

## Cu nanoparticle geometry as the key to bicolor behavior in Oregon sunstones: An application of LSPR theory in nanomineralogy

CHENGSI WANG<sup>1,†</sup>, ANDY H. SHEN<sup>1,\*</sup>, PETER J. HEANEY<sup>2,\*</sup>, AARON PALKE<sup>3</sup>, KE WANG<sup>4</sup>,  
HAIYING WANG<sup>4</sup>, AND LORE KIEFERT<sup>5</sup>

<sup>1</sup>Gemmology Institute, China University of Geosciences (Wuhan), Wuhan, Hubei 430074, China

<sup>2</sup>Department of Geosciences, The Pennsylvania State University, University Park, Pennsylvania 16802, U.S.A.

<sup>3</sup>Gemological Institute of America, Carlsbad, California 92008, U.S.A.

<sup>4</sup>Materials Research Institute, Pennsylvania State University, University Park, Pennsylvania 16802, U.S.A.

<sup>5</sup>Gübelin Gem Lab, Maihofstrasse 102, 6006 Luzern, Switzerland

### ABSTRACT

The coloration mechanism of Oregon sunstone is a classic and controversial topic in mineralogy because of the unique coexistence of anisotropic (green-red) and isotropic (red) color zones within single feldspar crystals. After nearly 50 years of research, no models proposed to date have satisfactorily accounted for all observed optical phenomena. Here, we present high-resolution transmission electron microscopy analyses of samples prepared by focused ion beam extraction along specific crystal directions. In both the anisotropic and the isotropic color zones, we observed Cu nanoparticles (NPs) included within plagioclase but with different geometries. In the isotropic (red) zone, NPs were randomly distributed nano-spheres or nano-ellipsoids (8.7–12 nm in diameter) with an aspect ratio of 1–1.3. In contrast, in dichroic (green/red) zones, NPs were directionally aligned nano-rods (8.5–21 nm along the long axis) with an aspect ratio of ~2.5. We applied localized surface plasmon resonance (LSPR) theory to simulate absorption spectra and developed a model to explain the observed optical properties. LA-ICP-MS and polarized UV-Vis spectroscopy were also performed to confirm our conclusions. This study systematically reveals the existence and optical influence of variably shaped metal-NP inclusions in feldspar crystals. Furthermore, it demonstrates the necessity of including LSPR in the canon of mineral coloration mechanisms. Cu-NP-bearing labradorite has been shown to exhibit third-order nonlinear optical properties, and approaches that incorporate NP shapes and sizes will assist in designing NP-embedded optical materials with tailored optical properties.

**Keywords:** Cu nanoparticles, Oregon sunstones, nano-inclusions, localized surface plasmon resonance, high-resolution transmission electron microscopy, plagioclase