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Filamentary native gold from the Elatsite porphyry Cu-Au deposit, Srednogorie zone, Bulgaria

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Abstract. Rare filamentary native gold is described from ores of the Elatsite porphyry Cu-Au deposit, Srednogorie zone, Bulgaria. The tortuous filaments formed bush-like clusters within small cavities of bornite-chalcopyrite ore. The filaments present euhedral single crystals, usually 2-3 to 10-15 µm wide and up to 1 mm long. Their stepped surfaces are bounded by alternating and unequally developed small flat faces of typical crystal forms of gold: *o*{111}, *d*{110}, *a*{100}, *f*{310}, *m*{311}, etc. The filamentary gold contains in average 18.7 wt.% Ag and 1.1 wt.% Cu. Au is closely related to the early high-temperature (600-530°C) magnetite-bornite-chalcopyrite mineral assemblage, most probably as Au-rich bornite solid solution, and is later re-deposited in native form in the close neighbourhood during formation of the main, lower-temperature (535-400°C) quartz-chalcopyrite-pyrite assemblage. The extremely anisometric and non-equilibrium filamentary crystals were grown from highly supersaturated heterogeneous boiling solutions.

Key words: native gold, filamentary crystals, crystal growth, Elatsite porphyry Cu-Au deposit, Srednogorie zone

Introduction

Native gold occasionally occurs as well shaped crystals and crystal aggregates, which always attract special attention and are a subject of considerable interest. The variety of crystal morphologies of native gold is its specific feature, widely discussed in the literature. Important generalisations are given by Goldschmidt (1918), Petrovskaya (1973), Abdulin et al. (2000), and other authors. Mostly, gold is known in octahedral, dodecahedral and more complex crystals, in crystals twins, in variable dendritic forms, prismatically elongated, flat or irregular crystal formations, etc. Much rarely, hair-like and fine fibrous gold formations are observed. Petrovskaya (1973) reported for wire-like gold (with thickness of 0.5-0.05 mm) from the Russian deposits Darasun, Lebedinoe, Baley, etc., and assigned it to a morphogenetic type of not well-bounded and rounded crystals. Novgorodova (1983) also mentioned for wire-like gold from the Urals golden deposits, with rounded cross-sections and without indications for crystallographic bounding. Fibrous ribbon-like gold (up to 2-3 cm long and to 0.3 mm thick) is known also from the classic Transilvanian gold deposits in Romania, and specimens of them are displayed in the famous Gold museum in Brad. However, the real crystal morphology especially of the fine filamentary gold is not well characterised and understood, although it is important in understanding the mechanisms and conditions of gold formation. Filamentary shapes are common for native silver and copper (Maleev, 1971).

We found well-formed aggregates of filamentary gold in the ores of the Elatsite porphyry Cu-Au deposit, not known earlier.

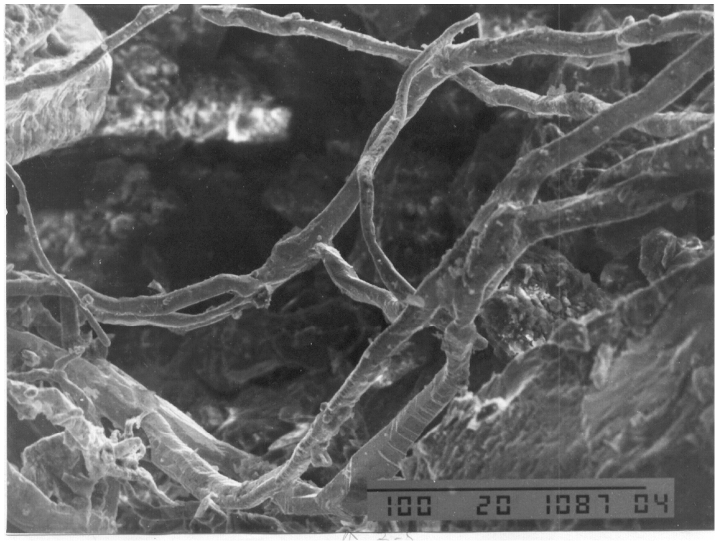

Fig. 1. SEM microphotograph of a group of not straight filaments of native gold in a small cavity of magnetite-bornite-chalcopyrite ore. The Elatsite deposit. Scale bar – 100 μm

Spatially and genetically this gold is closely related to the early magnetite-bornite-chalcopyrite assemblage (Fig. 1). Here are presented results of the morphological study, together with some discussion on the origin of this gold.

