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Size effect of the resistivity of thin epitaxial gold films

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The temperature dependence of the electrical resistivity of thin, epitaxial, and flat (111)-oriented gold films with thickness between 2 and 50 nm is investigated. The quality of these Au films is superior to epitaxial Au films grown on mica, therefore the investigation of well-defined thin films was possible. The experiments are analyzed within the frame of classical size-effect theories of the resistivity. It turns out that for thin films, the characteristic Debye temperature is strongly decreased as compared to the bulk value. In contrast to the more physically motivated approach of Soffer, the original model of Fuchs-Sondheimer describes the experimental data in a physically consistent way.

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I. INTRODUCTION

The minimum feature size in semiconductor fabrication still rapidly decreases without any nearby limits visible. In this context, also the behavior of miniaturized metallic structures such as thin interconnects is of special interest, since whenever their thickness approaches the length scale of the electron mean free path (∼40 nm at room temperature in Au), a strong deviation from the corresponding bulk electrical resistivity is expected. This so-called size effect of the resistivity can either be treated classically[1,2] or the quantization of electronic modes can be included (see, e.g., Refs. 3 and 4). If predictions on the resistivity of nanowires or thin films have to be made, the basic mechanisms contributing to scattering of electrons in reduced dimensions must be known.

While experimental work on quantum size effects is scarce, there is a large number of publications on the classical size effect of resistivity in thin films as well as on the question of which model might be most adequate to describe the experimental data.[5,6] For example, the influence of the surface on the resistivity can be masked by grain-boundary scattering.[7,8] A correct analysis should therefore be performed with a large number of parameters, which, however, in most cases cannot be determined independently. As a consequence, the results are usually not unequivocal. Furthermore, studying the thickness dependence of the resistivity is complicated by the fact that the sample structures change with increasing thickness. A well known example for this phenomenon is the average grain size, which tends to increase with increasing film thickness.

In addition to the direct influence of the sample structure on the thin-film resistivity, there are also indirect effects. For instance, if the Debye temperature is modified in a thin film with a high surface-to-volume ratio, the correspondingly changed electron-phonon interaction can also lead to a modified resistivity. Due to the lack of appropriate, ultrathin samples, this behavior could not yet be studied systematically, and so a decrease of the effective Debye temperature was either only assumed[9] or the resistivity in granular samples[10] or thin films[11] was simply interpreted within Bloch-Grüneisen theory,

$$\rho_{\mathrm{bulk}} = \rho_{0,\mathrm{bulk}} + K(T/\Theta_D)^5 \int_0^{\Theta_D/T} \frac{x^5 dx}{(e^x - 1)(1 - e^{-x})}, \qquad (1)$$

where Θ_D is the Debye temperature, K is the prefactor of the Bloch-Grüneisen law, and ρ_0 is the residual resistivity, which is related to the impurity mean free path l_{imp} by $\rho_0 = mv_{\mathrm{Fermi}}/ne^2 l_{\mathrm{imp}}$. Surface scattering of the conduction electrons or size-effect corrections to the resistivity were not considered in the analysis.

In the present experiments, the temperature dependence of the resistivity is investigated for thin, epitaxial, (111)-oriented gold films, which are grown on (0001) sapphire with a Nb seedlayer.[12] The epitaxial quality and flatness of these films allowed us to prepare samples down to a thickness of 2 nm. Epitaxial gold films of the same orientation, which are prepared according to a standard procedure on mica,[6] show a "channeled" structure already at a thickness below 50 nm, so that epitaxial and electrically conducting films with a thickness below 30 nm could not be obtained. This demonstrates the preparational improvement of the present approach. In addition, the grain size in the present samples is about one order of magnitude larger than the film thickness, so that grain-boundary scattering of the conduction electrons can be safely neglected. Thus, it is justified to analyze the size effect of resistivity exclusively in terms of surface scattering, reducing the number of unknown parameters in comparison to previous experiments.

