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## Lifting the Cloak of Invisibility: Gold in Pyrite from the Olympic Dam Deposit, South Australia

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"Invisible gold" in pyrite refers to gold either present within the sulfide lattice or as discrete nanoparticle (<100-nm-diameter) inclusions (NPs), making it undetectable by conventional optical and scanning electron microscopy. Investigation of "invisible gold" in chalcopyrite-pyrite ores from the Olympic Dam Cu-U-Au-Ag deposit, South Australia, confirms the presence of Au in some arsenic-bearing pyrites at concentrations measurable by laser ablation inductively coupled-plasma mass spectrometry (LA-ICP-MS). Arsenic-bearing pyrite in the studied sample shows As-Co-Ni-oscillatory zoning patterns with variable complexity, suggesting grain re-crystallization during replacement by chalcopyrite (Fig. 1a). LA-ICP-MS data obtained from 164 pyrite grains plot below the Au and As solubility limit (Fig. 1b), empirically defined from studies of epithermal and Carlin-type deposits.

Several As-rich pyrite grains were analyzed using Scanning Transmission Electron Microscopy (STEM) and EDX-STEM analysis of foils obtained by Focused Ion Beam methods. Micron-scale, oscillatory zoning patterns observed on back-scattered electron (BSE) images and LA-ICP-MS element maps extend down to the nanoscale. Decoupling between trace elements is common, for example, Ni depletion wherever As and Co are enhanced, with nucleation of discrete Co-As-bearing NPs (cobaltite/safflorite?; Fig. 1c).

Importantly, Au-bearing NPs are identified in all cases, in intimate association with other (sulfo)tellurides (Fig. 1d-f). In addition, abundant cassiterite and rare chalcopyrite NPs are also identified. Some of the largest Bi-Ag-telluride NPs contain electrum as tiny pore-attached NPs within the larger telluride (Fig. 1d, e). Nanometer-sized electrum NPs were also identified in association with chalcopyrite. Silver-Au-telluride NPs form mono- or bi-component NPs (Fig. 1f, g). These NPs occur along trails displaying As-Co-enrichment, or formation of nm-wide lamellae of Bi-Pb-sulfotellurides marking pyrite twin boundaries (Fig. 1h). One wider lamella was identified from the layer stacking as a strongly disordered member of the aleksite series. Coarser tellurobismuthite ( $\text{Bi}_2\text{Te}_3$ ), a few  $\mu\text{m}$  wide, is associated with altaite ( $\text{PbTe}$ ) at pyrite-chalcopyrite boundaries.

Pyrite displays kink- and screw-dislocations associated with trace element remobilization or NPs nucleation. These defects can be associated with either "marcasitization" or loss of Fe (formation of pyrrhotite) within nanoscale domains affected by fluid percolation and pyrite recrystallisation. Twin planes in pyrite enriched in heavy elements (Bi-Pb-Te) represent zones of weakness and assist element exchange between host mineral and percolating fluids during coupled dissolution reprecipitation reactions (CDRR), analogous to those known for hematite from Olympic Dam. Nanoscale textures in pyrite allow for interpretation of Au-NPs, as Au released from solid solution in pyrite during CDRR associated with marginal chalcopyrite replacement.

Nanoscale analysis lifts the cloak of invisibility for Au in pyrite at Olympic Dam. These results show that confirmation of whether gold occurs as NPs or in solid solution based solely on position above or below the solubility limit of Au in pyrite on a plot of Au vs. As (Fig. 1b) is impossible without corroborative studies at the nanoscale.

Fig. 1. (a) BSE image showing boundary between oscillatory zoned pyrite grains. (b) Plot of Au vs As. Yellow circles: grains analyzed by STEM. (c, e) EDX maps of pyrite and electrum. (d,f,g) Selected NPs. (h) Bi-Pb-Te-enriched twin plane in pyrite.

