Thermal behavior of $c(2\times2)$ layers in the Pb/Cu(110) system and the influence of surface defects on the order-disorder transition

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(Received 26 May 1999)

The thermal behavior of a $c(2\times2)$ Pb overlayer on Cu(110) surface is studied by thermal energy atom scattering. Diffraction analysis reveals a continuous reversible order-disorder transition with temperature at 437 ± 2.5 K. Two of the extracted critical exponents, γ and ν, are in good agreement with those of the two-dimensional Ising universality class. The third, β, associated with the disappearance of long-range order in the layer, is, however, smaller than the theoretical expectations. The observed transition is discussed in a framework of model, where the deviation of one of the exponents β, γ, and ν from its exact value depends in a different way on the kind of ''imperfection'' (mean terrace width, domain boundaries, ''frozen-in'' surface defects, local distortions, and surface alloying) in the real physical system. The annihilation of domain walls, created during the simultaneous growth of $c(2\times2)$ phase on two equivalent sublattices of {110} fcc surface, is found at 310 K. The experimental findings indicate also a noticeable change in the surface corrugation during the observed order-disorder transition.

I. INTRODUCTION

During the last few years, considerable interest was devoted to two-dimensional (2D) metal layers on metal substrates. Following the progress made in high-resolution diffraction techniques such as spot profile analysis low-electron energy diffraction (SPA LEED), thermal energy atom scattering (TEAS), and scanning tunneling microscopy (STM), many systems already studied in the early 1970s are now being reconsidered. One of the most studied is surely the case Pb/Cu. The low melting point of lead, its chemical inertness, and the lack of miscibility of the two metals are obvious reasons for this choice. A large variety of structural phase transitions on differently oriented copper substrates has been observed:[1] a succession of centered structures in the case of the Pb/Cu(100),[2–5] one centered and a series of quasi-unidimensional primitive structures for Pb/Cu(110),[6–9] incommensurate strongly compressed phase for Pb/Cu(111).[10] Most of the above structures are considered as a system with localized adsorption states. The theory predicts that the continuous order-disorder phase transitions for such systems may exhibit different universality class behavior based on the symmetry considerations.[26,11,12] Experimentally the critical behavior of 2D adsorbed phases has been studied extensively.[20,26] Obviously, the most studied case is that of the Ising universality class because of its simplicity and exact mathematical solution.[13,14] With respect to that, the detailed study of the system $O/W(112)$ reported by Wang and Lu[15] is considered a work that bridges the theory and experiment in the field of 2D critical phenomena.

Some attempts to study the critical behavior of Pb/Cu structures[1–3] by helium scattering have shown that the large Debye-Waller factor and the small corrugation of the surface potential are limiting the determination of the critical exponents associated with order-disorder transitions. In the particular case of the $c(2\times2)$ structure of Pb/Cu(110), however, the diffraction peaks are sufficiently intense[16,17] to enable a complete analysis of their temperature dependence. In this paper we present the results of such an analysis. As a second topic we discuss the nature of the surface imperfections and their influence on the 2D critical behavior of the system.

II. EXPERIMENT

The experiments were performed on the He-scattering apparatus described in detail elsewhere.[16] The wavelength, λ of the thermal energy He beam was equal to 0.57 Å, $\Delta\lambda/\lambda = 1.5\%$, and its transfer width was estimated 200 Å in all scattering experiments. The temperature was measured with a chromel-alumel thermocouple welded on the front side of the copper substrate, the temperature uncertainty being less then 1 K throughout the sample in the whole temperature range. The residual pressure during the experiments was less than 1.5×10^{-10} Torr. Lead was deposited from a home-built Knudsen cell. By monitoring the intensity of several diffraction peaks during lead deposition, a sharp maximum was observed at the completion of the $c(2\times2)$ layer.[16,17] This

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maximum, corresponding to a coverage $\theta=0.5$ was further used for the calibration of the outcoming Pb flux from the Knudsen cell versus cell temperature. The coverage θ is defined as the ratio of adsorbed atoms to the maximum number of substrate adsorption sites.

