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Site-specific modification of oxide nanoclusters: Towards atomic-scale surface structuring[1] KENNETH M. BECK, MATTHIAS HENYK, CHONGMIN WANG, WAYNE P. HESS, Pacific Northwest National Laboratory, PAOLO E. TREVISANUTTO, PETER V. SUSHKO, ALEXANDER L. SHLUGER, University College London — Atomic emission from MgO and CaO nanostructures is induced using laser light tuned to excite specific surface sites at energies well below the excitation threshold of the bulk material. Using selective 4.66 eV laser excitation of nanocrystalline thin films and nanocube metal oxide samples we have recorded a unique pattern of hyperthermal atomic desorption. Not only neutral O-atoms, but neutral Mg-atoms, with hyper-thermal kinetic energies in the range of 0.1–0.4 eV are readily observed. Our *ab initio* calculations suggest that metal atom emission is induced predominantly by electron trapping at surface 3-coordinated metal sites followed by electronic excitation at these sites- an 'electron plus an exciton' mechanism. The proposed elementary mechanism involves both sequential excitation and localization of excitons as well as electrons and holes at 3-coodinated surface sites. This mechanism differs from all previously suggested mechanisms for desorption induced by electronic transitions. This desorption process serves as an example of atomic scale modification of a nanostructured metal oxide using laser light tuned to excite specific surface sites at energies well below the excitation threshold of the bulk material.

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