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The influence of surface roughness on electrical conductance of thin Cu films: An *ab initio* study

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First-principles calculations show that atomic-scale surface roughness dramatically affects the electrical conductivity of thin films. Atomic clusters, 1–3 atoms high, deposited on the flat Cu(001) surface of an 11 monolayer thick film lead to a 30–40% reduction of its conductance. This is attributed to the destruction of isotropic Fermi surface sheets. We provide a simple parametrized formula, correlating the size of the surface added structures to the film conductance, and also demonstrate that Ta and Al surface monolayers on rough Cu surfaces cause a conductance decrease and increase, respectively. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2937188]

I. INTRODUCTION

The increase of Cu interconnect resistivity with decreasing wire cross section, typically referred to as "the size effect," has become a major challenge for modern integrated circuits technology.[1,2] This effect is observable when the interconnect width is reduced to below 100 nm, and it becomes dramatic below 50 nm, giving rise to a 100% increase in copper interconnect resistivity.[3–5] Both experimentally and theoretically, this problem is usually addressed by decomposing the total resistivity ρ_T into several components using Matthiessen's rule,

$$\rho_T = \rho_b + \rho_s + \rho_g + \rho_i, \qquad (1)$$

where ρ_b is a bulk resistivity that includes scattering from phonons, and ρ_s, ρ_g, and ρ_i are resistivity components due to scattering on surfaces, grain boundaries, and impurities, respectively. For each of these components several theoretical models have been proposed in the literature.[5–10] It is an extremely challenging problem to compute each term of Eq. (1) from atomic first principles. Therefore, the existing theoretical models are based on phenomenological considerations where various fitting parameters are used to characterize scattering properties of surfaces, grain boundaries, and impurities. The results are then fit to experimental data to obtain a relative contribution from each scattering mechanism.[3]

Surface scattering is considered to play a key role in the resistivity increase for thin films. Experiments by Rossnagel and Kuan[5] on 40–45 nm thick films, and recently by Plombon *et al.*[3] on 75 nm wide lines, show that approximately 50% of the resistivity increase is due to surface scattering effects. Surface scattering is typically discussed using the Fuchs-Sondheimer model,[7,8] which predicts the ratio between the thin film and the bulk resistivity ρ/ρ_0. Kuan and Rossnagel[5] revised this model to also account for surface roughness, proposing

$$\frac{\rho}{\rho_0} = 1 + 0.375(1-p)S\lambda/d, \qquad (2)$$

where d is the thickness of the film, λ is the electron mean-free path, p is the specularity parameter, and S is the surface roughness factor. The latter two are phenomenological parameters that describe surface scattering within this model. The specularity parameter $0 \leq p \leq 1$ characterizes the degree of diffusive scattering, i.e., $p=0$ for completely diffusive scattering, while $p=1$ for specular scattering. The roughness parameter $S \geq 1$ quantifies the contribution due to surface roughness, with a completely smooth surface exhibiting $S=1$. Clearly, these two parameters may be interrelated, as a rough surface is expected to cause diffusive scattering of electrons. Thus, in practice it is difficult, or even impossible, to independently determine p and S. As summarized in Ref. 11, most researchers conclude that their Cu layers exhibit completely diffuse surface scattering with $p=0$. The Monte Carlo simulation reported in Ref. 5 employs a sinusoidal nanometer-scale roughness to estimate a value of S. Based on their experimental roughness, the S value is only slightly larger than unity.

While classical phenomenological models have provided some useful understanding of interconnect resistivity, in our opinion it is also very important to employ *ab initio* approaches to address this problem. Using first-principles methods, one can investigate contributions to resistivity for each mechanism in Eq. (1) without phenomenological parameters and without fitting. To the best of our knowledge, no such study has been carried out and the present paper will serve as the first step to fill this gap.

