SN BL	Experiment title: High pressure investigation of the metastable beta-Ta phase	Experiment number : BM01-02-362
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Report:

Purpose: The structure of β -Ta electrolytic phase at room temperature belongs to the σ -type of Frank-Kasper phases, which are typical for binary intermetallic compounds and β -U. In comparison to the σ -type, additional intercalated atoms (Tapopulation factor ~0.01) have been detected between the atoms located in the channels of the structure [1]. The interatomic distances observed between the channel atoms are shorter than those of the atoms of the framework, this justify the "self-hosting" characteristic of β -Ta analogously to the complex tetragonal structures of the high pressure (HP) phases of the elements Rb, Ba, Sr, Bi, Sb [2,3,4,5]. The tetragonal β -Ta phase is absent in the p-T phase diagram [6]. High temperature (~1000 K) and the presence of electrical field under reducing conditions allow the formation of β -Ta by electrolytic crystallization on a cathode. According to [7] β -Ta irreversibly transforms to α -Ta when heated above 1273 K. In this work we present an investigation of β -Ta under high pressure.

Experimental method: Powdered sample was loaded in a diamond-anvil-cell (DAC) and its powder diffraction pattern collected on the MAR detector at a wavelength of 0.750Å. Using Fit2D [8], the raw images were integrated into 1D powder spectrum. The pressure was determined either from an internal standard (NaF or NaCl) or from the fluorescence line of a ruby chip. In total 6 HP runs (A, B, C in March 2002; D, E, F in July 2002) have been performed with different pressure medium conditions. Powder data have been collected between 0-11 GPa at room temperature.

Results: Depending on the pressure medium conditions, different scenarios have been observed;

Run (A) ground β -Ta powder, silicone oil as pressure medium, NaF as internal standard (quasi-hydrostatic conditions). The diffraction signal disappeared at 0.6 GPa (the first high pressure point reached after difficulties in the loading of the HP cell); the recorded image is again "amorphous-like".

Run (B) ground β -Ta powder, ethanol-methanol mixture as pressure medium, NaF as internal standard (hydrostatic conditions). The diffraction signal disappeared at 0.3 GPa (the first high pressure point reached after difficulties in the loading of the HP cell); the recorded image is again "amorphous-like".

Run (C) ground β -Ta powder, NaF as pressure medium and internal standard (non-hydrostatic conditions). The diffraction signal disappeared at 0.2 GPa (the first high pressure point); the recorded image is "amorphous-like".

Run (D) ground β -Ta powder, silicone oil as pressure medium, NaF as internal standard (quasi-hydro-static conditions). The diffraction signal didn't disappear and it was possible to collect data up to 6 GPa.

Run (E) ground β -Ta powder, silicone oil as pressure medium, NaF as standard (quasi-hydrostatic conditions). The diffraction signal didn't disappear and it was possible to collect data up to 11 GPa.

Run (F) ground β -Ta powder, no pressure medium and no internal standard (non-hydrostatic conditions). The diffraction signal disappeared; the recorded image is again "amorphous-like".

These results may be explained on the basis of the following simple picture, taking into account that in **Run A** and **Run B** the pressure medium probably went out of the hole of the gasket when the cell was closed (this is a problem commonly encountered in HP experiments). For true (or quasi) hydrostatic conditions, the powdered sample of β -Ta remains crystalline under pressures up to at least 11 GPa. Within this pressure range, it presents a diffusive-transition around 2.5 GPa (see below). While for non-hydrostatic conditions, the host-guest structure of β -Ta collapses and the sample either becomes "nanocrystal like" or "amorphous-like", producing a diffraction pattern with a halo characteristic for glasses or amorphous materials (see Fig 1a). The observation of two different behaviors, depending on the hydrostatic conditions, is not new and has been observed for many different systems.



Fig. 1. Raw diffraction patterns, (1a) recorded in Run F, (1b) in Run D. A typical 1D powder diagram is given in (1c).

The powder data collected in **Run D** and in **Run E** may be indexed by the tetragonal cell of β -Ta [1]. The metric remains tetragonal over the whole range of investigated pressure, but a discontinuity is observed as depicted in Fig 2a. Inspecting the raw data close to the singularity, one can show that at 2.3 GPa some of the powder rings present radial streaking that may even bridge together powder rings (Fig 2b). When the pressure is further increased to 3.4 GPa, this streaking diminishes (Fig 2c). This observation of streaking reveals the existence of disorder and allows establishing the diffuse character of the transition. A detailed analysis of these HP powder diffraction data is in progress.



Fig. 2. The evolution of the *c* lattice parameter is given in (2a). Azimuthal representations of the raw images collected at 2.3 GPa (2b) and 3.4 GPa (2c) showing a stronger streaking effect just before the transition. The vertical axis is the azimuthal angle, while horizontal one corresponds to 2-theta.

References

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