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Alluvial native gold, tetraauricupride and AuSn₂ from Western Switzerland

by Nicolas Meisser¹ and Joël Brugger²

Abstract

The chemical composition of gold, gold alloys and gold-bearing intermetallic compounds extracted from sediments collected in 16 rivers on the Molassic Plateau and in the Jura Mountains of Western Switzerland was analysed with an electron microprobe. Among the 70 particles studied, 68 consisted of Au–Ag alloys, one contained the mineral tetraauricupride (AuCu), and one the unnamed intermetallic compound AuSn₂. No correlation between the composition of the gold particles and the catchment basins could be recognised. Au–Hg amalgams were found sporadically as thin coatings around native gold particles, and formed by interaction between detrital gold and anthropogenic Hg. Metallic Hg was actually observed in some of the heavy mineral concentrates. The chemical composition of a tetraauricupride grain recovered from the Rio de Misery near Chandon (canton Fribourg), as well as the association with cupriferous native gold, indicate a natural origin for this mineral, whose occurrence is generally restricted to basic and ultrabasic-hosted Au mineralisations. The presence of an AuSn₂ intermetallic compound in the Boiron river (canton Vaud) is enigmatic. This compound has not yet been reported in nature, but the composition of the grain, its association with argentiferous native gold, the presence of frequent cassiterite grains, and the association with sediments highly enriched in humic acids, suggest a natural origin for this compound.

Keywords: alluvial minerals, native gold, tetraauricupride, AuSn₂, anthropogenic mercury, Switzerland.

1. Introduction

Due to their mechanical and chemical stability, native gold particles can be transported over large distances with clastic sediments. In Switzerland, gold has been found in the sediments of many rivers in the Jura Mountains and the Molasse Basin (e.g. HOFMANN, 1992; GLAUSER, 1999). Some alluvial gold particles may have an anthropogenic origin, but most were derived from primary Alpine deposits (e.g. JAFFÉ, 1989). The transport of gold from the lode deposits in the Alpine chain to the foreland (what is now the Molasse Basin and Jura chain) occurred at two different times. During the Tertiary, gold was transported in fluvial systems together with the clastic Molasse sediments (e.g. SCHMID, 1973). During the Quaternary glaciations, gold was transported by the glaciers (e.g. HOFMANN, 1992).

Historic exploitation of placer deposits in Switzerland took place both in relatively proximal (e.g. Napfgebiet) and distal (e.g. Rhine at Basel) locations relative to the primary deposits. The mineralogy of placer deposits in rivers from the Molasse Basin and Jura mountains in Western Switzerland has not been studied. Modern attempts of exploitation of such deposits occurred in the 1930's and 1940's around Geneva (PITTARD, 1936a, b) and Fribourg (RUTSCH, 1941). Later, François Maeder systematically sampled sediments from rivers of Western Switzerland (MAEDER, 1983; Fig. 1). At each locality, the heavy minerals were separated with a pan from about 12 l of sediment, previously screened over a 6 mm sieve. The concentrates were examined under a stereo-microscope and native gold and other alloys hand-picked. Between 2 to 10 gold particles were recovered by this procedure. This sampling