Geological setting and mineral assemblages

The Elatsite porphyry Cu-Au deposit is located in the northern part of the Panagyurishte ore district of the Central Srednogorie, considered as an Late Cretaceous calk-alkaline magmatic arc, which is a part of the Alpine-Balkan-Carpathian-Dinaride metalogenic province (Ciobanu et. al., 2002, and others). It is exploited since three decades in one of the largest open-pit mines in Eastern Europe.

Host rocks are Early Paleozoic metamorphosed green schists ('diabas-phyllitoid complex'), Paleozoic (314 Ma) granodiorites of the Vezhen pluton, and Late Cretaceous medium-acid subvolcanic bodies and porphyry dykes (Bogdanov, 1987; Dragov and Petrunov, 1996; Popov et al., 2000; Strashimirov et al., 2002; von Quadt et al. 2003; Georgiev, 2005; and others). The stockwork ore mineralisation is controlled by diagonal NW and NE faults. According to the U-Pb dating of von Quadt et al. (2003) the magmatic-hydrothermal ore-forming process extended between 92.1-91.84 and 91.42 Ma. Close are the Os-Re data of Zimmerman et al. (2003).

The main ore mineral assemblages (Petrunov et al., 1992; Petrunov and Dragov, 1993; Dragov and Petrunov, 1996; Tarkian et al., 2003; Kehayov and Bogdanov, 2005; Bogdanov et al., 2005) are as follows: (1) magnetite-bornite-chalcopyrite (*mt-bn-cp*), presented locally and associated with K alteration, containing also quartz, native gold, pyrrhotite, and minor Co-Ni tiospinels, molybdenite, PGE minerals, native Te, tellurides and selenides; (2) quartz-pyrite-chalcopyrite (*q-py-cp*), the main ore assemblage with gold, minor tennantite, molybdenite, etc.; (3) quartz-pyrite (*q-py*), (4) quartz-galena-sphalerite (*q-ga-sph*), and (5) late quartz. The temperature ranges according to micro thermometric data of Kehayov et al. (2003) are, respectively: 600-530°C, 535-400°C, 420-350°C, 364-200°C, 162-130°C.

Native gold is related to the sulphide

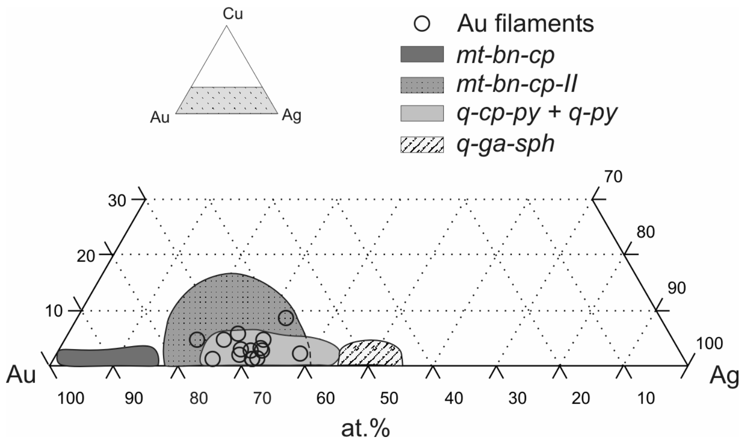

Fig. 2. Chemical composition of filamentary gold and compositional fields of gold from the main ore assemblages: *mt-bn-cp, q-cp-py + q-py and q-ga-sph* (after data of Strashimirov and Kovachev, 1996; Tarkian et al., 2003; Georgiev, 2005; and Kehayov and Bogdanov, 2005). In particular, *mt-bn-cp-II* concerns to visible gold cutting early bornite

assemblages. After Tarkian et al. (2003), highest is the content of Au in ore of the *mt-bn-cp* assemblage (19.3 ppm average from 8 analyses of ore samples), followed by the widest spread *q-cp-py* assemblage (3.7 ppm from 27 analyses), which is in fact the dominating Au carrier in the deposit.

