Supplementary information

Gallium-tin alloys as a low melting point liquid metal for repetition-pulse-laser-induced high energy density state towards compact pulse EUV sources

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(I) Experimental and characterisation conditions

(a) Alloy preparation and characterization

Ga:Sn alloys were synthesized using atomic ratios (at%) of gallium (99.99%, Zaiyouya Corp.) and tin drops (99.9%, Kanto Chemical Co. Ltd.). The gallium and tin were weighed using an accurate microbalance, then heated up to 200°C to induce formation of each alloy. Once the alloys were prepared, some of the alloy was used for DSC measurements, and some was applied to a glass substrate for EUV experiments. The alloy thickness ranged from 50-150 μm on the surface of the glass. The melting point of Ga:Sn 90:10% was obtained using a Seiko Instruments EXSTAR X-DSC7000 with a platinum (Pt) pan under nitrogen (N_2) flow with a temperature increase of +10°C per min. The melting point of the other Ga:Sn alloys were obtained using a Rigaku XRD-DSC Ultima IV with a ceramic pan (Al_2O_3) under N_2 flow with a temperature increase of +10°C per min. Accurate mass of each alloy were added to each DSC pan for characterization, then scanned from 0°C to +250°C. A Pt/ceramic pan was necessary because gallium has a tendency to form amalgams with metals commonly used for DSC pans such as aluminium. Scanning electron microscopy (SEM) was performed in the following manner; liquid alloys were applied to a glass substrate (approximately 1x1 cm^2) before mounting onto a target holder. A JEOL JSM 6610LA SEM with energy-dispersive X-ray Spectroscopy (EDS) probe was used to obtain micrograph of the Ga:Sn alloys, and elemental composition. The accelerating voltage was 20 kV and the SEM was in secondary electron mode. The EDS probe provided elemental mapping of Ga and Sn of corresponding SEM images. The lower melting point alloys required additional carbon tape to enable successful dispersion of the electron beam.

(b) Laser irradiation conditions

A 2 mJ 1064 nm Nd:YAG pulse laser (L11038-01, Hamamatsu Photonics) was used to ablate the target materials. More detail for the vacuum chamber experimental set-up used has been described elsewhere.[1] A focal spot size of 60 μm at full width half maximum (FWHM) was used to give a laser intensity of 7.1×10^{10} W/cm^2. The vacuum chamber pressure was in the order of $10^{-5}/10^{-6}$ Torr. EUV spectra was recorded by the use of a grazing incidence spectrometer (GIS) with a 5-20 nm grating fitted with a CCD (Tokyo Instruments, TIUV 235-920) set at a 45-degree angle with respect to the incidence of laser ablation. An energy calorimeter (Nova II, Ophir optics) was used to measure the energy of the EUV light at a 45-degree angle from the laser ablation incidence. The conversion efficiency (CE) was then estimated using a previously described technique.[1] The reference tin EUV data was obtained from ablation of a planar tin foil (100 μm, Nilaco, Japan). Reference gallium EUV data was obtained using melted gallium on a glass substrate. The calorimeter measured the energy of the tin EUV emission at 6.1 μJ. An in-band/out-of band factor of 1/1.8 [2] was applied to give the total in-band energy of 2.5 μJ. The imaging plate data were analyzed using a previously described technique.[1] Using the relative EUV intensities at 13.5 nm 2% bandwidth, the CE was then estimated for each material (table 1). A single laser pulse was used to ablate each material, with the exception of a 10 Hz emission on 70:30 at% Ga:Sn. An auto-stage was used to ensure fresh target was ablated throughout the duration of the 10 Hz exposure. In this case, a focal spot size of 100 μm FWHM ($2.5x10^{10}$ W/cm^2) was used. The captured EUV spectra were further analyzed by using Igor Pro software.

(c) Liquid alloy monitoring conditions

The injected Ga:Sn was monitored by a CCD camera (Keyence CV-V200) with a trigger period of 10 ms and exposure time of 100 μs, at the position of 100 mm below of the top of PTFE nozzle.

