U-Pb geochronology of zircon by femtosecond laser ablation

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Laser-induced down-hole fractionation of U and Pb is observed in ablation of zircon using nanosecond pulse width lasers (e.g. 213 nm Nd:YAG and 193 nm Excimer). The change in $^{206}Pb/^{238}U$ is greatest in the first 150-200 laser pulses (20 to 30 seconds of ablation at 5 Hz) and is controlled by laser fluence and laser spot size. Both parameters affect ablation rate and as the aspect ratio (depth:diameter) of the pit increases with the number of laser pulses, the signal intensity decreases as less material is removed from the ablation front. The combined effects of the strong fractionation at the start of ablation with the decay in signal intensity limit the accuracy and precision of the dating of small (<20 μm) zircons.

In this study the fundamental ablation behaviour of zircon has been investigated using a Photon Machines Analyte 198.fs femtosecond laser system (198 nm wavelength) and implications for U-Pb geochronology will be presented. Unlike with the nanosecond pulse laser systems there is no change in $^{206}Pb/^{238}U$ for the first 300 laser pulses (60 seconds of ablation at 5 Hz) at spot sizes in the range 20 to 60 μm. Although the beam energy distribution is Gaussian, the pits have relatively flat bottoms and near-cylindrical shapes at the larger diameters and become more conical with decreasing diameter. The ablation rate is slower than for nanosecond systems at similar spot sizes and fluences, and there is less condensation of ablated material and redeposition on the surface of the sample. These features result in a near steady-state signal intensity for a 60 second ablation at 5 Hz for a 40 μm spot size (shorter for smaller spots). The absence of down-hole fractionation and the efficient removal of material from the ablation site therefore enables higher spatial resolution.

The stoichiometric sampling of zircon using the femtosecond laser significantly improves data quality. Internal (single analysis) precision is typically better than 0.8% rsd for $^{206}Pb/^{238}U$ (compared to 1.2% rsd for the Excimer) and relative uncertainties on ages for zircon reference materials are ~0.9 to 1.3%: Plesovice: 336.1±3.5 Ma (2sd, n=25); Temora: 417.9±3.8 Ma (n=20); Seiland: 531.6±6.7 Ma (n=22); 91500: 1062.5±11.1 Ma (n=46).