Oscillatory lamellar microstructure in off-eutectic Al-Cu alloys

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We report results of a series of laser melting and resolidification experiments of hypoeutectic Al-Cu alloys designed to characterize the oscillatory lamellar eutectic morphology for the first time in detail. Oscillatory and nonoscillatory lamellar domains are observed to coexist over a wide range of compositions and growth rates. The dependence of the amplitude and frequency of oscillations is studied within this range. A mechanism for the propagation of the boundary line between the two different domains is proposed.

Eutectic growth is an important interfacial-pattern-forming system in the area of solidification. In this system two thermodynamically distinct solid phases are left behind a solidification front advancing in a melt of eutectic or near-eutectic composition. The solidification front is typically grown directionally in the presence of a temperature gradient. In the simplest situation where this front advances in a steady-state fashion the two solid phases form a spatially periodic structure of juxtaposed lamellas of the two solid phases (lamellar eutectics) or of rods of one solid phase imbedded in the solid matrix of the other phase (rod eutectics). The theoretical basis for the understanding of steady-state eutectic growth was provided by the pioneering analysis of Jackson and Hunt[1] who showed that both lamellar and rod morphologies result from a balance of diffusional undercooling and the undercooling due to interfacial curvature so as to maintain an isothermal interface. Both morphologies have also been well-characterized experimentally in both metallic systems and organic analogs.[2–4]

Although many aspects of steady-state growth seem by now to be reasonably understood, our experimental and theoretical knowledge of the range of stability of even the simpler lamellar structures at off-eutectic melt composition has remained very incomplete. On the theoretical side, Hurle and Jakeman[5] have treated the eutectic front as a single phase by averaging over the properties of the two solid phases. Their analysis however did not allow for local variations of the lamellar spacing which was shown subsequently by Datye and Langer[6] to play an important role in unstable modes. Datye and Langer based their stability analysis on a discrete model for displacements of triple points by averaging the composition field over individual lamellas and were the first to predict the presence of an oscillatory instability on twice the lamellar spacing at sufficiently off-eutectic melt composition. Since then this oscillatory instability has also been seen in numerical simulation of eutectic fronts by one of us.[7,8] On the experimental side, oscillatory eutectic microstructures have appeared in photographs taken of thin-film transparent organic materials[9] but to our knowledge no systematic study of this phenomenon has been performed. What has been more commonly reported in the metallurgical literature is a breakdown of steady-state lamellar structures accompanied by the appearance of dendrites of the growth governing phase and the coexistence of the two structures over some range of off-eutectic melt compositions.[10]

In this paper we report the results of a series of experiments conducted on an Al-Cu alloy system, which was designed in an attempt to characterize the oscillatory eutectic morphology in more detail. In particular, solidification experiments were performed by laser treatment at three increasingly more off-eutectic melt compositions in order to determine both the range of composition over which oscillations exist and the variations in amplitude and frequency of oscillations with composition and velocity. Using laser surface melting and resolidification techniques, the local solidification front velocity V can be directly related to the microstructure. This rate is calculated by taking a longitudinal cross section through the center line of the remelted trace and measuring the orientation of the microstructure which tends to be perpendicular to the local solid-liquid interface.[11]

The three hypoeutectic Al-Cu alloys with 25, 28, and 31 wt. % of copper (eutectic composition $C_E = 33$ wt. %) were produced by melting the pure Al and Cu components (99.99% purity) in a graphite crucible and then casting them into a copper mould. After machining off the surface that had been in contact with the mould, surface homogenization treatment were performed in order to reduce the scale of cast structure. This treatment consists of overlapping laser traces at a speed of 10 cm/s with a power density of 2.8×10^6 W/cm^2. The rapid solidification experiments were conducted using a CO_2

laser with a nominal power of 1500 W and a spot diameter of 420 μm, i.e., with a power intensity of 1.1×10^6 W/cm^2. Controlled-velocity surface melting was carried out by moving the specimen at various transverse speeds between 1 cm/s and 100 cm/s. During the laser treatment a continuous flow of helium was blown over the surface in order to reduce oxidation of the molten pool. Before laser treatments, the samples were polished using 1000-grit SiC paper in order to enhance absorption of the laser beam as well as to ensure the same surface quality for each sample. After laser treatment a nickel layer approximately 2-mm thick was electrolytically deposited onto the treated surface. This nickel deposit was necessary in order to permit the cutting of thin longitudinal sections from the middle of the laser trace for transmission electron microscopy (TEM) observations. A detailed description for the electrolytic thinning of these sections is given in another paper.[11]

