Carrier scattering and localization in nm-thick Al/Ru, Al/Co and Al/Mo superlattices

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Abstract

Thin films and superlattices are widely used in modern technologies. Certain metal superlattices with layer thickness between 1 to 10 nm have interesting magneto transport properties and unique applications in spintronics and data storage. We have studied electrical conductance of Al/Ru, Al/Co and Al/Mo superlattices with layer thickness between 1 to 2 nm. By monitoring the resistance changes during the growth of the superlattice, we are able to observe directly the effects of carrier localization and scattering when a highly disordered interface is being deposited.

We have observed a significant conductance drop when a transition metal layer is deposited on Al, suggesting a strong localization of Al carriers at the newly created interface. When Al is deposited on a transition metal, the rise of conductance with Al thickness is significantly delayed, indicating that carriers from the growing Al layer are filling the localized states at the new interface. No carriers from the transition metal are observed to be localized at the interface. Our measurements indicate the strongest localization occurs at the Ru/Al interface, followed by the Co/Al and Mo/Al interfaces. The Al/Ru, Al/Co and Al/Mo interfaces exhibit much weaker localization. Using Fuchs’ formula, we can also determine the carrier mean free path within each layer.

The abruptness and smoothness of the interfaces are evaluated by atomic force microscopy (AFM), which indicates that surface roughness of our superlattices is within one to two monolayers. Post-growth annealing shows that species intermixing at the interface only happens at temperatures higher than 150 °C.
Chapter 1

Introduction

Thin films play an important role in modern technology. They are widely used as contacts, barriers and plating seeds in semiconductor devices. Recent research has developed sequential deposition of multilayer superlattices. In 1970, Esaki and Tsu explored the transport properties of semiconductor superlattices [1], ushering in applications of semiconductor superlattices in a great variety of devices, such as high-electron-mobility transistors (HEMT) [2], heterojunction lasers [3], quantum cascade lasers [4], type-II superlattice photodetectors [5], etc. Quantum effects in metal superlattices have also been explored to achieve extreme sensitivity in signal detection, such as giant magnetoresistance (GMR) thin film heads [6–8]. Many more quantum mechanical phenomena in nm-thick superlattices still remain to be explored.

Carrier transport in a thin film is affected by scattering at surfaces, interfaces and grain boundaries. Fuchs in 1938 treated the role of surface scattering plays in thin film conductivity [9]. Kuan et al. investigated how surface scattering increases the resistivity in sub-0.1 μm Cu interconnects [10]. Y. P. Timalsima et al. studied how surface roughness contributes to an additional resistivity in Cu thin films with thickness down to ~4 nm [11]. When film thickness shrinks further down to 1 nm or smaller, classical criteria for weak scattering, i.e. \( \lambda_F \ll \Lambda \), where \( \lambda_F \) is Fermi wavelength and \( \Lambda \) is the mean free path, no longer applies, and quantum effects must be considered.

In this study, we set out to investigate the quantum effect involved in carrier transport in nm-thick superlattices. A slow deposition rate is used to grow nm-thick multilayer structures. Since thin films oxidize easily in air, in-situ conductance measurement is carried out during the
growth without breaking the vacuum. We can thus monitor the evolution of conductance in real time during the growth. Three different Al-based superlattices (Al/Ru, Al/Co and Al/Mo) are grown, measured and analyzed. The superlattices’ conductance is also monitored in a post-growth annealing experiment with the samples heating from room temperature to 300 °C and cooled down to room temperature. Atomic force microscopy (AFM) is used to study the roughness of superlattices’ top surfaces.

We will give a brief review in Chapter 2 on the classical Fuchs model and speculate on possible quantum effects on transport property of nm-thick films. We will then simulate the conductance of superlattices in Chapter 3 taking possible quantum effects on carrier localization into account. In Chapter 4, we describe the growth and current-voltage (I-V) measurement procedures used in this study. In Chapters 5-6, the measured data and analyses of three superlattices are presented, followed by conclusions and summaries in Chapter 7.

References


Chapter 2

Electrical conductivity in a nm-thick film

In a bulk metal, electric resistivity is governed mostly by phonon scattering. The dependence of resistivity on temperature is known as Matthiessen’s rule: $\rho_{tot}(T) = \rho_0 + \rho_p(T)$ [1]. The phonon scattering component $\rho_p$ depends on the density of phonons, which is a strong function of temperature. Scattering from impurity and crystal defects produces a residual resistivity, $\rho_0$, which is independent of temperature. Surface scattering is a part of $\rho_0$, which becomes most prominent in a thin film. In a film with thickness much smaller than the phonon mean free path, contribution of surface scattering to resistivity becomes so large that we can ignore the phonon scattering.

2.1 Conductivity in a bulk metal with weak scattering

The kinetic state of conducting electrons in a bulk metal is described by the Boltzmann distribution function $f(\vec{r}, \vec{v}, t)$, which specifies the number of particles in a volume $d\vec{r}d\vec{v}$ at time $t$, where $\vec{r}$ is the space vector and $\vec{v}$ the velocity vector. The effect of collisions with phonons or defects on the distribution function $f(\vec{r}, \vec{v}, t)$ is given by the Boltzmann transport equation:

$$\left(\frac{\partial f}{\partial t}\right)_{\text{coll}} = \frac{\partial f}{\partial t} + \frac{\partial f}{\partial \vec{r}} \frac{\partial \vec{r}}{\partial t} + \frac{\partial f}{\partial \vec{v}} \frac{\partial \vec{v}}{\partial t}. \quad (2.1)$$

The first term $\frac{\partial f}{\partial t}$ on the right side becomes zero when the system reaches equilibrium. The second term describes the diffusive and viscous properties of the electrons, and the third term denotes the effect of an external field.
Assume an electric field \( \vec{E} \) is applied in the \( x \)-direction, causing \( \frac{\partial \vec{v}}{\partial t} = -\frac{e\vec{E}}{m^*} \), where \( m^* \) is the effective mass of the conducting electrons. Suppose the electrons are in a uniform bulk medium, \( \frac{\partial f}{\partial \vec{r}} = 0 \), then the Boltzmann equation (2.1) at equilibrium becomes

\[
\left( \frac{\partial f}{\partial t} \right)_{\text{coll}} = -\frac{eE}{m^*} \frac{\partial f}{\partial v_x}.
\]

Upon the application of an electric field at \( t = 0 \), the distribution function will evolve from \( f_0 \) to a final distribution \( f_0 + f_1 \) exponentially with \( t \):

\[
f(\vec{r}, \vec{v}, t) = f_0(\vec{r}, \vec{v}) + f_1(\vec{r}, \vec{v})(1 - e^{-t/\tau}),
\]

where \( f_0(\vec{r}, \vec{v}) \) is the equilibrium distribution function in the absence of the field. The response of \( f(\vec{r}, \vec{v}, t) \) to the field is shown schematically in Figure 2.1 (curve (a)).

In reality, the effect of electric field saturates very quickly, i.e. \( \tau \) is of the order of microseconds. We can approximate the evolution of \( f \) with time as a linear rise within a time period \( \tau \), as depicted in Figure 2.1 (curve (b)). This approximation is called “relaxation time approximation”. Suppose we switch the electric field off, collisions with phonons or defects would return the system from \( f_0 + f_1 \) back to \( f_0 \) in roughly the same time period \( \tau \). This implies that collisions can bring about a changing rate of \( f \) given by

\[
\left( \frac{\partial f}{\partial t} \right)_{\text{coll}} = -\frac{f_1}{\tau}.
\]

If we consider the Boltzmann distribution \( f_0 \) in \( k \)-space at a low temperature, all electrons are located inside the Fermi surface, \( v \leq v_F \), where \( v_F \) is the Fermi velocity, and only electrons at the Fermi surface are allowed to be scattered. When an electric field is switched on, the distribution will evolve to \( f_0 + f_1 \) in a time period \( \tau \), and the Fermi surface will drift by an
Figure 2.1. The response of the distribution function $f$ to an electric field switched on at time $t = 0$ (curve (a)). In the relaxation time approximation, the full response is achieved linearly in a time period $\tau$ (curve (b)).

amount $\delta k$ in the direction against the field as shown in Figure 2.2.

According to the relaxation time approximation (2.4), and (2.2) with $\hbar \partial k = m^* \partial v$, we have

$$\frac{-f_1}{\tau} = -\frac{eE}{\hbar} \frac{\partial f}{\partial k}.$$  \hspace{2cm} (2.5)

Therefore,

$$\delta k = \frac{eE\tau}{\hbar}. \hspace{2cm} (2.6)$$

The average drifting velocity $v_d$ associated with $\delta k$ is

$$v_d = \frac{\hbar \delta k}{m^*} = \frac{eE\tau}{m^*}. \hspace{2cm} (2.7)$$
Figure 2.2. The drift of Fermi sphere in an electric field $\vec{E}$ in a time period $\tau$.

The conductivity $\sigma$ resulting from the drift velocity (2.7) is

$$\sigma = \frac{J}{E} = \frac{n e v_d}{E} = \frac{n e^2 \tau}{m^*}, \quad (2.8)$$

where $J$ is current density and $n$ the density of electrons. This expression of conductivity (2.8) is valid only in a weak scattering condition, i.e. $\delta k \ll k_F$, to fulfil the relaxation time approximation. In real space this means $\lambda_F \ll \Lambda$, where $\lambda_F$ is the Fermi wavelength and $\Lambda$ the mean free path of electrons.