III. RESULTS AND DISCUSSION

The very early stage of Pb adsorption on the Cu (110) surface was studied by monitoring specular (0,0), $(-1,0)$, and $(-1/2,-1/2)$ diffraction peaks. Below the coverage $\theta=0.35$ ML, we observed a lattice-gas formation where random adsorption of lead atoms takes place on fourfold symmetry sites on the Cu(110) crystal surface. Increasing the coverage, a two-level scattering was found in the range of $0.25<\theta<0.4$ ML. The corresponding height difference, $h=1.21\pm0.1$ Å , evaluated from so-called ''rocking curves'' of $(-1,0)$ diffraction peak (intensity versus normal component of the scattering vector $K_\perp$) indicates a process of intermixing, where adsorbed Pb atoms of the lattice gas replace Cu atoms of the outermost substrate layer.[17] This effect of surface alloying in the system Pb/Cu(110) was recently studied by Monte Carlo simulations in a model with a ''realistic'' tight-binding potential via coverage-dependent interactions.[18,19] Increasing the coverage the intensity of the $(-1/2,-1/2)$ diffraction peak, originating from the $c(2\times2)$ structure, grows continuously and reaches its maximum at $\theta=0.5$ where this structure is entirely completed. In accordance with the theory,[21–24] TEAS experiments clearly demonstrate that above the coverage 0.4 ML a process of demixing takes place, which leads to the formation of pure, not alloyed $c(2\times2)$ layer.[17] Thus the condition for a closed, one-level two-dimensional system is fulfilled. This is quite important since the system has to satisfy the requirements of the theory of 2D critical phenomena. We shall discuss later the problem of the conservation of the total number of adatoms within one layer, which also seems to be satisfied. It is important to be noticed that all experiments related to order-disorder transitions in the system $c(2\times2)$ Pb/Cu(110) are carried out on the complete $c(2\times2)$ layer at $\theta=0.5$ ML.

After the formation of the $c(2\times2)$ structure the system was heated up to 500 K. This annealing leads to relatively better thermodynamic equilibrium by elimination of some defects and domain walls. The latter are formed during the simultaneous growth of $c(2\times2)$ phase on two equivalent sublattices of {110} fcc surface. As it is shown on Fig. 1, the temperature of annihilation T_a of domain walls is about 310 K, where $(-1/2,-1/2)$ peak intensity starts to increase, contrary to Debye-Waller effect. Once the annihilation of the domain walls is completed the temperature variation of the intensity becomes again dominated by the Debye-Waller dependence. The result is a relatively well-ordered, $c(2\times2)$ structure with some distribution of domain walls due to the surface imperfection, ''frozen-in'' defects, and final width of terraces. The relaxed structure produces sharp $(-1/2,-1/2)$ diffraction spots, both LEED and TEAS.

The order-disorder transition of the $c(2\times2)$ phase was studied on the $(-1/2,-1/2)$ diffraction peak. Above $T=400$ K its intensity falls abruptly and reaches a value close to zero for $T\geqslant520$ K, the process being reversible. It has been checked that in the whole temperature range no lead

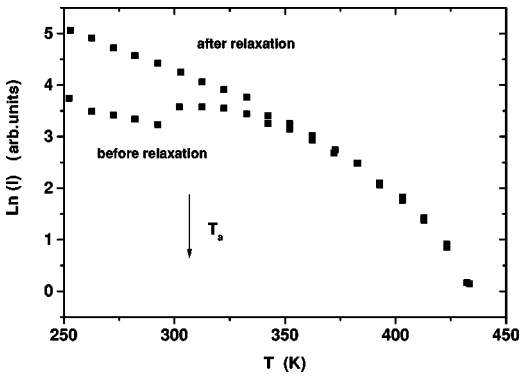

FIG. 1. Debye-Waller dependence of the normalized intensity of $(-1/2,-1/2)$ diffraction peak. The temperature of annihilation of domain walls is about 310 K. The upper curve corresponds to $c(2\times2)$ structure after a thermal relaxation of the lead layer.

desorption occurs. Furthermore, the specular (0,0) intensity of the clean Cu surface exhibits a standard Debye-Waller decrease for $T<550$ K.[25] Both normalized intensity I of the $(-1/2,-1/2)$ peak (after correction for background and Debye-Waller effect), and full width at half maximum (FWHM), versus temperature, are presented in Fig. 2. In the considered temperature range, the FWHM increases by a factor of 2.

