# Pyrite and Marcasite Intergrowths

## FROM NORTHERN ILLINOIS



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Excellent microcrystals of pyrite and marcasite are found in vugs in Ordovician limestone at several quarries in northern Illinois. The crystals include pyrite "bars" and marcasite blades which are epitactic on early-formed marcasite whisker crystals.

#### INTRODUCTION

Three quarries in northern Illinois (Fig. 1) have produced very similar microcrystals of marcasite and pyrite from rocks of the middle Ordovician Galena Group. The Irene quarry of the Rockford Sand and Gravel Company has provided the majority of the specimens described in this paper; it is located 2 miles north of Irene, Boone County, Illinois in the NW <sup>1</sup>/<sub>4</sub> of Section 9, T43N R4E (Cherry Valley 7.5' Quadrangle Map). The Mulford quarry of the Rockford Sand and Gravel Company is the source of many other specimens; it is located southeast of Rockford in Winnebago County (E <sup>1</sup>/<sub>2</sub>, SE <sup>1</sup>/<sub>4</sub>, Section 33, T44N, R34E, Rockford South 7.5' Quadrangle). The Mount Carroll quarry, owned by Wendling Quarries, DeWitt, Iowa, is located 3 miles southwest of Mount Carroll, Carroll County (SW <sup>1</sup>/<sub>4</sub>, Section 10, T24N R4E, Wacker 7.5' Quadrangle). Although it produces many collectible mineral specimens, the Mount Carroll quarry has produced only a

single specimen relevant to the subject of this paper. This specimen consists of one vug with numerous chains of pyrite cubes. Access to all of these quarries is restricted, and collecting visits must be arranged in advance with the quarry management; access is prohibited during times when the quarries are not in operation.

Though these quarries are not noteworthy for large crystals or rare species, a number of common carbonates and sulfides occur as small crystals generally not exceeding several millimeters. The pyrite/marcasite associations which occur here are interesting examples of pyrite/marcasite epitaxy. These associations form the focus of this paper.

#### **GEOLOGY**

The three quarries exploit the same stratigraphic units. The follow-

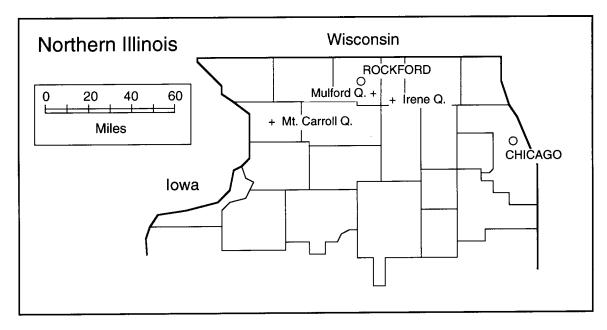


Figure 1. Map of northern Illinois showing the location of the three quarries.

ing description of the Irene quarry is applicable in general terms to all three. The strata are nearly flat-lying: Dips in the Rockford area generally are to the southeast and average 4 meters per kilometer (20 feet per mile).

The Irene quarry exposes the lowest 3.5 meters (12 feet) of the Dubuque Formation and about 17 meters (57 feet) of the underlying Wise Lake Formation, in which the minerals described in this paper are found. Both formations belong to the Galena Group of middle Ordovician age, which is approximately 450 million years old. The Galena Group is locally about 70 to 80 meters thick and consists primarily of dolostone (a rock composed mostly or entirely of the mineral dolomite) with occasional regions of limestone in the lower third, and with several thin, extensive beds of bentonite (altered volcanic ash) in the lower two thirds. In extreme northwest Illinois and southwest Wisconsin, the Galena Group and the underlying Platteville Group are host to the galena and sphalerite deposits of the Upper Mississippi Valley mining district, a leading lead and zinc-producing region for much of the 19th and early 20th centuries.

The Wise Lake Formation consists of about 22 meters of highpurity dolostone, formed by replacement of calcite by dolomite. The voids which usually host the minerals described below are of uncertain origin; some may result from the dissolution of massive fossils such as corals and stromatolites. The medium-grained pale gray to pale brown stone is very tough, and makes the collection and preparation of undamaged microminerals quite difficult.

Further information on the geology of this area can be found in Cote et al. (1969), Goodwin (1983), and Willman and Kolata (1978).

#### **MINERALOGY**

The mineral species attributed to the Wise Lake Formation in these quarries are described below, and are limited to simple carbonates and sulfides, and their alteration products. A somewhat larger species list characterizes the Dubuque Formation at the Mt. Carroll quarry, but is beyond the scope of this paper.