The systematic study of Kehayov and Bogdanov (2005) established evolution in the chemical composition of gold from the different assemblages, as shown on Fig. 2. The early gold from the *mt-bn-cp* assemblage occurs as isometric drop-like or irregular microscopic (5-40 µm) grains of high fineness (994-903) included in bornite and in smaller quantity in chalcopyrite. A second type of gold related to this assemblage includes larger in size macroscopic visible particles (from 100-300 µm to several mm) found in the upper horizons often as thin cracks in bornite and on its boundaries with K feldspar. This gold shows some lower fineness in the range 880-763 and some higher Cu content (up to 4-4.5 wt.% and even to 6.88 wt.%). On Fig. 2 it is marked as *mt-bn-cp-II.*

Gold in the *q-cp-py* and *q-py* assemblages is in rounded microscopic grains (5-40 µm and 5-30 µm, respectively) of lower fineness (853-753), and this in the late *q-ga-sph* assemblage reaches lowest fineness (682-594) and smallest size (5-15 µm). Similar data for gold of the first two assemblages gave also Strashimirov and Kovachev (1994), Tarkian et al. (2003) and Georgiev (2005).

As seen on Fig. 2, based on all these data, there is a clear trend of successive increase of Ag content in gold with decrease of *T*. Markedly is the *mt-bn-cp-II* field, which connects the fields of the first two assemblages, mostly overlaying the second one (*q-cp-py*). This is an indication for some later deposition of that gold, which corresponds and with its textural relations to bornite

Samples and methods

The filamentary native gold is presented as small bush-like bundles disposed within small open cavities in the massive sulphide ore of the early *mg-bn-cp* mineral assemblage (Fig. 1). The cavities are irregular in shape, up to several mm wide. The gold bundles, several in one cavity, are disposed on the uneven bornite surface. The size of the bundles is not larger than 1-2 mm. Included entirely in such ore cavities the very delicate golden aggregates are preserved intact in their original position and state.

The golden formations were studied by scanning electron microscopy with devices SEM Philips-515 and JEOL Superprobe-733 equipped with ORTEC-5000 EDS system. Because of the very small size and ductility of the golden fibers this was the only possibility for their observation. The samples were examined in their natural form, without coating. For crystallographic analysis of the SEM microphotographs, the SHAPE program of Dowty (1997) was used, with the additions of Kerestedjian and Atanassova (2000), very useful when applied to crystal views in accidental and changeable orientation.

Crystal morphology

The aggregates of native gold consist of numerous fine crystal gold filaments, emerging from separate golden grains on the bornite surface and developing roughly divergent in the free space of the enclosing small cavity. The single filaments are tortuous, changing gradually or more steeply and irregularly their direction (Figs. 1, 3, 4). Straight-linear segments of the filaments are rare.

The size of the single filaments in one and the same group is variable. Their length is usually below 1 mm. The thickness of fibres is more or less uniform, on places changeable. It varies between 2-3 µm and 0.3-0.5 mm, with prevailing values 10-20 µm (Figs. 1 and 3). Fibres of different size often coexist.

All filaments represent peculiar *euhedral* but highly anisotropic gold crystals (Figs. 3-5). Distinguishing feature is that they are bounded by alternating small flat faces of different crystal forms producing stepped and striated surfaces.

Prevailing are the faces of the main crystallographic forms of gold: octahedron *o*{111}, rhombic dodecahedron *d*{110}, and cube *a*{100}, with minor and locally occurring *m*{311}, *f*{310}, and some rarer forms. Elongation of filaments generally follows the <110> crystal directions, often developed in different planes. [110] is the only one PBC direction of strong bonds in gold (e.g., Fig. 3b, c).