In the classical models,[1,2] the resistivity of thin films ρ_{film} can be deduced from the resistivity of the corresponding bulk material ρ_{bulk} by

$$\frac{\rho_{\mathrm{bulk}}}{\rho_{\mathrm{film}}} = 1 - \frac{3}{2\kappa} \int_0^1 du (u - u^3)(1 - p) \frac{1 - e^{-\kappa/u}}{1 - pe^{-\kappa/u}}, \qquad (2)$$

where $\kappa = t/l$ is the ratio between the film thickness t and the mean free path of conduction electrons l of corresponding bulk material and p is the specularity parameter. Whereas p is a phenomenological parameter in the model of Fuchs-Sondheimer,[1] p is angle-dependent in the model of Soffer[2] and can be deduced from the surface morphology. In the case of vanishing lateral surface correlation, p is related to the surface roughness h by

$$p(u) = \exp[-(4\pi h/\lambda_F)^2 u^2], \tag{3}$$

where λ_F is the Fermi wavelength.

In the course of testing various classical size-effect models like Fuchs-Sondheimer and Soffer, it turned out that fixing the Debye temperature to the bulk value can lead to erroneous results. Particularly for ultrathin films, the influence of the surface leads to a softening of the phonon spectrum, which must be taken into account.

II. PREPARATION

In order to obtain samples combining some quite demanding properties such as being simultaneously ultrathin, epitaxial, flat, and electrically conducting, Au films were prepared on (0001)-oriented sapphire substrates with a thin Nb seedlayer. Details of the preparation are described in Refs. 12 and 13. First a Nb seedlayer (thickness 1 nm) was deposited at room temperature on c-cut sapphire substrates. Next, the Au films (thickness 7–46 nm) were grown on top of the seedlayer at a substrate temperature between 250 and 300 °C. For the ultrathin films (thickness 2–5 nm), the growth temperature had to be lowered to room temperature to guarantee continuous, electrically conducting samples. Epitaxial growth was monitored and verified during growth by reflection high energy electron diffraction (RHEED) and *ex situ* by x-ray diffraction (XRD). Atomic force and scanning tunneling microscopy (AFM and STM) confirmed the small roughness (±1–2 ML) even for the ultrathin Au films. The samples were patterned either by evaporation through masks or by optical lithography combined with wet chemical etching[14] resulting in bridges, which allowed four-probe measurements on different film segments. The length of each segment was 1.5 mm and the width was 30, 50, 100, or 750 μm. The temperature dependence of the sample resistance was measured applying a lock-in technique within a ^{4}He cryostat. To guarantee thermal equilibrium between the Au film and the sample holder, the temperature was ramped slowly (<0.3 K/min) during the resistance measurements.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the electrical resistivity for Au films of different thickness. The residual resistivity ρ_0 at liquid-helium temperature increases for decreasing sample thickness, or, expressed differently, the residual resistance ratio $R(300\text{ K})/R(4\text{ K})$ approaches 1 for that case. Without further analysis, at this point it is not clear, yet, whether the increase of ρ_0 is only due to enhanced surface scattering of the conduction electrons or whether structural defects built-in during the initial stages of film growth contribute additionally.

No explicit influence of the Nb seedlayer on the temperature dependence of resistivity down to $T=1.4$ K was observed. Even though the transition temperature to superconductivity of bulk Nb is $T_c=9.3$ K,[15] such a transition seems to be suppressed within our accessible temperature range. It is known from the literature that T_c decreases with decreasing film thickness t. Extrapolating the previously reported $\log T_c \sim t^{-1}$ dependence of Nb films[15] down to the film thickness of the Nb seedlayer (1.0 nm), one expects a decrease of T_c to 0.25 K, which is consistent with the present resistivity measurements.

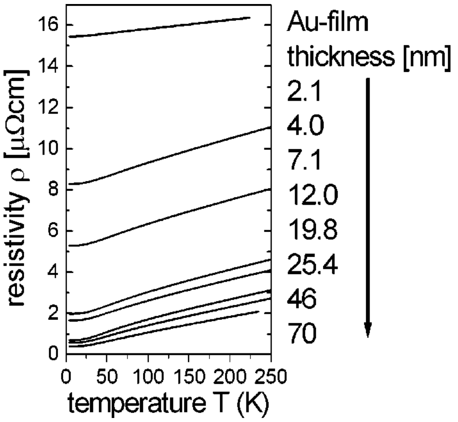

FIG. 1. Temperature dependence of the resistivity of epitaxial Au films on sapphire (thickness between 2 and 46 nm) and on mica (thickness 70 nm) revealing a strong increase of the residual resistivity at liquid-helium temperature for decreasing film thicknesses.