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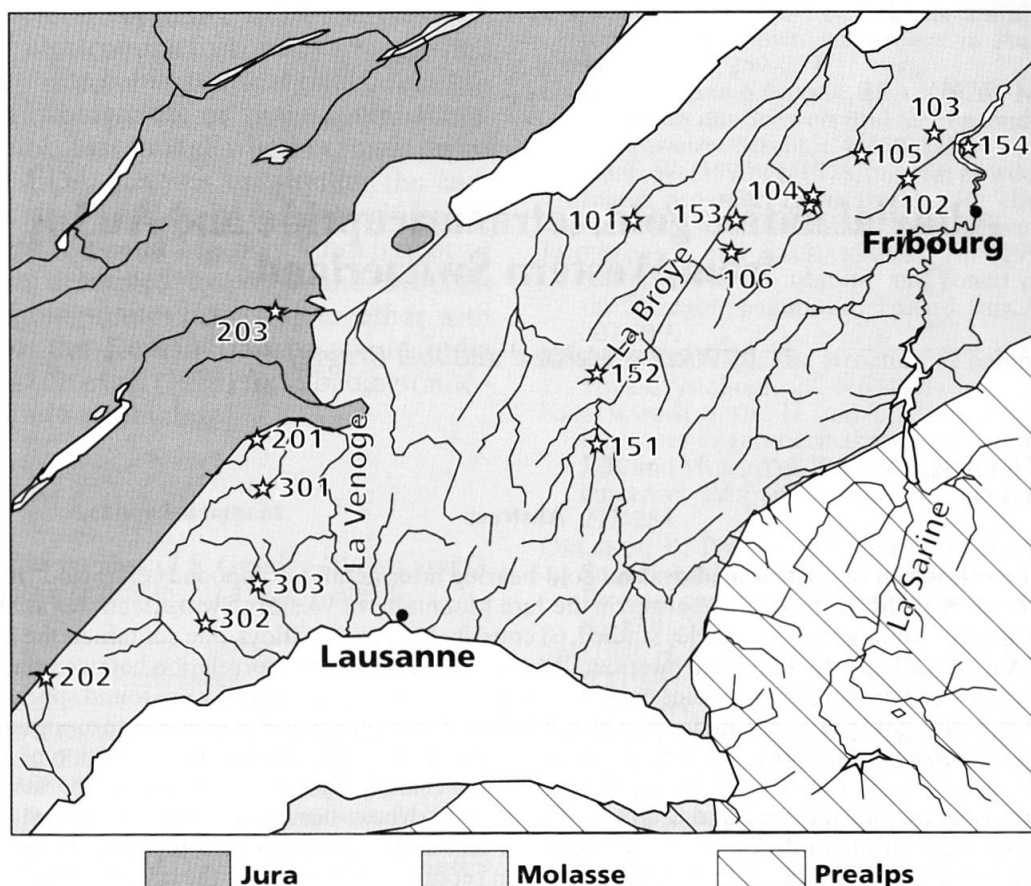


Fig. 1 Location of the studied samples in Western Switzerland in relation to the main geological features.

Tab. 1 List of sampled and studied localities.

Nr on Fig. 1	River	Locality, Canton	Swiss coordinates
<u>Molasse Basin</u>			
101*	La Petite Glâne	Franex, FR	553230/182500
102*	Le Tiguellet	Belfaux, FR	575140/185280
103*	Ruisseau de Pensier	Courtepin, FR	576140/188340
104	L'Arbogne	Mannens, FR	566930/183670
105	Rio de Misery	Chandon, FR	570730/189600
106	Ruisseau de Trey	Trey, VD	561900/179800
151	La Carrouge	Moudon, VD	550290/166490
152	La Cerjaule	Moudon, VD	550500/170700
153*	La Bioleyre	Fétigny, VD	560600/182300
154*	Le Lerapesson	Granges-Paccot, FR	577960/186010
<u>Jura Mountains</u>			
201	La Venoge	Moiry, VD	524670/166050
202	La Serine	Begnins, VD	507700/144600
203	L'Orbe	Les Clées, VD	525375/175880
<u>Lemanic region</u>			
301	Le Veyron	Chavannes-le-Veyron, VD	524230/161850
302*	Le Boiron	Yens, VD	521440/151610
303	La Morges	Vaux-sur-Morges, VD	524620/154600
<u>Others</u>			
	Rotache	Brenzikofen, BE	613500/184070
	Le Trient	Trient, VS	565440/101160
	Lonza	Kippel, VS	625900/138670

* Au amalgam occurrence.

All samples collected by F. Maeder, except for sample 203 collected by F. de Cocatrix.

has been completed with gold particles collected in the Orbe river by François de Cocatrix (Tab. 1), and we present here the results of mineralogical and chemical investigations on this material.