The above observations are consistent with a continuous phase transition of a 2D closed system. According to Landau's theory, the continuous transition in 2D phase with $c(2\times2)$ structure on a substrate of $p2mm$ symmetry, belongs to the Ising universality class.[26,27] The critical exponents of the function

$$A(T)=C_m|T-T_c|^m, \tag{1}$$

where C_m is a constant and $A(T)$ stands alternately for

(1) $I_{\mathrm{Br}}(T)$, Bragg's intensity as a long-range order contribution at $T<T_c$, $(m=2\beta)$;

(2) $I_D(T)$, diffuse-scattering intensity as the contribution of the short-range correlations at $T>T_c$, $(m=-\gamma)$;

(3) $\xi(T)$, inverse correlation length between fluctuations, proportional to FWHM at $T>T_c$, $(m=\nu)$, can be extracted from the analysis of the diffraction spectra. In that case the

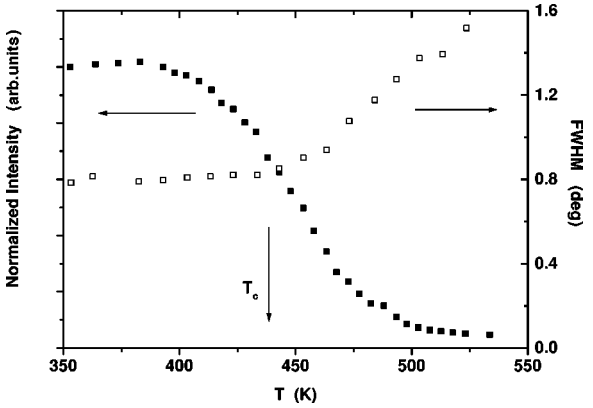

FIG. 2. Normalized intensity (divided by the Debye-Waller factor), filled squares, and FWHM, empty squares, of the $(-1/2,-1/2)$ diffraction peak vs temperature.

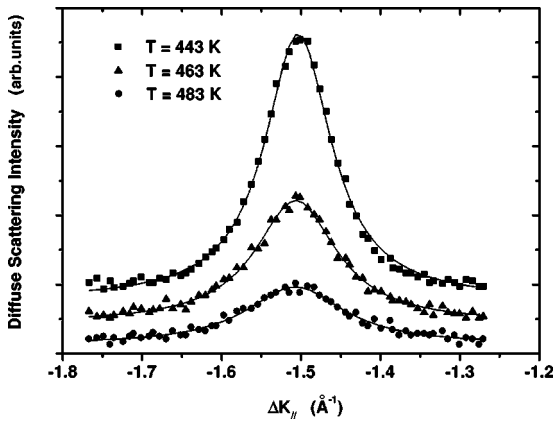

FIG. 3. $(-1/2,-1/2)$ peak profile at different temperatures. Solid curves: convolution of a parameterized Lorentzian function with instrument response function.

theory predicts the following exact values for the Ising model: $\beta=0.125$, $\gamma=1.75$, and $\nu=1$.

In order to evaluate the coefficients β, γ, and ν, we proceeded as follows.[2,3,15] The diffraction spectra were fitted by convolution of a Lorentzian function (using the experimentally measured parameters, I and FWHM) with our instrumental response function. The latter was obtained from the angular profile of the peak at low temperature (320 K). The results of the convolution procedure for three substrate temperatures are shown in Fig. 3. The temperature dependence of the FWHM and the intensity of the adjusted Lorentzian, are then fitted with the function $A(T)$ of Eq. (1).