In the following we investigate the influence of atomic-scale surface roughness on conductivity of thin copper films using state-of-the-art first-principles methods. We demonstrate that atomic-scale surface roughness has a rather dramatic effect on electrical conductivity of these films. In particular, we show that the presence of simple atomic roughness (1–3 atoms) on a perfect copper surface leads to a substantial (30–40%) reduction in the electrical conductivity of thin Cu films. We provide an explanation of the physical

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origin of the roughness-induced conductivity reduction, relating it to the intersheet electron scattering on the Fermi surface, considerably perturbed by roughness. We also propose a simple parametrized formula to describe the Cu layer conductivity as a function of the atomic-scale roughness amplitude. Finally, we theoretically show that the presence of metallic overlayers alters the surface scattering process, and therefore may be used to tune the conductivity of Cu films.

II. THEORETICAL METHOD

Our study was performed within a ballistic quantum transport point of view. In this approach the conductance is determined by ballistic motion of the electrons through the system. Although the electrons are not scattered out of their Blöch states, these states differ from those of the bulk material due to the altered electronic structure of the interface region which, in turn, is caused by the interface geometry, e.g., roughness, overlayers, etc. The electronic structure contribution to the electric conductivity is the dominant factor when sample dimensions are much smaller than the electronic mean-free path, which is 39 nm for Cu at room temperature. The conductance obtained in this framework, referred to as Sharvin conductance,[12] can be calculated using the Landauer-Büttiker formalism,[13]

$$G(\hat{n}) = \frac{e^2}{h}\sum_{\nu} N_{\nu}(\hat{n}) = \frac{e^2}{h}\frac{A}{4\pi^2}\frac{1}{2}\sum_{\nu} S_{\nu}(\hat{n}), \quad (3)$$

where $N_{\nu}(\hat{n})$ is the number of conducting channels for the transport in direction $\hat{n}$ for the band with index ν, and $S_{\nu}(\hat{n})$ is the projection of the Fermi surface on the plane, perpendicular to the transport direction. The above expression for Sharvin conductance via the Fermi surface projection is very attractive from a computational point of view, since the only input parameter is the Fermi surface, which can be accurately calculated from first principles using modern band-structure methods.

Our numerical calculations of the electronic structure were performed within the density functional theory with the local density approximation. We employed the SIESTA program package,[14] which uses norm-conserving pseudopotentials and numerical atomic orbitals as a basis set. Due to the large sizes of the systems under study, a single-zeta-polarized (SZP) basis set consisting of one *s*-, three *p*-, and five *d*-like orbitals per atom was used for Cu atoms in the interior of the film, while a double-zeta-polarized basis consisting of two *s*-, six *p*-, and ten *d*-like orbitals, was used to describe the surface adatoms. A SZP basis set was optimized for bulk copper using the procedure provided in Ref. 15. The quality of the optimized basis set as well as the quality of Cu pseudopotential was verified by performing a series of test calculations using a highly accurate linearized augmented plane wave method (LAPW),[16] which does not employ any pseudopotentials and uses a highly converged basis set. An excellent agreement with LAPW results was obtained for bulk copper both for the lattice parameter and for the electronic band structure up to 5 eV above the Fermi level.

Copper films were modeled by a supercell approach, keeping the periodicity in the *XY* plane, and constructing a slab in the *Z* direction, as shown in Fig. 1. The *x*- and *y* directions correspond to orthogonal ⟨110⟩ crystal directions, while the *z* axis is parallel to [001]. A 10 Å vacuum space was introduced between film surfaces in the *Z* direction to prevent them from interacting with each other. All systems under study were completely relaxed, keeping the in-plane lattice parameter equal to the theoretically calculated value of 3.60 Å, which is in excellent agreement with the experimental value of 3.61 Å.

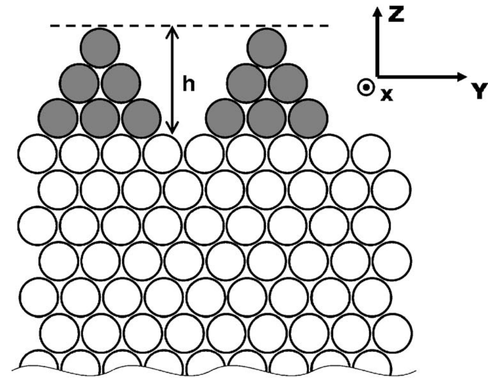

FIG. 1. Schematic representation of the copper slab structure used for the calculations. The unit cell and its first image in the transport direction are presented. The white and gray circles represent the Cu atoms of the perfect film (11 monolayers) and the roughness structures, respectively.