2. Analytical methods

Particles of native gold and unidentified Au alloys were embedded in epoxy resin and polished. Samples were examined with a Leitz Ortholux II POL-BK reflective light microscope equipped with a 50 W high pressure mercury arc lamp. Quantitative chemical analyses were obtained with a CAMECA SX 50 electron microprobe at the Université de Lausanne operated at 20 kV, 30 nA. Counting times were 20 s per element, and the standards (element, line) used were synthetic Au (Au, $L\alpha$), synthetic Ag (Ag, $L\alpha$), synthetic Cu (Cu, $K\alpha$), synthetic Fe (Fe, $K\alpha$), natural cinnabar HgS (Hg, $L\alpha$), synthetic Pd (Pd, $L\alpha$), synthetic Ni (Ni, $L\alpha$) and synthetic Sn (Sn, $L\alpha$). One Au–Ag amalgam fragment was studied by powder X-ray diffraction using a 114.6 mm diameter Gandolfi camera and Ni-filtered Cu $K\alpha$ X-radiation.

3. Mineral descriptions

3.1. NATIVE GOLD

Among the 70 gold particles analysed by electron microprobe, 68 consisted exclusively of Au–Ag alloys, one contained the mineral tetraauricupride (AuCu), and one a Au–Sn intermetallic compound not previously described in nature.

The composition of the native gold particles ranged from very pure native gold (99.9 at% Au) to auriferous native silver (47.60 wt% Ag, corresponding to 62.16 at%; Fig. 2). The native gold particles generally had low Cu contents (< 0.5 wt%), but some particles plotted along the Au–Cu join and contain up to 4.67 wt% Cu (Fig. 2). Thin coatings (< 5 μ m) of metallic white

amalgam (Au–Hg alloy) have been reported from six different rivers (MAEDER, 1983). The Hg contents measured in the core of the analysed particles were usually low, and only three particles displayed values higher than 1 wt% Hg. The Hg contents were usually highly variable within a single grain (Tabs 2 and 4). A brittle Au–Ag amalgam grain (120 \times 150 μ m) coating a native gold particle from Le Leraresson river (canton Fribourg), was analysed by X-ray powder diffraction, which showed that it was composed of two hexagonal phases: Au₂Hg and (Au,Ag)₃Hg₂ (Tab. 3). Some native gold particles also contain minor amounts of Ni (up to 0.33 wt%) and Pd (up to 0.06 wt%). Except for Hg, the investigated gold particles are chemically fairly homogeneous. A particle from La Cerjaule river (canton Vaud) is an exception, and shows a core to rim zoning (Tab. 2). The rim is enriched in Au and depleted in Ag relative to the core. Hg and Ni seem to show more variability in the rim than in the core of the particle.

3.2. TETRAAURICUPRIDE (AuCu)

Only one particle of alluvial tetraauricupride, the tetragonal $P4/mmm$ modification of (AuCu), was found during our study at Rio the Misery near the village of Chandon (canton Fribourg). This tetra-

Tab. 2 Electron microprobe analyses (wt%) of a chemically zoned gold particle from La Cerjaule river (canton Vaud).

	Core	Core	Rim	Rim
Au	50.24	51.15	71.28	96.36
Ag	45.15	47.60	27.58	3.02
Cu	0.05	<0.04	<0.04	0.04
Hg	0.56	0.88	0.48	<0.05
Ni	0.22	0.27	0.23	<0.04
Total	96.22	99.90	99.57	99.42

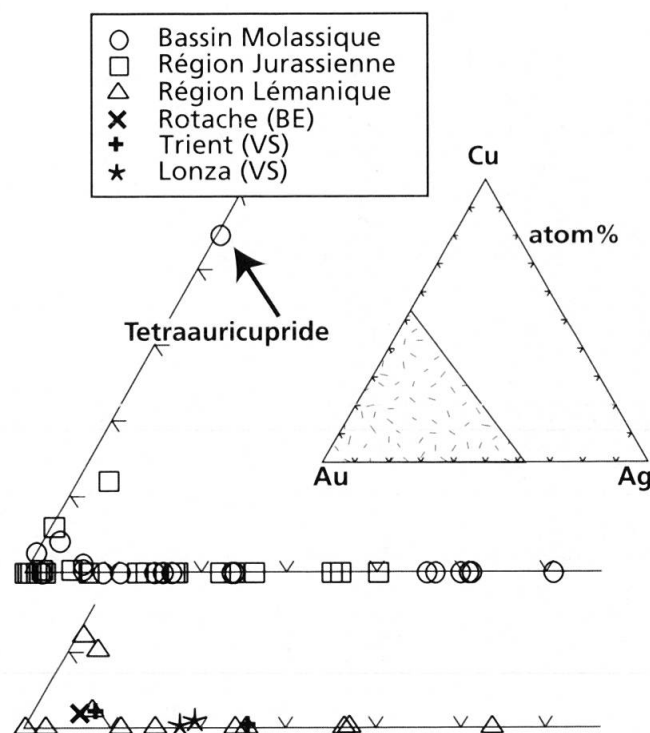


Fig. 2 Triangular Au–Ag–Cu (at%) diagram showing the chemical composition of native gold particles from alluvial sediments from Western Switzerland.