#### Calcite CaCO<sub>1</sub>

Calcite occurs occasionally as small, waxy lustered, colorless rosettes of intergrown rhombohedral crystals to 1.5 mm on edge, deposited upon drusy dolomite in vugs. The intergrown nature of these crystals prevents goniometric study. However, the crystals appear to be composed of the common rhombohedron {1011}, as judged by their shape and the orientation of the cleavage.

#### Chalcopyrite Cu,FeS<sub>2</sub>

Chalcopyrite occurs sparingly as sharp, striated, lustrous to lightly tarnished individual disphenoidal crystals to 2 mm, implanted on the drusy dolomite vug linings, usually without other sulfide minerals present in the same vug. In at least one vug from the Irene quarry, however, chalcopyrite occurs together with pyrite and marcasite. The disphenoid is {112}, which is the usually dominant form on chalcopyrite crystals (Palache *et al.*, 1944).

#### **Dolomite** $Ca,Mg(CO_3)_2$

Dolomite constitutes most of the rock in the quarries, and also occurs abundantly as tiny, sparkling rhombohedra of uniform size (0.4 mm on edge) densely lining irregularly shaped but well-defined vugs from 1 to 5 cm across. The dolomite is translucent, and sometimes has a colorless outer layer within which the translucent core is seen as a white phantom. Crystal faces are very flat and lustrous.

#### Goethite FeO(OH)

Goethite (or "limonite") replacements of chalcopyrite, marcasite and pyrite have been observed in vugs in locally weathered zones of yellowish dolostone.

#### Marcasite FeS<sub>2</sub>

Marcasite occurs in crystals of many different habits. Intermediate forms blur the distinctions between many of these habits, however. Marcasite crystals in these rocks tend to be flattened perpendicular to the b axis. Crystal habits range from diamond-shaped or rhombshaped plates through broad or narrow blades (Fig. 2) to elongated and commonly irregular-sided whisker-like crystals, which may occur as parallel growths. The edges of the diamond-shaped crystals, and the points of the blades, are composed of the prism {101} or sometimes a bipyramid such as {111}, based on measurements of angles formed by the edges, made using a petrographic microscope and in a few cases using scanning electron microscope images. These edge faces are small and extremely narrow, and are usually too narrow to be seen clearly under a stereoscopic microscope. Whisker-like crystals may be elongated parallel to either the a or the c axis, but elongation parallel to a is more common, based on the configuration of pyrite crystals oriented on them, as discussed below. Most of these crystals are too small and narrow to permit measurements of their terminations, thus goniometric study is impossible in most cases. Crystals are commonly transversely striated, so that the {010} "face" is actually composed of

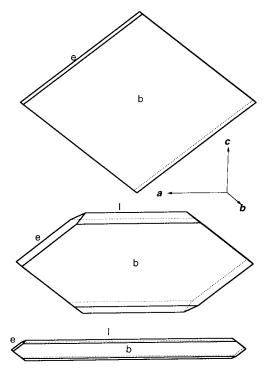


Figure 2. General habits of marcasite. Forms are  $b\{010\}$ , e(101), and  $l\{011\}$ .



Figure 5. Marcasite crystal composed of rhombic elements overgrown with irregular dendritic extensions, attached to a dolomite crystal and with an attached pyrite crystal. Compare with Figure 14. The field of view is about 0.3 mm wide. Irene quarry; R. Peter Richards specimen and SEM image.

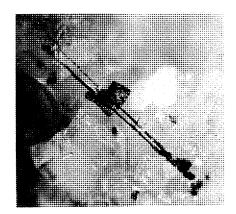


Figure 3. Hair-like acicular marcasite, a composite crystal composed of two nearly parallel branches which merge at one end. The crystal is 1.3 mm long. Irene quarry; Edwin L. Clopton specimen, Dan Behnke photograph.

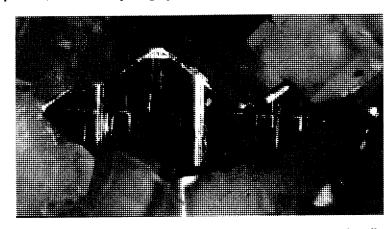


Figure 4. Rhombic marcasite crystal, heavily striated and with a horizontal "rib" which appears to represent an earlier, more acicular crystal, now partly overgrown. The crystal is 2 mm long. Irene quarry; R. Peter Richards specimen and photo.

Figure 7. Pyrite chains to 8.8 mm long. Irene quarry; Edwin L. Clopton specimen, Dan Behnke photograph.

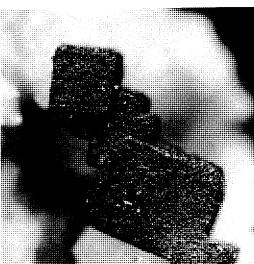


Figure 6. A chain of pyrite crystals, overgrowing an acicular marcasite crystal. The top pyrite crystal is 0.8 mm tall. Irene quarry; Dan Behnke specimen and photo.



alternating, inclined faces which only approximate {010} in aggregate.