For the analysis of the temperature dependence of the resistivity, the influence of the Nb seedlayer was neglected by arguing that the room-temperature resistivity of Nb ($\rho_{\text{bulk}}=14.5\ \mu\Omega$ cm) is almost an order of magnitude larger than the corresponding Au value ($\rho_{\text{bulk}}=2.2\ \mu\Omega$ cm) (Ref. 16) and therefore electron transport through the Nb seedlayer will be almost completely shorted by the Au film on top. To strengthen this argument, the resistivity of a 1-nm-thick Nb film was determined *in situ* after its growth at room temperature delivering a value of about 300 $\mu\Omega$ cm. This high value justifies neglecting the influence of Nb even for the thinner Au films. Naturally, it still would be desirable to prepare Au films without a Nb seedlayer. It must be pointed out, however, that ultrathin ($\geqslant 2$ nm) Au samples exhibiting the above-mentioned combined properties cannot be obtained without a Nb seedlayer on sapphire.

The experimental data were analyzed in terms of the classical size-effect models of the resistivity mentioned above. For this purpose, a fit program based on the Levenberg-Marquardt method[17] was written to compare the experimental data with those models. Several fit parameters can be varied or fixed to certain values depending on the actual aspect of interest. Electron scattering by phonons and bulk defects is characterized by the prefactor K in the Bloch-Grüneisen equation, the Debye temperature Θ_D, and the residual resistivity of the bulk material ρ_0, respectively. These parameters describe the electron transport in a corresponding bulk sample or a sample with negligible surface scattering. Surface scattering requires additional fit parameters, namely the specularity parameter p for the Fuchs-Sondheimer model, the surface roughness h for the Soffer model or the grain size g, and the reflection coefficient R for the Mayadas-Shatzkes model (see Refs. 7 and 8 for details). In order to reduce the number of fit parameters to get unambiguous results, the film thickness was determined by Laue oscillations from XRD measurements (see Fig. 2). Simulation of such spectra by the

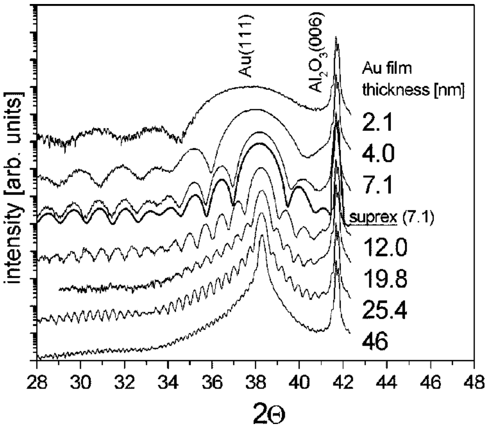

FIG. 2. $2\Theta/\Omega$ x-ray-diffraction scans of Au films with different thicknesses all show clear Laue oscillations of the Au(111) reflection. Even though the position of the Au(111) peak shifts to smaller values for the thinner films due to the influence of stress, the pronounced Laue oscillations indicate extremely flat and epitaxial films. A spectrum simulated with the SUPREX program is shown for the 7.1-nm-thick Au film (thick line).

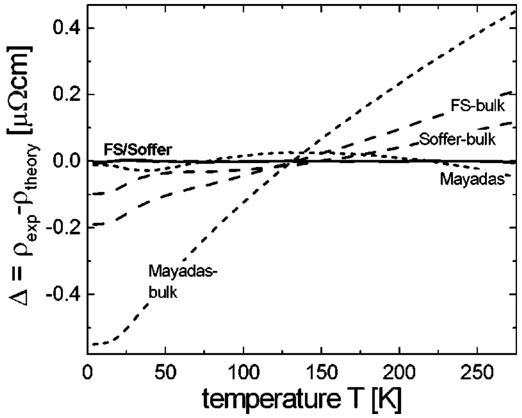

FIG. 3. Deviations between fits to various size-effect models and experimental data obtained for the temperature dependence of the resistivity for a 7.1-nm-thick epitaxial Au film. The different curves result from different models and their assumptions: For FS bulk, Soffer bulk, and Mayadas bulk, Θ_D and the prefactor K of Eq. (1) were fixed to the bulk values of Au, whereas for FS, Soffer, and Mayadas these quantities are used as fit parameters.

