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New interpretation of low binding energy of Au $4f_{7/2}$ in Au-bearing pyrite and arsenopyrite from Carlin–type gold deposits

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Gold in Carlin–type gold deposits exists in pyrite and arsenopyrite and is typically not visible using scanning electron microscopy (SEM) and electron probe microanalysis (EPMA), which is therefore called invisible gold. Transmission electron microscopes (TEM) reveal that the invisible gold in pyrite and arsenopyrite exists as gold nanoparticles or clusters. However, valence states of gold atoms in the naturally occurred sulfides are still far from full understanding. X-ray photoelectron spectroscopy (XPS) is a powerful analytical tool for determination of the chemical state of the elements. Binding energies of Au $4f_{7/2}$as determined by XPS are from 83.7 to 84.0 eV for Au^0, from 85.1 to 86.0 eV for Au^{+1}, and from 87.1 to 87.7 eV for Au^{+3}, respectively. In some synthetic gold compounds, such as KAu_5, gold exists as Au^{-1} and its binding energy of Au $4f_{7/2}$is 81.9 eV. The binding energies of Au $4f_{7/2}$from Carlin–type gold deposits are mostly around 83.0 eV with some measurements as low as 80.7 eV. Since the binding energies of Au $4f_{7/2}$from Carlin–type gold deposits are low and similar to those for negative Au^{-1} in synthetic gold compounds, it was suggested that valence states of gold in pyrite and arsenopyrite are also negative and that Au replaces S in the crystal structures of pyrite and arsenopyrite. In other words, the low binding energy of Au $4f_{7/2}$is attributed to existence of negative gold Au^{-1} in these minerals. Other studies attribute the low binding energy of Au $4f_{7/2}$ to surrounding chemical environment. For example, in a XPS study on Au-polymer hybrid composite, low binding energy of Au $4f_{7/2}$ obtained was attributed to the interaction between gold nanoparticles and polymers. We propose that the decreased binding energy of Au $4f_{7/2}$ observed in the sulfides from Carlin–type gold deposits is due to size changes in gold nanoparticles or clusters. Relationship between binding energy of Au $4f_{7/2}$, W, and sizes of gold nanoparticles or clusters, R, can be expressed as W = -a/R + b, where a and b are positive constants. The calculated binding energy of Au $4f_{7/2}$ is as low as 83.0 eV for 500 nm gold nanoparticles. With decrease in sizes of gold nanoparticles or clusters, the binding energy will further decrease. The low binding energy of Au $4f_{7/2}$in pyrite and arsenopyrite from Carlin–type gold deposits cannot be used as an evidence for negative gold Au^{-1}. And the invisible gold in sulfides likely exists as nano-scale gold particles or clusters.