The tips of filaments are commonly well seen, displaying clear crystal bounding. More regular or slightly distorted cubo-octahedral crystal shapes prevail (Figs. 3 and 4). Some tips are of larger thickness, presenting more separated distorted crystals (Fig. 4a). In other cases the terminals of filaments are divided into two, or three parallel branches (Fig. 4b, c, d). Transversal and inclined branches of different length also appear (Fig. 3a, b). The lateral faces of the filaments are formed by some of the main *d*, *o*, and *a* faces, forming complex striated structures of fine cross or oblique steps of these and some additional forms.

In some areas the cubic *a* faces are dominating, forming peculiar uniform pyramidal relief (Fig. 5a, b), in other places octahedral layered faces occur, sometimes with trigonal or strange ditrigonal vicinals (Fig. 5c). Common are the stepped *d:o, d:a*, or *o:a* surfaces (Figs. 3a, c, 4a-d). The parallelism of the stepped faces on great distances proves the single crystal nature of the filaments. Incomplete pyramidal forms are locally displayed (e.g., *f*{310} on Fig. 4a).

Rarely, some thin filaments are mechanically curved as seen from the change of the orientation of the steps. Twins along the typical for gold {111} twin planes are often present. They are displayed as longitudinal sutures between domains with striation at different angles (Fig. 5a).

It can be mentioned, that in the neighbour Chelopech high-sulphidation epithermal Au-Cu deposit, which is considered by Popov et al. (2000) as other part of the joint Elatsite-Chelopech ore field, the morphology of the native gold grains is different enough (Bonev et al., 2002). Only very rarely in Chelopech fine-fibrous gold can be observed (Fig. 3E in Bonev et al., 2002).

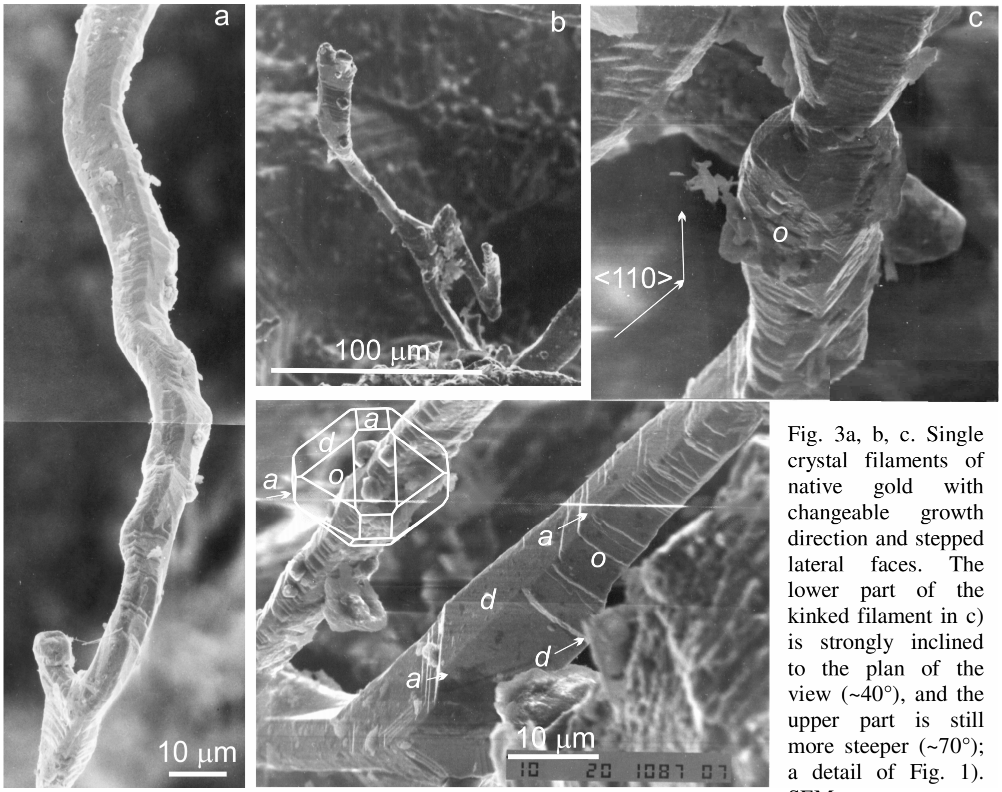

Fig. 3a, b, c. Single crystal filaments of native gold with changeable growth direction and stepped lateral faces. The lower part of the kinked filament in c) is strongly inclined to the plan of the view (~40°), and the upper part is still more steeper (~70°); a detail of Fig. 1). SEM