Applying the procedure described above we obtained the following values for T_c, γ, and ν: $T_c=437\pm2.5K$, $\gamma=1.81\pm0.12$, and $\nu=1.03\pm0.09$ [cf. Figs. 4(a) and 4(b)], the two critical exponents being in very good agreement with the theoretical values for the Ising universality class.

The determination of the long-range order coefficient β is a more difficult task and needs the real physical characteristics of the system to be taken into account in more detail. The deviation from a vertical tangent of the $I(T)$ curve at $T=T_c$ can be clearly noticed in Fig. 2. The rounding of the curve that indicates a ''smearing'' of the transition can be assigned to the presence of ''frozen'' antiphase boundaries[15] and to imperfections of the substrate. The relatively low transfer width of our TEAS instrument (200 Å) additionally restricts the experimental precision. Both contributions can be considered as finite-size effects affecting not only the coefficient β, and but also the critical temperature.[29] This problem will be discussed in detail later. Nevertheless, two different methods were applied to extract β.

First, for $T<T_c$ we used the functional form of Eq. (1), with $A(T)$ the intensity $I_B(T)$ of the $(-1/2,-1/2)$ diffraction peak and $m=2\beta$. We looked for a linear behavior of the double logarithmic plot at three different values of T_c [Fig. 4(c)]. Straight lines were obtained in the temperature ranges T_c-10 K to T_c-45 K for 435 K$<T_c<$440 K, with slopes, 2β, varying from 0.127 to 0.155. The as determined critical coefficient is then two times smaller than the expected theoretical value ($\beta_{\rm th}=0.125$).

Second, a procedure generally used, consisting in introduction of a Gaussian distribution of critical temperatures and fitting the measured peak intensity for $T<T_c$ with the formula:[2,3,15]

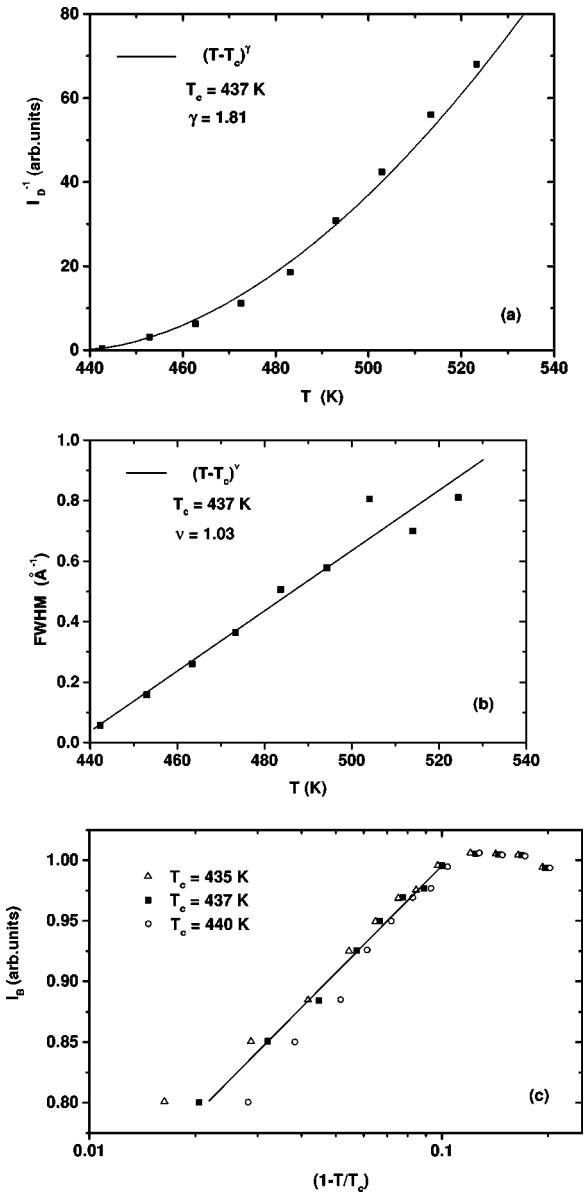

FIG. 4. (a) Reciprocal intensity of the Lorentzian function vs temperature. Solid line: power-law fit. (b) FWHM of the Lorentzian function vs temperature. Solid line: power-law fit. (c) $(-1/2,-1/2)$ normalized peak intensity vs logarithm of the reduced temperature. The slope of the straight line gives the value $2\beta=0.14$.

