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Gabriel Faivre: his legacy on eutectic solidification

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Abstract. In the honor of Gabriel Faivre (1944-2020), I will present a review of major scientific contributions to the understanding of the dynamics of eutectic growth patterns. From the end of the 1980s, Gabriel Faivre undertook a systematic research in solidification guided by the new concepts of the nonlinear physics of out-of-equilibrium pattern formation. Drawing on his outstanding capabilities as an experimentalist, he refined the method of in situ directional solidification of model alloys. With constant reference to physics and metallurgy, he succeeded in carrying out a high-level research, keen to reach strong qualitative impact and quantitative accuracy. Gabriel Faivre made key discoveries, together with coworkers and young researchers in Paris, and in collaboration with materials scientists and physicists in France and abroad. From symmetry breaking instabilities to eutectic cells and dendrites, over rod-like and labyrinth patterns, full light has been shed onto new phenomena, fascinating to the eye and the mind. During the last decade, Gabriel Faivre mentored an in-depth analysis of interfacial-anisotropy effects on coupled-growth patterns, thus reconciling the theories of regular eutectics and crystal-orientation dependent eutectic-grain growth. Being both a rigorous scientist and a generous colleague, he left us a vast legacy of prospective research topics in solidification and crystal-growth science. Sharing his knowledge of fine arts and humanities, Gabriel Faivre also instilled the best of intellectual thinking in those who were fortunate enough to work with him.

1. Introduction

Gabriel Faivre was born on December 16, 1944 in Nancy, France. After graduating at École des Mines de Paris (1966), he spent a few years as a metallurgical engineer at IRSID in Saint-Germain-en-Laye. He graduated a master degree (DEA) in Solid-state physics (Orsay, 1967), keeping unforgettable memories of the teaching by French physicists and metallurgists. Gabriel Faivre pursued his academic curriculum with a PhD in Physical Sciences, which he defended in 1974, under the direction of Georges Saada (University Paris XIII). He was hired by the CNRS as an associate researcher in 1971. He carried out a precursory work on the coherency constraints of precipitates [1], abnormal absorption in transmission electron microscopy of noncentrosymmetric crystals [2], and self-diffusion involving bivacancies at high temperatures [3]. Gabriel Faivre also studied the structures of dislocations in low-symmetry crystals [4,5]. He then turned to the equilibrium-polymer properties of liquid selenium [6,7], and the crystallization of spherulites [8,9]. In the late 1980s, Faivre moved to the Groupe de Physique des Solides (GPS) of University Paris VII (now Institut des Nanosciences de Paris, Sorbonne University) in the Jussieu campus, close to river Seine in the Quartier Latin of Paris, at the invitation of Christiane and Bernard Caroli [10,11]. From the beginning of the 1990's as a Directeur de Recherche at CNRS, Gabriel Faivre led an impressive, nearly 30-year journey through the study of the fascinating structures of solidification fronts. He was joined by myself (SA) as a young CNRS researcher in 1992, and then by Sabine Bottin-Rousseau in 2000. During his career, Gabriel Faivre supervised several doctorate students –Jacques Mergy (1992), Marie Ginibre (1997), Ghislain Boutet (1999) and Mikael Perrut (2007) as concerned solidification– and post-doc researchers –among them,



Thomas Ihle, Tamás Börzsönyi and Melis Şerefoğlu. Emeritus since 2012, he actively pursued his research until 2019.

Gabriel Faivre played an important role in the refoundation of the field of solidification that took place in the 1980s and 1990s. His research work on solidification put strong focus on the non-linear physics of self-organized structures in extended systems (nonequilibrium pattern formation), with constant reference to solid-state physics and metallurgy. His studies cast new light on the phenomenological equivalence between the dynamics of crystal melt-growth shapes –cellular, dendritic and eutectic growth– in metals and that of other well-studied nonequilibrium systems such as Rayleigh-Bénard convection [12,13]. He also provided renewed evidence of the overwhelming power of demonstration of in situ experimentation by real-time observation of the solidification front in model systems, and was keen to make it quantitative. This was achieved thanks to instrumental innovations developed with his collaborators in Paris. Faivre was one of the initiators of a microgravity project coordinated by the European space agency (ESA), thanks to long-term support by the French space agency (CNES) [14,15]. Fortunately, he had the opportunity to see the images of the first campaign in 2018. Gabriel Faivre also elaborated new theories, and inspired collaborations with physicists performing numerical simulations.