2.2 Conductivity in a thin film — classical Fuchs model

In a thin film with large grains and with a thickness smaller than the phonon mean free path $\Lambda_p$, surface scattering will be more frequent than the phonon scattering. Surface scattering
becomes the major source of electrical resistivity. Since the surface scattering and phonon scattering occur in sequence, their resistivities are added on each other:

\[ \rho_f = \rho_p + \rho_s, \quad (2.9) \]

or

\[ \frac{1}{\sigma_f} = \frac{1}{\sigma_p} + \frac{1}{\sigma_s}. \quad (2.10) \]

Surface scattering can be either specular or diffuse as shown in Figure 2.3. Specular scattering does not change the drift velocity component in the field direction and does not contribute to the resistivity. On the other hand, diffuse scattering at random angles can on average reduce the drift velocity component in the field direction to zero and contribute to the resistivity.

In 1938, Fuchs calculated the contribution of surface scattering to the film resistivity [2]. His model considers a thin metallic film with its surfaces perpendicular to the z-axis and an electric field \( \vec{E} \) applied in the x-direction, as shown in Figure 2.4. The distribution function \( f \) is uniform in the x- and y-direction, but not in the z-direction due to surface scattering occurring at \( z = 0 \) and \( z = t \), where \( t \) is a film thickness. According to the relaxation time approximation (2.5), the Boltzmann equation at equilibrium from (2.1) and (2.2) becomes

Figure 2.3. (a) A specular scattering and (b) a diffuse scattering at the surface.
Figure 2.4. Carrier scattering in a thin metallic film in an electric field.

\[- \frac{f_1}{\tau} = v_z \frac{\partial f}{\partial z} - \frac{eE}{m^*} \frac{\partial f}{\partial v_x}. \tag{2.11} \]

The general solution of (2.11) is

\[ f_1(\bar{v}, z) = \frac{eE\tau}{m^* v_x} \left[ 1 + \phi(\bar{v}) e^{-z/\tau v_x} \right]. \tag{2.12} \]

Assume that a portion \( p \) of electrons is scattered specularly at the surface, the total number of electrons leaving the surface is the same as those moving towards the surface, and the two surfaces at \( z = 0 \) and \( z = t \) are identical,

\[ \phi(v_z) = -\frac{1-p}{1-p e^{-t/\tau v_z}} \quad \text{for} \quad v_z > 0 \tag{2.13} \]

\[ \phi(-v_z) = -\frac{1-p}{1-p e^{t/\tau v_z}} \quad \text{for} \quad v_z < 0. \tag{2.14} \]

Since from (2.12), the carrier density change \( f_1 \) is smaller closer to surface, the drift velocity is also smaller next to a surface due to more frequent collisions. The drift velocity in the \( x \)-direction as a function of location \( z \) can be derived by
\[ \nu_d(z) = \frac{\int v_x f_1 d\vec{v}}{\int f_0 d\vec{v}}. \]  (2.15)

The drift velocity at different location \( z \) normalized by the bulk \( \nu_d \) in (2.7) is

\[ \frac{\nu_d(z)}{\nu_d(\text{bulk})} = 1 - \frac{3}{2} \int_0^\pi \sin^3 \theta \left( \frac{1-p}{1-p} e^{-z/\Lambda_p \cos \theta} \right)^\frac{(1-p)}{1-p} e^{-t/\Lambda_p \cos \theta} d\theta, \]  (2.16)

where \( \Lambda_p \) is the phonon mean free path and \( \Lambda_p \cos \theta = \tau v_z \). The normalized \( \nu_d(z) \) is plotted in Figure 2.5 for 1-nm-thick and 2-nm-thick films. Here it is seen that \( \nu_d(z) \) reduces to a lower value closer to a surface at \( z = 0 \) or \( z = t \). The thinner the film the bigger percentage of the population is affected, resulting in a smaller conductivity in the thinner films.

To get the conductivity along the \( x \)-direction,

\[ j_x(z) = e \int f_1 v_x(z) d\vec{v}. \]  (2.17)

Then the mean current density \( \bar{j_f} \) is calculated by integrating \( z \) value from 0 to \( t \),

\[ \bar{j_f} = \frac{1}{t} \int_0^t j_x(z) dz \]

\[ \bar{j_f} = \frac{4\pi e^2 E \tau}{3 m^*} \int \frac{\partial f_0}{\partial v} v^3 dv \left[ 1 - \frac{3\Lambda_p}{2t} \int_0^\pi \sin^3 \theta \cos \theta \left( \frac{1-p}{1-p} \left( 1-e^{-t/\Lambda_p \cos \theta} \right) \right) d\theta \right]. \]  (2.18)

For a bulk metal (\( t \to \infty \)), the second term in bracket of (2.18) reduces to zero and

\[ \bar{j_p} = \frac{4\pi e^2 E \tau}{3 m^*} \int \frac{\partial f_0}{\partial v} v^3 dv. \]  (2.19)

Since \( \bar{j} = \sigma E \), where \( \sigma \) is the conductivity, the ratio of the average conductivity in the thin film \( \bar{\sigma}_f \) to that in the bulk \( \sigma_p \) can be expressed as
Figure 2.5. Normalized drift velocity as a function of location $z$ for different $\Lambda$ in a thin film with thickness (a) 1 nm and (b) 2 nm. The average drift velocity throughout the film is given by the dashed lines.

\[
\frac{\overline{\sigma}_f}{\overline{\sigma}_p} = 1 - \frac{3\Lambda_p}{2t} \int_0^{\pi} \sin^3 \theta \cos \theta \frac{(1-p)(1-e^{-t/\Lambda_p \cos \theta})}{1-p e^{-t/\Lambda_p \cos \theta}} d\theta. \quad (2.20)
\]

Sondheimer in 1952 suggested the following approximations for this equation (2.20) [3]:

...
(1) For $t \ll \Lambda_p$,

$$\frac{\bar{\sigma}_f}{\sigma_p} = \frac{4}{3} \frac{1+p}{1-p} \frac{1}{\log\left(\frac{\Lambda_p t}{t}\right)}$$

(2.21)

(2) For $t \gg \Lambda_p$,

$$\frac{\bar{\sigma}_f}{\sigma_p} = 1 - \frac{3\Lambda_p}{8t} (1 - p).$$

(2.22)

The Fuchs model (2.20) and the Sondheimer approximations (2.21) and (2.22) for $p = 0$ and $\Lambda_p = 10$ nm are shown in Figure 2.6. Neither of the Sondheimer approximations is accurate for film thickness in the range of 1 to 10 nm. Therefore, the actual Fuchs integration (2.20) must be used for nm-thick films.

![Figure 2.6. The Fuchs model (2.20) (solid line) and the Sondheimer approximations (dashed lines) at $p = 0$. Neither approximation is satisfactory for 1 – 10 nm thickness range.](image)
2.3 Modified Fuchs model for a fine-grain thin film

In a polycrystalline film with both thickness and grain size smaller than the phonon mean free path, grain boundary scattering and surface scattering are equally dominant, and phonon scattering can be ignored. The Fuchs model needs to be modified by replacing $\sigma_p$ with a smaller $\sigma_g$ and replacing the phonon mean free path $\Lambda_p$ by the mean free path for grain boundary scattering $\Lambda_g$:

$$\frac{\bar{\sigma}_f}{\sigma_g} = 1 - \frac{3\Lambda_g}{2t} \int_0^{\pi} \sin^3 \theta \cos \theta \left( \frac{1-p}{1-p e^{-t/\Lambda_g \cos \theta}} \right) \, d\theta. \quad (2.23)$$

The average conductivities with different $\Lambda_g$ and $p$ are plotted in Figure 2.7 and Figure 2.8, respectively. The decreases in $\bar{\sigma}_f$ (the size effect) become prominent when the thickness is equal

![Figure 2.7. Sensitivity of thin film conductivity to $\Lambda_g$.](image-url)
or smaller than the mean free path $\Lambda_g$. It is noticed that both $\Lambda_g$ and $p$ can affect thin film conductivity at small thickness. Films with a larger $\Lambda_g$ or a smaller $p$ would experience a bigger size effect.

### 2.4 Effective surface scattering parameters

When a thin film is deposited on a substrate, the interface with the substrate can have a scattering parameter ($q$) different from that of the upper film surface ($p$), as shown in Figure 2.9. Lucas in 1964 has discussed this case and suggested a modification to the Fuchs model (2.23) [6]:

$$\frac{\bar{\sigma}_f}{\sigma_g} = 1 - \frac{3}{4\kappa} \int_0^\pi \sin^3 \theta \cos \theta \left( \frac{1-e^{-t/\Lambda_g \cos \theta}}{1-pq e^{-2t/\Lambda_g \cos \theta}} \right) \left[ 2 - p - q + (p + q - 2pq) e^{-t/\Lambda_g \cos \theta} \right] d\theta \quad (2.24)$$
Instead of using (2.24) with two parameters, we can use an effective \( p' \) applying to (2.23) and produce the same amount of surface scattering effect (Figure 2.10). A contour map of the effective parameter \( p' \) as a function of \( p \) and \( q \) is shown in Figure 2.11.

![Diagram of a thin film on a substrate with different scattering parameters](image)

**Figure 2.9.** A thin film on a substrate, with different scattering parameters for the upper surface and the bottom interface with the substrate.

![Graph showing the conductivity](image)

**Figure 2.10.** An effective scattering parameter \( p' = 0.22 \) produces the same conductivity in a film with upper parameter \( p = 0.5 \) and lower parameter \( q = 0 \) at \( \Lambda = 10 \) nm.
Figure 2.11. The contour map of effective $p'$ as a function of $p$ and $q$ at $\Lambda = 10$ nm.

