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LATTICE FITNESS OF NEIGHBOURING PYRITE GRAINS IN PYRITE PORPHYROBLASTIC DOMAINS IN CERTAIN ORES

by

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(with 2 tables and 7 text-figures)

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Abstract

Considerable attention has been given to pyrite porphyroblastic domains formed in certain ores. These domains are never more than a few centimetres in diameter, and consist of several hundred small pyrite grains showing strongly developed preferred orientation. It is concluded that the coordinating relationships observed among the grains of one domain are produced as a result of recrystallization and replacement of ores under hydrothermal metasomatic conditions. The following three types of preferred orientation have been observed: (1) parallel orientation of cube faces of grains, (2) parallel orientation of pyritohedral faces of grains, and (3) parallel orientation of the cube face of one grain and the pyritohedral face of another. Approximately 90% of all examined pairs show one of these types of parallel orientation. This very marked orientation of pyrite grains in a domain can hardly be interpreted as the result of coalescence of numerous grains which were crystallizing under conditions of stress. It is suggested that "fitness of lattice structure", a control mechanism proposed by Hunahashi and Hoe (1973), might have been a major factor at the embryonic stage of grain-growth under static field conditions and operated irrespective of whether or not the nuclei were in direct contact.

Introduction

During an underground survey of the Hitachi mine, Japan, one of the authors (J.W) was strongly impressed by the brilliant reflection of the light of his cap-lamp from innumerable crystal surfaces. No mineralogical terminology exists to describe this phenomenon, which could be likened to wolves eyes in the dark. This effect was produced by numerous pyrite porphyroblasts in the compact pyrite ores. Although various occurrences of large pyrite single crystal have been reported, little is to be found in the literature about porphyroblasts

of the type found here, which are fine-grained and composed of a great number of crystals arranged in similar orientation. This type of porphyroblast is not, however, so very rare. We have found similar specimens: registered as no.131 from Suvravar mine, Norway, nos.709 and 720 from Håkansboda mine, Sweden, no.719 from Riddarhyttan mine, Sweden and no.764 from Freiberg mine, Germany, all found among the registered specimens of the Department of Mineralogy, Stockholms University.

In the same time, feldspar porphyroblast, developing in various kinds of metamorphic rocks, is also one of the most compelling problems of metamorphic petrology.

Pyrite porphyroblastic domains studied here show fairly similar stereographic plots of coordinating relationships among the constituent grains in a domain to those of metamorphic rock, in spite of materials being chemically and crystallographically quite different. For this reason, a detailed study of the mode of the oriented growth of constituent ore grains in porphyroblastic domains and some considerations concerning their genesis will be presented here.

Samples

The samples investigated here have been obtained from the Hitachi mine, E. Japan, which is one of the cupriferous iron sulphide ore deposits. The geology of the area and the alteration associated with the ore bodies has been described in another paper (Watanabe 1974). Large crystals of pyrite have been reported from this mine (Akaoka 1920). Most of them occur as single crystals in compact copper ores such as chalcopyrite-pyrrhotite and/or chalcopyrite-sphalerite ores. They are commonly idiomorphic crystals (cube, octahedron etc.) in a poikilitic texture. Pyrite of this type (type 1) is not included in the present study. The pyrite studied here (type 2) occurs in the pyrite ores as porphyroblastic domains composed of a large number of small pyrite crystals. They are not ubiquitous but occur only in the following localities.

The Fujimi ore deposits consist of a number of ore bodies of irregular form but all in a nearly vertical direction. The various bodies show some characteristic features which indicate that they formed at different stages of mineralization. At the same time, each body also shows variation in the sulphide mineral associations with depth. It is not possible, because of these complexities to establish the complete mineralization succession of the ores but with regard to pyrite, we can at least recognize three generations in the main phase of mineralization: (Phase I) the earliest pyrites formed in massive pyrite ores have a rather distinctive clastic texture in polished section and are

Table 1

5F	500 ML	No. 10
5F	550 ML	No. 22, stope and west ore body, foot-wall
5F	550 SL	No. 17, stope
5F	600 ML	West, central and east ore bodies
5F	600 SL	No. 25, west and east stopes
5F	650 ML	No. 25, east. East and west sides of the Main ore body (Honko)
5F	650 SL	
5F	700 ML	West and east stopes
5F	750 ML	No. 4, hanging-wall, and west ore body
5F	750 SL	No. 5, west, foot-wall. West and east
4F	700 ML	
2F	550 2SL	East, stope
2F	550 SL	Stope
2F	700 ML	
2F	750 ML	
2F	750 SL	
2F	850 ML	