Two habits are of particular interest and are the main topic of this paper. Marcasite sometimes occurs as hair-like acicular crystals (Fig. 3), to several millimeters long, but less than 0.1 mm in cross-sectional dimension. These whisker-like crystals appear at first to be rare, but are often overlooked because of their small size. Also, they are the first generation of marcasite to form, and they are usually hidden by subsequent overgrowths of marcasite and/or pyrite.

Marcasite also occurs as thin diamond-shaped or rhomb-shaped plates attached to dolomite crystals. Some of these have a "rib" running down the middle (Fig. 4) between opposite corners of the plate, and with striations running perpendicular to it. This rib represents an earlier-generation, nearly overgrown crystal elongated parallel to the *a* axis.

Other diamond-shaped plates are of brilliant golden color and irregular outline, and many have one pyrite crystal (or rarely two) located in an interior position (Fig. 5), and usually projecting from both faces of the marcasite plate, as if penetrating the marcasite plate. Crystals with this morphology have been found only in one small boulder in the Irene quarry, and occur with lustrous chalcopyrite. The aggregates of platy marcasite and pyrite have a complex growth history, including epitactic growth of pyrite on marcasite, and several generations of both marcasite and pyrite growth. These interesting intergrowths will be discussed in more detail in the section below on "leaf epitaxy."

#### Pyrite FeS<sub>2</sub>

Pyrite occurs in microcrystals usually 2 mm or smaller, and showing a range of habits which combine the cube, pyritohedron and octahedron. The cube is nearly always present and is usually the dominant form. Pyritohedra are dominant on some crystals; these lack the octahedron entirely. Pyrite is found attached to dolomite crystals in the pockets, and commonly attached to marcasite crystals, in random or systematic orientations. Octahedral crystals are more abundant at Mt. Carroll than at the other two quarries.

In some of the most spectacular material from these quarries, pyrite occurs as very interesting, elongated bar-shaped aggregates of crystals of two types. In one type, referred to hereafter as "chains," cubic crystals are joined into chains up to 9 mm long (Figs. 6 and 7). The crystals are typically strongly striated and somewhat distorted, and pyritohedral faces often are present along the edges of the cubes. The cubes are joined along mutual edges, and appear to overlap somewhat, due to continued growth after they came in contact with each other. Striations on successive cubes usually run in opposite directions. In the other type ("bars"), crystals are joined together nearly parallel to faces of the rhombic dodecahedron {110} into bars up to 5 mm long. The crystals show combinations of cube, octahedron and pyritohedron, but are distinctly flattened, and elongated parallel to the length of the bar. This distortion leads to the total disappearance of some faces, systematic differences in the size of remaining faces of the same form, and a final shape which appears monoclinic rather than cubic (Fig. 8). In most cases, striations running in different directions along the bar and slightly non-parallel crystal edges demonstrate that the bar is actually composed of a number of crystals (Fig. 9). Some bars are decorated with a second generation of sharp, cubic crystals, apparently in random orientation. Some specimens (Fig. 10) show mirror-faced overgrowths of a second generation of pyrite along the crystal edges of a pyrite bar.

## Sphalerite ZnS

Sphalerite is occasionally encountered as crude black aggregates of crystals to 1 cm. These aggregates occur in fossil cavities and other dolostone openings which are not lined with drusy dolomite. More rarely, pinkish tan sphalerite has been found at Mulford quarry,

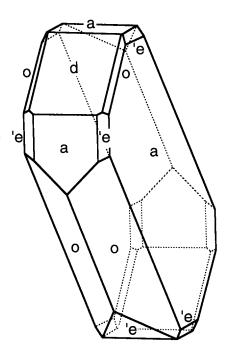
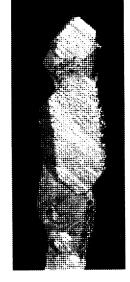


Figure 8. The pseudo-monoclinic symmetry displayed by crystals on pyrite bars. Faces shown belong to the isometric forms  $a\{100\}$ , " $d\{110\}$ ," ' $e\{210\}$ , and  $o\{111\}$ . The face labeled d (and its equivalent at bottom) is actually the contact surface with the adjacent crystal along the bar, and is not a true crystallographic face.

Figure 9. A pyrite bar, 4 mm tall, composed of four crystals. Irene quarry; R. Peter Richards specimen and photo.