2.5 Quantum effects on carrier transport in a nm-thick film

A. Diffuse scattering ($p = 0$) at a surface or grain boundary

If we assume that Bloch waves of conducting electrons in a fine grain film take a form of a wave packet with a phase coherence length comparable to grain size ($\sim 1$ nm) or film thickness ($\sim 1$ nm), then scattering at the surface or interface would involve diffraction from no more than a few atoms. Given the irregularity $\Delta t \gtrsim \lambda_F$ at a surface with steps, or at an interface with lattice mismatch, it is unlikely that specular scattering can ever occur. Therefore, a thin film with fine grains would favor diffuse scattering ($p = 0$). Furthermore, with mean free path $\Lambda_g$ of $\sim 1$ nm approaching $\lambda_F$ (0.36 nm for Al), we are entering a regime of strong scattering and (2.8) will no longer be valid.
B. Carrier localization from chemical bonds and from disorder scattering at the interface

When a transition metal is deposited on a simple metal surface, and if compound phases of the two metals exist, localized “covalent-like” bonds can form at the interface. As shown in Figure 2.12 (a), an adatom can land either on a flat terrace, at a surface vacancy or a surface step, resulting in different local stoichiometries. Stoichiometry determines the number of local bonds. As the surface coverage increases, overlapping of localized orbitals forms an energy band, as shown Figure 2.12 (b). This band of localized states is expected between the Fermi energies of the two metals. Furthermore, the total number of localized states should be proportional to the surface coverage and becomes saturated when the deposition of one monolayer is reached.

A new interface is formed when one monolayer of transition metal is deposited. Besides chemical bonds, the 2D interface can contain a high density of structural disorder, such as vacancies, steps, misfit dislocations, grain boundaries, etc., as shown in Figure 2.13. Scattering among these defects can produce additional localized states (Anderson localization [7]) at the interface.

In a superlattice consisting of layers of transition and simple metals, highly disordered interfaces with localized bonds can trap carriers. Effect of carrier localization can be comparable or even bigger than the effect of scattering at the interface. As will be discussed in Chapter 3, 2D interfaces can localize carriers from the simple metal layer until all localized states are filled. By monitoring the conductance drop associated with the creation of a new interface during growth, we can quantitatively measure the amounts of carriers localized as compared to those that are still conducting.
Figure 2.12. (a) Deposition of transition metal atoms on a simple metal surface. (b) Localized states due to chemical bonds.

Figure 2.13. Structural disorder at an interface.
References


Chapter 3

Computer simulation of conductance in a multilayer structure

The classical Fuchs model expresses how surface scattering reduces the conductance in a single metal layer. In this chapter we will extend the model to simulate the conductance evolution during the growth of a multilayer structure. In simulating nm-thick multilayers, quantum effects such as carrier localization are also included. Both interface scattering and carrier localization reduce the conductance. Simulations can show how these two effects bring about the conductance change when an interface is introduced during the growth.

3.1 Simulation of a single layer growth

We divide the growth of a layer into $n$ steps in the simulation. The growth of these successive steps (~0.1 nm) is supposed to be continuous, and no interface is created between the steps. The evolution of average conductivity $\bar{\sigma}_f$ and total conductance $G$ during the growth is given by the Fuchs model, assuming 100% diffuse scattering at the interface:

$$\bar{\sigma}_f(t) = \sigma_g [1 - \frac{3}{2\kappa} \int_0^\pi \sin^3 \theta \cos \theta (1 - e^{-t/\Lambda_g \cos \theta}) d\theta],$$  \hspace{1cm} (3.1)

and

$$G(t) = \bar{\sigma}_f(t)t.$$  \hspace{1cm} (3.2)

We can also calculate the differential conductance $\frac{dG}{dt}$, which represents the instantaneous conductivity $\sigma_f(t)$ during the growth.
Figure 3.1 shows normalized average conductivity $\bar{\sigma}_{f}(t)$, conductance $G(t)$ and differential conductance $\frac{dG}{dt}$ for $p = 0$ with different $\Lambda_g$. Here it is seen that the parameter $\Lambda_g$ has a big effect on transport properties. The size effect becomes prominent when the thickness is smaller than $\Lambda_g$. At a fixed thickness, e.g. 1 nm, the ratios to $\sigma_g$ of the average conductivity $\bar{\sigma}_{f}(t)$, the conductance $G(t)$ and the differential conductance $\frac{dG}{dt}$ all decrease significantly with increasing $\Lambda_g$.

3.2 Effect of carrier transmission between layers

For horizontal transport with $\vec{E}$ parallel to the film surface, a multilayer superlattice
behaves as a set of parallel resistors. If no carrier transmission across the interfaces is allowed, total conductance will be just the sum of the contributions from all layers:

\[ G = \sum_i \sigma_f(t_i)t_i, \]  

where \( \sigma_f(t) \) is given by (3.1).

If carriers are allowed to transmit into neighboring layers, those that cross the interface will have a longer mean free path and contribute more to the conductance, since to them the effective layer thickness \( t' \) is larger than a single layer thickness, as shown in Figure 3.2. The increase in effective layer thickness depends on the number of interfaces crossed. The fraction \( f \) of transmitted carriers and effective thickness \( t' \) in a two-layer structure are listed in Table 3-1.

The total \( G \) of a two-layer structure with carrier transmission becomes

\[ G = t_1[(1 - T_1)\sigma_f(t_1) + T_1\sigma_f(t_1 + t_2)] + t_2[(1 - T_2)\sigma_f(t_2) + T_2\sigma_f(t_1 + t_2)]. \]  

(3.4)

To demonstrate the effect of carrier transmission, the conductance of a two-layer structure with and without carrier transmission is calculated with \( p = 0, \Lambda_g = 1 \text{ nm} \) and \( t = 1 \text{ nm} \), and it is plotted in Figure 3.3. Comparing a transmission case \( (T_1 = T_2 = 0.5) \) and a non-transmission case \( (T_1 = T_2 = 0) \), the overall conductance increase due to carrier transmission is

---

**Figure 3.2. Carrier transmission in a two-layer structure.**
Table 3-1. Fraction of transmission and effective thickness in a two-layer structure.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Fraction $f$</th>
<th>Effective Thickness $t'$</th>
<th>Layer thickness $t$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2)</td>
<td>$1 - T_2$</td>
<td>$t_2$</td>
<td>$t_2$</td>
</tr>
<tr>
<td></td>
<td>$T_2$</td>
<td>$t_2 + t_1$</td>
<td>$t_2$</td>
</tr>
<tr>
<td>(1)</td>
<td>$1 - T_1$</td>
<td>$t_1$</td>
<td>$t_1$</td>
</tr>
<tr>
<td></td>
<td>$T_1$</td>
<td>$t_1 + t_2$</td>
<td>$t_1$</td>
</tr>
</tbody>
</table>

small (< 8%). The effect of carrier transmission can be larger for a larger $\Lambda_g$. In polycrystalline metal films, the $\Lambda_g$ is comparable to grain size, which is often no more than the film thickness. The simulation in Figure 3.3 thus reflects the realistic situation. We can therefore choose to ignore the complicated carrier transmission issue and accept the error of ~10% in multilayer conductance calculations. We will not consider the carrier transmission in our analyses of measured results in Chapter 6.

3.3 Simulation of carrier localization at an interface

When a new layer is grown on a substrate, the 2D interface can contain a high density of scattering centers (see Figure 2.13). At the interface, covalent-like chemical bonds can also form. Both disorder scattering and chemical bonds can create localized states and trap carriers from adjacent layers. The filling of localized states produces a drop in conductance. Since we are simulating the evolution of conductance during the superlattice growth, we can implement this localization-induced conductance drop as soon as a new transition metal layer is being
Figure 3.3. Conductance of a two-layer structure with layer thickness 1 nm, simulated with $p = 0$ and $\Lambda_g = 1$ nm. The dashed line is calculated with no carrier transmission between layers and the solid line is calculated with transmission $T = 0.5$.

deposited, as shown in Figure 3.4 (interface $(A)$). Free carriers from the layer underneath the new interface will fill instantly the localized states. The resultant conductance drop can be simulated by

$$-\Delta G \left( 1 - e^{-t/\ell^* (A)} \right),$$

(3.5)

where $t$ is the thickness of the new transition metal layer and $\ell^* (A)$ is a thickness constant. Here it is assumed that all localized states will be filled by existing free carriers instantly and the conductance drop $\Delta G$ will be saturated exponentially with a small thickness constant $\ell^* (A)$. Further growth of the transition metal layer will not inflict any more carrier localization. The evolution of conductance and differential conductance during the growth of the transition metal
Figure 3.4. Carrier localization can occur at a new interface formed upon the deposition of a new transition metal layer (interface (A)), or a new simple metal layer (interface (B)).

The size of $\Delta G$ reflects the total number of localized states filled at the interface. The localization produces a negative $\frac{dG}{dt}$ at the initial growth $t < \sim 2t_{(A)}^*$, and in subsequent growth the conductance drop $\Delta G$ stays intact.

When the transition metal layer growth finishes and a new layer of simple metal starts, a new interface with chemical bonds and disorder is formed, as shown in Figure 3.4 (interface (B)). Similar localized states are created at the new interface, but the filling of localized states will be different from the interface (A). When the interface (A) is created there are ample carriers from the underneath simple metal layer to fill the empty states. On the other hand, if we assume that carriers from the transition metal layer cannot fill the localized states, the filling of the interface (B) will rely on the carriers from the new simple metal layer above the interface (B). The filling of localized states in the interface (B) will be slower, and the conductance drops more gradual and commensurate to the available carriers in the simple metal layer being grown.