SUPREX program[18] gives an excellent agreement with the experimental data, as is demonstrated in Fig. 2 for the 7.1-nm-thick Au film (the quality of the fits to the other films is similar, but the curves are omitted here for clarity). As a result, additional information can be obtained on the roughness of the various films delivering a value of 1-2 monolayers (ML) for the standard deviation.

In a first step, the analysis of the resistivity data was performed with $K=5.2525$ $\mu\Omega$ cm and $\Theta_D=184.59$ K obtained from fitting the temperature dependence of the resistivity of Au bulk samples.[19] These values of K and Θ_D compare well with parameters found in the literature.[20–22] The temperature dependence of the resistivity for Au films with a thickness larger than 20 nm could be modeled fairly well with these values of K and Θ_D, but for thinner Au films there was a significant discrepancy between theory and experiment. An example of this disagreement is presented in Fig. 3, where the results for a 7.1-nm-thick Au film are shown. For that purpose, the differences $\Delta(T)$ between the fitted theoretical and experimental resistivity curves are plotted versus temperature. As demonstrated in the figure, $\Delta(T)$ deviates significantly from zero when K and Θ_D are fixed to the bulk values of Au (FS bulk and Soffer bulk). This result suggests a principal incompatibility between theoretical assumptions and experimental conditions. Additional support for this conjecture comes from the fact that both fits result in complete diffuse surface scattering ($p=0$), which is not compatible with previous reports on Au films.[23]

When K and Θ_D were used as additional fit parameters (FS and Soffer in Fig. 3), the pronounced deviations $\Delta(T)$ between theory and experiment disappear. This is not just the result of increasing the number of fit parameters, but rather due to a significant decrease of Θ_D.

One assumption common to both models, FS as well as Soffer, is usually not met by experiment. While in the models the film surface as well as its interface with the substrate are assumed to be identical with respect to roughness or specularity, this assumption cannot be guaranteed experimentally. By introducing two specularity or roughness parameters p_1 and p_2 (or h_1 and h_2) for the surface and the interface, respectively, the addressed experimental asymmetry can be taken into account.[24,25] In the present case, however, it turned out that p_1 and p_2 (or h_1 and h_2) converged to a common value, respectively, in the fits. To the best of our knowledge, in the literature two different specularity parameters were only used to interpret the thickness dependence of the resistivity of thin metal films[26,27] and not to describe their temperature behavior. The extracted results, however, appear quite arbitrary and are far from unambiguous. For that reason, and since p_1 and p_2 (or h_1 and h_2) always converged to a common value in the present experiments, we found it justified to describe the electron scattering at both sample boundaries by one single parameter.

A similar analysis was performed comparing the measurements to the Mayadas-Shatzkes model. Even though the number of fit parameters is larger in that case (average grain size g, electron reflection coefficient at grain boundaries R) than in the FS (p) or Soffer model (h), the disagreement $\Delta(T)$ between theory and experiment grew significantly in this case. Furthermore, the large scatter of the extracted fit parameters g and R, which even depended on their starting value in the fit algorithm, points to a negligible influence of grain boundary scattering. This conclusion is consistent with the large grain size in the present high-quality epitaxial Au films. This size can be estimated from the average terrace size measured by AFM or STM.[12] Since this experimentally determined grain size is at least an order of magnitude larger than the film thickness, the effect of grain-boundary scattering—if present at all—can therefore be safely neglected. For that reason, an analysis combining surface (FS or Soffer) with grain-boundary scattering[6,8] was not carried out. This is also justified by the results given in Fig. 3, where the excellent agreement between the classical size-effect model of FS or Soffer and the experimental data is demonstrated.

