembly

It has been shown that soft suspension systems effectively reduce high frequency vibrations, while the rigidity of the STM isolates efficiently against low frequency vibrations. The overall transmission of external vibrations to the tip-sample junction is given by the product of the transmission functions of the soft suspension system and the rigid STM assembly (Eqns. 2 and 8 or for multiple suspension systems, Eqns. 7 and 8):

$$\left| \frac{x_o}{z_o'} \right| = \sqrt{\frac{1 + \left(\frac{\xi v}{v_o} \right)^2}{\left(1 - \left(\frac{v}{v_o} \right)^2 \right)^2 + \left(\frac{2\xi v}{v_o} \right)^2}} \left[1 - \sqrt{\frac{1 + \left(\frac{v}{v_o' Q'} \right)^2}{\left(1 - \left(\frac{v}{v_o'} \right)^2 \right)^2 + \left(\frac{v}{v_o' Q'} \right)^2}} \right] \quad (9)$$

The overall transfer function is shown in Fig. 7 ($v_o = 1$ Hz, $\xi = .10$, $v_o' = 5000$ Hz, and $Q' = 30$) for both a single-spring and double-spring suspension system. For a single-stage vibration isolation system, displacements at the tip-sample junction will be on the

order of $0.01 \text{ \AA}$ for external vibrational amplitudes with characteristic frequencies of less than 1000 Hz (corresponding to the region where the amplitude transfer is less than 10^{-5}). At higher frequencies, between 1000 and 5000 Hz , external vibration amplitudes can not exceed $10 \text{ \AA}$ in order to meet a tip-sample amplitude of less than $0.01 \text{ \AA}$. The addition of a second spring-suspension system significantly relaxes this restriction, reducing the overall amplitude transmission to less than 10^{-8} for the entire frequency range from a few Hz to 10 KHz . At the resonance frequency of the soft spring, the overall amplitude transmission function has a local maximum of approximately 10^{-6} ; thus external vibration amplitudes occurring at or near this frequency must be held to less than $10,000 \text{ \AA}$ to keep the tip-sample displacement amplitude to less than $0.01 \text{ \AA}$.

6. Estimate of actual noise transferred to the tip-sample assembly

The actual amplitude of the tip-sample displacement for our instrument can be estimated from the accelerometer data collected for our laboratory (shown in Fig. 2). A model noise spectrum is constructed by adding the accelerometer data to an arbitrary $1/f$ background [2]. This is shown in Fig. 8. The accelerometer data used for this figure corresponds to that measured on the STM with the pumps running. It should be noted that these measurements were carried out in the vertical direction and thus would lead to tip sample displacements parallel to the sample surface (corresponding to the lateral resolution of the STM). Measurements in the horizontal direction were also carried out and are presented in the appendix. All displacements in the horizontal direction were significantly lower than those in the vertical direction and therefore would be expected to result in a

Fig. 4. Amplitude transmission functions are plotted for a variety of damping ratios. The eigenfrequency, ν_0 , of the spring is 1 Hz.

Fig. 5. Plot of the amplitude transfer functions for double- and single-stage vibration isolation systems.

Fig. 6. Amplitude transfer function for a rigid STM assembly.

Fig. 7. Overall amplitude transfer functions for a rigid STM supported by both single- and double-spring suspension are shown. For comparison, the transfer functions of the components are also plotted.

Fig. 8. Transmitted vibrational amplitudes based upon the measured displacements are plotted as a function of disturbance frequency.

smaller displacement of the tip-sample junction perpendicular to the sample surface (vertical resolution of the STM).

To determine the amplitude of the displacement of the tip-sample junction, the model noise spectrum is multiplied by the global amplitude transfer function (Eqn. 9). The results of this transformation are shown in Fig. 8 for both a single-spring and a double-spring suspension system (as in Fig. 6: $\nu_0 = 1$ Hz, $\xi = .10$, $\nu_0' = 5000$ Hz, and $Q' = 30$). A horizontal line is drawn to show the location of a displacement amplitude of $0.01 \text{ \AA}$; the transmitted amplitude is less than $0.01 \text{ \AA}$ over the entire frequency range for both the single- and double-spring suspension system.

7. Conclusions

Measurements of the vibrational amplitudes in the STM laboratory have been carried out using an accelerometer. These measurements include both horizontal and vertical displacements of the floor, of the support table, and of the STM. Estimates of resultant tip-sample displacements have been calculated for both a single- and double-spring suspension system, which have been used to model the spring suspension system of the Omicron STM and the combination of the Omicron spring suspension system with pneumatic legs, respectively. The results of these calculations indicate that tip-sample vibrations of less than $0.01 \text{ \AA}$ should be realized.

References:

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Appendix

For the following accelerometer measurements, the displacement, x (Å), is related to the acceleration, a (g), by:

$$x(\text{Å}) = (7.02 \times 10^9) a(\text{g})/f^2 \quad \text{and} \quad a(\text{g}) = (1.42 \times 10^{-10}) x(\text{Å}) f^2,$$

where f is the frequency (Hz). The acceleration and frequency may be taken straight from the graphs and plugged into the above equation.

The source of the above equation follows:

$$a(\sin 2\pi ft) = x = d^2 (x \sin 2\pi ft)/dx^2; a(\text{Pk to Pk}) = (2 \pi^2 x f^2);$$

$$a(\text{rms}) = 0.707 \pi^2 x f^2$$