Laser induced phase transition in iron thin films

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Introduction

High power laser pulse irradiation of material surfaces produces a wide spectrum of phenomena, all connected with emerging new technologies. Topics like: ablation, thin films deposition, laser induced chemical reaction are frequently studied. Few studies have been reported in the domain of laser induced solid state phase transitions at a relative low laser fluence. In this regime the laser pulse acting like a high power heat pulse, the evolution of the system is far from equilibrium and new structures or new mechanism of transformation is expected.

In the case of laser thin films heating, the heating rate is of the order of 10^{10}Ks^{-1} . If the laser pulse is absorbed only in the thin film and the film is laser irradiated while on the substrate, the rate of cooling can be equally fast, the cold substrate acting as a thermal sink. The thin film is suddenly heated and rapidly cooled, quenching the structure resulting after the laser pulse heating.

In a previous paper¹ the microstructural aspects of the b.c.c. - f.c.c. phase transition of iron thin films were studied by high resolution transmission electron microscopy. This transition of iron is one of the best known and studied allotropic transformation, mainly in the case of martensitic transformation². In the first paper¹, it was pointed out a new aspect of the phase transition process in the laser heating case, by revealing the appearance of a microscopic mixture of the b.c.c. and f.c.c. phases. This mixture represents an incomplete transformation in the same crystalline block and can be understood as a frozen-in stage of the transition. In this paper we report numerical analysis of the laser

heating and the correlation with the observed characteristic microstructure.

Experimental

Iron thin films have been deposited at RT by evaporation in a vacuum of 2×10^{-6} torr using 99.9% iron needles. Freshly cleaved KCl single crystals have been used as substrate. The thickness of the iron films was 40nm. To prevent oxidation of the iron film, before and after iron evaporation, two 5nm thick amorphous carbon layers were subsequently deposited while still in vacuum. A sandwich film, C-Fe-C, with 50nm total thickness was thus obtained.

KrF excimer laser irradiations ($\lambda=248\text{nm}$) have been performed in air. The thin films were irradiated by a single pulse on the KCl substrate. The laser fluence was in the range of 50 to $100\text{mJ}\cdot\text{cm}^{-2}$ and the pulse duration $\tau_{\text{FWHM}}=20\text{ns}$. The laser fluence was set in such a way that a minimum modification of the film could be observed by optical microscopy. After irradiation on the KCl substrate, the film is floated of in water and, the irradiated areas, mounted on TEM grids.

Numerical analysis

Fig. 1: Temperature evolution of points located at different depths from the surface of the irradiated sample.

Fig. 2: Evolution of the temperature derivative for points located at different depths.

We made a numerical analysis of the laser heating and subsequent cooling of the irradiated samples using a finite-difference method to

solve the heat-flow equation^{3,4}. In the case of the iron film deposited onto KCl substrate, we performed a 1-D numerical analysis. This option is based upon the observation that the heat diffusion length $L_T = \sqrt{\chi\tau}$ (χ is the thermal diffusivity) in the Fe film and in the KCl substrate on a time scale significant for our analysis ($\tau=100\text{ns}$) is about $L_T \approx 1\mu\text{m}$ (Fe) and $L_T \approx 250\text{nm}$ (KCl). Therefore, there is no significant lateral heat diffusion as compared to the dimensions of the laser spot, $d \approx 1\text{mm}$, and the heat-flow process is significant only over a direction normal to the sample surface. Relevant results of the 1-D numerical analysis are presented in figs. 1 and 2, presenting the temperature and its time derivative evolution in time for points located at different depths from the surface.

Results and discussions

Fig. 3. TEM and corresponding SAED images of the characteristic areas of the C-Fe-C irradiated thin films: (a) b.c.c. phase area, (b) mixture phase area, (c) f.c.c. phase area.

Fig. 3a illustrate the microstructure of the as deposited thin iron film in the b.c.c. phase with the lattice parameter $a=0.287\text{nm}$. In fig.3c the f.c.c. microstructure resulting after the phase transition is presented. Between the two phases the irradiated thin film shows a region with a mixed b.c.c. and f.c.c. phase as visible in fig. 3b. This mixed regions always appeared between the b.c.c. and f.c.c. areas and have an extension in the film plane of about 10 to 20 mm. High resolution images showed that during the laser heating a crystalline growth

process takes place. The dimensions of the crystallites grow from about 15nm to about 30nm, mainly by diffusion assisted crystal growth mechanism¹. No carbide reflections have been found in the transformed film. The passivating oxide layer which appeared in time on the iron film is not modified by the transformation process.

We assume that the transition temperature from the b.c.c. to the f.c.c. phase is similar in the thin film case as in the bulk (i.e. 1200-1300K). The numerical calculation in fig. 1 shows that in the thin film the temperature evolution is practically uniform in depth and the temperature peak is quite symmetrical in time. The detail of the temperature derivative in fig. 2 reveals some differences between the heating and the cooling rate of the film, but the order of magnitude is over 10^{10}Ks^{-1} for both cases.

The driving force for the transformation is proportional to the superheating of the film on the peak of the laser pulse. The superheating in the mixed phase areas of the irradiated films is low and with only 1 or 2ns time duration. In this case an incomplete transformation can be observed and the boundaries of the crystallites are mainly phase boundaries¹. The transformation takes place during temperature rise period and can be considered a reverse martensitic transformation. The transition is very fast, faster than processes like bulk diffusion or oxidation. The b.c.c. and the f.c.c. relative orientation most frequently present in the mixed phase area of the irradiated films is of the Bain type, which can be a characteristic of the reverse martensitic transformation. The laser pulse heating main characteristic is the ability to separate between the solid state transformation and the diffusive mechanism of structural or chemical transformation.

References

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