F: Fujimi ore bodies, ML: metre-level, SL: sub-level

traversed by a highly irregular net-work of fractures. These clastic grains seem to be replaced along the fracture by chalcopyrite-sphalerite of the main phase. (Phase II) pyrites of the main phase, which are commonly formed as large crystals at the hanging-wall side of the chalcopyrite-sphalerite ore bodies, are apparently replacing the copper-zinc ores. These grains tend to develop into large crystals (analogous to those of type 1). Pyrites of the final phase (Phase III) are investigated in the present work, they occur in massive pyrite ores which seem to be slightly later than those of phase II.

A typical sample belonging to phase III of the main mineralization is depicted in *Fig. 1*. About 80% of this ore is pyrite. Quartz, biotite, sphalerite, chalcopyrite and pyrrhotite form the bulk of the remainder. The grain size of pyrite individuals ranges from 0.3 to 2 mm.

The porphyroblastic domains, each composed of several hundred grains of pyrite, are packed tightly together in these ores (*Fig. 2*). As shown in *Fig. 2*, domains in contact show sutured margins as a result of scrambling for the controlling sphere of each domain. Domains showing idiomorphic boundaries were seldom observed. In rare cases, however, another type of porphyroblast is observed (*Fig. 3*). This tends to develop in areas rich in copper-zinc minerals. In this type the aggregates of pyrite grains tend to develop somewhat distinctive outlines: the cluster in the upper half of the figure seems to show a

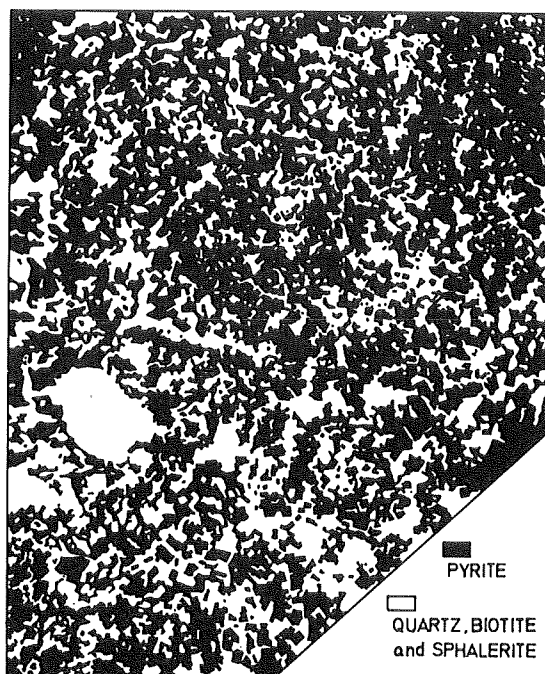


Fig. 1 A typical sample of pyrite ores forming porphyroblastic domains. Carefully oriented samples have been used to make the polished sections. (Loc. 2F 850ML)

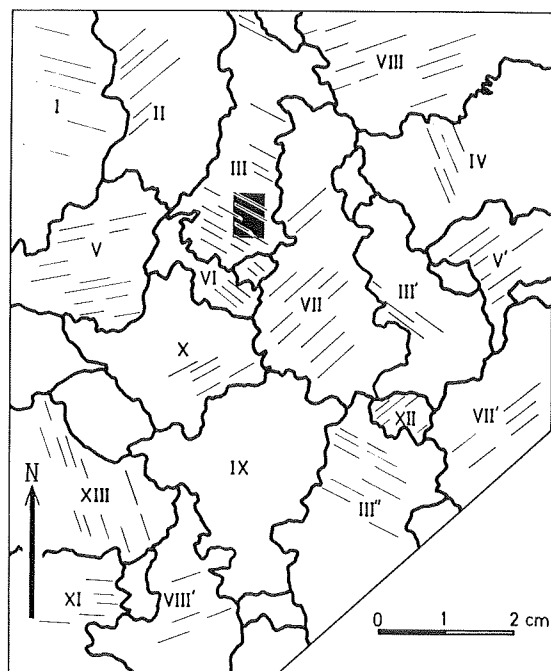


Fig. 2 Domain structure with closely packed porphyroblasts. (Analogues to Fig.1). Schematic shading within the domains indicates the direction of edges common to two intersecting surfaces; the polished surface of the material and the natural crystal surface bounding "pits" equivalent to an e-face. The rectangle in domain "III" is the area of the photomicrograph (Fig. 4a).