TABLE I. Fit parameters obtained from the temperature dependence of resistance for Au films of different thickness. The analysis was performed with the size-effect models of Fuchs-Sondheimer (FS) and Soffer.

	t (nm)	K ($\mu\Omega$ cm)	Θ_D (K)	l_{imp} (nm)	$p,h/\lambda_F$
FS	46.0	5.82	179.5	765	0.20
	25.4	6.23	187.3	1138	0.27
	19.8	5.88	168.9	207	0.16
	12.0	5.88	168.4	204.0	0.28
	7.1	5.84	158.2	54	0.15
	4.0	5.28	156.4	35	0.25
	2.1	5.09	129.0	3.9	0.05
Soffer	46.0	5.00	158.9	282	0.18
	25.4	4.75	154.0	316	0.22
	19.8	5.11	158.3	110.7	0.26
	12.0	4.70	155.5	111	0.27
	7.1	5.08	155.1	37	0.34
	4.0	4.30	154.6	27	0.34
	2.1	4.97	128.6	2.8	0.15

In the following, we will provide additional arguments allowing us to distinguish further between those two classical models. For this purpose, the values of the Debye temperature of our Au films obtained by fitting the experimental data to both classical size-effect models are analyzed (see Table I for the full set of parameters). The results are shown in Fig. 4 as a function of the thickness of the Au films. Clearly, for films with a thickness above 25 nm, the analysis within the FS model yields Debye temperatures close to the Au bulk value of ~185 K. For films with a thickness below 20 nm, however, the Debye temperature is strongly reduced.

It is well known from diffraction experiments on single-crystal surfaces that the surface Debye temperature is smaller than the corresponding bulk value.[28] In LEED experiments on Au crystals, the surface Debye temperature was found to be reduced to about 83 K.[29] Compared to Θ_D in bulk material, this means a reduction by about 50%. Physically, the depression of Θ_D is attributed to the lowered coordination number of surface atoms leading to their enhanced vibrational amplitudes. As a result, the spectrum of the surface phonons is softened as compared to the bulk behavior, thus giving smaller values of Θ_D.[30]

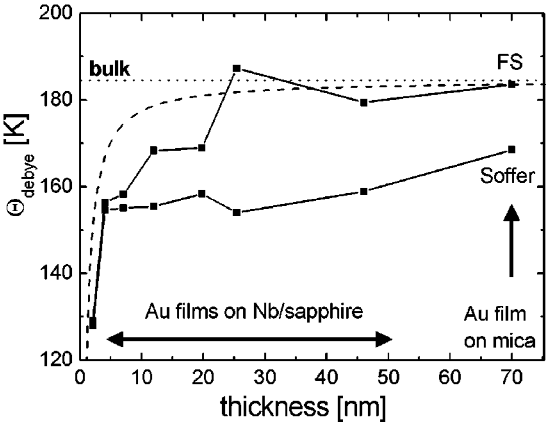

FIG. 4. Debye temperatures Θ_D as extracted from fitting experimental data to the models of Fuchs-Sondheimer and Soffer plotted vs film thickness (closed squares). The solid lines connecting the data points are just guides to the eye. The dashed curve represents the result of a simple averaging procedure assuming that 3 ML of the film contribute with a surface Debye temperature of 83 K rather than with the bulk value. The dotted horizontal line indicates the bulk value Θ_D of Au.

In addition to diffraction experiments, in metals Θ_D can also be extracted from the temperature dependence of the resistivity as a result of the electron-phonon coupling. Since in a thin film the corresponding electron-phonon scattering events occur at or close to the surface as well as within the film, the extracted Debye temperature is expected to represent an effective value with contributions from the bulk and the surface. In a first approximation, we estimate the effective value of Θ_D by assuming that three surface monolayers of an Au film[30] contribute with a value of 83 K and the remaining sample behaves like bulk. The resulting average values are given by the dashed line in Fig. 4. This rough estimate describes the Θ_D values as obtained from fitting to the FS model surprisingly well, adding additional confidence at least to the qualitative trends found for Θ_D.