Chemistry and genesis of filamentary gold

The filamentary native gold discussed here was found only within massive *mt-bn-cp* ore aggregates, which is strong evidence for their genetic link. However, the microprobe analyses of filamentary gold established Ag content in the range 12.85-20.69 wt.% (18.71 wt.% average from 14 analyses, i.e. 29.23 at.%) and Cu in the range 0.15-2.84 wt.% (average 1.11 wt.%, 2.95 at.%). With such fineness slightly lower than that of the early gold, on the diagram of Fig. 2, all these analyses fall in the field of *q-cp-py* assemblage (overlapped with *mg-bn-cp-II* field).

Besides, this specific gold commonly associates with later sericitic products of K feldspar alteration. The central parts of some small cavities, which include gold filaments, are filled by fine flakes of illite together with tiny prismatic (0.1 mm long) crystals of quartz. So, all these compositional, textural and paragenetical characteristics of the filamentary gold prove its later deposition during the main *q-cp-py* stage.

Important for understanding the gold behaviour in high-temperature porphyry Cu-Au deposits are the recent experimental data (Simon et al., 2000; Kesler et al., 2002) for the system Au-Cu-Fe-S. In these experiments, at 600°C bornite is the dominant collector for Au, with possibility to include 1,280 to 8,212 ppm (mean 4,875 ppm), against 104-125 ppm in chalcopyrite iss. At 400°C the Au content decreases to 13-80 ppm and 2-4 ppm, respectively, when the richest in Au natural bornite from porphyry deposits contains up to 1 ppm

