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Supplementary information for:

Monolayer doping of germanium with arsenic: a new chemical route to achieve optimal dopant activation.

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This file includes:

Figure S1 to S4

Keywords: Monolayer doping, arsenic, germanium, functionalisation, click chemistry, carrier concentration.

Supporting Figures

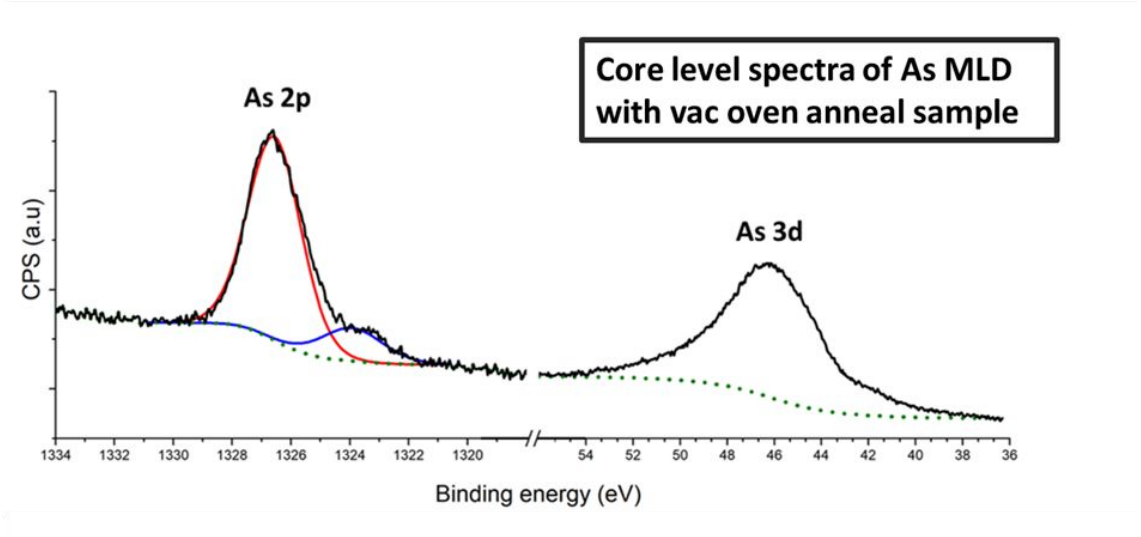
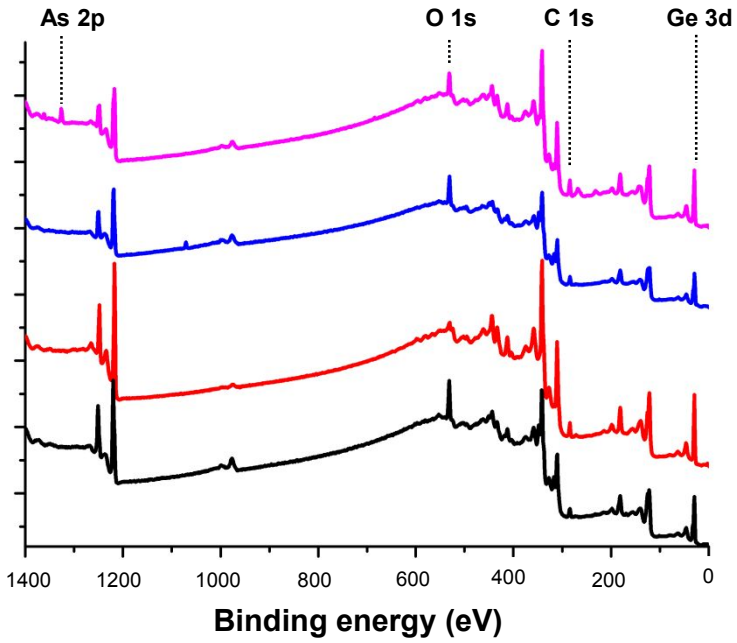


Figure S1: Core level XPS spectra of As 2p and As 3d peaks acquired from Ge functionalized with As-acid MLD using a vacuum anneal. As 3d overlaps with Ge plasmon peaks and therefore it is not possible to quantitatively analyse As presence from this peak. The As 2p peak is not capable of quantitative analysis either as it has no computed relative sensitivity factor (R.S.F). Two distinct peaks are noted in the As 2p signal. The shoulder peak at ≈ 1323 eV (blue) represents As in a +3 or +1 oxidation state while the peak at ≈ 1326.5 eV represents As in a +5 oxidation state which is the expected positioning from As-acid binding. It is possible that the shoulder peak represents As-acid which has decomposed during the functionalization procedure and now occupies this alternative oxidation state. As is also known to decay under X-rays from the +5 to the +3 oxidation state and may result in this shoulder artefact.