To compute the quantity $S_{\nu}(\hat{n})$ in Eq. (3), Fermi surface projections were carried out using the tetrahedron technique.[17] Finally, electric conductance was calculated in the *Y* direction (see Fig. 1) using Eq. (3). To test our method, the [110] conductance of bulk copper and of a completely smooth 11 monolayer thick copper film were calculated using the two band-structure methods mentioned above. We obtained a value of $0.58\times 10^{15}(1/\Omega\mathrm{m}^2)$ per spin, in excellent agreement with previous studies.[17]

III. RESULTS AND DISCUSSION

A *perfect* (i.e., atomically smooth) copper film conductance was calculated as a first step of our study. Increasing the number of copper monolayers, we observe that at 11 monolayers the film's conductance is converged to its bulk value of $0.58\times 10^{15}(1/\Omega\mathrm{m}^2)$. By comparing band structures for films of different thickness, we observed that 11 layers is the minimum thickness at which two film surfaces do not interact with each other across the film. In this regard, the bulk conductance of the 11 monolayer film seems to be evident. This result also demonstrates that at this thickness, the electron scattering from a perfectly smooth surface is completely specular, having a negligible effect on the resistivity.

To model rough surfaces, triangularlike atomic structures were added on top of the perfect copper film (see Fig. 1). Conductance was then calculated for a roughness amplitude *h* of 1, 2, 3, and 4 atomic monolayers, which corresponds to 1-, 3-, 6- (shown in Fig. 1), and 10 atom triangular steps, respectively. The triangle-triangle distance was kept constant at 1 atom in the base layer (Fig. 1), so that the roughness period increases with *h*. The results are presented in Table I, showing the absolute values of the electrical conductance as

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TABLE I. Conductance per spin of the Cu film in the Y direction as a function of roughness amplitude h.

h, layers	G_Y, $10^{15}(1/\Omega\text{m}^2)$	%
0	0.58	100
1	0.39	67
2	0.35	60
3	0.33	57
4	0.31	53

well as the percentage relative to the conductance of a perfectly smooth film. We observe that an array of rows of single Cu atoms on top of a perfect surface leads to a 33% conductance reduction. Increasing the roughness amplitude further reduces the conductance, however with a decreasing rate, up to ~50% at $h=4$.

In order to understand the physical origin of the drop in conductance when adding an array of (only) single-atom-high rows onto a smooth surface, we refer to Figs. 2(a) and 2(b), which are plots of the Fermi surface from a perfectly smooth film and from a film with an $h=1$ surface roughness, respectively. Exactly the same 1×2 supercell was used for the sake of comparison, and a periodic bands scheme was employed to see a general picture of the Fermi surface topology. We observe that even the Fermi surface of a perfect 11 monolayer thick film [Fig. 2(a)] is a complex multisheet surface due to the presence of multiple Cu layers. However, it exhibits well-developed perfect (free-electron-like) and distorted circular sheets, corresponding to the bottom and the middle of the s band, respectively. The distorted circle reflects the fourfold symmetry of the crystal lattice. Periodically distributed surface adstructures introduce a periodic perturbing potential in the Y direction with a period of at least two lattice parameters. In this case one or more Bragg planes cuts the Fermi surface in this direction, and gaps open in the Fermi surface in the intersection points.[18] The size of these gaps is determined by the strength of perturbing (surface roughness) potential. This effect should reduce the projected Fermi surface area, and therefore the conductance. This is exactly what we see in Fig. 2(b): The adatoms on top of the Cu surface cause a destruction of *isotropic* Fermi surface sheets, and new sheets are formed as a result of gap formation. Keeping in mind that Fermi velocity is always normal to the Fermi surface, we notice that for most of these sheets, the Fermi velocity loses its Y component. The electronic states being reflected into states on these sheets will lose their momentum in the Y direction, or will even be reflected back. On the other hand, the energy dispersion calculated for one of these states [Fig. 2(c)] shows that its parabolic dispersion (and therefore Fermi velocity) is well-preserved in the X direction; therefore, conductance in the X direction should not be altered significantly. This is indeed what we have found in the transport calculation. In particular, the conductance in the X direction of the rough layers deviates by less than 4% from that of a perfect film.