(II) Differential Scanning Calorimetry (DSC) curves

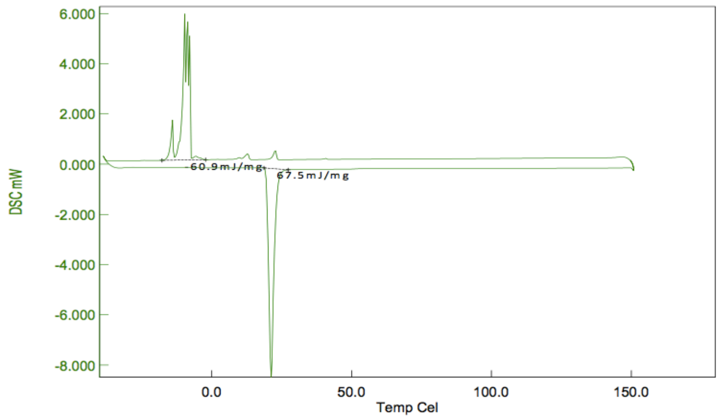

Fig S1. DSC curve for Ga:Sn 90:10%.

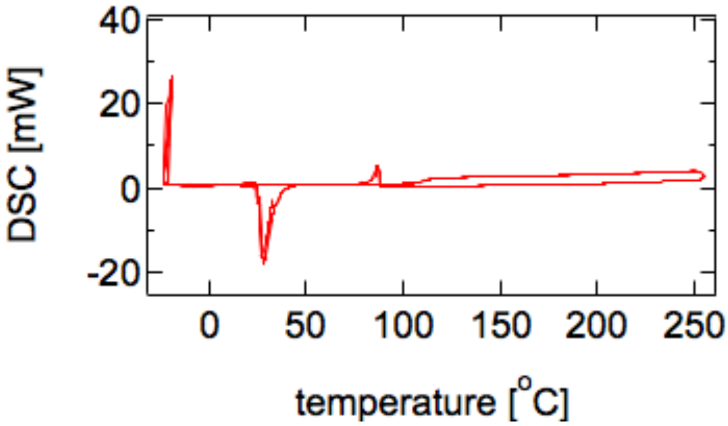

Fig S2. DSC curve for Ga:Sn 80:20%.

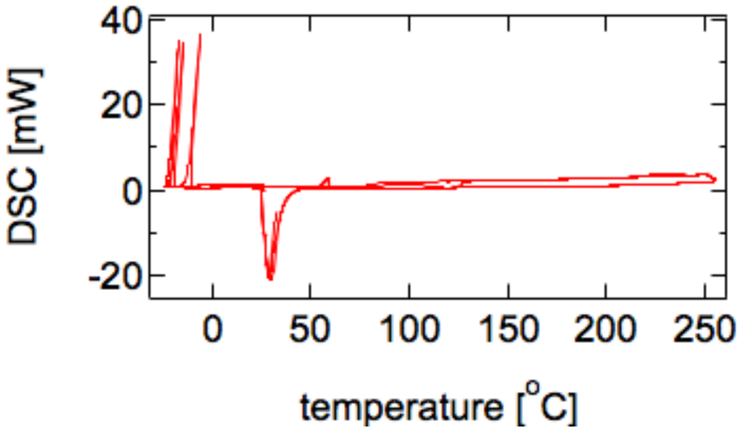

Fig S3. DSC curve for Ga:Sn 70:30%.

