

The processes of ordering and formation of two-dimensional glasses at metal surfaces

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Abstract

Using the experimental results obtained for the Dy–Mo(112) system, we discuss the possibilities and mechanisms of formation of two-dimensional (2D) glasses on metal surfaces. It has been found that in the coverage range $0.07 < \theta < 0.58$, ordered Dy superstructures formed and observed at $T < 400$ K are irreversibly destroyed by annealing to higher temperatures and turn into an amorphous (glass) structure on cooling. It is supposed that this conversion is caused by the formation, at $T > 400$ K, of a Dy–Mo surface alloy in which the rate of Dy surface diffusion is strongly reduced in comparison with its value in the absence of alloying. As a result, the mobility of Dy adatoms becomes too low at the temperatures corresponding to the ordered equilibrium state of the surface, and this state cannot be achieved in reasonable relaxation time. This interpretation is corroborated by the experimental data on substantial suppression of surface diffusion in some coadsorbed layers. Since surface glasses contact with ordered (crystalline) substrates, their structure may have a peculiar character different from that of “conventional” metal glasses. Surface glasses can find a number of applications as rather stable systems that combine low dimensionality, specific electronic structure of their constituents and extremely high density of defects.

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1. Introduction

Let us start with reminding definitions of some terms that will be used in this article. Solids with the lack of long-range order in the positions of atoms or molecules are called *amorphous*. They can be produced by many methods, e.g. by material deposition from the vapour phase onto a cooled substrate, by intense ion bombardment and strong plastic deformation of polycrystals or single crystals, etc. An amorphous solid is usually termed *glass* when it is obtained by cooling a liquid. Glasses represent a very important particular class of amorphous materials because of their wide practical use [1,2].

In previous papers [3,4], we reported the observation of two-dimensional glassy films produced by annealing of dysprosium submonolayers adsorbed on the (112) surface of molybdenum. This is a novel method allowing preparation of physical systems which combine the properties of lowered dimensionality, lack of long-range order, and of a high stability. The factor of stability is important both for the investigation of such systems and for their possible applications. Recall, for instance, that amorphous films obtained by vacuum evaporation onto cooled substrates are in most cases unstable against annealing, which results in their irreversible crystallization. The films considered in this work revert to the glassy state (vitrify) after annealing at temperatures which bring them into the molten state. Thus the experiments with a film once deposited onto the substrate can be repeated many times.

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The aim of this article is to discuss our experimental data in the light of some model considerations about two-dimensional (2D) glasses formed after annealing of metal submonolayers on metal substrates. The article is organized as follows. In Section 2 we describe experimental techniques used to prove the formation of the 2D glasses and to investigate the kinetics of surface diffusion. Experimental data are reviewed in Section 3. The results obtained and models used for their interpretation are discussed in Section 4, and Section 5 summarizes the work.

2. Experimental methods

2.1. LEED measurements

An atomic model of the Mo (112) surface is shown in Fig. 1. The (112) oriented Mo crystal sample used in this work had surface terraces with dimensions $L_{[1\bar{1}1]} \times L_{[1\bar{1}0]} \approx 350 \text{ \AA} \times 550 \text{ \AA}$. It could be cooled and heated in the temperature interval 90–2300 K. To investigate the order in adsorbed films, we used measurements of LEED peak intensities and full width of the peaks at half maximum (FWHM). An atomic beam of the adsorbate was deposited from a tiny metal slab (Dy) or from chemical reactors (Li and Sr evaporators designed as small Knudsen cells filled with chemicals of high purity). The ultrahigh vacuum chamber ($p \leq 1 \times 10^{-11}$ Torr) was equipped also with other experimental techniques: contact potential difference (CPD) measurements, Auger electron spectroscopy and mass spectrometry. The coverage was defined as the ratio of the adatom concentration to the surface concentration of the substrate atoms: $\theta = n_a/n_s$. For the Mo(112) surface, $n_s = 8.3 \times 10^{14} \text{ cm}^{-2}$. The film was adsorbed at the lowest (base) temperature $T_b = 90 \text{ K}$ available in the LEED chamber. Then it was annealed in a series of rising temperatures. The annealing at each temperature lasted 10 s, after which the sample was subjected to an intermediate slow cooling (5 K/s) down to T_b for the LEED measurements. The electron beam energy in LEED was kept constant in each series, usually in the range of 55–67 eV. Special attention was paid to precise determination of the LEED spot

positions. It was provided by computer processing of the LEED patterns using a preliminary calibration with reference to the well studied patterns of the system O–Mo(112) [5]. Other details of the structure determinations are given in Refs. [3,4].

