Thermal expansion of thin single-crystalline gold films

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The coefficients of thermal expansion of gold films ~ 4 nm thick and of the surface atomic layer are determined by the method of transmission high-energy electron diffraction.

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The thermal expansion of films with thickness $t < 10$ nm is determined by the contribution of layers not only in the bulk but also on the surface and near the surface. The coefficient of thermal expansion of thin films can be measured by the method of high-energy electron transmission diffraction (HEETO) from the thermal displacement of the diffraction peaks. Electron-diffraction pictures from very thin single-crystalline films under certain conditions can contain reflections from the surface atomic layers, in addition to reflections from the bulk. This situation arises, for example, with a noninteger number of units cells over the thickness of the film. This circumstance opens up interesting possibilities for studying the structure and properties of surfaces by the method of HEETO.

The coefficient of linear expansion of a thin film (α_f) and of an external atomic layer (α_s) were measured experimentally by the HEETO method. The objects of the investigations were very thin $t \sim 4$-nm single-crystalline Au films with (001) orienta-

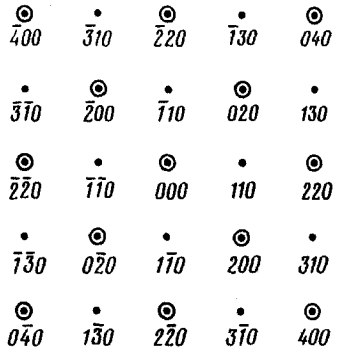

FIG. 1. Diagram of electron diffraction pictures for the (001) plane of a single-crystalline gold film ~4 nm thick. ○, ●—reflections from the bulk and surface, respectively.

tion, obtained by the method of laser evaporation of gold in a vacuum $\sim 10^{-5}$ Pa on (001) clevage surfaces of KCl at room temperature. After separation in water, the films were recovered with supporting meshes. In addition to the reflections characteristic for the (001) orientation, the electron-diffraction pictures contained reflections forbidden by the structure factor with indices of different parity, for example, (110), (130), and (330) (Fig. 1).

Inclined electron diffraction pictures established that these reflections in the reciprocal space are rods ~ 1 Å^{-1} long, which corresponds to diffraction by a layer ~0.5 of the gold lattice period (a_0). It can be shown that for the (001) orientation, which can be represented as a sequence of layers *AB AB*. . . in the [001] direction, the indicated forbidden reflections will appear with an odd number of layers, i.e., with a half-integer number of *fcc* cells over the thickness of the film. Electron-microscopic studies show that the films are continuous and contain packing defects (PD) along the (111) planes, whose density is $\sim 10^{10}$ cm^{-2}. Because of the steps on the surface of KCl, integer and half-integer numbers of fcc cells form during the condensation process in the film on both sides of PD (Fig. 2). We note that reflections of the type (110) from the surface of a single-crystalline Au film were observed, for example, by Krakow.[1]

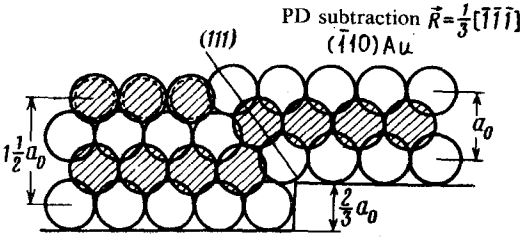

FIG. 2. Model of the formation of incomplete *fcc* cells in the region where the packing-defect surface intersects the film surface. The plane of the figure coincides with (110) Au.

The coefficients of linear expansion of the film (α_f) were determined from the displacement of the matrix reflections of the type (200) and (220), and the coefficient of linear expansion of the surface layer (α_s) was determined from the displacement of (310) reflections at temperatures ranging from room temperature to liquid-nitrogen temperature. The instrumental errors resulting from the change in the distance between the specimen and the photographic plate and from the possible instability of the accelerating voltage were taken into account with the help of a cathetometer and an etalon.[2] The value of α was determined to within $\pm 2\times10^{-6}$ K^{-1}.

It has been established that $\alpha_f \sim 31\times10^{-6}$ K^{-1}. This is 2.5 times greater than the value of the coefficient of thermal expansion of bulk gold (α_v) for the indicated temperature range (13×10^{-6} K^{-1}). We note that the values of α_f, determined from the (200) and (220) reflections are nearly equal. This indicates that the effect is due to thermal expansion, rather than the action of thermoelastic strain, since the elastic moduli in these directions differ approximately by a factor of two. The value of α_s for the (001) gold surface is $\sim 46\times10^{-6}$ K^{-1}.

In a first approximation, α_f can be represented in the form

$$\alpha_f = \alpha_v + 2(\alpha_s - \alpha_v)\frac{\Delta t}{t}, \qquad (1)$$

where Δt is the near-surface layer, whose lattice dynamics differs from the dynamics of the bulk layers.

It follows from (1) that for known values of α_f, α_v, α_s, and t, the value Δt is 1 nm. This value agrees with the theoretical estimates.[3] The coefficient of thermal expansion of the surface layer in a direction parallel to the surface of the film is approximately two times greater than the theoretical value.

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[1]W. Krakow, Surf. Sci. **111**, 503 (1981).
[2]A. T. Pugachev and N. P. Churakova, Zavodskaya laboratoriya, No. 8, 737 (1980).
[3]A. Maradudin, Defekty i kolebatel'nyĭ spektr kristallov (Defects and the Vibrational Spectrum of Crystals), Mir, Moscow, 1968.

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