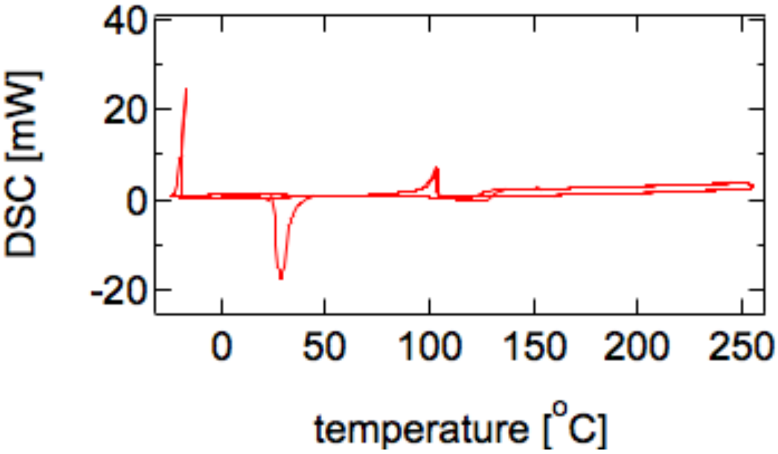

Fig S4. DSC curve for Ga:Sn 60:40%.

(III) Scanning Electron Microscope (SEM) data

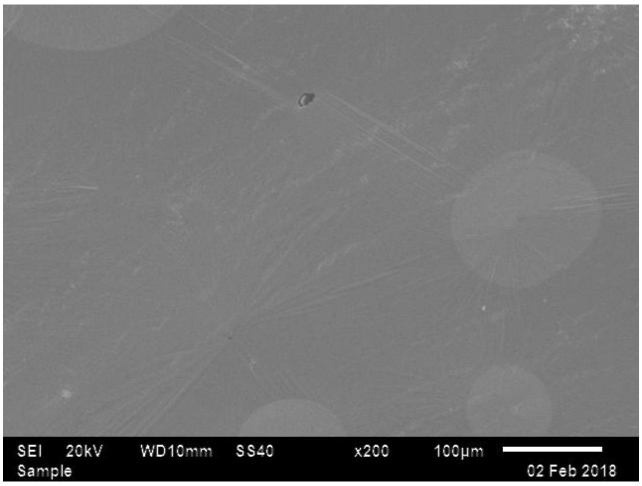

Fig S5. SEM image for Ga:Sn 90:10%.

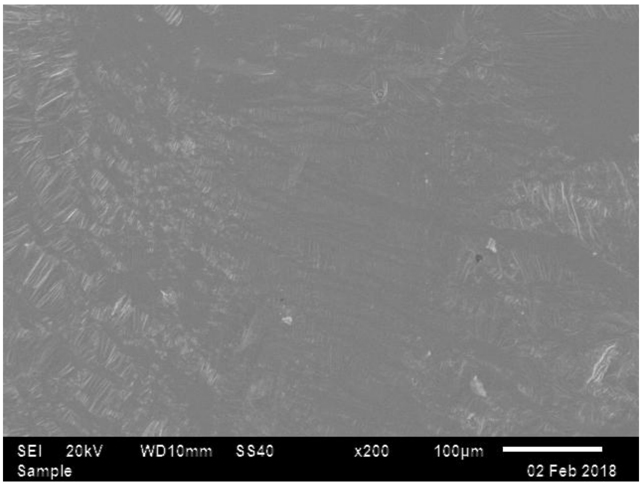

Fig S6. SEM image for Ga:Sn 80:20%.

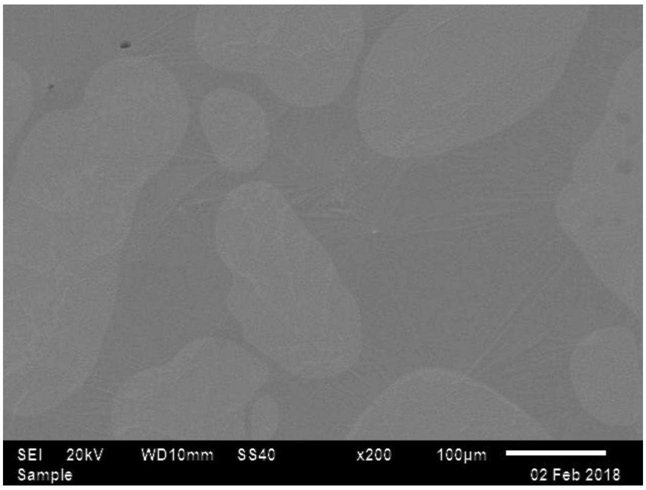

Fig S7. SEM image for Ga:Sn 70:30%.