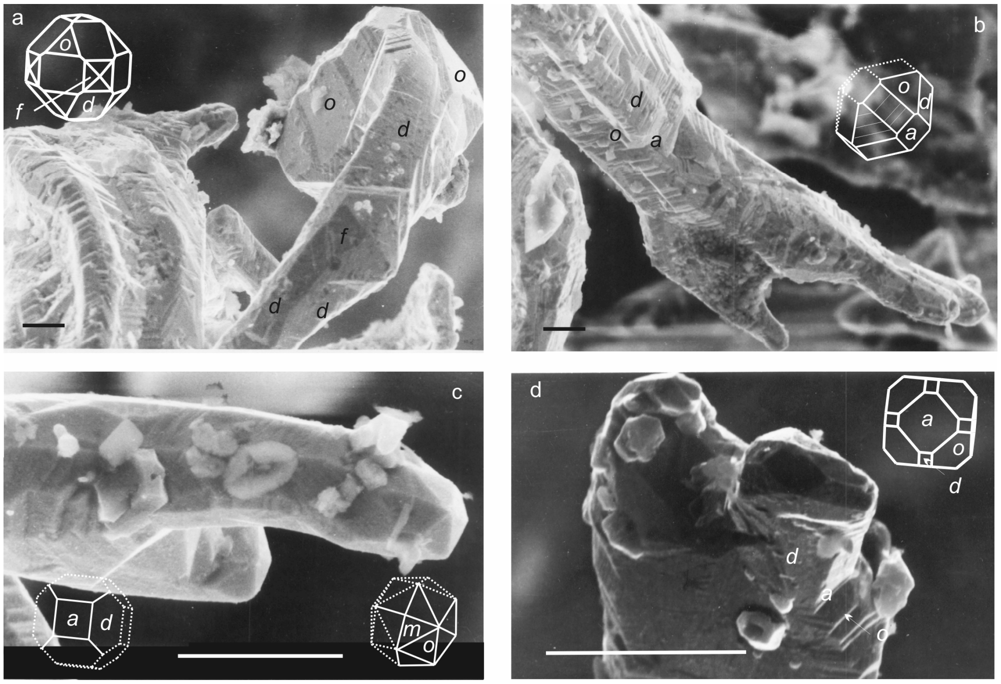

Fig. 4. SEM microphotographs of different tips of gold filaments: a) more individualised distorted crystal termination; b and c) splitted terminations with complex shapes (triple in b, and double in c); d) twin-peaked termination of cubo-octahedral habit; enlarged detail of the top of Fig. 3. Crystal forms: *d*{110}, *o*{111}, *a*{100}, *m*{311}, *f*{310}. Scale bar for all pictures are 10 µm

and chalcopyrite about an order of magnitude less. Applied to Elatsite, these results well explain the early introducing (at 600°C) of Au in bornite solid solution (probably not in fully saturated state) with its removing and re-deposition as native gold during the later quartz-chalcopyrite-pyrite stage from sulphide-rich solutions, when also partial converting of bornite and magnetite performed.

The chemical mobility of gold in similar environment was extensively studied by many authors, including Gammons and Williams-Jones (1997), Stefánsson and Seward (2003, 2004) and others, which show that the aqueous speciation of gold is very sensitive to fluid composition and temperature. The dominant transporting complex for Au at high-*T* (400-600°C), in oxidized, acidic, highly saline, and potassium-rich fluids of low H_2S activity (magnetite stable), is $AuCl^{2-}$ and metal may precipitate as Au-rich copper sulphides (bornite, iss). With decreasing *T* and increase of H_2S activity (pyrite stable) the gold sulphide complexes, became predominating for concentrated acidic chloride solutions and then remobilization of Au can be realised. In such condition boiling is important factor for gold precipitation. The thermodynamical analysis of Heinrich (2005) also substantiate the differences between these two successive fluid regimes, the second of which at excess of sulphur species over chalcophile metals (Cu and Fe) ensures efficient Au transport.