Faivre liked to quote Jacques Friedel, Raimond Castaing and André Guinier among his admired masters. His work also owes much to sustained discussions he had with non-linear physicists. He greatly appreciated meetings organized in the 1990's by the European research network *Pattern Formation, Noise and Spatio-Temporal Chaos in Complex Systems* (L. Kramer). Gabriel Faivre enjoyed fruitful collaborations, noticeably those with Alain Karma (Northeastern University, Massachusetts) and Mathis Plapp (Ecole Polytechnique, Palaiseau), but also Laurent Limat (CNRS, Paris) and Efim Brener (FZ Jülich, Germany), as well as with Stephan Rex, Ulrike Hecht and their colleagues (Access e.V., Aachen, Germany). He also established links with research groups in Marseille (Bernard Billia; Alain Pocheau) and Grenoble (Denis Camel) in France, and Zürich (Jörg Bilgram) and Lausanne in Switzerland –he frequently remembered the Zermatt workshops organized by Wilfried Kurz and Michel Rappaz (EPFL).

Gabriel Faivre's contributions to spherulite growth [8,9], dendritic/cellular solidification [16-18], subboundaries [19] and faceted growth [20,21] are all of great value. This makes too many topics for the present paper. I shall limit myself to a brief account of Gabriel Faivre's research activity in the field of eutectic growth [22] during, say, the latest 20 years of his career, and focus on (i) the spatio-temporal dynamics of thin and bulk eutectics, and (ii) the effects of the free-energy anisotropy of interphase boundaries on the microstructures of eutectic grains.

2. Eutectic solidification patterns

2.1. Morphology diagram

Binary eutectic alloys present a three-phase equilibrium between the liquid and two different solids at a given eutectic temperature T_E . This makes possible a two-phase growth from the melt upon cooling. In directional solidification at imposed velocity V in a fixed temperature gradient G , eutectic growth occurs in the form of two-phase patterns. In nonfaceted alloys, the dynamics is primarily determined by solute diffusion in the liquid and by capillary effects. The resulting composite microstructure is a frozen-in trace left behind in the bulk solid by the coupled-growth patterns. A reference in the field of eutectic growth is the theoretical and experimental work by Jackson and Hunt (JH) in the 1960's [23]. These authors established the existence of a scaling length, the so-called minimum-undercooling spacing λ_m , which is typically of order $10 \mu\text{m}$ for V in a $1\text{-}\mu\text{ms}^{-1}$ range, and varies with V as $V^{-1/2}$. Experimentally, Jackson and Hunt showed the general-demonstration power of thin-sample directional solidification using model transparent nonfaceted alloys for in situ observation. It should be mentioned that Gabriel Faivre, who suspected the extraordinary potential of the model eutectic $\text{CBr}_4\text{-C}_2\text{Cl}_6$ alloy, was confronted to the detrimental fact that the CBr_4 molecule reacts with foreign impurities, and degrades in the liquid state. In the late 1980's, he initiated a collaboration with Robert Mellet (CNET), who was an expert on high-level purification of organic systems by zone-refining. Purified CBr_4 and

