Two-color wavemixing in Secondary Ion Mass Spectrometry

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Synopsis An alternative method for laser postionization in SIMS analysis is introduced. By mixing the fundamental laser field with an intensity of 1×10^{15} W/cm^2 with its second harmonic frequency component a control is attained over the ion yield of specific fragments.

Secondary ion mass spectrometry [1] (SIMS) is a proven and extremely valuable tool providing comprehensive chemical and spatial analysis of a broad range of materials. However the usefulness of the SIMS technique is severely limited by the low ion yield (~1%) and matrix effect [1] turning the quantitative analysis into a very difficult procedure. In this work the fundamental laser field of an infrared femtosecond postionization source is mixed with its second harmonic (SH) frequency in interaction region located above the sample surface. The resulting symmetry broken field is subsequently used to control the ionization yield in SIMS analysis. Although, the wavemixing was formerly broadly employed to demonstrate and explore symmetry breaking and/or coherent control, here, for the first time, we extend the method to SIMS analysis.

The MS part of the setup employed the time-of-flight SIMS IV (ION-TOF GmbH, Germany). The sample surface was bombarded with 25 keV Bi_3^+ primary ions with the pulse duration of 1 μs and the repetition rate of 50 Hz. An ultrafast three-stage Cr:forsterite fs IR MOPA (Master Oscillator Power Amplifier, 100 fs, 4.5 mJ, 1240 nm, photon energy ~ 1eV) was used as the laser source for postionization. A typical peak focal intensity (for the transform limited pulse) was of the order of 5×10^{14}-1×10^{15} W/cm^2 with the Keldysh parameter reaching 0.2, suggesting that the experiment was performed in the transition region between multiphoton and tunnel ionization. The laser beam from the amplifier passed through a f=300mm (f30) lens and a DKDP crystal cut for the type-I SH generation and the p-polarized fundamental field along with the second harmonic was subsequently focused into the target chamber of SIMS analyzer approximately 500 microns above the sample surface.

Native silver and nanosilver samples were chosen as a model system for the analysis. The yield of selected fragments was thereby enhanced/suppressed by a factor of 8 and 23 respectively. The influence of the symmetry broken field on the ion yield was confirmed by angular turning of the SH crystal (Fig. 1) as well as by revealing the phase dependence of the wavemixing process.

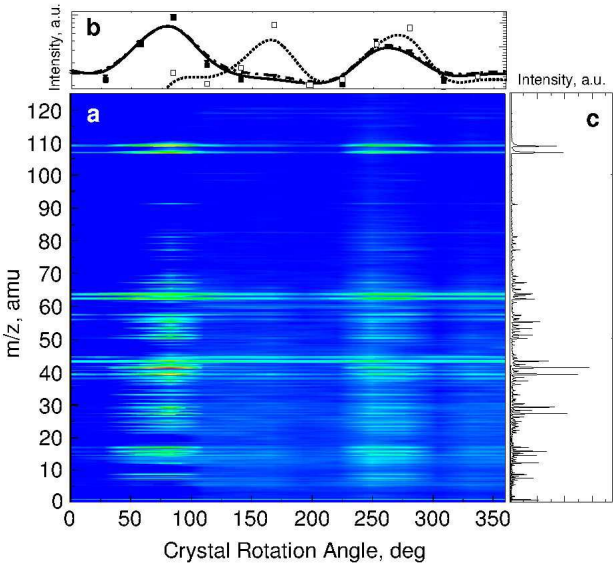

Figure 1. Nanosilver mass spectra as a function of the SHG crystal angular detuning

The method therefore opens additional degree of freedom for the control over the ion yield in SIMS analysis and is a further small step *en route* towards the ultimate chemical coherent control.

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References

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