The amount of conductance drop $\Delta G$ vs. new layer thickness is complicated, but our measurements (Chapter 5 and 6) show that the function
Figure 3.5. Evolution of (a) conductance and (b) differential conductance during a transition metal layer growth. The solid lines demonstrate the effect of carrier localization at the interface formed at \( t = 0.2 \text{ nm} \).
\[-\Delta G \left[ \text{erf} \left( \frac{t}{t^*_{(B)}} \right) \right]^2 \] (3.6)

can describe our observed drop most accurately.

The evolution of \( G \) and \( \frac{dG}{dt} \) during the growth of the simple metal layer under the influence of localization at the interface \((B)\) is shown in Figure 3.6, which shows a much slower rise in \( G \) and no negative \( \frac{dG}{dt} \) at the small thickness.

### 3.4 Simulation of multilayer growth

Combining the effects of carrier scattering (the Fuchs model section 3.1) and carrier localization at the interfaces (section 3.3), we can simulate the conductance evolution in a ABAB superlattice, consisting of transition metal \( A \) layers and simple metal \( B \) layers, using

\[
G_A = \bar{\sigma}_f(\sigma_g, \Lambda_g) t - \Delta G \left( 1 - e^{-t/t^*_{(A)}} \right)
\]

\[
G_B = \bar{\sigma}_f(\sigma_g, \Lambda_g) t - \Delta G \left[ \text{erf} \left( \frac{t}{t^*_{(B)}} \right) \right]^2.
\] (3.7)

Here \( \bar{\sigma}_f \) is sensitive to \( \sigma_g \) and \( \Lambda_g \), and \( \Delta G \) depends on the strength of localization at the interface. Figure 3.7 shows a simulated conductance evolution in a nm-thick superlattice. We assume \( A \) and \( B \) layers have the same \( \sigma_g \), and we also set \( \Lambda_g = 5 \) nm for all layers. The instant fill interface \((A)\) has \( t^*_{(A)} = 0.1 \) nm and slow fill interface \((B)\) has \( t^*_{(B)} = 1.2 \) nm. Here it is seen that a small \( t^* \) brings about a sharp drop in conductance and a larger \( t^* \) a more gradual drop. The two interfaces are chemically the same, small \( t^* \) occurs when the interface is filled by the simple metal layers underneath, while big \( t^* \) occurs when the interface is back filled by the simple metal layer above the interface.
Figure 3.6. Evolution of (a) conductance and (b) differential conductance during a simple metal layer growth. The solid lines demonstrate the effect of carrier localization at the new interface underneath the simple metal layer.
Figure 3.7. Evolution of (a) conductance and (b) differential conductance during multilayer growth. The solid lines show the effect of carrier localization, and the dashed lines are without localization.
Chapter 4

Growth of Al-based superlattices and in-situ conductance measurement

4.1 Thin film deposition and calibration of deposition rate

In this study, thin film superlattices are deposited using a DC magnetron. In this process, an Ar plasma is generated in a low pressure ambient Ar gas by applying a potential between the electrodes, as shown in Figure 4.1. An electric field created by the potential accelerates the energetic Ar ions in the plasma towards the cathode target and they collide with the target atoms. Some target atoms are dislodged from the surface and if they are ionized, are then accelerated toward and condensed onto the anode substrate surface.

Figure 4.1. A simple DC diode discharge setup.
Figure 4.2. Magnetron cathode configuration and $\vec{E} \times \vec{B}$ electron drift paths created by $\vec{E}$ and $\vec{B}$ field.

In a DC magnetron an array of magnets is set behind the cathode, which produces a magnetic field perpendicular to the electric field, with the $\vec{E} \times \vec{B}$ electron drift paths configured into a circular, closed loop parallel to cathode surface [1] (see Figure 4.2). The secondary electrons from the target will also join in the cycloid motion. The magnetic field in effect increases the rate of collision between electrons and Ar gas and traps the plasma near the cathode.

To deposit nm-thick films, we need a low deposition rate. Deposition rate is usually linear with the power input to the DC magnetron [2]. To calibrate the deposition rate, the thickness of deposited film can be measured by the atomic force microscope (AFM) or cross-sectional transmission electron microscopy (XTEM). In this study, ex-situ AFM is used to measure the deposited thickness. To prepare calibration samples, a mask is used to cover a half of the substrate. After deposition, the mask is removed, and the step-height of the film is measured by AFM. Figure 4.3 shows a step profile measured by AFM. More than 10 measurements are carried out at different step locations to ensure accuracy. Table 4-1
summarizes the measured deposition rates at a 14.5 cm throw distance for different targets, all 5 cm in diameter, under the deposition conditions used in this study.

![Graph showing deposition rate](image)

**Figure 4.3.** A step-height of Al film measured by ex-situ AFM for calibrating the deposition rate. The step profile shows an Al film deposited on a SiO$_2$/Si substrate for 10 minutes at 200 mA/79 W.

**Table 4-1.** Deposition conditions and rates for 5-cm-diameter metal targets at a 14.5 cm throw distance.

<table>
<thead>
<tr>
<th>Material</th>
<th>Conditions mA/V/W</th>
<th>Calibration deposition time (min)</th>
<th>Deposition rate (nm/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ta</td>
<td>200/230/46</td>
<td>10</td>
<td>3.4 ± 0.1</td>
</tr>
<tr>
<td>Ru</td>
<td>200/330/66</td>
<td>10</td>
<td>3.6 ± 0.1</td>
</tr>
<tr>
<td>Mo</td>
<td>200/280/56</td>
<td>10</td>
<td>3.9 ± 0.2</td>
</tr>
<tr>
<td>Co</td>
<td>200/315/63</td>
<td>10</td>
<td>2.3 ± 0.1</td>
</tr>
<tr>
<td>Al</td>
<td>200/430/79</td>
<td>10</td>
<td>3.9 ± 0.2</td>
</tr>
</tbody>
</table>
In this work, we grow Al/transition metal superlattices with Al thickness shown in Figure 4.4, and thickness of transition metals listed in Table 4-2. Substrates used in this study are 725-μm-thick Si wafers coated with a 2.4-μm-thick thermal oxide (SiO$_2$). We measure the resistance during the growth using a procedure described in the next section.

### 4.2 In-situ resistance measurements

To carry out I-V measurement while the film is being grown, we deposit four Ta contact

<table>
<thead>
<tr>
<th>Transition metal</th>
<th>Ru</th>
<th>Co</th>
<th>Mo</th>
</tr>
</thead>
<tbody>
<tr>
<td>TM (1)</td>
<td>1.2</td>
<td>1.5</td>
<td>1.8</td>
</tr>
<tr>
<td>TM (3)</td>
<td>1.3</td>
<td>0.8</td>
<td>2.2</td>
</tr>
<tr>
<td>TM (5)</td>
<td>1.3</td>
<td>0.8</td>
<td>2.2</td>
</tr>
<tr>
<td>TM (7)</td>
<td>0.8</td>
<td>2.2</td>
<td></td>
</tr>
</tbody>
</table>
pads on a 2.9 cm by 2.9 cm SiO$_2$/Si substrate. Their size is 0.5 cm in diameter and ~140 nm in thickness, as shown in Figure 4.5. The square substrate with Ta contact pads is mounted on a heating stage (Figure 4.6 (b)), and four I-V leads are affixed to the Ta contact pads (Figure 4.6 (c)). A circular PVD aperture, 1.8 cm in diameter, is mounted in front of the substrate to define the deposited film area partially overlapping with the Ta pads (Figure 4.5 and Figure 4.6 (d)). The PVD chamber is then pumped down to the pressure of ~5 × 10$^{-7}$ Torr.

We use two different methods to measure the resistance: 4-point and 2-point measurements. The 4-point probe method, as shown in Figure 4.7(a), monitors the relative resistance during the growth. The 2-point probe method, as shown in Figure 4.7(b), measures the absolute value of film resistance.

The resistance of the deposited film is measured in-situ automatically once every 2 seconds. Film growth is interrupted periodically with the incoming beam blocked by a shutter in front of the gun, so that I-V reading can be recorded clear of any interference from the charges in the beam. The resistance of a circular-shaped film during the deposition is approximately [4]:

Figure 4.5. Schematic of a 2.9 cm by 2.9 cm SiO$_2$/Si substrate with four Ta contact pads on top. The dashed circle is the deposited area of the test film defined by an aperture.
Figure 4.6. (a) Schematic of magnetron, substrate stage, contact leads and aperture, (b) a square substrate with pre-deposited Ta contact pads mounted on the heating stage, (c) isolated I-V leads attached to contact pads, and (d) circular PVD aperture [3].

\[ R \approx 1.4 \frac{\rho}{t}. \]  \hspace{1cm} (4.1)

where \( \rho \) is the resistivity of the film.

The superlattice is deposited and measured at room temperature. After the superlattice growth and transport measurement are completed, the superlattice is heated to 300 °C with the resistance monitored during the heating and cooling periods without breaking the vacuum. The 25 Ω heating coil is powered by an external source.

4.3 Characterization of surface roughness

In this study, an XE-100 AFM is used to probe the surface roughness of the superlattices after the resistance measurement and annealing experiment. As shown in Figure 4.8, a sample is
Figure 4.7. (a) Four-point probe measures relative resistance changes, and (b) two-point probe measures absolute resistance of the film.

set on a piezoelectric scanner stage, which moves the sample in the $x$- and $y$-direction and records the sample position. A probe tip is mounted at the end of a cantilever, which can move up and down ($\Delta z$) as the tip tracks the sample surface. A laser beam incident on a cantilever and reflecting to a photodiode measures the bending angle of the cantilever. The force between tip and the surface (the bending angle) is maintained to be fixed during the scanning by the $\Delta z$ corrections. The $\Delta z$ and ($x$, $y$) position signals generate a 3D image of the film surface.