2.2. Surface diffusion

In parallel experiments, we investigated the kinetics of surface diffusion of lithium on the Mo(112) surface precovered with dysprosium. Such data are important for the understanding of the conditions and mechanism of surface vitrification.

The technique developed and used by us to study surface diffusion is based on recording the diffusional evolution of adsorbate coverage profiles (Fig. 2a) [6]. The initial profiles shaped as a coverage step were created by vacuum evaporation of the adsorbate through an appropriate mask. By recording the current–voltage curve of the electron current collected by the crystal sample, one can determine the contact potential (work function) difference between the crystal surface and the cathode of a movable electron gun (Anderson's method [7]).

The calibration of the Mo(112) work function versus adsorbate coverage was carried out in separate experiments. These data were used to convert the work function distribution over the surface into the coverage distribution (θ -profile). The evaluation of the coverage profiles recorded after annealings for different times and at different temperatures allows extraction of values of the surface diffusion coefficients $D(\theta, T)$ corresponding to various coverages and temperatures (the Boltzmann–Matano method [6,8–10]). In the next step of the data evaluation, the Arrhenius plots $\lg D(\theta, T) = f(1/T)$ were used to obtain the activation energy of diffusion E_d and the pre-exponential factor D_0 in the equation $D = D_0 e^{-E_d/kT}$.

The Mo(112) surface has a highly anisotropic atomic corrugation: it consists of the close-packed rows of Mo atoms separated by channels (troughs) (Fig. 1). The distance between Mo atoms in the rows equals 2.73 Å, the distance between the rows 4.45 Å, and the depth of the channels 1.29 Å. The anisotropy of the surface atomic relief

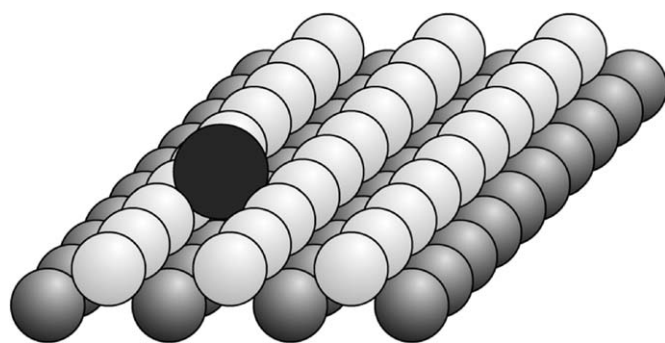


Fig. 1. A model of the Mo(112) surface. Mo surface atoms are shown as light balls and Mo atoms of the first subsurface layer as grey balls ($r_{\text{Mo}} = 1.36 \text{ \AA}$). A Dy adatom is shown as a black ball ($r_{\text{Dy}} = 1.77 \text{ \AA}$).

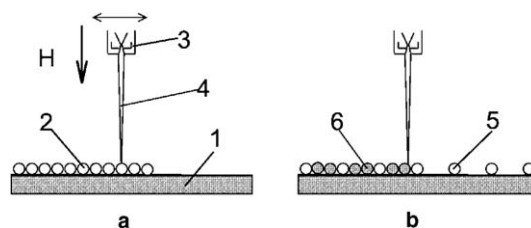


Fig. 2. Schematic of recording the adsorbate coverage profiles using the method of contact potential difference. (a) 1 is the substrate, 2 a step-like initial profile of a one-component adlayer, 3 the electron gun, 4 the electron beam, H is the focusing magnetic field and (b) 5 is a pre-adsorbed base layer (light balls) and 6 is a coadsorbed (two-component) layer.

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