ELECTRONIC SUPPLEMENTARY INFORMATION (ESI)

ESI1. Determination of the average ablation rate of SiC

For the determination of the ablation rate of SiC, additional experiments with 5, 10, 20, 30, and 40 laser shots per position were conducted (see Fig. S1). To simplify the profilometer measurements with the "DektakXT" (Bruker, MA, USA) of the craters, the spot size was increased to 40 µm for this experiment. For each number of laser pulses, four profilometer measurements were conducted. The results indicate a linear relationship between ablation depth and the applied number of laser shots, as can be seen in Fig. S2.

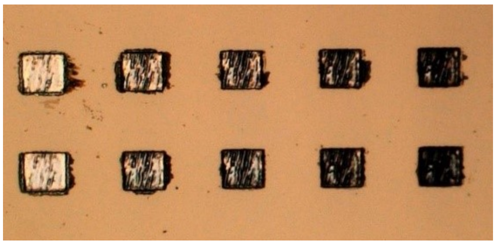

Fig. S1: 300x magnification of the ablation craters used for the determination of the ablation rate. From left to right, the amount of the laser pulses was increased to 5, 10, 20, 30, and 40 pulses, using a 3.5 J/cm^2 fluence, 40 µm spot size, 20 Hz frequency, 450 ml/min chamber He flow and 500 ml/min sniffer He flow. Each number of laser pulses was conducted on two different sample positions, one on top of the other.

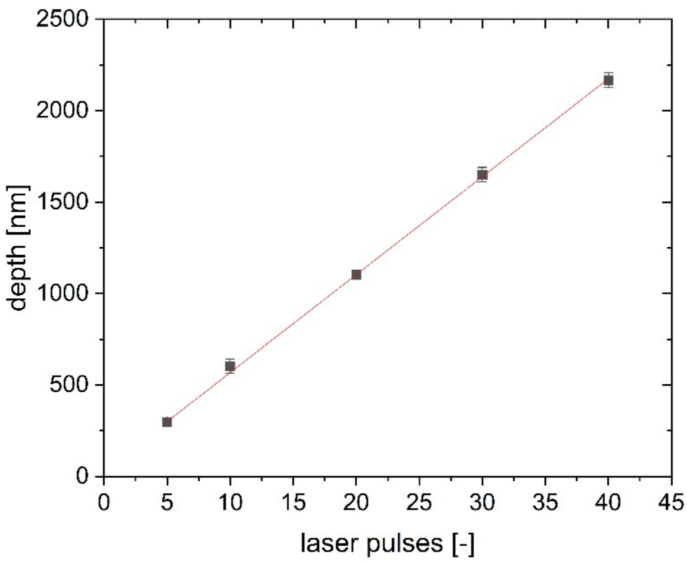

Fig. S2: Determination of the average ablation rate in SiC using 5, 10, 20, 30, and 40 laser pulses.

ESI2. Detailed information on the online-LASIL measurements

Online-laser ablation of solids in liquids (LASIL) measurements have been performed for a better comparison of the LA-ICP-MS and ToF-SIMS measurements. Therefore, online-LASIL has been applied to determine the overall Al-contents of the investigated SiC samples, a precondition for the calculation of the quantitative depth profiles.

For the online-LASIL analysis, all solutions and dilutions were prepared freshly with deionized water (18.2 MΩcm^{-1}) from a Barnstead EASYPURE II water dispenser (ThermoFisher Scientific, Germany). The concentrated hydrochloric acid was at least trace element grade, and the ICP single-element standards for Al, Si, and In had a 1000 mg/kg concentration and were purchased from Merck (Germany).

The ablation of the sample was conducted by a J200 LIBS system (Applied Spectra, Inc., Fremont, CA) with a pulse length of four ns and a 266 nm wavelength. The online-LASIL cell can be placed inside the XYZ stage of the laser system, enabling spatially resolved analysis as described by previously conducted works.[1, 2] For the sample analysis, a controlled segmentation of the carrier solution using an Arduino controller and a six-port-valve, as stated by Podsednik et al.,[3] was utilized. The segmentation of the carrier solution was achieved using Ar-gas; every 10 s, one bubble was introduced for 0.3 s with a pressure of 0.8 bar.

An iCAP TQ ICP-MS (ThermoFisher Scientific, Germany) in oxygen reaction mode was used as a detection system. For the sample introduction into the ICP-MS, a peristaltic pump (Perimax 12, SPETEC, Germany), a PFA nebulizer, and a cyclonic spray chamber were used. The ICP-MS was tuned for maximum intensity of the ^{115}In signal and CeO^+/Ce^+ ratio below 2 % before all experiments with a solution provided by the manufacturer. The data were recorded and evaluated by the instrument software Qtegra (Version: 2.10.4345.236). Detailed ICP-MS and laser parameters are listed in Table S1.