Figure 4.8. A schematic of the AFM feedback system [5].
The AFM can be operated in two different modes: contact mode or non-contact mode. In this study, we use the contact mode, which works in the repulsive regime and the tip is in contact to the sample surface all the time during scanning.

References

Chapter 5

*Measured conductance in Al-based superlattices*

In this work, thin Al/transition metal superlattices are deposited on a SiO$_2$/Si substrate to study their transport behavior. A transition metal layer is deposited on the SiO$_2$/Si-substrate first, followed alternatively by Al and transition metal layers. The thickness of each layer is of the order of 1 – 2 nm. Growth of each layer is interrupted after every 2 or 3 seconds deposition, so that I-V measurement can be carried out without the interference of incoming beam. Relative resistance values are measured *in-situ* using the 4-point probe method. After completing the superlattice, 2-point probe method is used to measure the absolute resistance values of the superlattice. The 2-point probe value is then used to normalize the 4-point probe relative values.

After growth of the superlattice is complete, the samples are heated from room temperature to 300 °C and cooled down to room temperature. Resistance of samples during the heating and cooling is also monitored using 4-point probe. After annealing, roughness of the top surface of superlattice is measured by AFM.

5.1 Al/Ru superlattice

Figure 5.1 shows the relative resistance of Al/Ru superlattice measured once every 2 seconds by the 4-point probe during the whole superlattice deposition. Each layer is grown through 11 separate depositions, except the first Ru layer. Each deposition is 3 seconds for Al and 2 seconds for Ru. During the deposition, resistance reading is overwhelmed by the charges in the beam. During the ~40 seconds growth interruption, a shutter is used to block the beam,
and a stable resistance value is obtained. Thicknesses of the Al and Ru layer are listed in Figure 4.4 and Table 4-2. The normalized resistance and conductance measured during the growth are plotted versus thickness in Figure 5.2 and Figure 5.3, respectively. The measured resistance and conductance are related to $\rho$ and $\sigma$ (see section 4.2):

$$ R \approx 1.4 \frac{\rho}{t} $$

and

$$ G \approx 0.72 \sigma t. $$

It is noticed in Figure 5.3 that a sharp drop in conductance occurs at the beginning of every Ru deposition on Al. The conductance drop suggests a significant number of carriers from the Al layer is localized at the new Ru/Al interface. Only a very small negative drop is observed when an Al layer is deposited on Ru, but the rise in Al conductance is delayed by ~1 nm of deposition. This suggests that carriers from the Ru layer are not localized at the new Al/Ru interface, but
Figure 5.2. *In-situ* resistance of the Al/Ru superlattice during deposition. Each data point represents the resistance reading at each growth interruption.

carriers from the Al layer are localized at the Al/Ru interface. The small error bars shown in Figure 5.2 and Figure 5.3 indicate stable transport behavior observed at every steps of grain growth.

The differential conductance $\frac{dG}{dt}$ is plotted in Figure 5.4, which shows a sharp drop at the beginning of Ru deposition similar to the simulation shown in Figure 3.5 (b). It is noted that $\frac{dG}{dt}$ in Al layers also shows feature of localization depicted in Figure 3.6 (b). Table 5-1 lists the conductance drop $\Delta G$ upon the beginning of Ru layer deposition and maximum $\frac{dG}{dt}$ value reached in each Ru and Al layers. It is noted that the instantaneous conductivity $\frac{dG}{dt}$ of the Al and Ru layer both saturate at $\sim 0.008 \mu \Omega^{-1} \text{cm}^{-1}$.
Figure 5.3. Observed conductance of the Al/Ru superlattice during growth.

Figure 5.4. Differential conductance $\frac{dG}{dt}$ of the Al/Ru superlattice during growth.
Table 5-1. Maximum conductance drop $\Delta G$ and $\frac{dG}{dt}$ in each layer of the Al/Ru superlattice.

<table>
<thead>
<tr>
<th>Layer</th>
<th>$t$ (nm)</th>
<th>$\Delta G_{\text{max}}$ (k$\Omega^{-1}$)</th>
<th>$\frac{dG}{dt}_{\text{max}}$ ($\mu\Omega^{-1}\text{cm}^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al (2)</td>
<td>2.2</td>
<td>—</td>
<td>0.0048</td>
</tr>
<tr>
<td>Al (4)</td>
<td>2.2</td>
<td>—</td>
<td>0.0068</td>
</tr>
<tr>
<td>Al (6)</td>
<td>2.2</td>
<td>—</td>
<td>0.0080</td>
</tr>
<tr>
<td>Ru (3)</td>
<td>1.3</td>
<td>0.37</td>
<td>0.0090</td>
</tr>
<tr>
<td>Ru (5)</td>
<td>1.3</td>
<td>0.68</td>
<td>0.0083</td>
</tr>
</tbody>
</table>

5.2 Al/Co superlattice

Figure 5.5 shows the relative resistance of Al/Co superlattice measured during deposition. Similar to the Al/Ru superlattice, the layers are deposited through eleven 3 seconds periods for Al and eleven 2 seconds periods for Co. Resistance values are normalized through a
2-point measurement carried out at the end of deposition and plotted in Figure 5.6. The evolution of conductance is shown in Figure 5.7. The overall behavior of conductance is very similar to that of the Al/Ru superlattice: a significant drop at the beginning of each Co layer deposition, and no drop but a significant delay in conductance rise in Al layers.

The differential conductance $\frac{dG}{dt}$ in Figure 5.8 shows a sharp drop at the beginning of each Co deposition on Al layer. It is noted that $\frac{dG}{dt}$ shows the characteristic “s” shape behavior in Al layers. Table 5-2 lists the conductance drop $\Delta G$ at the beginning of Co deposition and maximum $\frac{dG}{dt}$ values for each Co and Al layer. It is noted that $\frac{dG}{dt}$ of Al and Co layer saturates at ~0.013 and ~0.0075 $\mu\Omega^{-1}cm^{-1}$, respectively. The thickness of Co layer (0.8 nm) are much thinner than Al layer (2.2 nm). If their thicknesses are the same, they may reach similar $\frac{dG}{dt}$ values.

Figure 5.6. In-situ resistance of the Al/Co superlattice during deposition. Each data point represents the resistance reading at each growth interruption.
Figure 5.7. Observed conductance of the Al/Co superlattice during growth.

Figure 5.8. Differential conductance $\frac{dG}{dt}$ of the Al/Co superlattice during growth.
### Table 5-2. Maximum conductance drop $\Delta G$ and $\frac{dG}{dt}$ in each layer of the Al/Co superlattice.

<table>
<thead>
<tr>
<th>Layer</th>
<th>$t$ (nm)</th>
<th>$\Delta G_{\text{max}}$ (k$\Omega^{-1}$)</th>
<th>$\frac{dG}{dt}_{\text{max}}$ ((\mu\Omega^{-1}\cdot\text{cm}^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al (2)</td>
<td>2.2</td>
<td>—</td>
<td>0.0062</td>
</tr>
<tr>
<td>Al (4)</td>
<td>2.2</td>
<td>—</td>
<td>0.0106</td>
</tr>
<tr>
<td>Al (6)</td>
<td>2.2</td>
<td>—</td>
<td>0.0125</td>
</tr>
<tr>
<td>Co (3)</td>
<td>0.8</td>
<td>0.26</td>
<td>0.0076</td>
</tr>
<tr>
<td>Co (5)</td>
<td>0.8</td>
<td>0.70</td>
<td>0.0075</td>
</tr>
<tr>
<td>Co (7)</td>
<td>0.8</td>
<td>0.89</td>
<td>0.0072</td>
</tr>
</tbody>
</table>

#### 5.3 Al/Mo superlattice

Figure 5.9 shows the relative resistance of Al/Mo superlattice. The layers are deposited through eleven 3 seconds periods for both Al and Mo layers. The normalized resistance and

![Graph of resistance over time for Al/Mo superlattice](image)

**Figure 5.9.** *In-situ* relative resistance during deposition of the Al/Mo superlattice.
conductance are plotted in Figure 5.10 and Figure 5.11. The behavior of conductance is very similar to that of the Al/Ru and Al/Co superlattices: a significant drop at the beginning of each Mo deposition and a slight delay in conductance rise in Al layers. The total advance in conductance in Al layers and the amount of drop in conductance at the Mo/Al interface in this superlattice are much larger than in previous two superlattices, even though the Al layer thickness is the same.

The differential conductance $\frac{dG}{dt}$ is plotted in Figure 5.12, which shows a sharp drop at the beginning of each Mo deposition. Table 5-3 lists the conductance drop $\Delta G$ at the beginning of Mo layer deposition and maximum $\frac{dG}{dt}$ value achieved in each layer. It is noted that even though the Al and Mo layers are of the same thickness, the Al layers reach a much higher (~3×) instantaneous conductivity than the Mo layers.

![Figure 5.10. In-situ resistance of the Al/Mo superlattice during deposition. Each data point represents the resistance reading at each growth interruption.](image-url)
Figure 5.11. Observed conductance of the Al/Mo superlattice during growth.

Figure 5.12. Differential conductance $\frac{dG}{dt}$ of the Al/Mo superlattice during growth.
Table 5-3. Maximum conductance drop $\Delta G$ and $\frac{dG}{dt}$ in each layer of the Al/Mo superlattice.

