Quantum-chemical Design of Improved Photoemissive Materials

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Improved photoemissive materials are needed for many applications, ranging from future electron and x-ray sources, such as free-electron x-ray lasers or energy-recovery linear accelerators, through night vision devices, scanners and detectors to solar cells. Several photoemissive properties need to be tailored for optimal applications. Such properties are the workfunction of the surface of the material, the quantum-efficiency (percentage photons turned into emitted electrons) and the angle of the emission cone. Furthermore, optimal devices are expected to be chemically stable under the application conditions. In a series of recent studies [1-3] we have designed improved photoemissive materials with applications-tailored properties using electronic structure calculations.

Motivated by recent results in surface catalysis, we have pointed out by electronic structure calculations that the angle of the emission cone can be tuned by the number of oxide monolayers deposited on metal surfaces [1], such as in MgO:Ag(001). The emission cone is determined by the ratio of the surface parallel and surface perpendicular momenta of the emitted electrons, and these are, in turn, determined by the extent and depth (below the Fermi-level) of the occupied portion of the surface bands in the momentum space (Brillouin-zone) of oxide-covered metals. The emission cone angle is smallest when 3 monolayers of MgO are deposited on Ag(001). Also note that the addition of a few MgO monolayers decreases the workfunction of Ag from 4.6 eV to 3.0 eV (experimentally confirmed).

In another study, we have designed [2] a chemical modification of the seasoned photoemissive material, Cs_2Te, such that the modification will preserve the exceptionally high (~20%) quantum-efficiency of Cs_2Te, while it decreases its workfunction from about 3.0 eV to ~2.4 eV. This is achieved by turning Cs_2Te into a conjugated pi-electron system through acetylation, leading to a new, designer compound of Cs_2TeC_2 in which a parallel arrangement of rod-like 1D polymers of $[TeC_2^{2n-}]_n$ is embedded in a Cs^+ matrix. Analogous compounds, such as Cs_2PdC_2 or Cs_2PtC_2 are known from the inorganic chemistry literature and we have successfully synthesized Cs_2TeC_2, as well. The lower workfunction of Cs_2TeC_2 is advantageous as it allows for using visible laser light instead of ultraviolet one (no photon loss due to wave-length down-conversion) and better pulse-shape control of the laser photons and the emitted electrons.

While searching for low-workfunction alternatives of Cs_2Te, such that preserve its high quantum-efficiency, we became aware of Cs_2Te_5, an existing phase of the Cs-Te system for which large single crystals have also been synthesized in the chemical literature. Also this material has polymeric 1D substructures embedded in Cs^+ matrix, due to polytelluride ions that form infinitely long wavy ribbons of ~4 Å diameter and are arranged parallel to each other. As low as 1.9 eV workfunction value has been calculated for the (010) surface of Cs_2Te_5 and even higher quantum efficiency than that of Cs_2Te is predicted for its illumination by visible photons.

Results from the experimental investigation of the above designer photoemissive materials will also be reviewed.

References:

1. K. Németh*, K.C. Harkay, M. van Veenendaal, L. Spentzouris, M. White, K. Attenkofer, G. Srajer; Phys. Rev. Lett., 104, 046801 (2010).
2. J.Z. Terdik, K. Németh*, K.C. Harkay, J.H. Terry Jr., L. Spentzouris, D. Velázquez, R. Rosenberg, G. Srajer; Phys. Rev. B., 86, 035142 (2012).
3. A. Ruth, K. Németh*, K.C. Harkay, J.Z. Terdik, L. Spentzouris, J. Terry; J. Appl. Phys., accepted for publication (2013).
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