The extreme symmetrical distortion of the

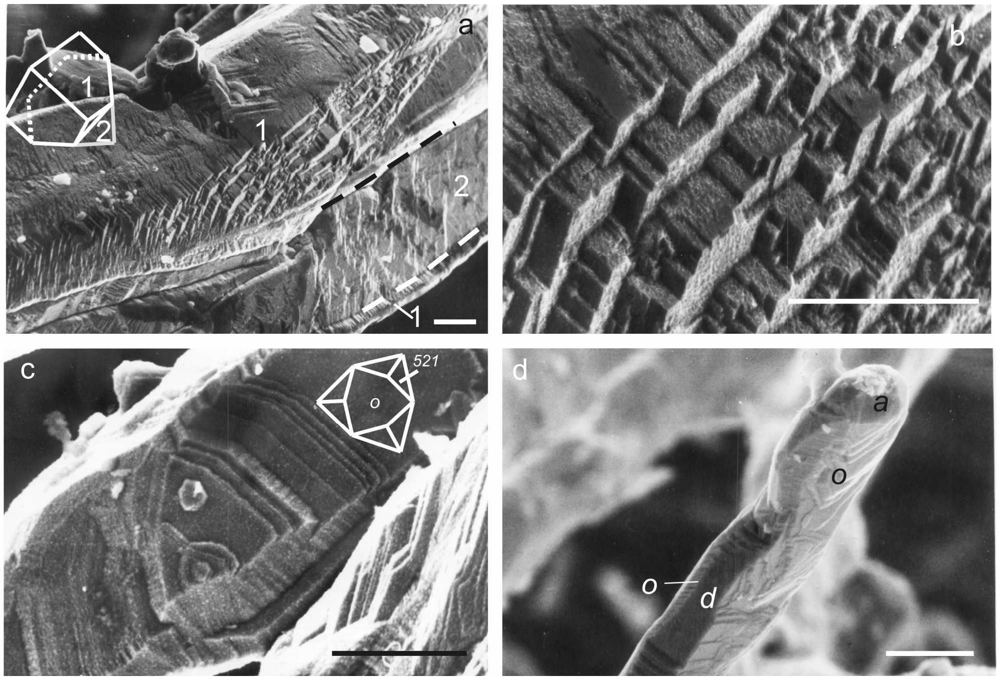

Fig. 5. SEM microphotographs showing the surface morphology of some filaments. a) a bundle of fibers in twinned positions, bounded by cubic faces; 1 and 2: twinned along (111) crystals in two orientations, the twin boundaries between them being marked by dotted lines; b) pyramidal surface, a detail of a); c) octahedral *o* (111) surface with layered structure, the layers being outlined by the {521} vicinal form; d) nearly straight [110] filament with stepped faces. Scale bars for all pictures are 10 μm

filamentary gold crystals with their cubic structural symmetry (*Fm3m*) is evidence for specific growth conditions far from the equilibrium.

The changeable growth directions and stepped and striated surfaces of filaments are not consistent with the dislocation growth model, often supposed for straight whiskers, whereas the high melting point of the Ag-bearing gold (~1040°C) exclude the possibility for a VLS (vapour-liquid-solid) growth mechanism (Maleev, 1971; Givargizov, 1986; Sunagawa, 1997) under the real geological conditions discussed.

The specific morphology of gold crystals can be explained by growth from *highly heterogeneous supersaturated fluids*, such as the boiling fluids. Evidence for such fluids is given by Kehayov et al. (2003). The beginning of filamentary growth was related to moment of arising of morphological instability on the surface of a fine-grained gold aggregate-seed (e.g., as this shown on Fig. 1 of Kehayov and Bogdanov, 2005), followed by fast growth of its most protruding parts at diffusional regime. Such mechanism, not controlled by axial dislocations, can be prevailing for growth of straight whiskers in solutions (Bonev, 1990). In the two-phase liquid-vapour fluids, the local heterogeneity of environment strongly influences the changeable fibre growth direction oriented towards the highest super-saturated areas of the high salinity liquid. The presence of vapour bubbles of low-density in the crystallisation space is also a strongly disturbing factor, influencing the direction of crystal growth. The gravitation and convective flows are other disturbing factors influencing the movement and distribution of bubbles.

It is important to mention that similar highly anisometric filamentary crystals are known and for other cubic minerals, even if the morphological information for them is quite scarce. Remarkable is the case with *galena*, also of *Fm3m* symmetry. Many peculiar galena crystals associated with whiskers, as described by Bonev (1993, see Figs. 1a, 3g, h, 5, 6a, c) have morphology similar to this of the filamentary gold, with changing growth directions and stepped faces. Similar are and the conditions of growing of these galena crystals, formed from highly supersaturated hydrothermal fluids in small cavities of polymetallic ore.

It can be expected that such specific highly anisometric crystal growth is widerspread in nature, even though and still not well evaluated.

Conclusions

- The rare in nature, filamentary native gold occurs in the Elatsite porphyry copper-gold deposit as small bush-like groups within small cavities of the early magnetite-bornite-chalcopyrite ore.
- The euhedral, not straight-linear and tortuous filaments of gold, 2 to 10 μm wide, are bounded by small alternating faces of the main crystal forms of gold: *o*{111}, *d*{110}, and *a*{100}, minor *m*{311}, *f*{310}, etc. Elongation generally follows the <110> directions and their sides have stepped surfaces with cross or oblique oscillation striation.
- The chemical composition of filamentary gold with 18.7 wt.% Ag and 1.1 wt.% Cu is comparable with this of gold from the main quartz-chalcopyrite-pyrite assemblage.
- Most probably, the high-temperature (600-530°C) Au-rich bornite solid solution is early collector for gold, by cooling of which (down to 400°C) gold is separated and re-deposited in the immediate neighbourhood. This explains the spatial and genetic link of gold with bornite, but also and with later illite-quartz alteration products.
- It is supposed that the extremely non-equilibrium filamentary crystals of gold formed in highly supersaturated heterogeneous boiling two-phase fluids of high-salinity liquid and low-density vapour phase. The gas bubbles influenced and disturbed the directed crystal growth.

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