Tab. 3 Powder diffraction data for Au–Ag amalgam grain from Le Lerapesson river (canton Fribourg) compared with (Au, Ag)₃Hg₂ (weishanite) and Au₂Hg.

Au–Ag amalgam Lerapesson (FR)		(Au, Ag) ₃ Hg ₂ , weishanite JCPDS card Nr 43-0694			Au ₂ Hg JCPDS card Nr 19-0522		
d _{obs} [Å]	I _{vis.}	d _{obs} [Å]	I	h k l	d _{obs} [Å]	I	h k l
2.87	30				2.867	20	0 0 6
2.60	30				2.590	20	2 0 6
2.53	20	2.5293	40	1 0 0			
2.430	60				2.421	100	5 0 0
2.398	50	2.4067	30	0 0 2			
2.338	40				2.330	20	3 3 0
2.290	30				2.278	20	2 0 7
2.233	100	2.2432	100	1 0 1	2.226	100	5 0 3
2.060	10				2.047	10	3 3 4
2.019	20				2.011	20	2 2 7
1.945	10				1.941	20	5 1 4
1.895	20				1.892	20	5 2 2
1.746	10b	1.7504	50	1 0 2	1.747	20	4 4 0
1.656	20				1.654	20	2 0 10
1.522	20b				1.521	20	5 2 7
1.507	20b				1.500	20	5 0 9
1.461	60b	1.4609	60	1 1 0			
1.442	40				1.437	20	6 3 4
1.401	60				1.398	50	5 5 0
1.355	50	1.3593	80	1 0 3	1.358	20	7 3 1
1.342	40				1.339	20	8 0 6
1.323	30				1.319	20	3 1 12
1.301	60				1.298	50	3 3 11
1.282	20				1.282	20	5 3 9
1.266	20				1.267	20	7 2 7
1.249	70	1.2509	80	1 1 2			
1.226	70	1.2255	60	2 0 1	1.223	100	1 0 14

auricupride grain measured $400 \times 180 \mu\text{m}$ (Fig. 3), and showed a lower reflectivity than native gold under the metallographic microscope. Under plane-polarised reflected light in oil, its colour was copper red with a slightly yellow tint. Pleochroism is not observed. Under completely crossed polarizers in oil immersion, tetraauricupride displays a weak anisotropy with grey to

brownish tints. The tetraauricupride from Rio de Misery was composed almost exclusively of Au and Cu (Tab. 4), with only traces of Ag. Au was more abundant than Cu (Au:Cu = 53.5:46.5 at%). The native gold particle included in the tetraauricupride contained significant but variable contents of Cu and Hg (Tab. 4), and was poor in Ag.

Tab. 4 Electron microprobe analyses of a tetraauricupride particle associated with mercurian native gold from Rio de Misery near Chandon (canton Fribourg).

	Tetraauricupride				Native gold			
	#1	#2	#3	Mean	#4	#5	#6	Mean
Au [wt%]	79.65	77.44	78.70	78.59	97.17	98.05	97.17	97.46
Ag [wt%]	0.07	<0.05	<0.05	<0.05	0.08	<0.05	0.08	0.05
Cu [wt%]	22.08	22.00	21.82	21.97	1.59	0.97	1.59	1.38
Hg [wt%]	<0.05	<0.05	<0.05	<0.05	0.44	1.17	0.44	0.68
Total	101.80	99.44	100.52	100.56	99.28	100.19	99.28	99.57
Au [mol%]	53.74	53.18	53.78	53.58	94.64	95.93	94.64	95.09
Ag [mol%]	0.09				0.14		0.14	0.09
Cu [mol%]	46.17	46.82	46.22	46.42	4.80	2.94	4.80	4.17
Hg [mol%]					0.42	1.12	0.42	0.65