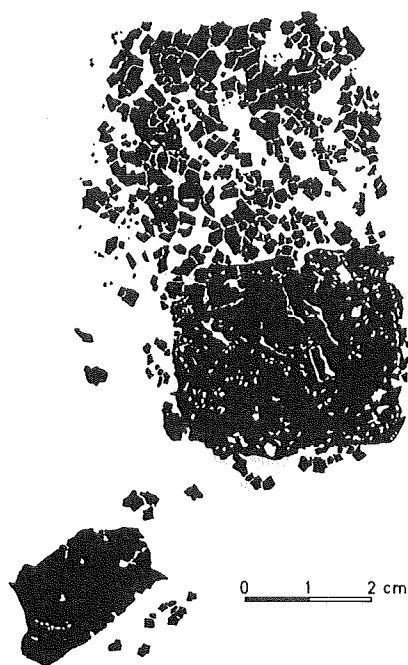


Fig. 3 Another type of pyrite porphyroblast in a matrix rich in chalcopyrite-sphalerite-pyrrhotite. (Loc. 5F 550ML).

fairly clear cubic outline. This is a glomero-porphyritic domain, in which most of the pyrite grains have same orientation: the lower half of the figure shows an even more distinct cube form which can be regarded as the same phenomenon in a more advanced state. Although predominantly pyrite, it contains lots of minute ramifying inclusions of copper-zinc minerals.

In almost all cases, pyrrhotite is associated with the pyrite porphyroblastic domains. This raises some interesting questions concerning the partition of sulphur and iron during porphyroblastesis.

Under the microscope, pyrite in the porphyroblastic domain (phase III) has no clastic texture and shows no evidence of replacement by sphalerite, chalcopyrite or pyrrhotite. Most of the individual pyrite grains are idiomorphic, having cubic form, with embayments or "pits" (*Figs. 4a and 4b*) developed at the grain boundaries. When the pyrite shows very marked preferred orientation, the regular parallel arrangement of well-developed cubes sometimes produces a tessellated texture. This texture suggests growth in a static environment.

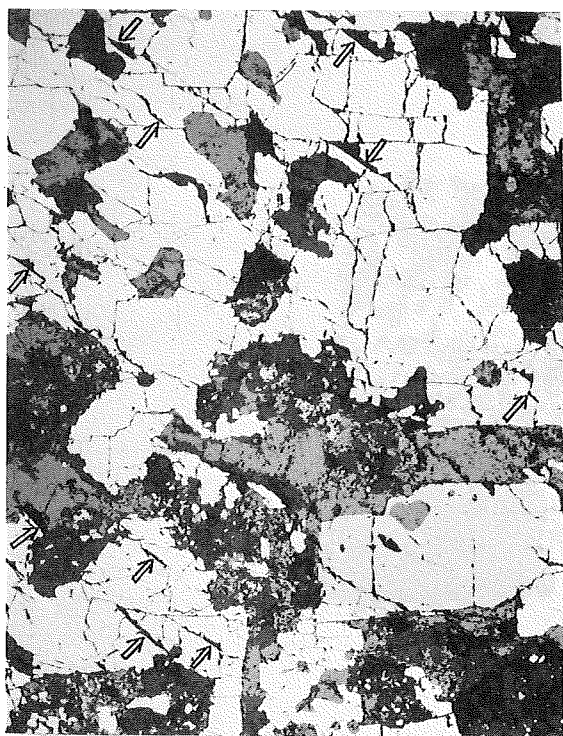


Fig. 4a A typical photomicrograph of pyrite showing ore texture and "pits" (arrow head) from domain III of Fig. 2. The pyrite shows very marked preferred orientation. The "pits" are aligned and the direction is indicated by the shading in domain III of Fig. 2. Pyrite of this sort is almost idiomorphic with major development of cube faces and minor e-faces. Sphalerite (grey) containing abundant chalcopyrite exsolution dots. Quartz (dark grey) and minor amount of pyrrhotite. X 15