<table>
<thead>
<tr>
<th>Layer</th>
<th>$t$ (nm)</th>
<th>$\Delta G_{\text{max}}$ (k$\Omega^{-1}$)</th>
<th>$\frac{dG}{dt}_{\text{max}}$ ($\mu\Omega^{-1}\text{cm}^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al (2)</td>
<td>2.2</td>
<td>—</td>
<td>0.012</td>
</tr>
<tr>
<td>Al (4)</td>
<td>2.2</td>
<td>—</td>
<td>0.024</td>
</tr>
<tr>
<td>Al (6)</td>
<td>2.2</td>
<td>—</td>
<td>0.024</td>
</tr>
<tr>
<td>Mo (3)</td>
<td>2.2</td>
<td>0.94</td>
<td>0.0069</td>
</tr>
<tr>
<td>Mo (5)</td>
<td>2.2</td>
<td>1.98</td>
<td>0.0062</td>
</tr>
<tr>
<td>Mo (7)</td>
<td>2.2</td>
<td>2.07</td>
<td>0.0064</td>
</tr>
</tbody>
</table>

5.4 Annealing and cooling of superlattices

Figure 5.13 shows the normalized $\textit{in-situ}$ resistance of the Al/Ru and Al/Mo superlattices during the annealing from RT to 300 °C and then cooling down to 100 °C. We have also annealed the Al/Co superlattice, but an unstable connection failed the resistance measurement. The resistance of the Al/Ru and Al/Mo superlattices starts to increase at $T \gtrsim 150$ °C, indicating onset of intermixing between layers. The resistance of the Al/Ru superlattice increases by $1.45 \times$ during annealing to 300 °C, whereas the resistance in the Al/Mo superlattice increases by $1.13 \times$. A larger resistance increase may indicate more intermixing. During cooling from 300 °C to 100 °C, the resistance is still increasing, suggesting that intermixing remains active during cooling. The resistance increases $1.1 \times$ during cooling in both Al/Ru and Al/Mo superlattices. As the superlattices are deposited at room temperature, below the intermixing temperature, we expect the interface in the superlattices to be abrupt prior to annealing.
Figure 5.13. Normalized resistance of the (a) Al/Ru and (b) Al/Mo superlattices during the annealing and cooling.
5.5 AFM of superlattice surfaces

Figure 5.14 shows the *ex-situ* AFM surface morphology of three Al-based superlattices after annealing. The very top surface of the Al/Ru superlattice is Al, while the Al/Co and Al/Mo superlattices have Co and Mo as the top surface, respectively. The surface roughness is characterized by the step height and correlation length [1]. The root-mean-square (RMS) of measured surface step height is listed in Table 5-4. The RMS values of the three superlattices

![AFM images of Al-based superlattices](image)

Figure 5.14. *Ex-situ* AFM surface images of Al-based superlattices.

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Table 5-4. RMS and correlation length of superlattices measured by AFM.

<table>
<thead>
<tr>
<th>Superlattice</th>
<th>RMS (nm)</th>
<th>Correlation length (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al/Ru</td>
<td>0.40</td>
<td>16</td>
</tr>
<tr>
<td>Al/Co</td>
<td>0.43</td>
<td>30</td>
</tr>
<tr>
<td>Al/Mo</td>
<td>0.47</td>
<td>49</td>
</tr>
</tbody>
</table>

are very similar, all within one to two monolayers thickness. We have also analyzed the correlation lengths for the three superlattices, as listed in Table 5-4. It is noticed that Mo surface has a larger correlation length.

References

Chapter 6

Analyses of measured conductance in Al-based superlattices

6.1 Analyses of the Al/Ru superlattice

To fit the measured conductance, we adopt the simulation scheme discussed in section 3.4:

\[ G = \sum_i \left[ \sigma_f(\Lambda_g, \sigma_g, t_i) t_i - \Delta G f(t_i) \right], \]  

(6.1)

where

\[ \sigma_f(\Lambda_g, \sigma_g, t_i) = \sigma_g \left[ 1 - \frac{3\Lambda_g}{2t_i} \int_0^{\pi/2} \sin^3 \theta \cos \theta \left( 1 - e^{-t_i/\Lambda_g \cos \theta} \right) d\theta \right], \]  

(6.2)

\[ f(t_i) = 1 - e^{-t_i/t^*} \quad \text{for transition metal layer}, \]  

(6.3)

\[ f(t_i) = \left[ \text{erf} \left( \frac{t_i}{t^*} \right) \right]^2 \quad \text{for simple metal layer}. \]  

(6.4)

Even though the above equations include four parameters \( \Lambda_g, \sigma_g, \Delta G, \) and \( t^* \), the curve fitting can be unique if we follow these steps:

(a) \( \Lambda_g \) and \( \sigma_g \) are linked in (6.2). Therefore, both parameters represent only one variable.

(b) \( \Lambda_g \) is determined by the curvature of \( \frac{dG}{dt} \) prior to localization, as shown in Figure 3.1. The assigned \( \Lambda_g \) value must conform to the shape of the measured \( \frac{dG}{dt} \) curve. Also the \( \frac{dG}{dt} \) curve calculated from the \( \sigma_f t \) term alone in (6.1) should coincide with the measured \( \frac{dG}{dt} \) at large \( t \), as shown in Figure 3.5 (b) and Figure 3.6 (b).

(c) Once \( \sigma_f t \) is determined by (a) and (b), \( \Delta G \) and \( t^* \) are determined directly from fitting \( \Delta G f(t) \) vs. \( \sigma_f t - G \) using a curve fitting program in MATLAB.
(d) Many other functions were tried for \( f(t) \) in (6.3) and (6.4), including
\[
1 - e^{-\frac{t^2}{2\tau^2}}, \sin \left( \frac{t}{\tau^*} \right)
\]
and \( \tanh \left( \frac{t}{\tau^*} \right) \). The functions in (6.3) and (6.4) gave the most satisfactory fit to experiment.

Following these steps, a unique and optimized fitting of (6.1) to the measured Al/Ru conductance \( G \) is achieved and shown in Figure 6.1 (solid lines) using the fitting parameters listed in Table 6-1. The fitting of differential conductance \( \frac{dG}{dt} \) is also plotted in Figure 6.2. In the Ru layer, measured \( \frac{dG}{dt} \) exhibits a large curvature, indicating a small mean free path \( \Lambda_g \) (~1 nm).

Upon Ru deposition on Al, a significant conductance drop is observed, suggesting a strong localization of carriers from the Al layer at the newly formed Ru/Al interface. The fitting \( \Delta G \) value reflects the strength of localization and the \( t^* \) in (6.3) indicates how rapid the localized states are formed and filled.

![Figure 6.1. Simulated (solid lines) and measured (symbols) conductance of the Al/Ru superlattice during growth.](image-url)
Figure 6.2. Simulated (solid lines) and measured (symbols) differential conductance $\frac{dG}{dt}$ of the Al/Ru superlattice during growth.

In the Al layer, the small curvature of $\frac{dG}{dt}$ suggests a large mean free path $\Lambda_g$ (~5 nm).

The fitting $\sigma_g$ values listed in Table 6-1, which should be equal to the differential conductance $\frac{dG}{dt}$ at large $t$, also agree with the measured $\frac{dG}{dt_{max}}$ listed in Table 5-1. The $\sigma_g$ values in the second and third layer of Al are twice as large as that in the first layer, probably due to larger grain size. Only a very small conductance drop is observed when the second and third Al layers are deposited on Ru, indicating that Ru carriers are not localized at the new interface. This may suggest that the energy of localized states is higher than the Fermi energy $\varepsilon_F$ of Ru [see Figure 2.12 (b)]. The small drops may be due to localization of carriers from the first Al layer. The conductance increase in the Al layer above the interface is delayed by ~1 nm, strongly suggesting that carriers of Al are back filling localized states at the underneath interface. The $\Delta G$
Table 6-1. Fitting Parameters of the Al/Ru superlattice.

<table>
<thead>
<tr>
<th>Materials</th>
<th>$\sigma_g$ ($\mu\Omega^{-1}\text{cm}^{-1}$)$^\dagger$</th>
<th>$\Lambda_g$ (nm)</th>
<th>$t^*$ (nm)</th>
<th>$\Delta G$ (k$\Omega^{-1}$)</th>
<th>$R^2$ (%)$^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al (2)</td>
<td>0.007</td>
<td>5</td>
<td>0.93</td>
<td>0.24</td>
<td>99.8</td>
</tr>
<tr>
<td>Al (4)</td>
<td>0.010</td>
<td>5</td>
<td>0.74</td>
<td>0.36</td>
<td>99.2</td>
</tr>
<tr>
<td>Al (6)</td>
<td>0.012</td>
<td>5</td>
<td>0.81</td>
<td>0.42</td>
<td>99.7</td>
</tr>
<tr>
<td>Ru (3)</td>
<td>0.009</td>
<td>1</td>
<td>0.10</td>
<td>0.47</td>
<td>99.2</td>
</tr>
<tr>
<td>Ru (5)</td>
<td>0.009</td>
<td>1</td>
<td>0.12</td>
<td>0.86</td>
<td>99.9</td>
</tr>
</tbody>
</table>

$^\dagger$ The actual material conductivity is given by $1.4 \sigma_g$ (see equation 5.2).

$^*$ $R^2 = \frac{\Sigma_i (\hat{y}_i - \bar{y})^2}{\Sigma_i (y_i - \bar{y})^2}$, where $y$ is the data point and $\hat{y}$ is the fit.

realized from the back filling of Al carriers is smaller, suggesting a weaker localization at the Al/Ru interface than the Ru/Al interface.