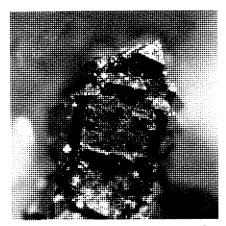


Figure 10. Mirror-smooth second-generation pyrite overgrowths along the edges of rough faces of first-generation pyrite. Second-generation growth occurs on both cube and octahedron faces. Visible length of the specimen is 1.5 mm. Irene quarry; Edwin L. Clopton specimen, Dan Behnke photograph.

implanted on drusy dolomite, sometimes as reasonably sharp euhedral crystals showing combinations of positive and negative tetrahedra and a trapezohedron.

### PYRITE-MARCASITE INTERGROWTHS

Because of the unusual nature of several types of pyrite/marcasite intergrowths from these quarries, we have focused our efforts on an understanding of the development of these intergrowths, the presentation of which forms the rest of this paper. Our hypotheses were formed through:

- (1) A study of the literature on pyrite and marcasite associations.
- (2) Thorough study of numerous specimens with a stereoscopic microscope
- (3) Detailed study of representative specimens with a scanning electron microscope.
- (4) Study of polished sections of selected specimens in reflected polarized light with a petrographic microscope.
  - (5) Goniometric measurement of selected pyrite crystals.
- (6) Comparisons of the crystal structures of pyrite and marcasite, facilitated by development of oriented stereoscopic drawings of these structures using the program ATOMS.
  - (7) Morphological analysis and modeling using the program SHAPE.
  - (8) X-ray and electron diffraction study of selected specimens.

#### **Bars and Chains**

Pyrite bars are of mineralogical interest because they so obviously deviate from the equant habit expected of an isometric mineral. Pyrite bars have been reported from many localities. Most are microcrystals with extreme length/width ratios: lengths may reach several millimeters but widths are generally less than 0.1 mm. These crystals are often so small in cross section that they are nearly invisible, even under a microscope at 30x, unless the microscope light is reflecting from a line-like crystal face. These tiny acicular crystals, often called whiskers, are known from low-temperature hydrothermal deposits at a number of localities, and are highly elongated single crystals. The distorted habit has been attributed to screw dislocations (Henderson and Francis, 1985, 1989; Murowchick and Barnes, 1987; Richards, 1985).

Pyrite bars large enough to be easily seen with the naked eye are far less common, but are known from several localities (listed in Table 1) in addition to the quarries described in this paper. They are commonly more irregular than the "whisker" crystals, and many are not single crystals. They share several characteristics which separate them from other distorted pyrite crystals or crystal aggregates.

Where such crystals occur, the bar-shaped habit is common among crystals at the locality, at least in a particular vein or pocket. The bars are dramatically more elongate than "normal" crystals of the same species with which they may occur. While these elongated crystals are common, there are, conversely, no crystals which are shortened to create a platy habit. These characteristics indicate that a mechanism other than "accidents of chance" is at work, and separates bars from slightly elongated crystals which result from irregularities of growth, either accidental or imposed by a non-uniform supply of atoms to the growing crystal.

Bars are composed of one or only a few crystals, in a non-random arrangement, and are found in cavities in the host rock. Thus they are distinct from polycrystalline pyrite aggregates of linear form (for example, pyritized worm burrows or linear pyrite concretions), which contain thousands of randomly oriented pyrite crystals, generally forming an overall cylindrical shape. Many of these polycrystalline aggregates are found embedded within the host rock, whereas pyrite bars are typically found free-growing in cavities in the host rock.

While the bars from the northern Illinois quarries initially appear to be similar to those from the localities listed in Table 1, careful examination shows that they are distinctly different from crystals from

Table 1. Previously known localities for macroscopic pyrite bars.

Location		crystals Cross-sectio	Comments n
Amex mines Boss, Missouri	to 32 cm	to 7.6 cm	Pitted but apparently single cuboctahedral crystals or parallel aggregates with elongated rectangular axial cavity parallel to the <i>a</i> axis. See White (1975).
Naica Chihuahua, Mexico	to 5 cm	2.1–2.4 mm	Morphologically complex single crystal overgrowths on pyrite whiskers elongated parallel to the <i>a</i> axis. See White (1973).
Rensselaer and Pleasant Ridge, Indiana	to 3 cm	to 1 cm	Combinations of cube, octahedron and pyritohedron. Single crystals or several subparallel crystals. A central tubular cavity is present in most specimens. Some are elongated along <i>a</i> and probably epitactic on pyrite; others may be epitactic on marcasite. See Brock (1986).
Stillwater, Arkansas	10–20 mm	1 mm	Cuboctahedral crystals of uncertain size and highly variable shape. Elongated parallel to the <i>a</i> axis. See Shannon (1923).
Rondout, New York	(?)	1 mm	Cubic crystals elongated parallel to the <i>a</i> axis, size uncertain. See Whitlock (1905).

all of the localities with the exception of some from Rensselaer, Indiana. In the other occurrences, the pyrite bars are elongate parallel to an a axis of the pyrite, and are either apparently single crystals or are composed of a few parallel or nearly parallel crystals. These crystals are typically nearly square in cross-section. By contrast, the bars from the Illinois localities are usually composed of a larger number of crystals. More importantly, the a axes of all crystals are aligned in one of two directions, both of which are inclined to the long axis of the bar (Figs. 6, 7 and 9). They sometimes deviate from square cross-section, being flattened parallel to the coplanar a faces of the component crystals.