C_2Cl_6 with a good chemical stability could be mixed, and used for both precise thermodynamic assessment [24] and unprecedented solidification studies –including that of ternary eutectics (see, e.g., [25] and [26]). During the doctorate work by Jacques Mergy, and then by Marie Ginibre, major aspects of the dynamics of lamellar-eutectic patterns in two dimensions (2D), in thin samples, were explained. Basic theories (the JH analysis of regular eutectics, but also the Mullins-Sekerka theory of cellular patterns) predict the existence of continuous branches of steady periodic structures with the spatial periodicity λ varying over a finite range at fixed control parameters. The problem is to determine the interval of accessible λ values, and to identify the mechanisms that limit that interval. In thin lamellar-eutectic samples, secondary instabilities occur, on the large- λ side, via spontaneous breaking of basic symmetries. The term spontaneous means, as a general feature of pattern formation in spatially extended systems [12], that the morphological instabilities are not imposed from outside, but are made possible by the nonlinear coupling between the shape of the interface and the diffusion field. Faivre and Mergy discovered the tilt bifurcation, corresponding to a change from steady-state, mirror-symmetry patterns to travelling, tilted ones. It can occur uniformly [27,28] or in the form of finite domains [29]. Interestingly, the drifting motion of tilt-domain walls entails a dynamic lamellar-spacing selection [29,30]. The tilt bifurcation is independent of any crystallographic effect. During Marie Ginibre's thesis, a comprehensive description of 2D symmetry-broken shapes could be achieved [31] –including some oscillatory patterns that had been observed previously in Al-Cu [32]. In addition, a comparison of experimental observations and time-resolved numerical simulations by Karma and Sarkissian was achieved, with quantitative agreement [33]. A few years later, the question of how incipient spacing modulations in a lamellar eutectic pattern smooth out and evolve towards a fully periodic state was addressed by Gabriel Faivre and myself, in collaboration with Alain Karma and Mathis Plapp [34]. This led to revisit the so-called “phase-diffusion” phenomenon in reference to the previous work by Langer [35].

In 2000, Bernard Zappoli at CNES brought a strong support to our project dedicated to in situ observation of bulk eutectics, with the prospect of a science-in-microgravity campaign aiming at studying eutectic growth in purely diffusion-controlled conditions [15]. That was a challenging task, both theoretically and experimentally, as regards the number of degrees of freedom, the possibility of forming lamellar, rod-like and more complex shapes, and the presence of thermo-solutal convection. The problem of how to observe eutectic growth patterns in situ over the whole solidification front was not obvious as well. With Sabine Bottin-Rousseau, it was decided (i) to use flat-wall samples of the $CBr_4-C_2Cl_6$ alloy with a useful thickness in the millimeter range, and (ii) to observe the solidification front from the exterior, at an oblique view. With some improvements –mainly a diaphragm for dark-field viewing– the method (DIRSOL) worked fine [36]. With the new instrument, the zig-zag instability of lamellar eutectics was discovered [37]. It was also observed that, in fully isotropic conditions, a lamellar eutectic initially forms “labyrinth” patterns that evolve slowly, and appear almost “frozen” [38]. These observations also prepared us to address an even more complex question: that of the so-called lamellar-to-rod transition –a typically 3D morphological change that obviously cannot occur in thin samples. For this purpose, Mikael Perrut studied a rod-like eutectic, namely the succinonitrile-d,camphor (SCN-DC) alloy [39,40], with the help of Victor Witusiewicz and Laszlo Sturz at Access e.V. in Aachen (Germany) [41]. Thanks to Mikael Perrut's remarkable thesis work, the decision was made at ESA to engage a collaborative project (Paris, Aachen, Leoben) and build a new apparatus, starting from the DIRSOL method, for the study of solidification processes in transparent alloys by real-time optical monitoring. After a long maturation process –let us also mention the work by Melis Şerefoğlu during that period of time [42]– the apparatus (TRANSPARENT ALLOYS) was built by ESA contractors, sent to the ISS (Dec. 2017), and installed in the Microgravity Science Glovebox (MSG) facility. The first scientific campaign onboard the ISS in January-March 2018 brought full evidence of a lamellar-to-rod transition in the SCN-DC alloy [43].

2.2. Anisotropy effects

Faivre and Mergy made several observations during thin-sample directional solidification showing that, in a given sample, the lamellar-growth dynamics was varying from one eutectic grain to another

[11]. This indicated that the orientation of the crystals could influence the coupled-growth dynamics. In particular, in certain eutectic grains, the lamellae were growing tilted with respect to the main solidification axis, and, simply speaking, followed an inclination, which did not change whatever the value of V and λ . This dynamics was called “locked-lamellar” growth by Christiane Caroli, which illustrates well that, in the anisotropic case, the tilt dynamics is imposed by the crystallography, as opposed to a spontaneous breaking of the symmetry. In Ref. [11] the term “floating” dynamics was also introduced for designating regular lamellar eutectic grains within which the interphase boundaries can freely rotate (Fig. 1a). The question of the formation, growth and competition of eutectic grains has been formulated and extensively documented in the 1970’s in the metallurgy literature. The main background at that stage was that of the identification (in particular by electron microscopy) of (heterophase) coincidence lattice planes, thus defining crystal orientation relationships (ORs) [44]. As a common wisdom, it was stated that lamellae were driven to grow along a coincidence plane corresponding to a low-energy inclination of the interphase boundary. The observations were convincing, and the intuition inspiring, but no rigorous argument was advanced. Gabriel Faivre, with Sabine Bottin-Rousseau and myself, decided to reconsider the question on the following bases.