Fig. 4b This is an enlargement of the upper right corner of Fig. 4a. The angle between e- and a-faces is approximately $28-30^\circ$ in almost all cases measured, while the theoretical angle is 26° . On the right of the picture there is a crack which shows slight offsetting at grain boundaries and which has not been filled by later mineral. This means a post-ore origin for this cracking. X 87

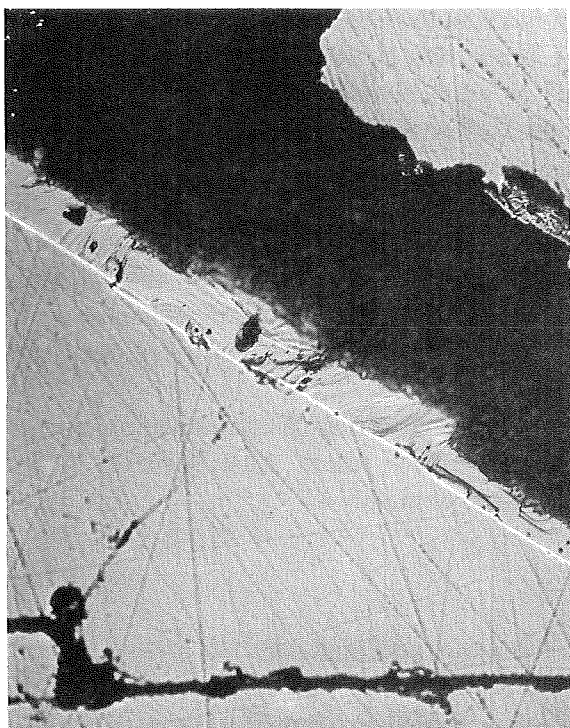


Fig. 4c This is an enlargement of a portion of Fig. 4b showing layering-growth on the e-face which forms the boundary surface of a "pit". The angle between a- and e-faces is 31° . (Interference contrast by Nomarski method). X 450

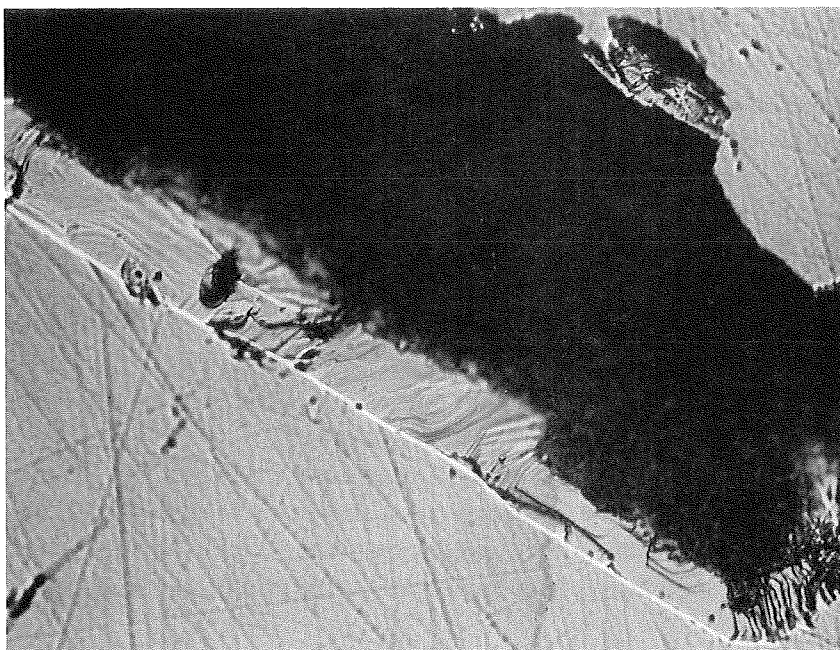


Fig. 4d Even greater enlargement of the same area of Fig. 4c, showing layering-growth hillocks (Interference contrast by Nomarski method). X 710

A diagnostic feature of a domain is a simultaneous glittering over the whole area of the domain when a light beam hits it at a certain angle due to reflections of light from the "pits" at grain boundaries. Different domains, generally, produce this reflection at different angles. The "pits" are not analogous to triangular cleavage pits sometimes found in galena, where plucking along the cleavage during grinding of the sample, produces cleavage "holes". These hollows are, on the contrary, bounded by natural faces (*Figs. 4a-d*). Detailed observation shows a micro-texture of layered growth on the sides of the "pits" which is characteristic of the pyritohedral faces of pyrite and a sure indication that these "pits" are natural growth features mainly bounded by pyritohedral, e-faces (*Figs. 4a-d*).