3.3. AuSn₂

This compound, not yet described as a mineral species, was found in the Boiron river near the village of Yens, Morges district (canton Vaud). The Boiron river drains a large forest and transports large quantities of vegetal debris. The studied heavy minerals – including native gold and AuSn₂ – were sampled in sediments showing an alternation of black brownish layers rich in organic matter and grey sands. *In situ* pH measurements of intergranular water were conducted with a pH-meter WTW 323-B in December 1999, and showed values between 5.6 and 6.0. 100 g dried, homogenised gold-bearing sediment contained 32 wt% of humic acids soluble in 1 N aqueous NaOH.

AuSn₂ appears as a 65 × 20 µm particle at the border of an argentiferous native gold particle (Fig. 4). A systematic search for Sn in the Boiron river heavy mineral concentrates allowed to identify frequent cassiterite grains, but also rare anthropogenic Sn phases like metallic tin and Sn–Pb alloys. Under the metallographic microscope, AuSn₂ is white and more reflective than native gold. Pleochroism is not observed under plane-polarised reflected light in oil immersion. Under completely crossed polarisers in oil, AuSn₂ displays a strong anisotropy with blue to brown yellow tints. The grain is an aggregate of monocrystalline acicular domains, measuring about 1 µm in length. The studied AuSn₂ phase contains only Au and Sn. In particular, Ag, Cu, Hg, Pd, Pt and Ru

were analysed with the electron microprobe, and found to be below detection limits (about < 0.05 wt% each).

4. Discussion

Native gold with variable Ag content is common in primary Au deposits. Although most argentiferous gold particles probably have a natural origin, many Au–Ag alloys are used in jewellery, and an anthropogenic origin for some of the studied particles cannot be excluded (DEVISMES, 1978). No specific chemical typology as a function of catchment basin is evident over the studied area (Fig. 1). In the studied area, on the Swiss Molasse

Tab. 5 Electron microprobe analyses of AuSn₂ associated with argentiferous gold from the Boiron river near Yens, canton Vaud. Hg, Pd, Pt, Ru, Fe and Ni were below detection limits (about 0.05 wt%).

	AuSn ₂				Argentiferous gold
	#1	#2	#3	Mean	#4
Au [wt%]	55.73	55.44	55.55	55.57	90.73
Ag [wt%]	< 0.05	< 0.05	< 0.05	< 0.05	9.30
Cu [wt%]	< 0.05	< 0.05	< 0.05	< 0.05	0.06
Sn [wt%]	43.03	43.04	43.07	43.05	
Total	98.76	98.48	98.62	98.62	100.09
Au [mol%]	31.76	32.05	31.85	31.89	84.09
Ag [mol%]					15.73
Cu [mol%]					0.18
Sn [mol%]	68.24	67.95	68.15	68.11	

