Type of presentation: Oral

MS-9-O-1620 Chemical analyses of Ga segregation at Pb nanoparticles using ChemiSTEM Technology

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Al-Pb composites consisting of nanometer-sized Pb inclusions embedded in a polycrystalline Al matrix serve as model systems for size-dependent melting studies [1]. To analyze the impact of the surrounding matrix on the melting and solidification temperatures of the Pb nanoparticles, Ga was added to the matrix within the solute solution regime. The alloying of Ga expands the lattice of the Al matrix and thus reduces the mismatch between matrix and particle. Although Ga is immiscible with Pb, the melting and solidification of the Pb nanoparticles was affected by the Ga addition. A large undercooling of the solidification onset of up to 100K with respect to melting was found with increasing Ga content [2]. Chemical analyses (EDX) were performed using a double-corrected, monochromated Titan 60-300 with ChemiSTEMTM technology. The results of this investigation are shown in the Figures. The Pb nanoparticles were oriented along the <110>-direction of the fcc matrix and imaged by HAADF-STEM (Fig. 1). The chemical analysis shows that Ga segregates at the particle-matrix hetero-interface (Fig. 2). The Ga concentration has been traced in the form of a profile across the Pb nano-particle (see boxed area in Fig. 2). It shows peaks in the Ga signal at the interfaces to about 7.4 at.% (Fig. 3). The level across the Pb nanoparticle is about 1.2 at.% higher on average than the surrounding matrix concentration of 4.7 at.% due to the other Ga enriched interfaces which are projected in plan view. The width of the Ga enriched zone around the Pb nanoparticle is about 2.2 nm. This Ga enriched zone acts as a buffer between the nanoparticle and the nominal $Al_{94}Ga_6$-matrix inhibiting the nucleation of the liquid Pb. It has been shown that vacancies play a key role in accommodating the misfit strain upon solidification of nanoparticles embedded in Al [1,3]. Presumably the increased number of vacancies available around the Pb nanoparticles relative to the matrix lead to an accelerated Ga diffusion in this region and thus to the observed Ga segregation. The Ga segregation, in turn, leads to solid solution hardening of the buffer phase, which then becomes more rigid with the Ga increase. This hinders an elastic accommodation of the mismatch strain from the matrix side. The scenario of a suppressed nucleation upon cooling is hence explicable since the liquid Pb nanoparticles contract upon solidification and thus bring a negative pressure on the Pb particles. Thus the undercooling of the Pb nanoparticles can be explained by thermodynamics according to Clausius-Clapeyron.

[1] E. Johnson, H.H. Andersen, U. Dahmen, Microsc. Res. Techniq. 64 (2004), p. 356.
[2] A. Moros et al. to be published.
[3] L.H. Zhang, E. Johnson, U. Dahmen, Acta Mater. 53 (2005) p.3635.

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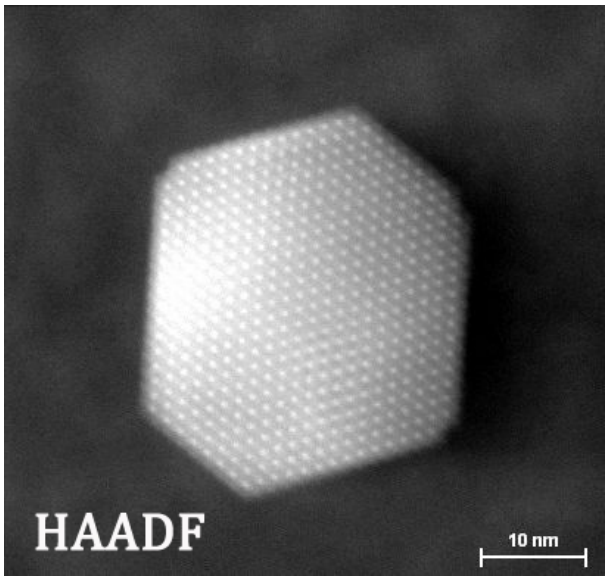

Fig. 1: HAADF-STEM image of a faceted Pb nanoparticle embedded in an $Al_{94}Ga_6$-matrix viewed along the <110>-direction. Six out of fourteen interfaces are imaged in edge-on conditions.

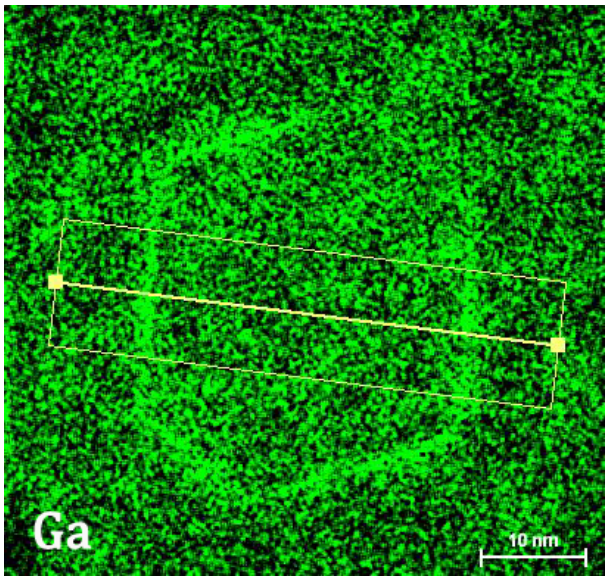

Fig. 2: EDX Ga map using a probe current of 450 pA and an acquisition time of 454 s.

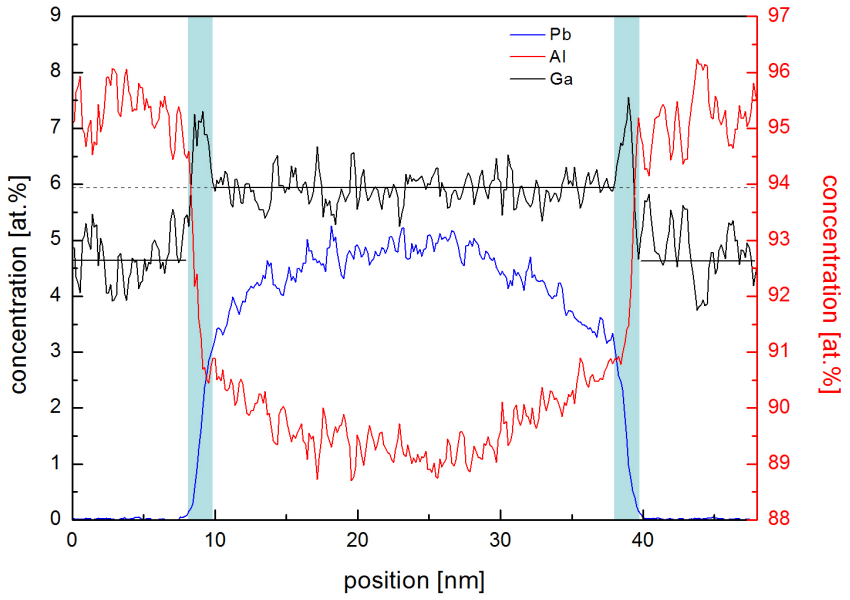

Fig. 3: Al, Ga and Pb profiles corresponding to the boxed area in Fig.2.