It can be concluded that the simultaneous glittering of numerous grains in a domain is due to reflection from these e-faces which must have the same orientation in all the individual grains of the domain.

Techniques of measurement

Korn (1934) was the first to investigate systematically the possibility of determining the orientation of opaque minerals in polished sections on the basis of cleavage planes, twinning planes, exsolution lamellae, crystal boundaries, zones and so on. In the present case the orientation of pyrite grains in the oriented polished sections, which were obtained by serial sectioning from a large slab, was determined by optical measurement of a-faces on the U-stage of an ore microscope setting the instrument for reflectivity measurement (Leitz Mikroskop-Photometer, MPE).

In the case of pyrite showing cube form, the a-axis can be determined stereographically by measuring two a-faces intersecting at right angle each other. The direction of all three a-axes in a grain of pyrite can be determined in this way. It is then possible to deduce the position of e-faces even though one cannot actually observe them in a grain. A diagram to illustrate this is shown in *Fig. 5*. Comparison of the position of a- and e-faces in neighbour-grains enables one to ascertain how they are related crystallographically.

When a domain exhibits pitting of the type previously described, due to the development of small e-faces, the orientation of these can be determined by measuring the U-stage position which gives maximum reflectivity from the "pits". At that position an e-face is exactly normal to the microscope tube.

Results

The mode of crystal-combination, by which the individual grains in a

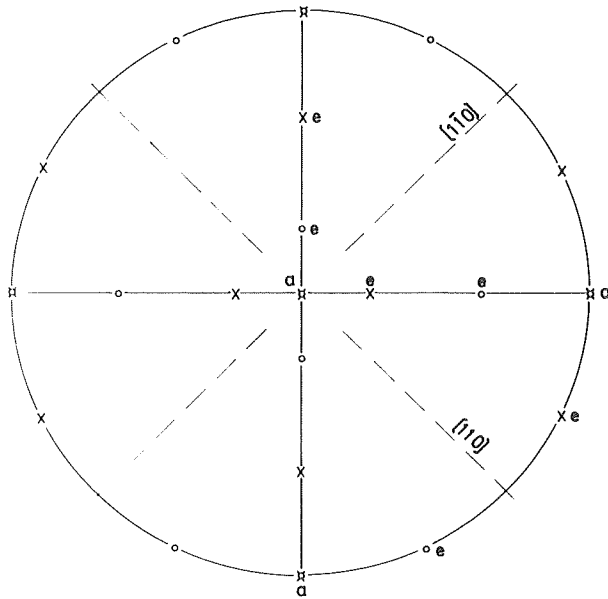


Fig. 5 Estimation of the symmetrical component of pyrite deduced from the actual measurement. e: pyritohedral face, a: cube face. $[110]$ and $[1\bar{1}0]$ are twin axes (broken lines) of so-called Iron Cross Twin of pyrite. This diagram includes two sets of component faces after twin disposition (open circle and cross). Solid line: a-face.

domain achieve a definite crystallographic relationship to neighbour-grains, was examined. The results obtained are summarized in *Table 2*. Approximately 90% of the examined pairs forming one domain are related in one of the following three ways; (1) $a\parallel a$, (2) $e\parallel e$, or (3) $a\parallel e$ (where a is a cube face, e is a pyritohedral face and the symbol ' \parallel ' denotes complete parallelism). There is thus no room for doubt concerning the strong degree of parallel orientation exhibited by such domains. Attention is also drawn to the fact that the pyrite grains do not need to be in contact in order to display this parallelism. It is also worthy of note that these relations seem to be established regardless of the nature of the interstitial minerals. The results are tabulated in detail in *Table 2*, and the conclusions can be summarized as follows: i) The data indicate that there is a strong degree of preferred orientation among the examined pyrite grains regardless of whether they are in direct or indirect contact with each other. ii) The coordination relationships of $e\parallel e$ and $a\parallel e$ are far more common than those of $a\parallel a$, both in direct and indirect contact pairs. iii) The percentage of coordination relationships in the direct contact pairs is slightly higher than that observed in the indirect contact pairs (95% in the former 90% in the latter). iv) In the indirect pairs, however, data are not enough to say exactly, it is noteworthy that 85-90% of the examined pairs participate in one of the three

types of coordination. The nature of the interstitial minerals (sulphides or silicates) does not appear to affect the degree of parallelism. v) A cluster of grains which are characterized by a high percentage of coordination constitute a domain structure.