$$I_{\rm Br}(T)=\frac{I_0}{\sqrt{2\pi\Delta t}}\int_{T-T_c}^{\infty}\left(\frac{T_c-T+t}{T_c+t}\right)^{2\beta}\exp\left[-\frac{1}{2}\left(\frac{t}{\Delta t}\right)^2\right]dt, \tag{2}$$

where Δt is the FWHM of the Gaussian distribution for T_c and I_0 is a constant, allowed us to enlarge the fitting region near T_c where the rounding of the $I(T)$ curve appears, but does not permit its full fit. This procedure led to similar values of $2\beta=0.128$, as previously obtained, with reasonable estimates for Δt (4 K).[15,28] Thus our experimental findings demonstrate very good agreement with theoretical predictions of Ising universality class for the critical exponents γ, ν, and deviation for the critical exponent β. To explain this discrepancy it is appropriate to divide the influence of system ''imperfections'' on the critical behavior in two parts:

(i) contribution of large scale finite-size effects: mean terrace width, antiphase domain boundaries, transfer width of the analyzer;

(ii) contribution of local imperfections: point defects, lattice distortions, surface alloying.

Considering the nature of the coefficients β, γ, and ν, we have to take into account that the last two, γ and ν are extracted at $T>T_c$ and they reflect the short-range order fluctuations in the system. On the contrary, coefficient β, associated with the long-range order parameter, is defined below the critical point at $T\leqslant T_c$ where the system is ordered. It is well known that finite-size effects, e.g., mean terrace width, antiphase boundaries, point defects, etc., influence the behavior of the real system in the critical region. That is the case when the correlation length is comparable with the system size.[29,30] Apparently, the relatively small mean terrace width of 100 Å of the crystal used in the experiments may substantially influence the behavior of the critical exponent at $T\leqslant T_c$. On the contrary, in the region $T>T_c$, i.e., in the disordered state of the 2D layer, the correlation length is the distance over which ordered regions extend, since the range it measures refers to fluctuations about domains with no long-range order. Hence, when the correlation length becomes smaller than system size, the short-range order fluctuations will not be strongly affected by boundary effects. Accordingly, our experimental findings gave for the critical exponents that describe the fluctuation of the short-range order γ and the decay of the correlation length ν, values close to those appropriate for the Ising universality model. Thus the first group of the system ''imperfections,'' defined as large-scale finite-size effects, influence essentially the β coefficient, but have less effect on γ and ν.