The first impression given by these bars, after one realizes that the component crystals are not parallel to each other or to the bar axis, is that they are composed of alternating crystals twinned on {110}. However, this geometric configuration would result in crystals of the more complex habit having parallel edges, and striations on differently oriented crystals would be perpendicular. Neither configuration is observed; rather, the striations make angles of about 75° and 105° with each other, and the crystal edges which would be parallel under twinning instead make angles of about 15° with each other (see Figs. 6 and 14).

A hypothesis which explains the geometric observations is that the pyrite is epitactic on an acicular marcasite crystal, which is completely overgrown by the pyrite. Under this hypothesis, a cube face of the

pyrite would be parallel either to the (101) structure plane of marcasite, or to the crystallographically equivalent (101) plane. These two planes make an angle of 74°38′ with each other. In order to fully specify the epitactic relationship, a direction in each parallel face must also be specified. Two possibilities would explain the observed morphology; either [100] or [010] of pyrite is parallel to [010] of matcasite. These two epitactic models can be summarized in crystallographic shorthand as:

A: (001)[100] pyrite  $\parallel (101)[010]$  or (101)[010] marcasite, or B: (001)[010] pyrite  $\parallel (101)[010]$  or (101)[010] marcasite.

The two models are very similar. The only difference is a  $90^{\circ}$  rotation of the pyrite about the b axis of the marcasite. This difference, while subtle, is crystallographically significant. Recall that cube faces of pyrite have two-fold symmetry, not four-fold symmetry. Thus the [010] and [001] directions are not equivalent in pyrite. This fact is reflected in the orientation of striations on cube faces, and by the observation that the pyritohedron  $\{210\}$  is very common, while the pyritohedron  $\{120\}$ , which is morphologically identical but rotated by  $90^{\circ}$  relative to the structure, is extremely rare. Furthermore, the directions of striations and the positions of  $\{210\}$  faces of pyrite in epitactic associations formed according to model A will be different from those in associations formed according to model B.

When the interfacial angles between adjacent crystals on pyrite bars are measured, these two models predict the measured interfacial angles, within the measurement error. Thus either is a satisfactory description of the observed morphology. Only model A is recognized in the literature, however, and it is the more probable of the two when the crystal structures are considered (Brostigen and Kjekshus, 1970; Fleet, 1970; Brock and Slater, 1978). This can be understood by studying Figure 12, which presents stereopair drawings of slices of the marcasite and pyrite structures, superimposed according to each model. Model A causes entire sheets of the two structures to coincide almost perfectly. The misfit between the two structures in this orientation is minimal, and intergrowths with this orientation would be expected, according to the laws of crystal physics. Model B causes only alternating chains of the two structures to coincide. The misfit between the two structures is much larger, and intergrowths with this orientation would therefore be much less likely to occur.

Examination of a number of bars shows that all are consistent with respect to placement of the {210} faces. Thus only one of these two models is realized in this material. The question is, which one? If the direction of elongation of the marcasite were known, the correct model could be deduced from the placement of the {210} faces of the pyrite. Unfortunately, determining the direction of elongation of the marcasite is not a trivial matter. Most bars show no external evidence of a marcasite crystal. Studies of polished cross-sections of bars reveal that marcasite is present near the center of the bar, but the areas of marcasite are very small and the orientation of the marcasite cannot be deduced from these sections. The mass of the pyrite is much greater

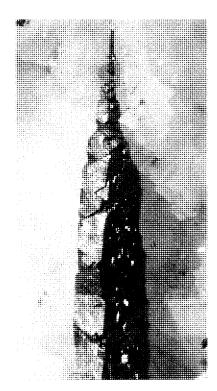


Figure 11. Pyrite bar with acicular marcasite crystal projecting from one end. The part of the aggregate shown is 3 mm long. Irene quarry, R. Peter Richards specimen and photo.