Table 2

Neighbour-grain relationships of pyrite	Total number of pairs examined	Direct contact pairs	Indirect contact pairs				
			(1)	(2)	(3)	(4)	Total
a a	50 pairs 14.2%	16 pairs 14.0%	22 pairs 34.4%	8 pairs 23.5%	2 pairs 2.5%	2 pairs 3.4%	34 pairs 14.3%
e e	131 pairs 37.3%	47 pairs 41.2%	17 pairs 26.6%	12 pairs 35.3%	36 pairs 43.9%	19 pairs 32.8%	84 pairs 35.3%
a e	140 pairs 39.7%	46 pairs 40.4%	17 pairs 26.6%	9 pairs 26.5%	38 pairs 46.3%	30 pairs 51.7%	94 pairs 39.5%
Un-coordinated	31 pairs 8.8%	5 pairs 4.4%	8 pairs 12.4%	5 pairs 14.7%	6 pairs 7.3%	7 pairs 12.1%	26 pairs 10.9%
(Total)	352 pairs 100.0%	114 pairs 100.0%	64 pairs 100.0%	34 pairs 100.0%	82 pairs 100.0%	58 pairs 100.0%	238 pairs 100.0%

(1) Separated by 1 grain of pyrite

(2) Separated by 2 grains of pyrite

(3) Separated by 1 grain of quartz

(4) Separated by 2 grains of quartz

The orientation relationships between the various domains have also been studied. Most domains show strong reflectivity at a given angle which indicates that a large number of the pyrite grains within the domain have developed a face parallel to this common direction. The orientations of surfaces bounding "pits" in each domain were measured by MPE. All the plots for one domain fall in a cluster deviating from a point by only a few degrees. The statistically central values of cluster from 18 domains are presented in *Fig. 6*. The domains plotted here correspond to those of *Fig. 2*. In this figure, the measured $e(210)$ faces, the calculated a -faces and the measured a -faces do not project randomly but lie within a definite zone, which is within $30\text{--}40^\circ$ of the central pole (vertical direction). This direction corresponds to the plunge of the ore bodies. This suggests that, at some embryonic stage when the nuclei of the domains were forming there may have been some orientation of grains due to stress. Later development of domains during porphyroblastesis may well have taken place under static conditions. *Fig. 7* shows partial stereographic plots (see *insert Fig. 6*) of the crystallographic relations between pairs of domains. It is a noteworthy fact that these domains all have some crystallographical components in common regardless of whether they are in direct or indirect contact. On a rough estimate it would appear that the commonest types of domain relationships are those of a||a and a||e, whilst e||e is very rare. It should be

Fig. 6 A diagram showing orientation of boundary “pits” of pyrite in each domain. Roman numerals are comparable to those in Fig. 2. 1: measured $e(210)$ pole, 2: calculated a -axis, 3: measured a -axis.

The term porphyroblast has seldom been used in the literature of ore geology (Schouten 1934, Edwards 1947, Bastin 1960, Cameron 1961 and Oelsner 1966), probably because the term has genetical implications and the genesis of ore bodies is often difficult to establish. Three textbooks have dealt with this term (Ramdohr 1950, Freund 1954 and 1966). Quoting from them, Ramdohr (p.49-51) points out: Mit den Rekristallisationserscheinungen eng verknüpft ist die Bildung der Porphyroblasten, ---. Besonders neigen in rekristallisierten Erzmengen der Magnetit, Pyrit und Ilmenit zur Porphyroblastenbildung, ---. Am bekanntesten ist wohl der Fall der zugerundeten Pyrite in Kupferkieserz von Sulitelma, ---. --- diese Kristalle sind ein wandfreie Porphyroblasten und die Rundung eine Folge "gehemmten Kristallwackstums