When doing the same analysis as above but assuming the model of Soffer, qualitatively the same trend is observed for the Debye temperature. Quantitatively, however, there is a significant difference. Even for Au films as thick as ~70 nm, Θ_D is still well below the bulk value. Quite generally, the Soffer model delivers too small Θ_D values for films thicker than ~20 nm. Furthermore, the values extracted from the same fits for the surface roughness h ranged between 0.78 Å and 1.7 Å, which, being significantly smaller than the interatomic distance of Au ($d=2.35$ Å), are physically unreasonable. At this point, one might argue that the assumption[25,31] in Eq. (3) of a surface roughness profile with a vanishing correlation length L is not suitable for the present Au films. To exclude that possibility, 25-nm-thick Au films were exposed to oxygen/hydrogen plasmas before measuring the temperature dependence of resistivity. Due to these plasma treatments, the film surface is oxidized first to a depth of ~3−4 nm by the oxygen plasma and subsequently reduced back into the metallic state by the hydrogen plasma. These consecutive oxidation and reduction steps result in a noticeable roughening of the Au surface.[32] But even for these films with their surface morphology artificially changed towards a higher roughness, the resistivity—if analyzed within the Soffer model including $L=0$—delivers almost the same values for Θ_D and h as without the artificial roughening. Thus, it is concluded that the parameters extracted from the Soffer model do not sensitively depend on the details of the assumed surface roughness profiles. As a consequence, the clearly too small Θ_D values delivered by this approach point to a principal weakness of the model. There are, on the other hand, previous experiments[6] on epitaxial Au films on mica (film thickness between ~50 and 200 nm) leading to the opposite conclusion that the model of Soffer is more appropriate to describe the temperature dependence of resistivity. Since, however, in that case grain boundary scattering could not be excluded and a value of $\Theta_D \sim 160$ K is used, which for the range of film thicknesses analyzed in that study is too small compared to the bulk value, this conclusion is not convincing. Furthermore, too small values were obtained for the surface roughness.[31] In contrast to those previous results, the present experiments on epitaxial Au films lead to the conclusion that the contribution of surface scattering to their resistivity is consistently described by the original model of Fuchs-Sondheimer, even though the model of Soffer is physically more appealing.

While the comparison between the classical size-effect theories and the experimental data leads to an unequivocal conclusion in favor of the FS model, the influence of electronic quantization on the temperature dependence of resistivity remains unclear. Even though the motion of electrons perpendicular to the ultrathin and flat Au films is highly restricted, no obvious related deviation of the experimental behavior from the classical FS size-effect model was observed. Whereas the expected stepwise change of the density of occupied states should lead to observable effects in the thickness dependence of the resistivity, a related influence of level quantization on the rather smooth temperature dependence of resistivity is supposedly less distinct. In addition, small thickness fluctuations might lead to smearing of any small characteristics in the experiment,[4] and the low probability of specular scattering $(5-28\%)$ possibly hinders the development of deviations from the classical behavior. At this point it remains an open question whether the absence of a fingerprint of quantization in the temperature dependence of resistivity is either due to the smallness of the correction or to experimental reasons leading to blurring of distinct features.

IV. CONCLUSION

The temperature dependence of resistivity was measured for thin epitaxial Au films with thicknesses between 2 and 70 nm. Due to the significantly improved structural quality of these samples, for the first time the contribution of grain boundary scattering to the experimental resistivity behavior can be safely excluded. As a consequence, the number of parameters necessary to describe the results theoretically is reduced, making such an analysis much more meaningful. Consistently, applying the Mayadas-Shatzkes model, which is based on this type of scattering, to fit the experimental results led to the strongest deviations between experiment and theory. Analysis based on the models of Fuchs-Sondheimer and Soffer, respectively, clearly revealed the necessity to include a thickness-dependent decrease of the Debye temperature for Au films below 30 nm. Quantitatively, however, it turned out that the Soffer model delivers values for both the Debye temperatures as well as the roughness parameters, which are too small to be physically acceptable. Thus, the experimental results lead us to conclude that among the various size-effect theories, the model of Fuchs-Sondheimer is most appropriate to describe the effect of thickness on the temperature dependence of our high-quality Au films.