6.2 Analyses of the Al/Co superlattice

Optimized fittings of observed conductance and $\frac{dG}{dt}$ in the Al/Co superlattice are shown in Figure 6.3 and Figure 6.4 (solid lines). The fitting parameters are listed in Table 6-2. Similar to the Al/Ru superlattice, $\sigma_g$ values in Co and Al layers are close to the value of $\frac{dG}{dt}_{max}$ listed in Table 5-2. Upon Co depositing on Al, the occurrence of a large conductance drop $\Delta G$ indicates strong localization of Al carriers at the newly formed Co/Al interface. For Al layers, the small curvature in $\frac{dG}{dt}$ indicates a large mean free path $\Lambda_g$ (~5 nm). No instant conductance drop is observed when Al is deposited on Co, indicating that the Co carriers are not localized at the Al/Co interface. The delay (~1 nm) of conductance rise in the top Al layer suggests that Al carriers are back filling the localized states at the underneath interface. Conductance drop $\Delta G$ in
Figure 6.3. Simulated (solid lines) and measured (symbols) conductance of the Al/Co superlattice during growth.

Figure 6.4. Simulated (solid lines) and measured (symbols) differential conductance $\frac{dG}{dt}$ of the Al/Co superlattice during growth.
Table 6-2. Fitting Parameters of the Al/Co superlattice.

<table>
<thead>
<tr>
<th>Materials</th>
<th>$\sigma_g$ ($\mu\Omega^{-1}\text{cm}^{-1}$)†</th>
<th>$\Lambda_g$ (nm)</th>
<th>$t^*$ (nm)</th>
<th>$\Delta G$ (k$\Omega^{-1}$)</th>
<th>$R^2$ (%)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al (2)</td>
<td>0.0087</td>
<td>5</td>
<td>1.32</td>
<td>0.25</td>
<td>97.7</td>
</tr>
<tr>
<td>Al (4)</td>
<td>0.015</td>
<td>5</td>
<td>1.00</td>
<td>0.63</td>
<td>99.8</td>
</tr>
<tr>
<td>Al (6)</td>
<td>0.018</td>
<td>5</td>
<td>0.95</td>
<td>0.67</td>
<td>99.8</td>
</tr>
<tr>
<td>Co (3)</td>
<td>0.0085</td>
<td>1</td>
<td>0.08</td>
<td>0.33</td>
<td>99.3</td>
</tr>
<tr>
<td>Co (5)</td>
<td>0.0085</td>
<td>1</td>
<td>0.08</td>
<td>0.84</td>
<td>99.8</td>
</tr>
<tr>
<td>Co (7)</td>
<td>0.0085</td>
<td>1</td>
<td>0.08</td>
<td>1.05</td>
<td>99.3</td>
</tr>
</tbody>
</table>

† The actual material conductivity is given by $1.4 \sigma_g$ (see equation 5.2).

* $R^2 = \frac{\sum(y_i - \hat{y})^2}{\sum(y_i - \bar{y})^2}$, where $y$ is the data point and $\hat{y}$ is the fit.

Al is smaller, suggesting a weaker localization at the Al/Co interface.

6.3 Analyses of the Al/Mo superlattice

Optimized fittings of the conductance and $\frac{dG}{dt}$ of the Al/Mo superlattice are shown in Figure 6.5 and Figure 6.6 (solid lines). The fitting parameters are listed in Table 6-3. Similar to the Al/Ru and Al/Co superlattices, the $\sigma_g$ values in Mo layers ($0.008 \mu\Omega^{-1}\text{cm}^{-1}$) are the same as the $\frac{dG}{dt}_{\text{max}}$ values listed in Table 5-3. The fitting requires a larger $\Lambda_g$ (~5 nm) in Mo. Mo deposition also produces a larger conductance drop $\Delta G$ than the depositions of Ru and Co.

The $\sigma_g$ in the second and third Al layer are twice of that in the first Al layer, possibly due to grain size increase in upper layers. For Al layers, the small curvature of $\frac{dG}{dt}$ indicates a large mean free path $\Lambda_g$ (~5 nm). The $\sigma_g$ values in Al layers are larger than the observed $\frac{dG}{dt}_{\text{max}}$ listed in Table 5-3. This may be due to the fact that instantaneous conductivity $\frac{dG}{dt}$ has not reached the maximum value at the 2.2 nm Al thickness. Smaller conductance drop $\Delta G$ in Al also indicates a
Figure 6.5. Simulated (solid lines) and measured (symbols) conductance of the Al/Mo superlattice during growth.

Figure 6.6. Simulated (solid lines) and measured (symbols) differential conductance $\frac{dG}{dt}$ of the Al/Mo superlattice during growth.
Table 6-3. Fitting Parameters of the Al/Mo superlattice.

<table>
<thead>
<tr>
<th>Materials</th>
<th>$\sigma_g$ (µΩ·cm$^{-1}$)$^\dagger$</th>
<th>$\Lambda_g$ (nm)</th>
<th>$t^*$ (nm)</th>
<th>$\Delta G$ (kΩ$^{-1}$)</th>
<th>$R^2$ (%)$^\ast$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al (2)</td>
<td>0.017</td>
<td>5</td>
<td>0.98</td>
<td>0.31</td>
<td>99.7</td>
</tr>
<tr>
<td>Al (4)</td>
<td>0.033</td>
<td>5</td>
<td>0.93</td>
<td>0.54</td>
<td>99.7</td>
</tr>
<tr>
<td>Al (6)</td>
<td>0.034</td>
<td>5</td>
<td>0.96</td>
<td>0.58</td>
<td>98.8</td>
</tr>
<tr>
<td>Mo (3)</td>
<td>0.008</td>
<td>5</td>
<td>0.10</td>
<td>1.02</td>
<td>99.8</td>
</tr>
<tr>
<td>Mo (5)</td>
<td>0.008</td>
<td>5</td>
<td>0.15</td>
<td>2.21</td>
<td>99.5</td>
</tr>
<tr>
<td>Mo (7)</td>
<td>0.008</td>
<td>5</td>
<td>0.15</td>
<td>2.30</td>
<td>99.5</td>
</tr>
</tbody>
</table>

$^\dagger$ The actual material conductivity is given by $1.4 \sigma_g$ (see equation 5.2).

$^\ast R^2 = \frac{\sum (\hat{y}_i - y)^2}{\sum (y_i - y)^2}$, where $y$ is the data point and $\hat{y}$ is the fit.

weaker localization. Lack of instant $\Delta G$ drop in Al suggests again that Mo carriers are not localized at the Al/Mo interface.

6.4 Comparison of $\sigma_g$ in three superlattices

The intrinsic conductivity $\sigma_g$ depends on carrier density $n$ and mobility $\mu$:

$$\sigma_g = ne\mu. \quad (6.5)$$

Carrier density $n$ is proportional to atomic density and valency, and $\mu$ is sensitive to mean free path $\Lambda_g$ and surface roughness.

Values of $\sigma_g$ in Al layers in three different superlattices are listed in Table 6-4 for comparison. Despite identical atomic density, valency and $\Lambda_g$, the $\sigma_g$ of Al layer is smaller in the Al/Ru and Al/Co superlattices than in the Al/Mo superlattice, indicating that the former two superlattices must have rougher interfaces. A rougher interface means larger step height or smaller correlation length. According to AFM measurements presented in Chapter 5, the step
Table 6-4. Comparison of $\sigma_g$ in three superlattices.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Atomic density [1] ($10^{22}$ cm$^{-3}$)</th>
<th>Valency</th>
<th>$\Lambda_g$ (nm)</th>
<th>Observed $\sigma_g$ ($\mu\Omega^{-1}$cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al on Ru</td>
<td>6.02</td>
<td>3</td>
<td>5</td>
<td>0.012</td>
</tr>
<tr>
<td>Al on Co</td>
<td></td>
<td></td>
<td></td>
<td>0.018</td>
</tr>
<tr>
<td>Al on Mo</td>
<td></td>
<td></td>
<td></td>
<td>0.034</td>
</tr>
<tr>
<td>Ru on Al</td>
<td>7.36</td>
<td>$\leq$ 8</td>
<td>1</td>
<td>0.0090</td>
</tr>
<tr>
<td>Co on Al</td>
<td>8.97</td>
<td>$\leq$ 9</td>
<td>1</td>
<td>0.0085</td>
</tr>
<tr>
<td>Mo on Al</td>
<td>6.42</td>
<td>$\leq$ 6</td>
<td>5</td>
<td>0.0080</td>
</tr>
</tbody>
</table>

heights are similar in all three superlattices, but the Mo surface has a larger correlation length.

Smaller grain size (1 nm) in Ru and Co may be responsible for the rougher interfaces.

Conductivities $\sigma_g$ in three transition metals are also listed in Table 6-4. The three transition metals have very similar $\sigma_g$ values and atomic densities. Since Mo has a much larger mean free path $\Lambda_g$ ($5\times$) and a smoother surface, the valency of Mo must be smaller than Ru and Co by at least a factor of 5. Therefore, it is likely that most of $d$ electrons in Mo are localized.