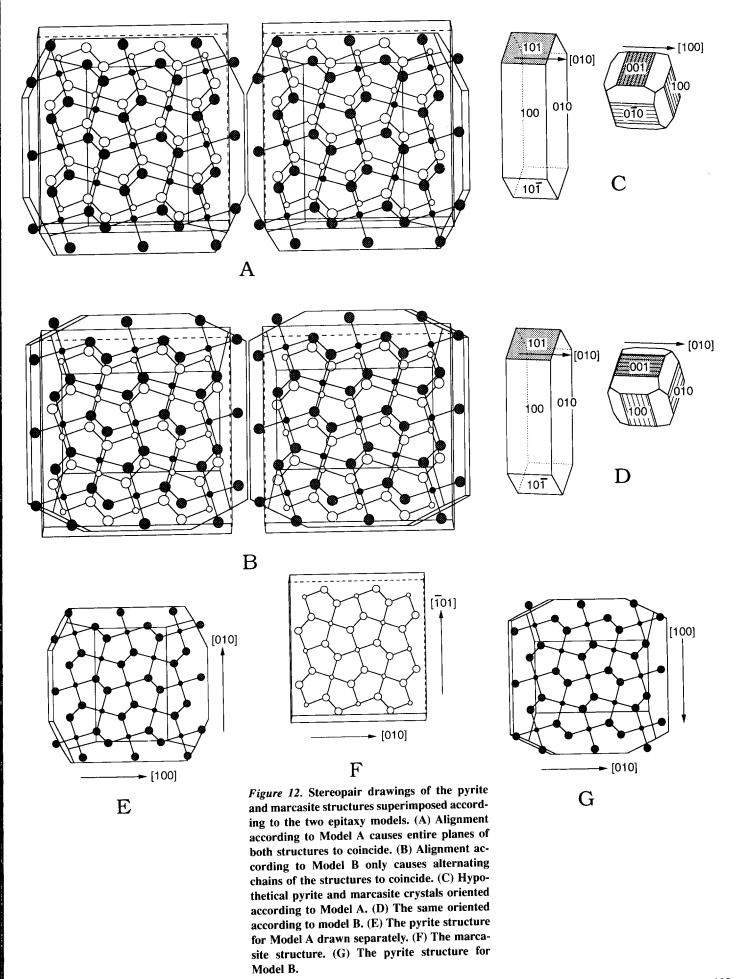
than the mass of marcasite, preventing X-ray examination of the marcasite in typical bars.

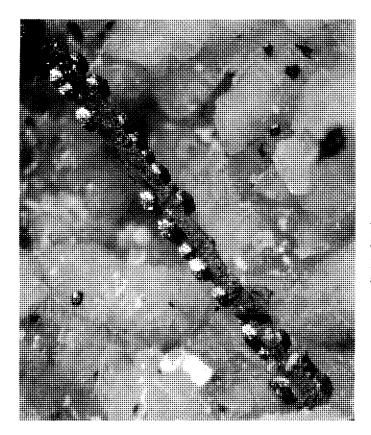
A few specimens have been found with acicular marcasite projecting from one end (Fig. 11). Acicular marcasite crystals with isolated epitactically oriented pyrite have also been found, and these pyrite crystals show the same orientation as those which form the bars, as determined by the placement of the  $\{210\}$  faces. These associations are equivalent to more typical bars in the early stages of their development. X-ray study of the marcasite projecting from one bar showed that it was elongated parallel to the a axis. This result, in combination with the orientation of the pyrite  $\{210\}$  faces, indicates that the bars have developed according to model A.

Several remarkable specimens have also been located which have parallel marcasite crystals projecting *between* the pyrite crystals which form the bar (Fig. 13). These marcasite crystals are also overgrowths on the original acicular crystal, and apparently grew simultaneously with the pyrite, or nearly so. Their habit sometimes includes the bipyramid {111}, the orientation of which is also consistent with an original marcasite crystal elongate along the *a* axis and with epitactic model A.

Our ideas and observations are summarized in Figure 14. Initially, acicular marcasite crystals grew in vugs in the dolostone. These crystals were elongate parallel to their a axes. Thereafter, pyrite crystals nucleated epitactically on the marcasite whiskers, in one of two equivalent orientations. Further growth led to the partial or complete coalescence of adjacent like-oriented pyrite crystals, and contact between opposite-oriented crystals. Partial coalescence creates crystals with notches (see Fig. 14D, lower). Complete coalescence results from filling of the notches, and produces crystals such as those in Figure 14E and Figure 8. Coalescence can sometimes be recognized by irregularities in the surface texture or slight misfit of the coalesced crystals. Extensive coalescence seems to be associated with the habit involving larger octahedron and pyritohedron faces; aggregates with these properties form the pyrite bars. Pyrite chains typically have

The first of these epitactic relationships is geometrically equivalent to one listed, with several others, by Palache *et al.* (1944) in reference 11 to the section on marcasite: {001}[100] pyrite parallel to {010}[101] marcasite. While our statement of the relationship is geometrically identical to Palache's in terms of the external morphology, ours is preferable because marcasite (101), not marcasite (010), is the structural plane which is similar to pyrite (001) (Brostigen and Kjekshus, 1970; Brock and Slater, 1978). What may be the same relationship is listed by Palache *et al.* in the section on pyrite on page 284. However, on page 284 two relationships (including this one) are listed as involving {001}[001] of pyrite. We note that these are in error, because the direction [001] does not lie in the plane {001}. Possibly {001}[100] was intended instead.