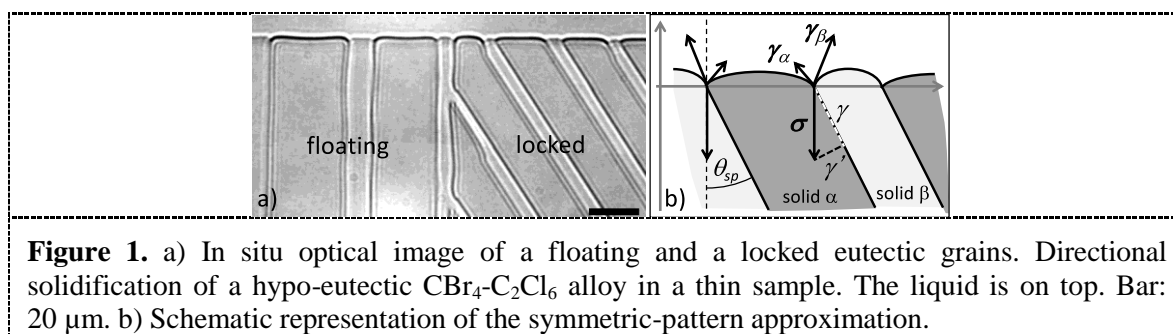


Figure 1. a) In situ optical image of a floating and a locked eutectic grains. Directional solidification of a hypo-eutectic $\text{CBr}_4\text{-C}_2\text{Cl}_6$ alloy in a thin sample. The liquid is on top. Bar: $20\ \mu\text{m}$. b) Schematic representation of the symmetric-pattern approximation.

The locked-lamellar dynamics, and, more generally, the eutectic-grain dependence of coupled-growth patterns, is not described by the JH theory of regular eutectics, which assumes fully isotropic interfaces. Generally speaking, it has been shown that, with some anisotropy of solid–liquid or solid–solid interfaces, lamellar eutectic patterns must travel laterally in steady-state, and deliver tilted-lamellar microstructures in the solid. Gabriel Faivre carried out an analysis “à la JH” of tilted-lamellar patterns, and brought a demonstration that a weak solid-liquid anisotropy, to be expected in nonfaceted eutectics, can entail the formation of tilted-lamellar patterns with tilt angles not larger than a few degrees [45]. In contrast, the free energy γ of the interphase boundaries in the solid can be substantially anisotropic, in particular in eutectic alloys that present special ORs with dense coincidence planes. Lamellar tilt angles can then be large. Two questions were open: 1- how decisive progress could be made on a theoretical level –that is, how a realistically large anisotropy of γ could be incorporated in the equations? and 2-what could be done experimentally to inspire and/or support that theory?

The anisotropic quantity γ enters the condition of local equilibrium at the trijunctions. It can be expressed by introducing the Hoffmann-Cahn surface-tension vector σ in the Young-Herring equation [46]. The vector σ is the sum of a component parallel to the boundary, of value γ , and a component normal to the boundary, of value γ' (the angular derivative of γ). In two dimensions, an anisotropic surface tension of the interphase boundary means that, at fixed relative orientation of the two solids, the value of $\gamma = \gamma(\theta)$ depends on the inclination angle θ of the interphase boundary measured from a fixed axis –e.g., the axis of the temperature gradient \mathbf{z} in directional solidification. The $\gamma(\theta)$ polar plot (γ -plot) is a circle for an isotropic system, or a slightly deformed circle (with a centrosymmetric shape) for a mildly anisotropic system. In contrast, the γ -plot exhibits deep, singular minima in strongly anisotropic cases –for example in a eutectic grain with an OR with dense lattice planes in coincidence. At the trijunction, except for special configurations, the σ vector is no longer parallel to the interphase boundary, and the lamellae can be expected to grow tilted. The question then is to determine the value