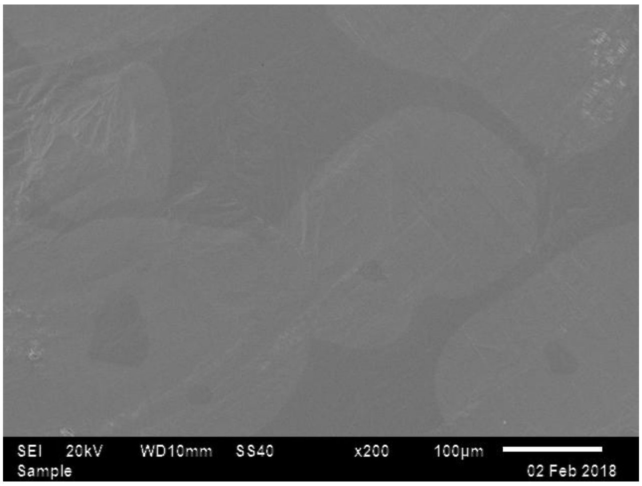

Fig S8. SEM image for Ga:Sn 60:40%.

(IV) Energy-Dispersive X-ray Spectroscopy (EDS) data

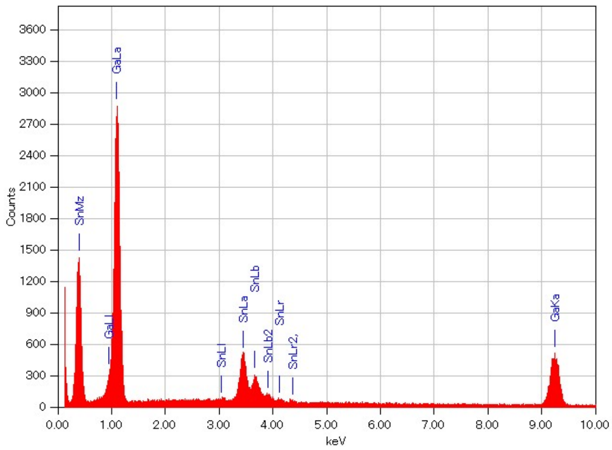

Fig S9. EDS graph for Ga:Sn 90:10%.

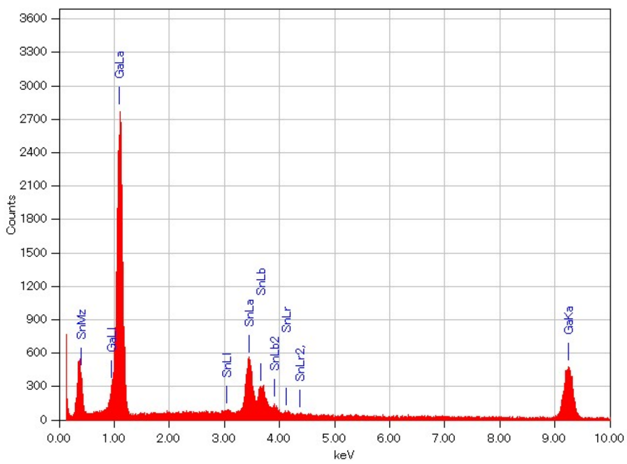

Fig S10. EDS graph for Ga:Sn 80:20%.