Next, we represent our numerical results in an analytical form. We note that our approach is different from that of Kuan *et al.*[1] in two aspects. First, we are fitting the results of accurate parameter-free quantum-mechanical calculations to a mathematical formula. Second, since our first-principles

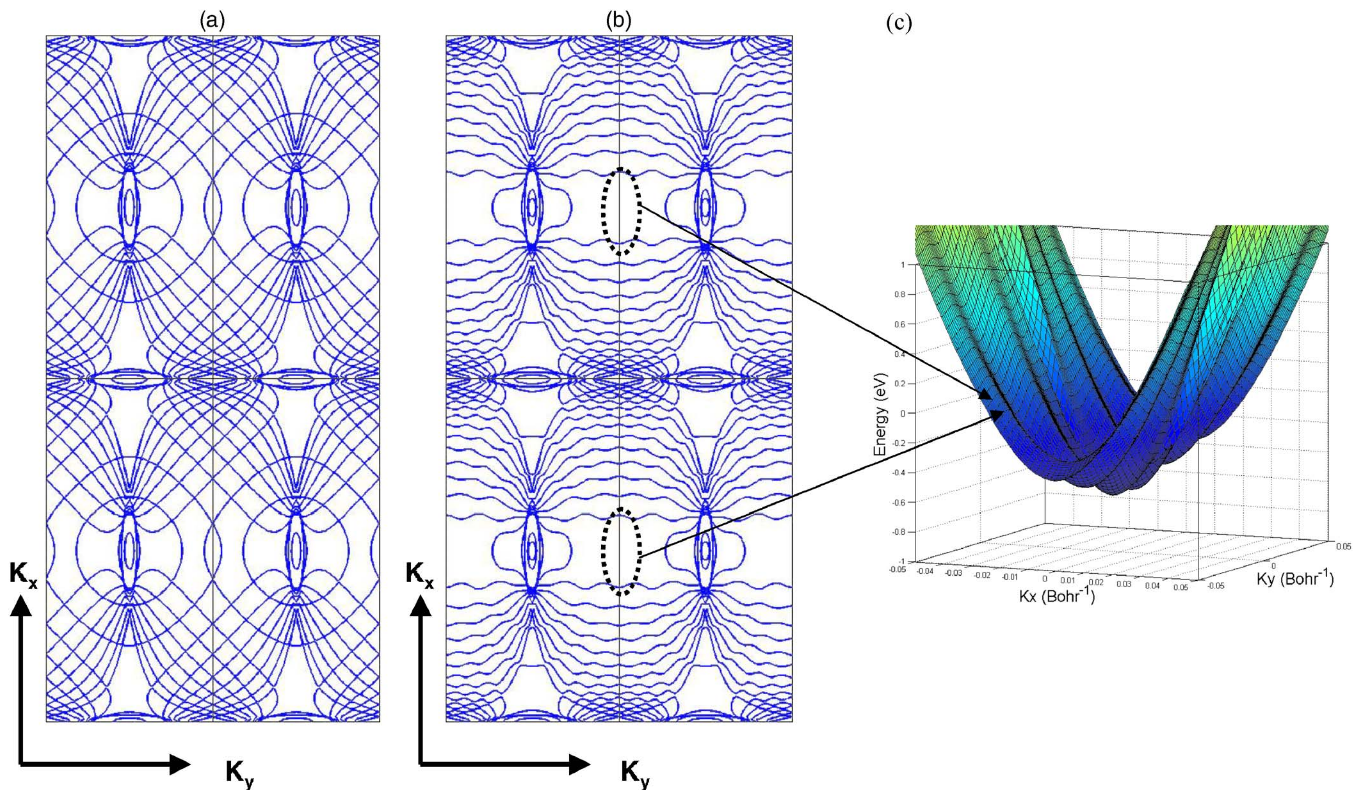

FIG. 2. (Color online) Calculated Fermi surfaces for (a) a smooth, 11 monolayer thick Cu film, and (b) the same film including a 1 atom high surface roughness. Panel (c) shows the energy dispersion for one of the sheets shown in (b).