	Ge 3d %	C 1s %	Core level area ratio Ge 3d: C 1s
As-rec	76.8	23.2	4.6:1
Cl-terminated	71.2	28.8	3.8:1
As functionalised with no vacuum	71.1	28.9	3.3:1
As functionalised with vacuum	64.8	35.2	2.9:1

Figure S2: Survey spectra analysis of as-received Ge (black), Cl-terminated Ge (red), As functionalised with no vacuum Ge (blue), and As functionalised with vacuum Ge (pink). Peaks are indicated for Ge 3d, C 1s, O 1s and As 2p. Quantification of the Ge 3d and C 1s components enabled an understanding of carbon (C) content after each process. A degree of C contamination is noted on the as-received sample. This content increases to approximately the same value for Cl-terminated and As functionalised with no vacuum samples. It is possible that some of the solvents (acetone, ipa, etc.) used in processing are still present on the Ge surface in these samples and lead to this increase. The As functionalised with vacuum sample both demonstrates the presence of the As 2p peak and a significant increase in C 1s %. This is as expected given that monolayer formation with As-acid would lead to the introduction of 1 x As and 6 x C atoms per molecule. The peak at 1070 eV in the As functionalised with no vacuum sample is likely a sodium related contamination which can result from manual handling during processing and is deemed inconsequential.

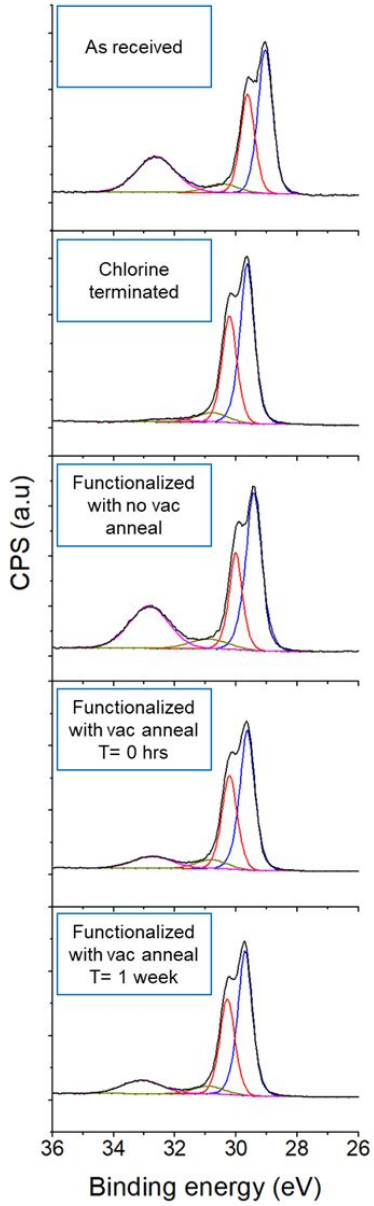
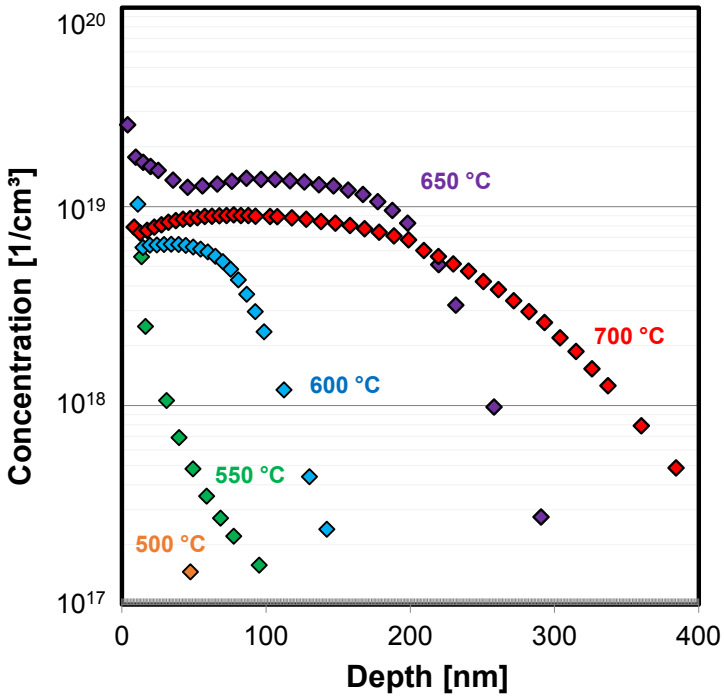


Figure S3: Core level XPS spectra of the Ge 3d peak after As-acid MLD processing steps. This spectra is used to both track oxidation levels and give tentative evidence of monolayer formation. The GeO_2 component of the signal at ≈ 33 eV is removed with HCl treatment to leave a pristine (Cl-terminated) Ge surface. After carrying out As-acid MLD functionalization procedures, without the use of a vacuum anneal, the GeO_2 component shows virtually complete re-growth to a condition similar to the as-received sample. The sample which has been functionalized with As-acid MLD using a vacuum oven anneal displays a minimal amount of GeO_2 re-growth in comparison to the as-received sample. Storage in ambient conditions for 1 week does not lead to further re-growth of this oxide in the As-acid MLD functionalized with vacuum oven anneal sample. This apparent stability to oxidation also represents tentative evidence of monolayer formation. The grown As-acid monolayer acts as a blocking layer to prevent re-oxidation.



RTA temperature (°C)	Dose (cm^{-2})
400	8.4e10
500	8.6e11
550	1.2e13
600	6.1e13
650	2.5e14
700	2.1e14

Figure S4: ECV analysis of As-MLD doped samples with varied RTA temperatures at a constant time of 10 seconds. All samples were capped with a 50 nm sputtered SiO_2. Data shown demonstrates that optimal activation is achieved with RTA temperature of 650 °C. Increasing RTA temperature leads to increased activation up to 650 °C but also leads to increased diffusion and resulting junction depths.