6.5 Comparison of carrier localization in three superlattices

As discussed in previous sections, carriers from transition metal layers are not localized at the interfaces above or beneath the layer. All conductance drops are due to localization of Al carriers. The conductance of Al layers prior to any localization is given by

$$G_{Al} = \bar{\sigma}_{f_{Al}} t_{Al} = ne\mu_f t_{Al}.$$  \hspace{1cm} (6.6)

If $\Delta n$ is the amount of localized carriers, then the conductance drop $\Delta G$ is
\[ \Delta G = \Delta ne\mu t_{Al}. \quad (6.7) \]

The percentage of carriers localized at the interfaces is

\[ \frac{\Delta n}{n} = \frac{\Delta G}{G_{Al}}. \quad (6.8) \]

The observed \( \frac{\Delta G}{G_{Al}} \) values for the three superlattices are plotted in Figure 6.7. Here it is seen that the percentage of Al carriers localized at the interface below is significantly smaller than the percentage localized at the interface above. It is also noticed that almost all Al carriers are localized at both interfaces: Figure 6.7 indicates \(~100\%\) in the Al/Ru superlattice, \(~95\%\) in the Al/Co superlattice, and \(~80\%\) in the Al/Mo superlattice. The percentage of localization is observed to be zero for the transition metal carriers. This means that the conductance in the superlattice is carried out almost exclusively by the transition metal layers.
Figure 6.7. The percentage of carriers $\frac{\Delta G}{G_{Al}}$ localized at the interface above the Al layer (solid symbols) and at the interface below the Al layer (open symbols) observed in the three superlattices.
We can calculate the density of localized states $\Delta n$ at the interface from (6.8) using the density of carriers in the Al layer,

$$n = (\text{atomic density}) \times (\text{valency}) \times (\text{thickness})$$

$$= (6.02 \times 10^{22}\text{ cm}^{-3}) \times (3) \times (2.2 \times 10^{-7}\text{ cm}) = 3.97 \times 10^{16}\text{ cm}^{-2},$$  \hspace{1cm} (6.9)

and the measured $\frac{\Delta G}{G_{Al}}$ values. The results are listed in Table 6-5. Assuming close-packed planes at the interface: (0001) for hcp and (110) for bcc, we can determine the density of surface atoms and the number of localized carriers per surface atom, which are also listed in Table 6-5. The localized states are less at the interface ($B$) than at the interface ($A$). These localized states can come from either covalent bonds or disorder scattering.

The known compounds between Al and the three transition metals are listed in Table 6-6. Not all compounds can form at the interface. The number of bonds per surface atoms depends on local stoichiometry. For instance, the number of bonds per atom at a planar close-packed interface is 3, at a surface step is 6, and at a vacancy is 9. Assuming 2 electrons per covalent

<table>
<thead>
<tr>
<th>Interface</th>
<th>$\Delta G_{Al}$</th>
<th>$\Delta n_{Al}$ ($10^{16}\text{ cm}^{-2}$)</th>
<th>Density of surface atoms ($10^{16}\text{ cm}^{-2}$)</th>
<th>Localized carriers per atom</th>
</tr>
</thead>
<tbody>
<tr>
<td>($A$) Ru on Al</td>
<td>0.79</td>
<td>3.14</td>
<td>0.16</td>
<td>20</td>
</tr>
<tr>
<td>($B$) Al on Ru</td>
<td>0.34</td>
<td>1.33</td>
<td>0.16</td>
<td>8</td>
</tr>
<tr>
<td>($A$) Co on Al</td>
<td>0.56</td>
<td>2.22</td>
<td>0.18</td>
<td>12</td>
</tr>
<tr>
<td>($B$) Al on Co</td>
<td>0.39</td>
<td>1.55</td>
<td>0.18</td>
<td>9</td>
</tr>
<tr>
<td>($A$) Mo on Al</td>
<td>0.66</td>
<td>2.62</td>
<td>0.14</td>
<td>19</td>
</tr>
<tr>
<td>($B$) Al on Mo</td>
<td>0.16</td>
<td>0.64</td>
<td>0.14</td>
<td>5</td>
</tr>
</tbody>
</table>
Table 6-6. Compounds between Al and three transition metals.

<table>
<thead>
<tr>
<th>Interface</th>
<th>Compounds</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al/Ru</td>
<td>Al₆Ru, Al₁₃Ru₄, Al₂Ru, Al₃Ru₂, AlRu</td>
</tr>
<tr>
<td>Al/Co</td>
<td>Al₉Co₂, Al₁₃Co₄, Al₅Co, Al₅Co₂</td>
</tr>
<tr>
<td>Al/Mo</td>
<td>Al₅Mo, Al₄Mo, Al₈Mo₃</td>
</tr>
</tbody>
</table>

bond, we expect 6 localized carriers per atom at a smooth interface. The number can increase to ~12 at a surface step. Accordingly, the localized states at the interfaces (B) are likely from chemical bonds. The number of localized states at the interfaces (A) are much larger. Those excessive localized states must come from disorder scattering, which means the interfaces (A) contain more defects.

As listed in Table 6-7, Al has a much smaller bulk modulus than transition metals, therefore, deposition of Al on a thick substrate would cause less stress in the film and fewer defects at the interface, and consequently less localized states. Ru has the largest modulus, followed by Mo and Co. The density of localized states at the three interfaces (A) also follows

Table 6-7. Correlations between localized states and bulk modulus.

<table>
<thead>
<tr>
<th>Interface</th>
<th>Δn (10¹⁶ cm⁻²)</th>
<th>Localized carriers per atom</th>
<th>Bulk modulus [2] (N/m²)</th>
<th>epitaxy</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A) Ru on Al</td>
<td>3.14</td>
<td>20</td>
<td>3.2 (Ru)</td>
<td>?</td>
</tr>
<tr>
<td>(B) Al on Ru</td>
<td>1.33</td>
<td>8</td>
<td>0.72 (Al)</td>
<td>?</td>
</tr>
<tr>
<td>(A) Co on Al</td>
<td>2.22</td>
<td>12</td>
<td>1.9 (Co)</td>
<td>?</td>
</tr>
<tr>
<td>(B) Al on Co</td>
<td>1.55</td>
<td>9</td>
<td>0.72 (Al)</td>
<td>?</td>
</tr>
<tr>
<td>(A) Mo on Al</td>
<td>2.62</td>
<td>19</td>
<td>2.7 (Mo)</td>
<td>?</td>
</tr>
<tr>
<td>(B) Al on Mo</td>
<td>0.64</td>
<td>5</td>
<td>0.72 (Al)</td>
<td>Yes</td>
</tr>
</tbody>
</table>
the same order, suggesting that density of defects is likely correlated to the film stress during growth. It is noticed that epitaxial growth may produce less defects and localized states. Al is known to grow epitaxially on Mo [3], and we have observed the least amount of localized states at the Al/Mo interface.

6.6 Comparison of average conductivity in three superlattices

The conductance of three superlattices are compared in Figure 6.8. Average conductivity of the Al/Mo superlattice (0.0033 \( \mu \Omega^{-1} \text{cm}^{-1} \)) is 38% higher than the Al/Ru and Al/Co superlattices (0.0024 \( \mu \Omega^{-1} \text{cm}^{-1} \)). Recall that most conduction is carried out by the transition metal layers, and all three transition metals have similar intrinsic conductivities. The higher

![Figure 6.8. Linear fitting of conductance in three superlattices. The Al/Mo superlattice has a higher average conductivity.](image-url)
conductivity in the Al/Mo superlattice can be attributed to the 20% Al carriers that are not localized, and the fact that interfaces in that superlattice are smoother.

References

Chapter 7

Conclusions

The effects of interface scattering and carrier localization on the conductance of superlattices have been investigated using in-situ I-V measurements during superlattices growth and computer simulation. Through analyzing the measured conductance rise during each layer growth, we have obtained information on carrier mean free path $\Lambda_g$ and intrinsic conductivity $\sigma_g$.

The carrier mean free paths are found to be small (~1 nm) in Ru and Co, indicative of a small grain size. The mean free paths in Mo and Al are found to be uniformly larger (~5 nm), which may indicate a large grain size or weak grain boundary scattering. The intrinsic conductivities of Ru, Co and Mo layers derived from our measurements are very similar ($\sigma_g \sim 0.008 - 0.009 \, \mu\Omega^{-1} \text{cm}^{-1}$). Since Mo has a much larger mean free path $\Lambda_g$ (5x) than Ru and Co, most of $d$ electrons in Mo are likely to be localized. The $\sigma_g$ values of Al layers are higher than the three transition metals, particularly in the Al/Mo superlattice ($\sigma_g \sim 0.03 \, \mu\Omega^{-1} \text{cm}^{-1}$), which may be due to smoother interfaces in that superlattice.

We have detected instant conductance drops upon deposition of transition metals on Al, indicative of Al carriers filling the localized states at the new interface. When Al is deposited on transition metals, no instant conductance drop is observed, suggesting that transition metal carriers are not localizable at the new interface. The new interface, however, is found to trap carriers of the above Al layer, resulting in a delay in Al conductance rise. Both inter-metallic bonds as well as structural defects can produce localized states at the interfaces. The density of localized states at the interface is found to correlate with the bulk modulus of the top growing
layer. Large modulus produces more disorder and localization. Therefore, deposition of low-modulus Al produces less localized states at the interface than the deposition of high-modulus transition metals.

Our measurements indicate that most Al carriers are localized at the interfaces: ~100% in the Al/Ru superlattice, ~95% in the Al/Co superlattice and ~80% in the Al/Mo superlattice. All carriers in the transition metal layers are not localized. Therefore, electrical conduction in the superlattices is carried out mostly by the transition metal layers. The conductance of the Al/Mo superlattice is ~38% higher because only ~80% of Al carriers are localized.

Post-growth annealing experiments indicate that inter-species mixing occurs only at $T \geq 150$ °C. AFM surface analyses of the superlattice surfaces confirm that the surface roughness at the interface is within one to two monolayers.