A quantitative study of microstructures was then performed directly by taking a large number of photographs of different parts of the longitudinal sections at various magnifications. A summary of our experimental observations is shown in Table I. The quantity $U_\infty=(C_E-C_0)/\Delta C$ was chosen as a measure of the degree of departure from the eutectic composition.[6] Here C_0 is the alloy composition and ΔC the difference in composition between the two solid phases: Al and Al_2Cu. The values of λ reported in Table I were obtained by averaging the lamellar spacing over more than 20 successive Al and Al_2Cu lamellas. The amplitude and frequency of oscillations of lamellas are characterized by two quantities ϵ and Ω. The quantity $\epsilon=(S_{max}-S_{min})/S_{min}$ is chosen as the relative difference of the maximum and minimum width of Al lamellas as shown in Fig. 1. The amplitude ϵ, so defined, has the advantage of being a scale-independent dimensionless quantity with $\epsilon=0$ for steady-state lamellar structures. The dimensionless frequency of oscillations was chosen as:[6] $\Omega=\omega\lambda/V=\pi\lambda/L$

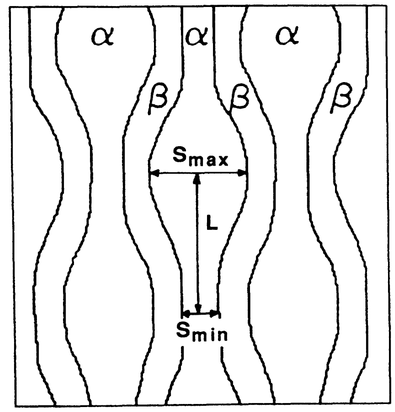

FIG. 1. Simulated oscillatory eutectic morphology for $U_\infty=0.17$ (after Ref. 8). S_{max} and S_{min} are the maximum and minimum widths of an α lamella. L corresponds to the half wavelength of one oscillation.

where L, the half wavelength of one oscillation, is shown in Fig. 1.

The oscillatory phenomenon generally does not extend over the whole sample but remains confined to regions extending over several lamellas. The values of ϵ and Ω given in Table I were measured only on well-developed oscillations within these regions. Oscillatory and nonoscillatory lamellar domains coexist and the boundaries between these two different domains propagates along the solidification front. This propagation phenomenon together with the advance of the solidification front generates oblique boundary lines as shown in Fig. 2. The angle θ between these boundary lines and the growth direction was found to remain approximately constant ($\theta=35°$) for very different growth conditions (e.g., $V=4.8$ cm/s, $U_\infty=0.1$, and $V=9.6$ cm/s, $U_\infty=0.04$). A similar propagation phenomenon associated with the tilting of

TABLE I. Summary of the results as a function of U_∞ and growth rate.

U_∞	V (cm/s)	λ (nm)	λ^2V (μm^3/s)	L (nm)	Ω	ϵ
0.04	7.1	33.6	79.7	50.0	2.11	0.95
0.04	7.8	30.0	69.9	51.4	1.83	0.71
0.04	8.7	28.6	70.7	42.9	2.09	1.07
0.04	9.6	25.0	59.8	42.1	1.87	0.93
0.04	15.1	20.8	65.5	37.5	1.75	1.10
0.10	0.6	70.8	31.2	95.8	2.32	1.60
0.10	1.7	45.8	35.9	72.9	1.98	1.50
0.10	2.1	66.7	93.9	93.8	2.23	1.80
0.10	3.5	35.4	43.9	62.5	1.78	1.67
0.10	3.6	39.6	56.4	62.5	1.99	2.33
0.10	3.8	50.0	95.8	93.8	1.67	1.55
0.10	4.8	35.4	60.7	64.6	1.72	2.00
0.10	4.9	35.4	60.9	62.5	1.78	2.56
0.10	5.2	31.2	50.3	54.2	1.81	1.50
0.10	6.7	28.6	54.6	47.1	1.91	1.71
0.10	6.9	27.1	51.0	47.3	1.80	2.71
0.10	7.5	25.0	47.2	48.6	1.62	2.09
0.16	0.5	66.7	22.2	100.0	2.09	3.00
0.16	1.7	37.5	23.9	83.3	1.41	1.10

