Photoablation with time-evolving polarization states

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Over the past several decades, advances in direct surface structuring have been accomplished using a variety of ultrafast laser approaches. Micromachined structures show dependence on the pulse length, chirp, polarization, vector polarization, and topological states of the laser pulses. All of these parameters allow one to control material modification, thereby introducing new machining capabilities. Of particular interest in the past decade is the use of cylindrical and vortex vector beams. One area that remains relatively unexplored is the use of pulses with time-evolving polarization states.

Here, we explore the use of optical centrifuge pulses in photoablation of silica and silicon surfaces. Optical centrifuge pulses have linear polarization that undergoes angular acceleration to frequencies with $\Omega_{oc} > 6 \times 10^{13}$ rad/s over the duration of the pulse. [1,2] My research group has used an optical centrifuge to prepare gas-phase molecules in extreme rotational states in order to investigate their collisional energy transfer and bimolecular reaction dynamics. [3-7] Here we turn our attention to the effect of such pulses on photoablation. The optical centrifuge pulses are created by splitting Ti:sapphire laser pulses into pairs of pulses with $\lambda_0 = 805$ nm. One pulse ω_1 has positive chirp and the other pulse ω_2 has negative chirp. Opposite circular polarization is induced in the pair of pulses before they are recombined in time and space to form the angularly accelerating linear optical field. The instantaneous angular frequency Ω_{oc} is determined by the time-dependent frequency difference of ω_1 and ω_2, where $\Omega_{oc}(t) = \frac{1}{2}[\omega_1(t) - \omega_2(t)]$. This research compares photoablation for six polarization schemes: the optical centrifuge, a dynamic polarization grating, positively chirped pulses with linear or circular polarization, and negatively chirped pulses with linear or circular polarization.

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References:

[1] J. Karczmarek, J. Wright, P. Corkum and M. Ivanov, *Phys. Rev. Lett.* **82**, 3420 (1999)
[2] D. M. Villeneuve, S. A. Aseyev, P. Dietrich, M. Spanner, M.Y. Ivanov and P. B. Corkum, *Phys. Rev. Lett.* **85**, 542 (2000)
[3] L. W. Yuan, S. W. Teitelbaum, A. Robinson and A. S. Mullin, *Proc. Natl. Acad. Sci. USA* **108**, 6872 (2011)
[4] M. J. Murray, H. M. Ogden, C. Toro, Q. N. Liu, D. A. Burns, et al, *J. Phys. Chem A* **119**, 12471 (2015)
[5] H. M. Ogden, T. J. Michael, M. J. Murray, Q. N. Liu, C. Toro, et al., *Phys. Chem. Chem. Phys* **21**, 14103 (2019)
[6] M. R. Laskowski, T. J. Michael, H. M. Ogden, M. H. Alexander and A. S. Mullin, *Faraday Discuss.* **238**, 87 (2022)
[7] M. E. Ritter, S. A. DeSouza, H. M. Ogden, T. J. Michael and A. S. Mullin, *Faraday Discuss.* (2024) in press