On the other hand, it is evident that not only large-scale finite-size effects and low transfer width of the experimental analyzer contribute to deviation from the ''ideal'' order-disorder transition behavior. The influence of local surface imperfections should be taken into account, since they act in the same range as short-range fluctuations in the system above T_c. As a typical example of such local imperfections consider 2D surface alloying. This process of intermixing between Pb and Cu atoms at the system interface was experimentally observed[9,17] and may affect the critical exponents too. Recently, Monte Carlo simulations on Pb/Cu(110) demonstrated that partial surface alloying could take place even at the completed $c(2\times2)$ phase ($\theta=0.5$ ML) (Refs. 18 and 19) being not completely reversible with temperature process. Once the system has been thermally treated to reach the equilibrium state, on going back to low temperature some part of the Pb atoms remains embedded in the topmost Cu layer. Thus additional ''frozen-in'' distortions and local defects due to alloyed atoms change the ''perfection'' of both substrate and 2D adsorbed layer. In some particular cases (high concentration of alloyed atoms) the presence of local surface defects may dominate the effect of finite system size on the critical behavior. The Monte Carlo results show a very small shift (-0.25%) of the ''effective'' transition temperature obtained in simulations at different system sizes.[18] This strongly suggests that the finite-size effects imposed by the relatively small mean terrace width in the experimental study cannot only explain by itself the observed different decay of a long-range order. Nagl *et al.*[31] have shown by STM measurements that Pb atoms predominately decorate the step edges and incorporate (alloy) through these edges in the Cu matrix of the upper terraces. This process can influence the decay of long-range order in two ways. First, it may lead again to an additional effective smearing out of the power of steps to disrupt the critical fluctuation across them. This can lower the symmetry from that of the flat surface, thus, critically violating the theoretical expectations based on the symmetry considerations. Second, preserving part of the Pb atoms on the terraces in substitutional positions of Cu atoms even after high-temperature annealing. That may change the energetics in the adsorbate layer and in that way influence the nature of long-range order decay.[32] Unfortunately STM measurements have not been carried out on the high-temperature disordered state of $c(2\times2)$ phase of Pb/Cu(110). Thus there is not an experimental evidence for surface alloying for the complete $c(2\times2)$ phase. For the time being only Monte Carlo results indicate the presence of some ''frozen-in'' defects in the disordered state for the above system. This effect, in general, can be due to the approximation done in the many-body potential parameters, used in the simulation. In the specific case of Pb/Cu(110), the good experimental values obtained for the critical coefficients γ and ν speak in favor of absence or very small amount of defects or alloyed atoms in the disordered state. That indicates that the system most probably conserves the total number of atoms within one layer during the order-disorder transition.

It is evident that local surface imperfections described above, can affect mainly the critical coefficients γ and ν. Apparently, this is not the case in our experimental study since the extracted γ and ν are found to be in very good agreement with theoretical expectation for the Ising universality class. The above considerations suggest that the deviation of the critical exponents from their exact values depends in a different way on both the kind and size of ''imperfections'' in the real physical system.

Finally, taking into account the general behavior of the system $c(2\times2)$ Pb/Cu(110), the experimentally determined critical exponents γ and ν being in very good agreement with theoretical expectations and the above-described model in which the extracted critical exponents are influenced mainly by large-scale finite-size effects (in a different way below and above T_c) it could be concluded that Pb/Cu(110) layer with $c(2\times2)$ structure reflects the basic characteristics of the 2D Ising-type continuous order-disorder transition.

The results of Fig. 5(a), displaying the temperature variation of intensity and the FWHM of the $(-1,0)$ diffraction peak, indicate, however, that the order-disorder transition is accompanied by some secondary effects. Breaks in the curves are clearly seen, whereas in the case of Ising transition, the existence of two $c(2\times2)$ sublattices should not have any influence on this particular diffraction peak, because both of them are in phase. No variation of the peak position was detected and the transition is reversible within the experimental accuracy. By contrast, the FWHM of the (0,0) peak stays constant up to 550 K, while its intensity is described by two successive linear Debye-Waller plots, the break occurring at about $T=430$ K [Fig. 5(b)]. Such a phenomenon has been already observed for the (2×1)–(1×1) transition on the (110) surface of gold, and attributed to the