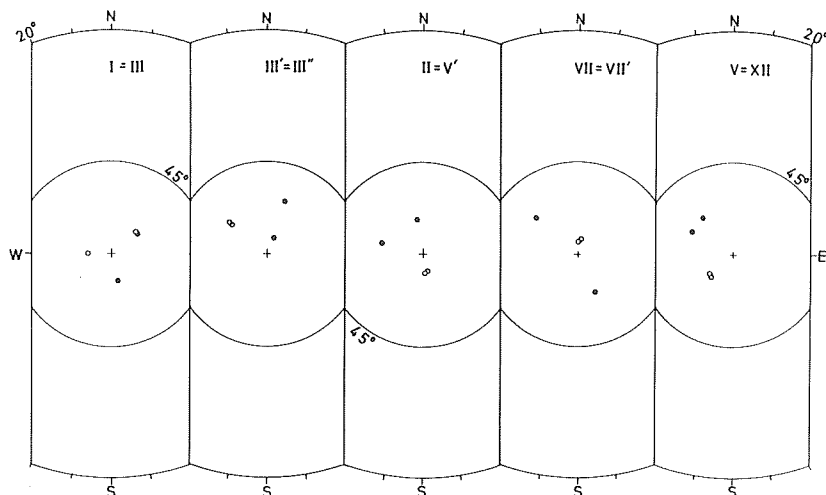


Fig. 7 Nearly parallel disposition of domains, represented by the orientation of measured e-face poles (solid circle) and calculated a-axes (open circle). Roman letters are the number of domain in Fig. 2.

in Sinn von O. Mügge" ---. And also Freund (p.280-281 in 1966) says: Some ore minerals such as pyrite, arsenopyrite, magnetite tend to occur among the recrystallized other products in the form of "porphyroblasts", commonly as euhedral "idioblasts" and more rarely as anhedral "zenoblasts". Porphyroblasts often contain from the times antedating deformation, ---.

In both statements, the authors are referring to a single crystal of pyrite porphyroblastesis, which has been produced as a result of recrystallization in the ores. However, the material investigated in the present study is not a single grain but a poly-grain domain showing strong preferred orientation, in our opinion however the term "porphyroblast" should be equally acceptable here. To the best of our knowledge intergranular coordination (Hunahashi 1973) of poly-grain ore masses of the type described here has not been reported previously.

A number of authors, from mineralogical point of view, have discussed epitaxis among sulphide minerals (Zimányi 1925, Royer 1928, Dana 1944 and Grigorév 1965). However, epitaxial intergrowth means that the incrustation of new mineral coincides with the termination of growth of the earlier mineral. They are not only found among substances of different chemical composition, but also in polymorphic varieties of the same compound. According to Grigorév, regular orientation of crystallites occurs not only during the free crystallization of minerals, but also during metasomatic replacement when homoaxial pseudomorphs are obtained, i.e., enstatite-antigorite, etc., during the

unmixing of solid solutions, e.g., perthites, bornite-chalcopyrite and in other intricate exsolution structures, during recrystallization and various other processes. According to Dana (1944), pyrite has been found in oriented growths with sphalerite, marcasite, arsenopyrite, galena, tetrahedrite and pyrrhotite.

pyrite	{001} [110] sphalerite	{001} [110]
pyrite	{001} [110] pyrrhotite	{10 $\bar{1}$ 0} [0001]
pyrite	{001} [001] galena	{111} [110]
pyrite	{001} [110] arsenopyrite	{001} [010]
pyrite	{111} [001] tetrahedrite	{111} [001]
pyrite	{001} [001] marcasite	{010} [101]
pyrite	{001} [110] marcasite	{010} [100]

"Syntaxis" is occasionally used as a synonym for epitaxis, but Grigorév (1965) uses this term in its broader sense, as defined by G. Donnay and J.D.H. Donnay (1953). Grigorév said (p.47): Syntaxis designates an oriented intergrowth between two substances during crystal growth and is based on the crystallochemical similarity of both substances. The resulting intergrowths are termed polycrystals. Syntactic intergrowths between one mineral and another may occur on separate areas. However the zoned growths of two different minerals were described, and their syntactic nature assumed (King 1957).