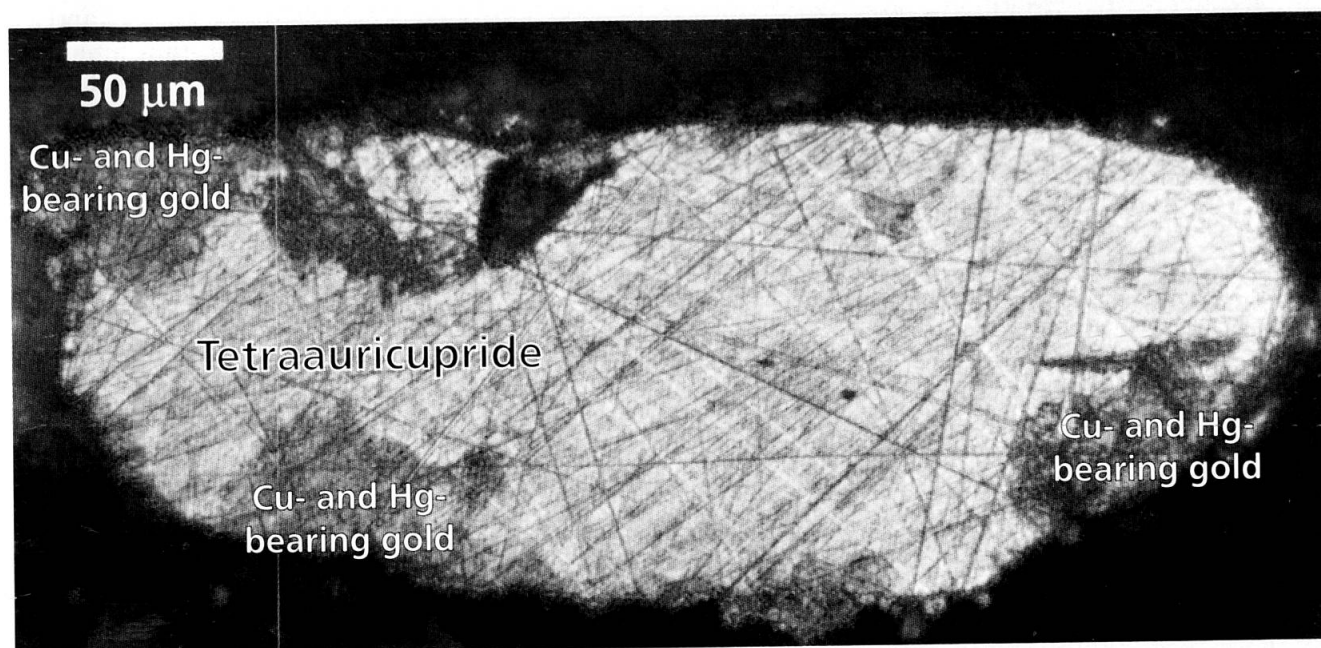


Fig. 3 Microphotograph of tetraauricupride particle with a rim of cupriferous and mercurial native gold. Rio de Misery near Chandon, canton Fribourg. Partially crossed nicols; oil immersion. The colour microphotograph was digitally treated to enhance the contrast for black and white publication. The shown picture is the blue layer of the CMYK picture. The dark regions (native gold) appear darker, although they have a higher reflectivity.