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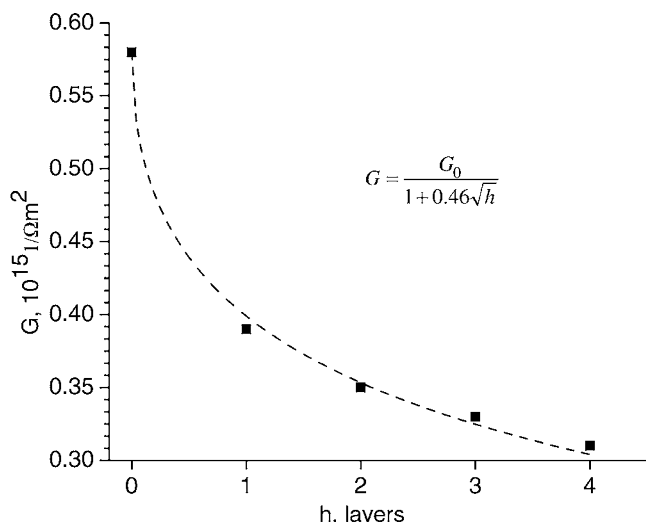

FIG. 3. Conductance as a function of roughness amplitude h. A simple analytical fit is shown.

results were collected from model films in the forms shown in Fig. 1, care must be taken when using the resulting formula for arbitrary forms of surface roughness. A simple analytical fit for the triangularlike model roughness is given in Fig. 3. We note that our formula can be reduced to the one used by Kuan *et al.*[1] if $\sqrt{h}$ is replaced by a surface roughness factor S [see Eq. (2)]. Therefore, both approaches appear to be consistent, provided that an S factor for our triangularlike surface has a $\sqrt{h}$ dependence.

Finally, we complete our study by taking a first step in investigating the possible influence of metallic adlayers on electric conductivity of rough copper films. We start by calculating the electric conductance of a rough Cu film, covered by a Ta monolayer. Due to larger sizes of Ta atoms, a 1×4 supercell is used where the Cu roughness is represented by 1 Cu adatom ($h=1$ structure), and the adlayer is a monolayer of (4) Ta atoms. The surface was completely relaxed and the structure was shown to be energetically stable. The calculated conductance in the Y direction of the Ta-covered rough copper film is $0.39\times10^{15}(1/\Omega\text{m}^2)$. This is smaller than the conductance for exactly the same Cu surface ($h=1$, 1×4 supercell) but without Ta coverage, which was calculated to be $0.41\times10^{15}(1/\Omega\text{m}^2)$. This small decrease with Ta addition is in good agreement with experimental data,[5] where a 10% conductance decrease was found for a thin Ta coverage. Exactly the same calculation was also performed for an Al adlayer (instead of the Ta adlayer). In this case, the calculated conductance is $0.42\times10^{15}(1/\Omega\text{m}^2)$, which is slightly larger than the conductance of the rough surface without coverage. We emphasize that these results are of a more qualitative character, and systems with a thicker surface coverage should be considered whn making any quantitative conclusions. Nevertheless, these qualitative results are consistent with experiments where the opposite effect of Ta and Al coverage on electrical conductivity was observed.[5]

IV. CONCLUSION

In conclusion, we performed *ab initio* calculations of electrical conductance of very thin copper films with triangular surface roughness, where the roughness amplitude was varied from 1 to 4 atomic layers. We found that the presence of very small roughness structures, consisting of only a few atoms, leads to a dramatic reduction of the electrical conductivity. This is attributed to intersheet electron scattering at the Fermi surface, which is considerably perturbed by roughness. We further provide a simple analytical formula that relates the conductivity to the roughness amplitude, and demonstrate that Al and Ta surface coverages have opposite effects on the electrical conductivity of thin copper films.

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