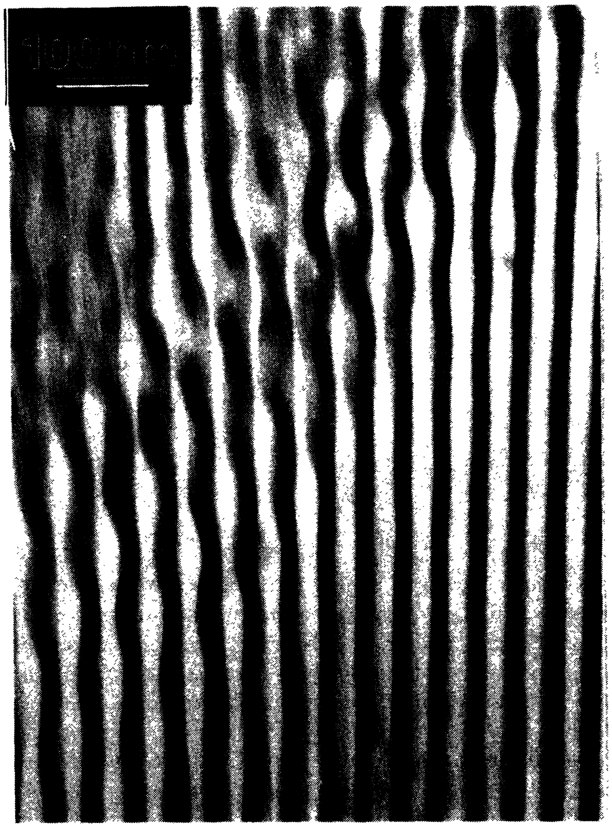

FIG. 2. General TEM view of the transition between oscillatory and nonoscillatory lamellar domains. White and dark lamellas correspond to the Al and Al_2Cu phases, respectively ($U_\infty = 0.10$, $V = 4.8$ cm/s).

FIG. 3. Enlarged TEM view of a well-developed domain of oscillations ($U_\infty = 0.10$, $V = 5.2$ cm/s).

lamellas has been observed recently in a transparent organic system.[12]

The boundary layer of excess composition ahead of the growing interface present at off-eutectic composition[6–8] preferentially favours the growth of one solid phase. At hypoeutectic concentrations in the Al-Cu system this phase is the Al phase. The oscillatory phenomenon has been understood theoretically to be governed by this favored phase. This understanding is now confirmed experimentally as shown clearly in Fig. 3. The almost parallel flat sides of lamellas at minimum amplitude appearing in this photograph are a finite amplitude signature of the oscillatory morphology. This departure from purely sinusoidal oscillations is in good agreement with the results of numerical simulations as shown in Fig. 1.

The relation between the interlamellar spacing and growth rate follows approximately the $\lambda^2 V = \text{const}$ relationship[1] as shown in Fig. 4. This relationship which describes steady-state lamellar structures remains valid for the observed nonsteady-state oscillatory lamellar structures. The $\lambda^2 V = \text{const}$ value decreases with increasing U_∞ as observed previously by Jordan and Hunt.[13] The scatter observed for $V < 3$ cm/s in Fig. 4 is due to melt pool instabilities inherent in surface melting and resolidification techniques.[14] The oscillatory phenomenon is observed to exist over a finite range of velocity as shown in Fig. 4. The minimum velocity for the appearance of oscillations is close to $V = 7$ cm/s for $U_\infty = 0.04$ and for the two other alloys, this limit cannot be determined experimentally because of melt pool instabilities. At high velocity the oscillatory phenomenon becomes perturbed by the appearance of other morphologies: a cellular-dendritic structure for $U_\infty = 0.16$ and for the two other alloys a wavy eutectic morphology similar to that observed previously in Al-Cu alloy at $U_\infty = 0$.[11]

The concentration range over which the oscillatory phenomenon exists appears to be close to the range studied experimentally. For $U_\infty = 0$ no oscillations are observed. As U_∞ increases, the amplitude ϵ of oscillations increases as shown in Fig. 5. For the largest value of U_∞