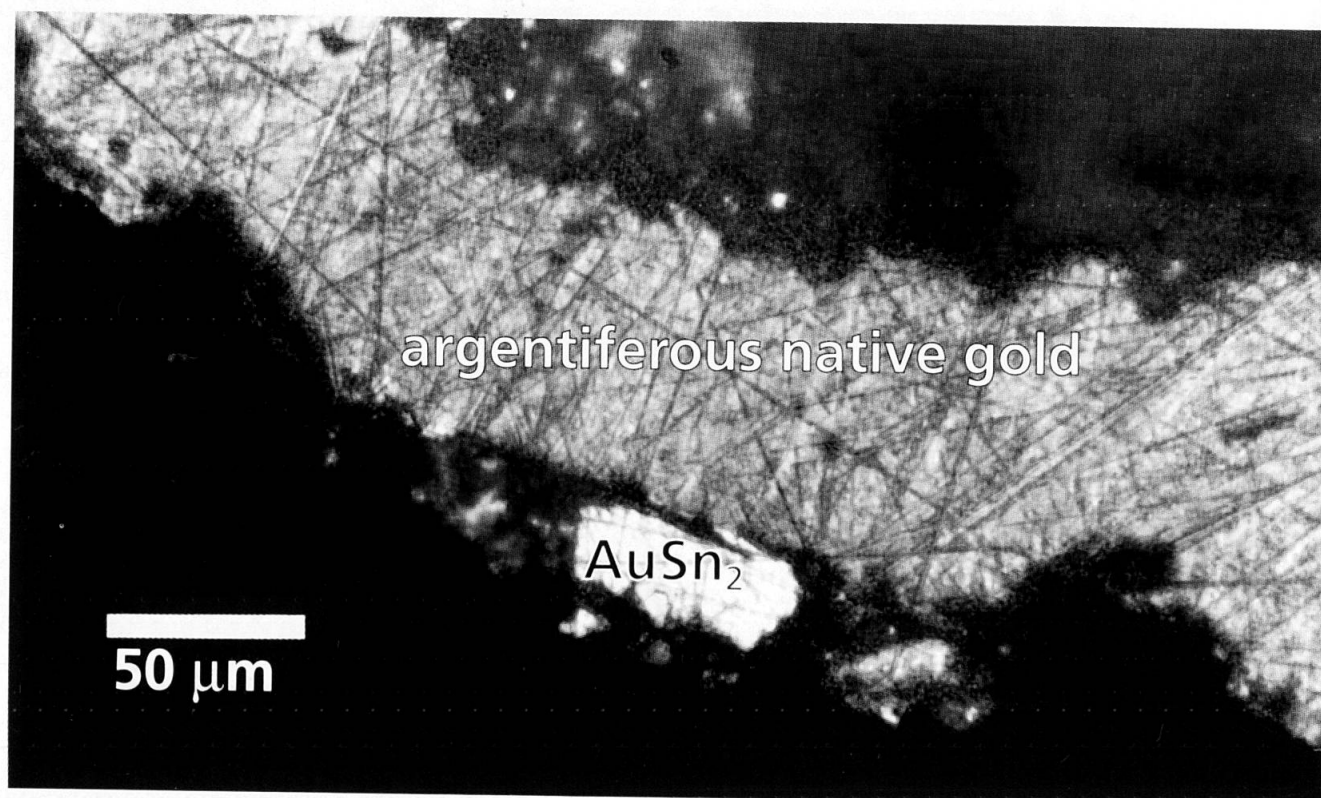


Fig. 4 Microphotograph of a AuSn_2 grain at the rim of an argentiferous native gold particle. Le Boiron river near Yens, canton Vaud. Partially crossed nicols; oil immersion.

Plateau, rivers always cut through large amounts of late Wurmian sediments left by the Rhône glacier, and some gold obviously was transported during the Quaternary. On the other hand, with the exception of the Orbe river where the sampling area is located on Cretaceous limestone, the sampled rivers run over Oligocene–Miocene Molasse, and Tertiary transport of some of the recovered native gold particles is probable.

Ag-rich rims along Au–Ag particles similar to that observed in a particle from Cerjaulle river (Tab. 2) have been described in natural nuggets from many localities (e.g. MANN, 1984; GROEN et al., 1990). Several mechanisms have been proposed for their formation: under reducing conditions at ambient temperature, Au^+ complexes with different inorganic (e.g. Cl^- , SO_4^{2-} , HS^-) and organic (e.g. humic acids) ligands are usually much less soluble than Ag^+ complexes (e.g. COLLIN and VIEILLARD, 1991). On the other hand, under relatively oxidising conditions, the silver component in the Au–Ag solid solution is unstable with respect to AgCl and AgSO_4 (e.g. KRUPP and WEISER, 1992).

Au amalgams often form within sediments in rivers by reaction between detrital gold particles and mercury of anthropogenic origin (PALACHE et al., 1944; CHEN et al., 1981). Indeed, weishanite ($\text{Au, Ag}_3\text{Hg}_2$) is the only Au amalgam accepted as a mineral species, i.e. of certain non-anthropogenic

origin. This alloy was found *in situ* in the silicified zone of the Ag-rich part of an Au–Ag deposit in the Poshan mining district, Tongbai, Henan Province, China (LI et al., 1984). Alluvial native gold with low Hg content is widespread in the studied area. Particle with elevated Hg content in the core may have a natural origin (Tabs 2 and 4). In contrast, mercury occurring in thin amalgam coatings around gold particles certainly has an anthropogenic origin. In rare cases, Energy Dispersive X-Ray Spectroscopic qualitative chemical analyses revealed the presence of microscopic ($< 5 \mu\text{m}$) mercury droplets at the surface of these grains. In the Aubonne river (canton Vaud), metallic Hg was found in heavy mineral concentrates. Several anthropogenic sources of Hg in non-industrial area can be cited:

- sink mud with low Hg contents used as fertilisers;
- antiseptics (merbromin, thiomersal, phenyl mercuric borate, etc.) and diuretics (meralluride, mercaptomerine, mersalyl, etc.) in human and animal medicine;
- agricultural fungicides (HgCl_2 , Hg_2Cl_2 , phenyl mercuric chloride and nitrate) and herbicides (phenyl mercuric acetate). The use of these substances is now prohibited;
- mercury fulminate as old constituent of priming melange in cartridges and shells;

– historic Au panning using metallic Hg for the recovery of native gold particle (not documented in the studied area).