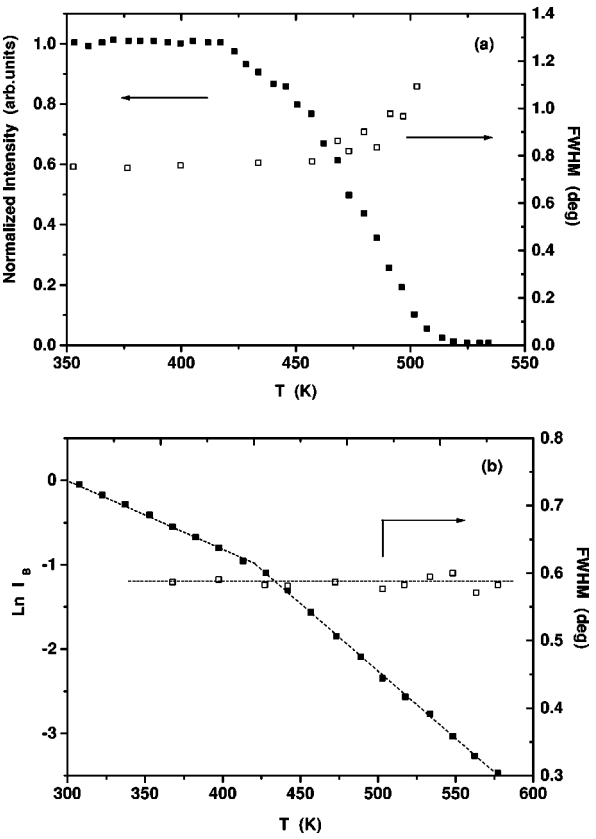

FIG. 5. (a) Normalized intensity divided by the Debye-Waller factor and FWHM of the $(-1,0)$ diffraction peak vs temperature. (b) Logarithm of the specular intensity and FWHM vs temperature.

increase, during the Ising-type transition, of the density of phonons polarized perpendicularly to the surface.[33]

The peculiar behavior of the $(-1,0)$ peak could not be explained by melting of the $c(2\times2)$ layer. Against this hypothesis speaks the lack of displacement of this peak, which indicates that the disordered structure is governed by the substrate potential.

The decrease of intensity of the $(-1,0)$ peak may be due to change in the surface corrugation of the $c(2\times2)$ layer during the order-disorder transition. This assumption was checked by comparison of the rocking curves of the (0,0) diffraction peak at the completion of the $c(2\times2)$, below and above the critical temperature (cf. Fig. 6). The important intensity oscillations at $T=373$ K at large incident angle disappear completely at $T=453$ K. The peculiar features in the specular intensity below the critical temperature are interpreted by the presence of selective absorption resonances

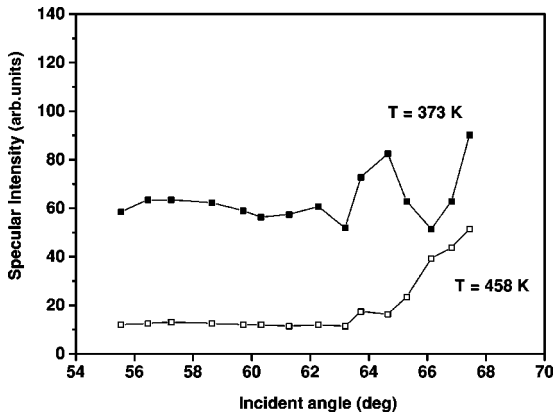

FIG. 6. Specular intensity vs incident angle at the completion of the $c(2\times2)$ layer, below and above the critical temperature.

that are typical for strongly corrugated surfaces.[34,35] Thus, increasing the temperature, the order-disorder transition of the $c(2\times2)$ structure seems to be associated with another transition from a strongly corrugated to a fairly flat surface.

IV. CONCLUSIONS

The temperature behavior of the ordered 2D phase $c(2\times2)$ in the system Pb/Cu(110) is studied by thermal-energy atom scattering. During the thermal relaxation of the layer domain-wall annihilation is observed in the range of 310 K. Increasing the temperature an order-disorder transition at 437 (±2.5) K of the 2D phase takes place. The general behavior of the system indicates that this transition reflects some basic characteristics of 2D Ising universality class. In particular, the values of critical exponents $\gamma=1.81\pm0.12$ and $\nu=1.03\pm0.09$ being in very good agreement with the theoretical prediction, while the discrepancy with β value could be due to the ''finite-size effects'' such as finite terrace width and domain boundaries. Besides the finite-size effects some additional deviation from the exact Ising model behavior could be due to the enhanced surface corrugation of the adlayer. It is shown that the influence of large-scale finite-size effects is more pronounced below T_c where the long-range order dominates over the system. Conversely, in the disordered state above T_c, the short-range fluctuations cannot be affected critically by large-scale system size limitations. In that case, local ''frozen-in'' defects or lattice distortions may modify the exact values of critical exponents γ and ν. The experimentally observed strong decrease of $(-1,0)$ diffraction peak intensity may be considered as a change in the surface corrugation during the order-disorder transition from a strongly corrugated to a flat surface.

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