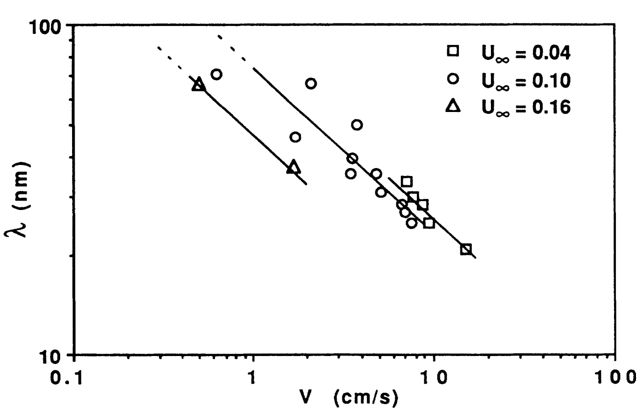

FIG. 4. Interlamellar spacing λ as a function of velocity V for the three hypoeutectic alloys. The straight lines correspond to the $\lambda^2 V = \text{const}$ relationship with const=23, 56, and 69 $\mu m^3/s$ for $U_\infty = 0.16$, 0.10, and 0.04, respectively.

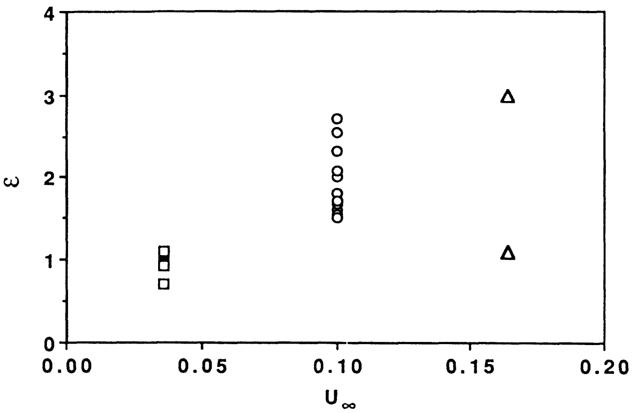

FIG. 5. Amplitude ϵ of oscillations as a function of U_∞ for the values given in Table I.

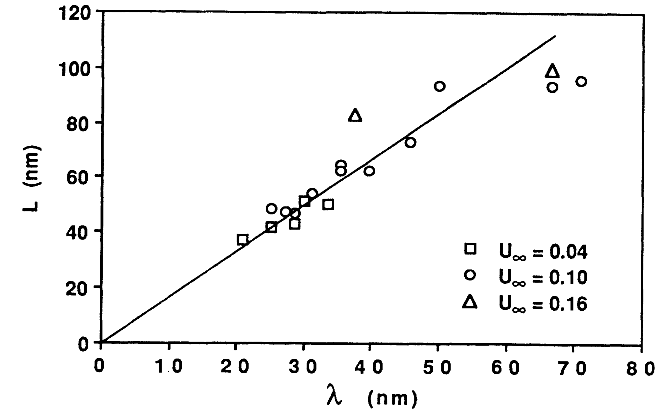

FIG. 6. Half wavelength of oscillation L as a function of λ. The slope of the line gives the value of π/Ω ($\Omega=1.9\pm0.2$).

($U_\infty=0.16$) the oscillations of lamellas become less correlated in the transverse direction. It should be noted that oscillations are observed for values of U_∞ much lower than the threshold value $U_\infty=0.21$ predicted by linear stability analysis.[6] This threshold value was calculated in Ref. 6 for a symmetrical phase diagram which represents a good approximation to the experimental situation encountered here. In addition, all theoretical investigations of oscillations to date have been restricted to two-dimensional situations and to small Péclet P number limit ($P=\lambda V/2D$, where D is the diffusion coefficient of solute in liquid).[6-8] In this experiment, values of P range between 0.05 and 0.5. A preliminary three-dimensional experimental study shows a relatively stable lamellar structure in transverse sections. This indicates that two-dimensional modeling should provide a good description of the oscillatory phenomenon and points out the necessity of longitudinal sections to observe this phenomenon.

The frequency Ω given in Table I was observed to remain reasonably constant over the whole range of compositions and velocities. This can be seen in Fig. 6 where the experimental values for the half wavelength of oscillation L vs λ fall on a straight line (by definition the slope of this line is equal to π/Ω). The mean value of Ω is 1.9 ± 0.2, in good agreement with the value of $\Omega=1.93$ found in numerical simulation for $U_\infty=0.17$ in Ref. 8 (see Fig. 1).