The intergrowth of tetraauricupride with cuprian (and mercurian) native gold observed in the grain from Rio de Misery (Fig. 3) is typical of natural deposits. The composition of the tetraauricupride (excess Au) from Rio de Misery is very close to that reported for tetraauricupride from Riam Kanan, Borneo (STUMPFL and CLARK, 1965) and from the Kerr mine, Ontario (KNIPE and FLEET, 1997). Therefore, we conclude that a natural origin of the tetraauricupride from Rio de Misery is likely. Tetraauricupride was first reported from serpentinised pyroxenites associated with PGM mineralisation along with native gold, native silver, magnetite, chromite, pyrrhotite, pyrite and chalcopyrite at Sardala, Xinjiang, China (CHEN et al., 1983). Further occurrences were reported from other PGM-bearing mafic/ultramafic rocks displaying more or less advanced alteration (Talnakh, Siberia; Karabash, Urals; Troodos ophiolitic sequence, Cyprus); from some alluvial PGE deposits derived from the weathering of ophiolitic units (Riam Kanan, Borneo); or from retrograde assemblages in the komatiite-hosted hydrothermal deposit at the Kerr Mine, Ontario (see KNIPE and FLEET, 1997 and references therein). In the Swiss Alps, an *in situ* occurrence was reported in Alpine stage ilmenite–pentlandite–djurleite–native copper–andradite–forsterite±chrysotile veins cutting some parts of the Geisspfad ultramafic complex, Binntal, Valais (MEISSER, 1999). The association with basic rocks is a common feature of all these occurrences. Tetraauricupride most likely forms at relatively low temperature by the interaction of Cu-rich fluids derived from the mafic rocks with Au mineralisation pre-existing in these rocks. At atmospheric pressure, tetraauricupride is stable up to 400 °C (OKAMOTO et al., 1987).

AuSn₂ has not been previously reported in nature. Synthetic AuSn₂ is orthorhombic with space group *Pbca*, and melts incongruently at 315 °C in the binary Au–Sn system (OKAMOTO and MASALSKE, 1984; LEGENDRE et al., 1985). The synthetic phase has a very restricted composition range (LEGENDRE et al., 1985). Yuanjiangite, AuSn, is the only natural Au–Sn intermetallic compound documented as a mineral species. This rare mineral has been described by LICHANG et al. (1995) from placer deposits in the Hunan province (China). Yuanjiangite is seen as having formed by *in situ* replacement of placer gold within Sn-rich sediments. The recovered AuSn₂ forms a partial rim around a gold particle. This suggests an origin similar to that of yuanjiangite, i.e., by interaction between a gold particle and a Sn-rich mineral in

acidic sediments. Sn-minerals are not common in the Swiss Plateau alluvial deposits, and cassiterite (SnO₂) was previously reported only in heavy mineral concentrates from the Napf area (STALDER et al., 1998). The native argentiferous gold associated with the AuSn₂ grain contained 9.30 wt% Ag. This composition seems natural, as only the artificial alloy called “Pale yellow gold” with 92 wt% Au, 0–8.3 wt% Ag and 0–8.3 wt% Fe approaches this composition. The only anthropogenic source for Au–Sn compounds is found within the micro-electronic industry. Experimental Au–Sn solder joints for microprocessors are initially loaded with large quantities of the brittle AuSn₄ phase, which is then gradually decomposed to the more reliable AuSn₂ by thermal treatment (LINDER et al., 1996). No microprocessor manufactory is known within the Le Boiron river basin. Moreover, these materials would be produced from high purity gold devoid of Ag. Au–Sn alloys may also form during gold-plating of bronze objects; however, the absence of detectable Cu and Hg in the native gold associated with AuSn₂ does not support such an origin. We conclude that the AuSn₂ grain from the Boiron river most probably formed by natural chemical exchange processes directly in the river. Because of the complex chemical environment characterising the hosting sediments (low pH, high concentrations of humic acids, possible involvement of biological or photochemical reactions; e.g. WOOD, 1996), and in the absence of experimental data about the environmental geochemistry of the Au–Sn system, the nature of the AuSn₂-forming process remains speculative.

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