It might be possible to explain the formation of the porphyroblastic domains described here in terms of epitaxis or syntaxis in the light of parallel growth (Buckley 1951, esp. p.405-415), although this involves invoking mechanisms, such as simultaneous nucleation, growth and regular arrangement of pyrite grains in a poly-grain domain of the same compound. Penetrate twin of pyrite is rather common on the Iron Cross Law with twin axis [110], combining two pyritohedron crystals, but contact-twinning of pyrite is rare. The three types of the coordination observed in the present porphyroblasts, i.e., a||a, e||e and a||e, are not in the relations of penetrate-twins but in those of parallel growth.

Marked orientation of the pyrite described here for pyrite has been identified previously in a number of silicate minerals (in particular, quartz and plagioclase). And there has been much discussion concerning the factors controlling such alignments (Koark 1956, Trommsdorff and Wenk 1963, Wenk 1965, Sander 1970, Watanabe 1971, Ohta 1972, Hunahashi 1973, Hunahashi and Hoe 1973, Watanabe 1974). The coordination of coexisting grains has mainly been interpreted as representing a stage in the development of "super-individuals (überindividuer)" by aggregation, or as the result of twinning

of relations among neighbouring grains. Some domain theories, in which the nuclei in the embryonic stage develop afterward into porphyroblasts in a metamorphic or hydrothermal environment, were presented in the latter five papers. The authors have suggested that there are certain preferred crystallographic relationships which are often achieved in a domain, or among domains, e.g., very common twin relations among plagioclase, and Japanese and Estérel twin relations among quartz grains.

Disposition of porphyroblastic domains in certain crystallographic affinity is common in some silicate minerals, such as quartz and plagioclase, and pyrite porphyroblasts might be expected to behave in the same way, in spite of quite different composition and crystal structure. Some definite crystallographically affined relationships among porphyroblastic domains of pyrite were found: the most common types are $a||a$, $a||e$, whilst $e||e$ is very rare.

Variations in crystal habit of pyrite have been studied by Mügge 1925 and Sunagawa 1957. Mügge has described a famous rounded-form of porphyroblast consisting of a large single grain of pyrite from Sulitelma, Norway, and explained it as a result of "gehemmtes Kristallwackstum". Most pyrite porphyroblastic domains of the present study show very irregular outline (=type 2). Mügges interpretation may be applied to the type 1 (idiomorphic large crystal in a poikilitic texture), but not to the type 2 domains.

One of the present authors noted in another paper (Watanabe 1974) that the pyrite grains in the altered rocks surrounding the ore bodies occasionally have a definite crystallographic relationship to adjacent grains of quartz. As far as we know, this phenomenon has not been reported in previous literature. As a result of close examinations and on the basis of theoretical considerations, Watanabe reached the conclusion that common angles between c-axes of quartz and the poles of pyritohedral- or cube-faces of pyrite are 38° , 26° , 12° and 0° . Unfortunately, the present authors could not establish the existence of such a relationship between quartz and pyrite grains in the present study, although it might be expected to exist, since the quartz occurs interstitially and in very minor amounts.

The genetic interpretation of the observed coordination will have to await further studies, however, it is only possible at present to summarize as follows.

- (1) Small grains of pyrite constituting a porphyroblastic domain exhibit coordinated relationships formed through recrystallization and replacement of ores under hydrothermal metasomatic condition.
- (2) Parallel growth existing among the pyrite grains in one domain and between the porphyroblastic domains may be attributed to a control mechanism termed the "fitness of lattice structure" by Hunahashi and Hoe (1973).

(3) In all cases, small amounts of pyrrhotite tend to crystallize around or in the porphyroblastic domains. Thus, some chemical adjustments were involved during porphyroblastesis.

(4) The fact that domains are oriented with respect to each other (Figs. 6 and 7) suggests that nuclei of the embryonic stage of recrystallization have been affected by stress. However, observations under the microscope indicate that the subsequent growth of pyrite took place in non shear stress field.

Some essential problems still remain unsolved.

(A) What is the "fitness of lattice" which is established among crystals irrespective of similar or dissimilar crystal-chemistry (pyrite-pyrite and pyrite quartz)? Why and how was the "fitness of lattice" control established during the recrystallization of ore minerals and not at other stages?

(B) What factors produced such regularity of orientation among pyrite grains, in the indirect contact pairs of pyrite and between porphyroblastic domains where the control mechanism of "lattice fitness" is less likely to have operated?

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