We propose a mechanism that relates the frequency Ω to the motion of the boundaries between the oscillatory and nonoscillatory lamellar domains described above. A steady-state lamella begins to oscillate when the neighboring oscillating lamella reaches its maximum amplitude as can be seen in Fig. 2. Therefore the angle θ between a boundary line and the growth direction is related to the frequency of the oscillation via the relation $\Omega=\pi\cdot\tan\theta$. With the mean value Ω found experimentally ($\Omega=1.9\pm0.2$), this gives $\theta=31.2°\pm2.6°$ which is in reasonable agreement with the experimental value of 35°. Also, the observation that both Ω and θ remain constant over the whole range of composition and velocity studied is consistent with the fact that both quantities are directly related.

In conclusion, the breakdown of steady-state lamellar eutectic structure at off-eutectic compositions is not necessarily associated with the appearance of dendrites of the growth governing phase. Instead, in the metallic alloy system studied here, the destabilization is first caused by the occurrence of an oscillatory morphology which persists over a wide range of compositions and growth rates. As this morphology has already been observed in organic systems, it should be evident in all eutectic systems and describes both further theoretical and experimental investigations.

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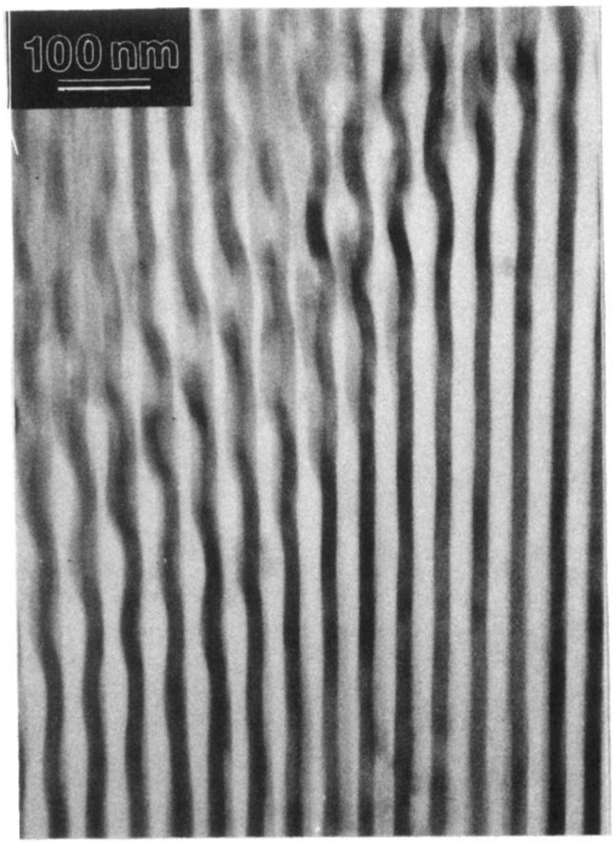

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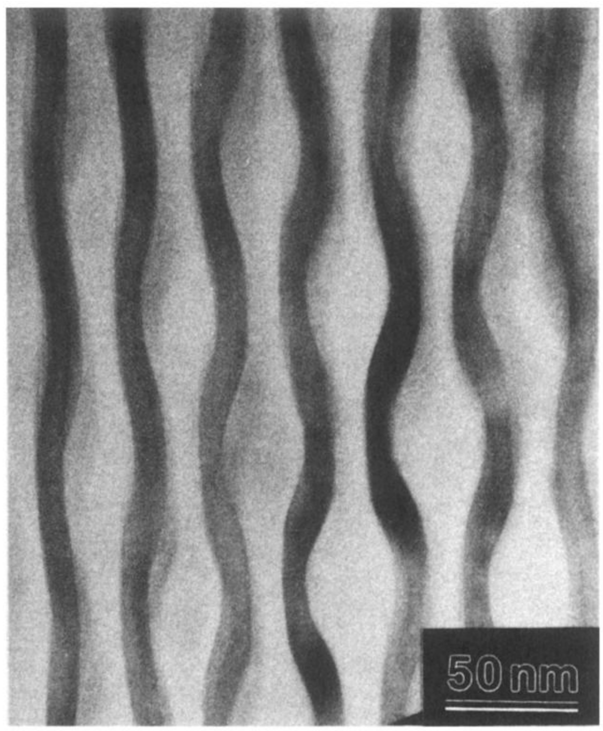

FIG. 3. Enlarged TEM view of a well-developed domain of oscillations ($U_\infty = 0.10$, $V = 5.2$ cm/s).