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[1] E. Sondheimer, Adv. Phys. **1**, 1 (1952).
[2] S. Soffer, J. Appl. Phys. **38**, 1710 (1967).
[3] G. Palasantzas and J. T. M. DeHosson, Phys. Rev. B **63**, 125404 (2001).
[4] A. E. Meyerovich and I. V. Ponomarev, Phys. Rev. B **65**, 155413 (2002).
[5] D. Schumacher, Thin Solid Films **152**, 499 (1987).
[6] J. Sambles, K. Elsom, and D. Jarvis, Philos. Trans. R. Soc. London, Ser. A **304**, 365 (1982).
[7] A. Mayadas, M. Shatzkes, and J. Janak, Appl. Phys. Lett. **14**, 345 (1969).
[8] A. Mayadas and M. Shatzkes, Phys. Rev. B **1**, 1382 (1970).
[9] P. vanAttekum, P. H. Woerlee, G. C. Verkade, and A. A. M. Hoeben, Phys. Rev. B **29**, 645 (1984).
[10] M. Mukherjee, S. Saha, and D. Chakravorty, Appl. Phys. Lett. **63**, 42 (1993).
[11] S. Kim, H. Suhl, and I. K. Schuller, Phys. Rev. Lett. **78**, 322 (1997).
[12] G. Kästle, H. Boyen, B. Koslowski, A. Plettl, F. Weigl, and P. Ziemann, Surf. Sci. **498**, 168 (2002).
[13] G. Kästle, *Electron Transport in Nanomodulated, Epitaxial Gold Films*, Forschritt Berichte VDI, Reihe 9 (VDI Verlag, Düsseldorf, 2003).
[14] W. Eidelloth and R. Sandstrom, Appl. Phys. Lett. **59**, 1632 (1991).
[15] S. Wolf, J. Kennedy, and M. Nisenoff, J. Vac. Sci. Technol. **13**, 145 (1976).
[16] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1996).
[17] W. Press, B. Flannery, S. Teukolsky, and W. Vetterling, *Numerical Recipes in Pascal* (Cambridge University Press, Cambridge, 1989).
[18] E. E. Fullerton, I. K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, Phys. Rev. B **45**, 9292 (1992); E. E. Fullerton, J. Guimpel, O. Nakamura, and I. K. Schuller, Phys. Rev. Lett. **69**, 2859 (1992); E. E. Fullerton, J. Pearson, C. H. Sowers, S. D. Bader, X. Z. Wu, and S. K. Sinha, Phys. Rev. B **48**, 17 432 (1993).
[19] J. Mydosh, P. Ford, M. Kawatra, and T. Whall, Phys. Rev. B **10**, 2845 (1974).
[20] N. Karpe, G. Lapogian, J. Bottiger, and J. Krog, Philos. Mag. B **71**, 445 (1995).
[21] G. White and S. Woods, Philos. Trans. R. Soc. London, Ser. A

251, 273 (1959).

[22] J. Jacobs, R. Birtcher, and R. Peacock, J. Vac. Sci. Technol. **7**, 339 (1969).

[23] J. W. F. Egelhoff, P. Chen, C. Powell, D. Parks, G. Serpa, R. McMichael, D. Martien, and A. Berkowitz, J. Vac. Sci. Technol. B **17**, 1702 (1999).

[24] M. Lucas, J. Appl. Phys. **36**, 1632 (1965).

[25] J. Sambles and K. Elsom, J. Phys. D **15**, 1459 (1982).

[26] G. Reeves, M. Lawn, and R. Ełłiman, J. Vac. Sci. Technol. A **10**, 3203 (1992).

[27] M. Lucas, Appl. Phys. Lett. **4**, 73 (1964).

[28] C. Waldfried, D. McIlroy, J. Zhang, P. Dowben, G. Katrich, and E. Plummer, Surf. Sci. **363**, 296 (1996).

[29] M. Kostelitz and J. Domange, Solid State Commun. **13**, 241 (1973).

[30] A. Al-Rawi, A. Kara, and T. Rahman, Phys. Rev. B **66**, 165439 (2002).

[31] U. Jacob, J. Vancea, and H. Hoffmann, Phys. Rev. B **41**, 11 852 (1990).

[32] B. Koslowski, H.-G. Boyen, C. Wilderotter, G. Kästle, P. Ziemann, R. Wahrenberg, and P. Oelhafen, Surf. Sci. **475**, 1 (2001).