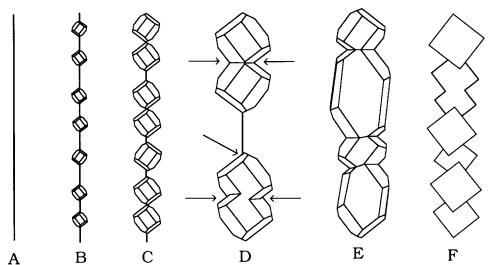


mineral, or a different direction of elongation for an acicular marcasite host. The bars (and other materials) from Rensselaer, Indiana offer an excellent opportunity for further study of pyrite-marcasite epitaxy, particularly if some bars are identified in which the mineral of the central cavity is still present.

Pyrite occurs in epitactic orientation on acicular marcasite from other localities as well, but in these occurrences the pyrite crystals do not merge to form bars. Instead, they form chain-like configurations or remain isolated from each other on the host marcasite. These localities include Pint's quarry, Raymond, Iowa; Hamilton, Illinois; Keokuk, Iowa; Milan, Ohio; and possibly Rockbridge County, Virginia (Dietrich,

Figure 13. Pyrite bar with marcasite crystals projecting between individual pyrite crystals. The portion of the bar shown is 5 mm long. Mulford quarry; R. Peter Richards specimen and photo.

Figure 14. Sequence of development for pyrite bars and chains. When individual epitactic pyrite crystals begin to contact each other, like-oriented adjacent crystals may merge by reentrant filling, leading to the bar form. If such crystals do not merge to any great extent, the chain form results.



crystals of more cubic habit and with limited coalescence.

In crystals in which pyrite and marcasite were developing simultaneously (or nearly so), new marcasite growth occurred in parallel with the acicular marcasite. Geometric competition (Grigor'ev, 1965) between growing crystals favored continued growth of marcasite only in "notches" between the pyrite crystals (if at all), where pyrite growth was slowest.

Armed with an explanation for these pyrite bars, we return briefly to the pyrite bars from elsewhere. Some bars from Rensselaer and Pleasant Ridge, Indiana are clearly polycrystalline, with the individual pyrite axes inclined to the length of the bar. They also typically have an elongate cavity parallel to the length of the bar, which may represent a now-dissolved acicular marcasite crystal. In these characteristics they are comparable to the crystals from northern Illinois. However, the orientation of the pyrite relative to the bar axis is different in many specimens; this may indicate an overgrowth not involving epitaxy, a different epitactic relationship, a different host

1985; reported as a marcasite twin). In all these instances, the orientation of the pyrite to the marcasite host appears to be the same as described in this paper.

What about occurrences of pyrite bars elongate parallel to an *a* axis, such as those listed in Table 1? Hansen (in White, 1975) reported that the bars from the Amex mine had a rectangular hole at the bar axis, and speculated that they represented second-generation pyrite overgrowths on pyrite or marcasite whiskers. While marcasite whiskers are unlikely to provide the right crystallographic orientation for these bars, pyrite whiskers are usually elongate parallel to an *a* axis, and would provide an appropriate substrate. White (1973) showed elegantly that the pyrite bars from Naica are overgrowths on pyrite whiskers of square cross-section and about 0.03 mm thick. Overgrowth on a pyrite whisker probably also explains the materials from the other two localities. The Amex crystals are truly remarkable, because the original whiskers must have been as much as a foot (30 cm) long, which is extremely long for naturally occurring whiskers.