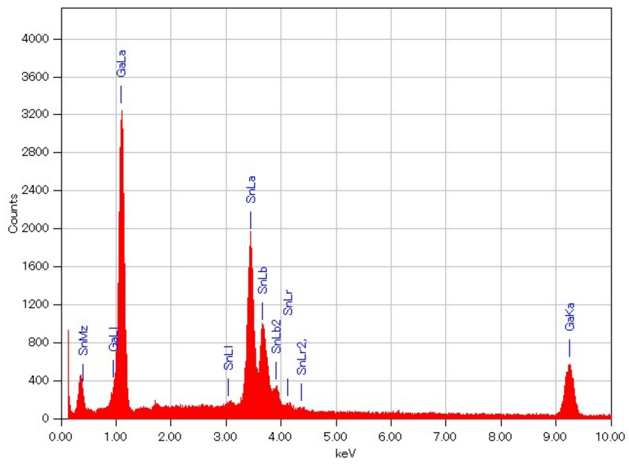

Fig S11. EDS graph for Ga:Sn 70:20%.

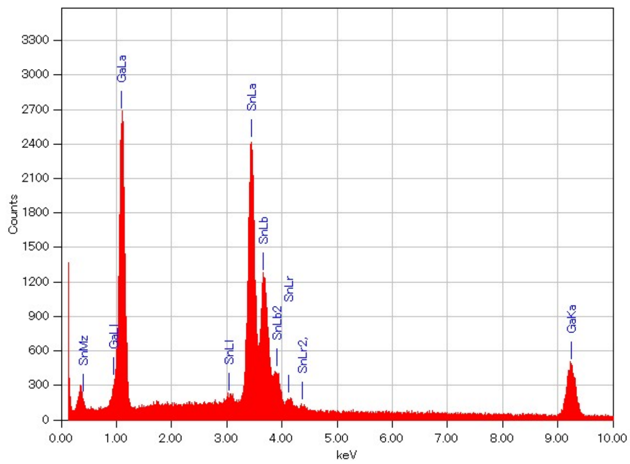

Fig S12. EDS graph for Ga:Sn 60:40%.

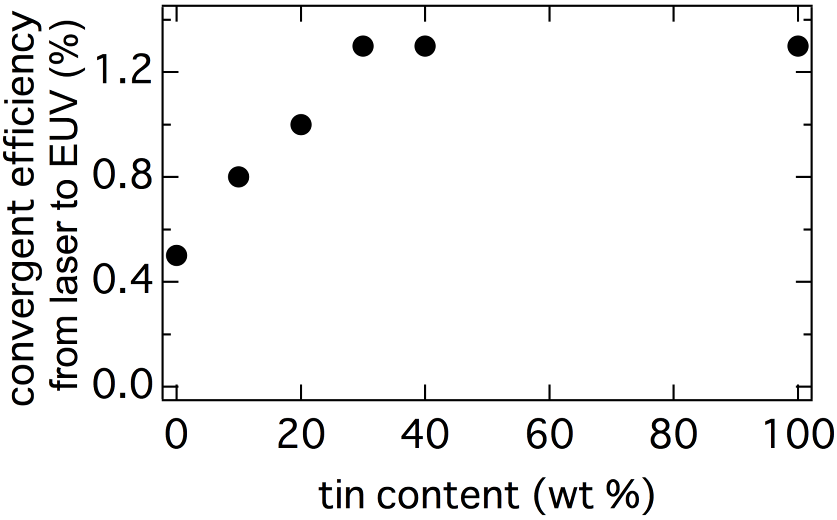

Fig. S13 Tin content dependent conversion efficiency of EUV emission in 2% bandwidth at 13.5 nm

References

(1) C. S. A. Musgrave, T. Murakami, T. Ugomori, K. Yoshida, S. Fujioka, H. Nishimura, H. Atarashi, T. Iyoda, K. Nagai, *Rev. Sci. Instrum*. **2017**, 88, 033506.

(2) H. Sakaguchi, S. Fujioka, S. Namba, H. Tanuma, H. Ohashi, S. Suda, M. Shimomura, Y. Nakai, Y. Kimura, Y. Yasuda, H. Nishimura, T. Norimatsu, A. Sunahara, K. Nishihara, N. Miyanaga, Y. Izawa, K. Mima, *Appl. Phys. Lett*. **2008**, 92, 111503.