of the lamellar tilt angle θ_t in steady-state. As a key point, in situ observations during thin-sample directional solidification of the $\text{CBr}_4\text{-C}_2\text{Cl}_6$ alloy were made, evidencing that in locked-lamellar patterns, the shape of the solid-liquid interfaces was keeping (nearly) the same mirror symmetry as in a regular, isotropic case. This indicates a weak coupling between the lateral drift motion of the pattern and the diffusion field. It can then be realized that a strict symmetric-pattern condition imposes that the σ vector is parallel to the axis of the temperature gradient (Fig. 1b) [47]. In the symmetric-pattern (sp) approximation, the steady-state tilt angle $\theta_t \equiv \theta_{sp}$ of the lamellae is fully determined. It can be calculated from a nonlinear equation for the inclination θ of the interphase boundary for an arbitrary anisotropy function $\gamma(\theta)$, knowing the orientation of the eutectic grain. The contribution of Gabriel Faivre to this analysis was determining. The qualitative relevance of the sp-approximation was confirmed numerically [48] (also see [49] and refs. therein). A similar approach was used to analyse the growth of anisotropic subboundaries [50].

Aiming at a better understanding of the mechanisms via which eutectic grains form during early solidification stages, a careful experimental study of the so-called eutectic invasion was carried out by Gabriel Faivre and myself with Sébastien Moulinet [51]. This process occurs without nucleation of new crystals, and can deliver floating, isotropic eutectic grains with no special OR.

The rotating directional solidification (RDS) method was then developed (see [52] for a previous attempt, and [21] for the first use of the RDS apparatus in Paris). During RDS, a standard directional-solidification configuration is used, and a thin sample, instead of being translated, is rotated about an axis perpendicular to the sample plane. On one side of the sample, the solid grows (it melts on the other side) while the orientation of the crystals is continuously varied. In an isotropic eutectic grain, the interphase boundaries remain locally perpendicular to the isotherm, and describe mere circles in the sample reference frame. In an anisotropic grain, it can be intuited that the shape of lamellae in the solid reflects the γ -plot. Indeed, within the symmetric-pattern approximation, the RDS microstructure is homothetic to the shape of the interphase boundaries predicted by the Wulff construction [53]. A more recent RDS setup has been built by Melis Şerefoğlu in her laboratory in Turkey, and used, in particular, for the study of interfacial-anisotropy effects in ternary, three-phased eutectics [54].

The next step was to establish a clear link between the locked-lamellar growth dynamics, the presence of deep minima in the γ -plot, and coincidence planes in anisotropic eutectic grains. For this purpose, in situ optical observations were coupled with ex situ crystallographic measurements. The thin-sample technique was adapted to metallic eutectic alloys. Using first the low-melting $\text{In-In}_2\text{Bi}$ system, Gabriel Faivre helped us –with Oriane Senninger and Laurent Carroz– to define a new experimental protocol including the analysis of x-ray diffraction pole figures, and to establish a method for identifying dense coincidence planes, and ORs [55]. More recently, this methodology was applied to thin $\text{Al-Al}_2\text{Cu}$ samples [56].

3. Conclusion

Gabriel Faivre was a gifted physicist, a man of rare intelligence. From the reading of the above text, it should be clear that he was viewing laboratory experimentation and theoretical analysis as intimately linked and interdependent activities. He accomplished outstanding achievements on both levels.

Gabriel Faivre was a discreet person, sometimes even a little secretive. He was uncompromising and rigorous in his scientific arguments, but very attentive in his relations with his peers – at the same time demanding and benevolent. Being indefatigable at passing his knowledge to his collaborators, Faivre also knew how to leave the initiative to his close collaborators and thus bring out new ideas.

Gabriel Faivre was also a man of great culture. During his whole life, he has been immensely fond of books, history, music and fine arts. He also learned a lot, according to his own words, from rock climbing, which he practiced intensely in his youth, mainly in the French Alps.

Gabriel Faivre passed away on April 17, 2020 in Paris, in the protective presence of his beloved family. His colleagues are deeply, and forever indebted to him, being conscious how fortunate they are to have known him, and worked with him.

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