#### Leaf Epitaxy

Careful study of numerous examples of the relationship shown in Figure 5 reveals a history of development similar to that of the bars and chains, and involving the same epitactic relationship, but one in which marcasite deposition dominated over pyrite deposition, and in which the initial marcasite crystal was elongate along the c axis (but not nearly so extremely as the marcasite within the bars). This growth history is diagrammed in Figure 15. Its development from an elon-

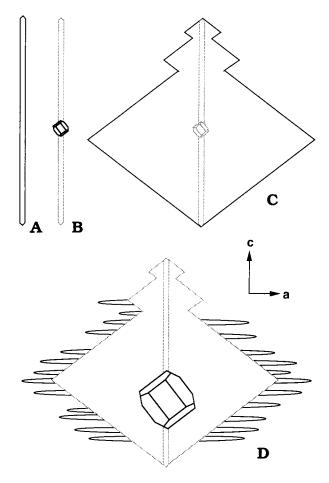


Figure 15. Sequence of development for "leaf" epitaxy. Marcasite growth dominates over pyrite growth. The initial acicular marcasite crystal is elongated in the c direction.

gated marcasite crystal is supported by a faint textural difference in the final marcasite crystal, running along the middle of the crystal and passing though the pyrite; by the observation that one end of the midline of the marcasite is always in contact with the matrix, but the pyrite often is not (thus marcasite preceded pyrite); and by the fact that the pyrite is invariably located near the midline of the marcasite crystal, which suggests that was the only part of the crystal present when the pyrite began to grow.

The conclusion that the first generation marcasite crystal is elongated parallel to the c axis is based on the orientation of the laterformed rhombic marcasite crystals. In these later crystals, the short dimension of the rhombic shape is parallel to the c axis (Fig. 2, top). This dimension is consistently oriented parallel to the long dimension of the first-generation crystal, which indicates that the long dimension of the first-generation crystal must also be parallel to the c axis. It is important to note that these crystals were found in a single pocket together with chalcopyrite, and do not occur together in the same vug with the bars discussed earlier. One would not expect to find marcasite

elongated along a together with marcasite elongated along c in the same pocket (unless they were of two different generations); finding different directions of elongation in different pockets is less problematic

The formation of one or a few small pyrite crystals as a second stage is supported by their location only along the midline of the marcasite crystal, and by the fact that many of these pyrite crystals are complete, with half on each side of the marcasite leaf. If the leaf were completely developed, there would be no reason for the pyrite to be confined to the middle, and no way for the pyrite to "penetrate" the marcasite.

The idea that the pyrite enlarged after the marcasite reached full or near-full size is supported by the observation that the pyrite breaks away from the marcasite very readily, leaving no hole in the marcasite and a flat surface on the pyrite where it had been in contact with the marcasite. Only a very tiny spot near the center of this surface appears broken. The pyrite did not bond strongly to the marcasite along much of this contact surface, perhaps because of a thin film of impurities on the marcasite, or perhaps simply because the contact surface is not the one involved in the epitactic relationship.

The development of the fringed edges is a late step, as indicated by the presence of a fine line where the fringes join to a much more regular diamond-shaped crystal edge. Often the fringes merge, forming an irregular margin. There is no evidence to indicate whether the pyrite enlarged before the fringes formed, or vise versa.

Study of thin edges of these fringes with transmission electron microscopy has produced selective area diffraction (SAD) images with a known orientation to the long direction of the fringe crystals, and hence a known orientation to the entire marcasite plate. Indexing these SAD images reveals that the flat faces are (010) and that the fringe crystals are elongate parallel to the a axis. This verifies the morphological interpretation of the orientation of these plates, and the c-axis elongation of the nucleus crystal.

In the leaf epitaxy, much more of the mass of the aggregate is composed of marcasite, and less of pyrite, than is true of the bars and chains. While these leaves have a more complex growth history, they are comparable to examples of pyrite epitactic on marcasite from many other localities, in which bladed marcasite crystals are decorated with one or a few pyrite crystals. We have personally seen such aggregates from Bellevue, Ohio and Rossie, New York; other localities are mentioned in the literature.

#### **CONCLUSIONS**

- (1) Bar-like and chain-like polycrystalline aggregates of pyrite from northern Illinois formed by epitactic overgrowth on acicular marcasite crystals. Some bars also have marcasite crystals alternating with the pyrite crystals. Bar habits of these morphologies have not been described previously.
- (2) The relationship we have described as "leaf epitaxy" also involves epitactic overgrowths of pyrite on marcasite, but with a more complex history of formation, and one which involved more marcasite deposition and less pyrite. Leaf epitaxy is morphologically related to pyrite epitactic on bladed marcasite from other localities.
- (3) Pyrite bars described in the literature can be divided into (microscopic) pyrite whiskers; overgrowths on pyrite whiskers; overgrowths on acicular marcasite crystals, some with a different direction of elongation than reported here; and possibly overgrowths on acicular marcasite governed by different epitactic relationships than reported here.

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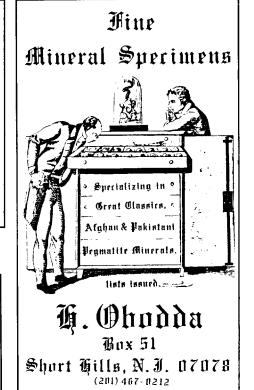
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