Table S1: ICP-MS and laser parameters for the online-LASIL measurement.

ICP-MS detection system	
RF Power	1550 W
Nebulizer gas flow	1.05 l/min
Cool gas flow	14 l/min
Auxiliary gas flow	0.80 l/min
Oxygen flow rate	0.23 ml/min
Measured Isotopes	^{27}Al, ^{27}Al^{16}O, ^{28}Si^{16}O, ^{29}Si^{16}O, ^{115}In
Dwell time	0.01 s (0.05 s for ^{27}Al and ^{27}Al^{16}O)
J200 Tandem LA/LIBS	
Laser fluence	7.2 J/cm^2
Spot size	150 µm
Scan speed	0.1 mm/s
Repetition rate	20 Hz
Liquid carrier solution flow	0.2 ml/min

The online-LASIL measurement aimed to ablate the whole Al-doped region with a single ablation layer. Therefore, the ablation depth required to ablate the whole Al-doped region must be evaluated. Thus, the cumulative Al-signal (normalized to Si) for the LA-ICP-MS depth profile (see Fig. 6 in the manuscript) has been calculated. The results demonstrate that 95% of the Al signal derives from the upper 600 nm for all the analyzed samples, indicating the minimal ablation rate for the online-LASIL measurement. To ensure the ablation of the whole doped region, the online-LASIL measurement's laser parameters were optimized to result in an ablation depth between 700 nm and 800 nm. The depth of the craters was evaluated with a "DektakXT" stylus profilometer (Bruker, USA).

The online-LASIL results were converted to atom density in solid (cm^{-3}), a standard Al-implantation specification, for combining the qualitative and quantitative data. This will further be important to compare the homogenous doped regions of the quantitative depth profiles with the reference values. The atom density calculation can be verified in Podsednik et al.'s[3] work (see Formula (1)). The needed parameters are Si and Al concentration in the carrier solution, the molecular weights, the density of silicon carbide (3,21 g/cm^{-3}), and the Avogadro constant

($6.022*10^{23}$ mol^{-1}). The results can be seen in Table S2; besides the atom density, the concentration in solid $c_{Al,solid}$ (µg/g) is displayed, the calculation can be seen in Formula (2).

$$C_{Al} = \frac{X_{Al}\rho_{SiC}N_A}{\sum_{i=1}^{n}(X_i * M_i)} \quad (1)$$

$$C_{Al,solid} = \frac{X_{Al} * M_{Al} * 10^6}{\sum_{i=1}^{n}(X_i * M_i)} \quad (2)$$

In this formula, X_i is the substance amount fraction calculated by the concentrations measured in the carrier solution.

Table S2: Results of the bulk online-LASIL measurements.

Dopant concentration of the constant region (cm^{-3})	Al concentration in solid $c_{al,solid}$ (µg/g)	Al atom density in solid $\rho_{N,Al}$ (cm^{-3})
3E+20	1691 ± 223	1.21E+20 ± 1.60E+19
1E+20	598 ± 161	4.28E+19 ± 1.15E+19
3E+19	153 ± 29	1.09E+19 ± 2.10E+18
1E+19	53 ± 23	3.77E+18 ± 1.62E+18

Looking at the online-LASIL results (see Table S2), one thing becomes evident: the measured Al atom density in solid does not align with the known atom density of the constant region. This is expected as only the upper 250 nm of the SiC samples are homogenously doped. Moreover, when the Al atom density in solid is plotted against the calculated Al-atom density of the constant doped region, a linear dependency with the formula y=0.41x+5E+16 and a coefficient of determination of 0.992 are achieved. This strongly indicates that the whole Al-implanted area of the samples was ablated.

For the combination of the data from the online-LASIL and LA-ICP-MS measurements, the correct alignment of the ablation depths is crucial to achieve accurate quantitative depth profiles. In this study, the depth of online-LASIL measurements serves as the alignment's reference point since it represents the analyzed volume's bulk concentration. For this purpose, the bulk Al atom density $\rho_{N,Al}$ measured by online-LASIL is split up based on the intensity values of the qualitative depth profile, as illustrated below:

$$c_i = \frac{\frac{I_{Al,i}}{I_{Si,i}} * \rho_{N,\ Al}}{\sum_{i=1}^{n}\left(\frac{I_{Al,i}}{I_{Si,i}}\right)} \quad (3)$$

In this equation, ci represents the quantitative result for a specific depth, and I_{Al} and I_{Si} are the Al and Si intensity of the corresponding qualitative depth profile. This equation can be seen as the distribution of the quantitative bulk measurement according to the normalized Al intensity of the qualitative analysis. It is essential to emphasize that this method is only valid when the summation of depth values l_i extends to the depth of the ablation crater corresponding to the online-LASIL measurement.

REFERENCES

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