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Report:

We have undertaken a series of elevated temperature *in-situ* high-resolution powder diffraction experiments on Sn nanocrystals embedded in a thin-film silica matrix. High temperature Brillouin spectroscopy had shown an abrupt red-shift in the inelastic peaks, which was attributed to mode softening due to the melting of the nanoparticles of Sn [1]. The temperature at which the anomaly occurred depended on the nominal particle size. Preliminary parallel beam powder diffraction measurements carried out at the Daresbury SRS after submission of the experiment proposal showed that melting occurred in the region of the anomaly [1]. However, the beam intensity resulted in poor signal to noise, inadequate for us to measure the transition temperature or particle size precisely. Using the furnace on BM16 in association with the high-resolution Ge analysers, we have been able to measure the melting temperature of the Sn nanocrystallites as a function of size and confirm that the acoustic mode anomaly does occur at the melting temperature. We were also able to determine the thermal expansion coefficients of the particles.

Measurements were performed on SiO_x films 150 nm thick on (001) oriented silicon. Films contained approximately 25% Sn in the form of nanoparticles nominally 2.5, 10 and 20 nm diameter. Furnace calibration was undertaken by simultaneous measurement of the lattice parameter of the silicon substrate from which the true sample temperature was deduced

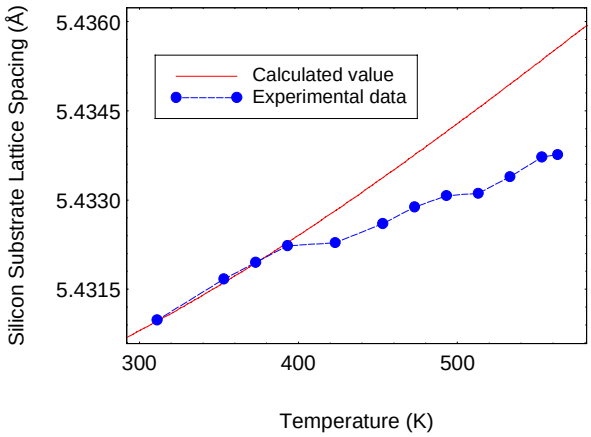

Figure 1

Particle Diameter (nm)	Brillouin anomaly temperature (^{o}C)	Melting temperature from XRD (^{o}C)
4.5 ± 2	135 ± 15	135 ± 15
11 ± 1	190 ± 10	180 ± 10
18 ± 1	210 ± 10	200 ± 10

Bulk melting temperature 232^{o}C

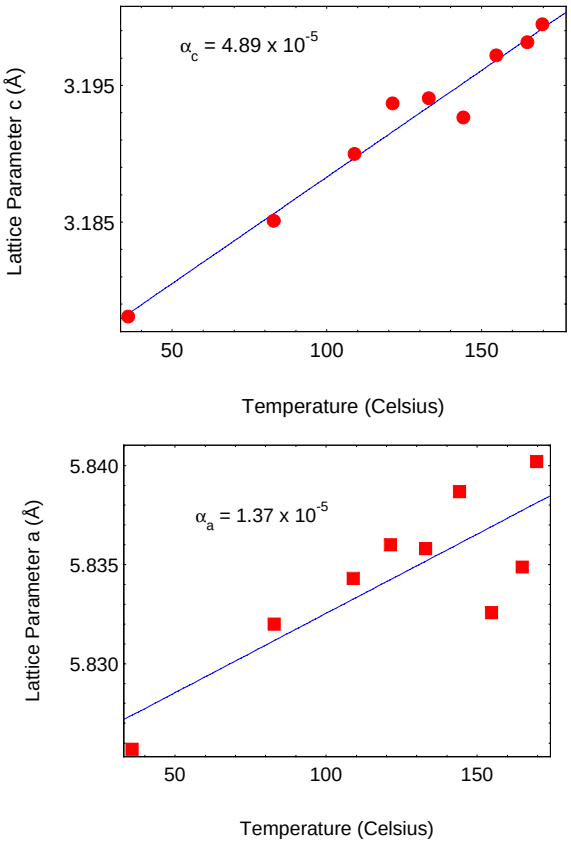

Figure 2

the published thermal expansion coefficient [2]. As evident in Fig 1, the sample temperature differed from the furnace thermocouple reading by nearly 100^{o}C in the region of interest. The particle size, determined from the diffraction peak widths agreed with nominal particle sizes to within 2nm. The disappearance of the Bragg peaks, corresponding to particle melting occurred within 10^{o}C of the temperature of the Brillouin scattering anomaly, well inside the experimental error. Effective medium theory, incorporating the observed particle melting, explains the redshift. Lattice parameter measurements of the Sn nanoparticles gave values of the thermal coefficients (Fig 2) in the *c* and *a* directions which were, within the experimental precision, equal to those of bulk material, indicating that no significant levels of strain exist within the nanoparticles. Hysteretic behaviour was observed in the nanoparticle melting, crystallization taking between one and two hours after cooling the melted particles back to room temperature.

[1] C E Bottani, A Li Bassi, B K Tanner, A Stella, P Tognini, P Cheyssac and R Kofman, Phys Rev B: 59 (1999) R15 601- 604
[2] B. K. Tanner, J. Clarke, A. Li Bassi, T.P.A.Hase, B.D.Fulthorpe and E Dooryee, to be submitted to J Sychrotron Radiation
[3] A Li Bassi, C E Bottani, B K Tanner, A Stella, P Tognini, P Cheyssac and R Kofman, to be submitted to Phys Rev B