Generalized length scales for three-dimensional dendritic growth

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The crucial length scale of dendritic growth is the tip radius. Usually it is determined by fitting the data to a theoretical function. We present a method of a generalized tip radius which is entirely based on geometric considerations and is not dependent on an underlying assumption of the shape of the tip. Furthermore the results are stable and the average change of the tip radius between successive images is less than 6%.

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Pattern formation is of great interest in materials science as well as in theoretical physics as it is found everywhere in nature [1]. The growth of a crystal into a supercooled melt may lead to dendritic patterns, which are considered to be archetypes for pattern formation far from equilibrium starting from initially homogeneous conditions. A typical result of such a process is the microscopic structure of cast metals. There is great theoretical and technical interest in modeling these processes. Usually models are based on certain assumptions, and therefore it is necessary to verify models by comparing them with experiments. The tip radius R_{tip} is the most important intrinsic length scale of dendrites; therefore, it is a suitable quantity for such a comparison. In this paper we will show why procedures used in the literature to determine R_{tip} might lead to results which are difficult to compare. We propose a procedure to determine R_{tip}, which can be applied easily and is independent of any fitting procedures.

A three-dimensional dendrite as observed in experiments is shown in Fig. 1. Details of an experimental setup to observe the development of dendritic structures *in situ* are given in Ref. [2]. The contour, as extracted from such an image [3], is the basic information to analyze the shape of a dendrite.

A convenient measure to characterize the tip and its behavior is the tip radius, which is defined as

$$R = \frac{1}{\kappa} = \frac{(1+f'^2)^{3/2}}{f''}, \tag{1}$$

where R is the radius of curvature and κ the curvature for a given analytical function $f(x)$ describing the tip in a two-dimensional projection of the dendrite. f' and f'' denote the first and second derivatives, respectively. It is important to note that a radius of curvature is only defined if both f' and f'' are continuous and f'' is nonzero in the point of interest. In case of a parabolic solution $y=f(x)=ax^2$ in two dimensions it is found that

$$R_{tip} = R|_{x=0} = \frac{1}{2a}. \tag{2}$$

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Brener and Temkin [4] however have shown analytically that the intermediate region behind the tip of the dendrite, where no side branches grow yet, is not described by a parabola but rather by a hyperbolic function

$$y = f(x) = a|x|^b \quad \text{with} \quad b = \frac{5}{3}. \tag{3}$$

In the general case $y=f(x)=a|x|^b$ the radius of curvature is given by

$$R|_{x\to 0} = \frac{1}{b(b-1)a}(1+a^2b^2x^{2b-2})^{3/2}x^{2-b}. \tag{4}$$

The second derivative of $f(x)$ is diverging in $x=0$ for $b \neq 2$. Notice that for the determination of the tip radius the case $b=2$ is special:

$$R_b|_{x=0} = \begin{cases} 0, & 1<b<2 \\ \frac{1}{2a}, & b=2 \\ \infty, & b>2. \end{cases} \tag{5}$$

Obviously the tip radius is not defined for $b=\frac{5}{3}$. In the range $y<2R$, it was found in Ref. [2] that a parabola, a power law,

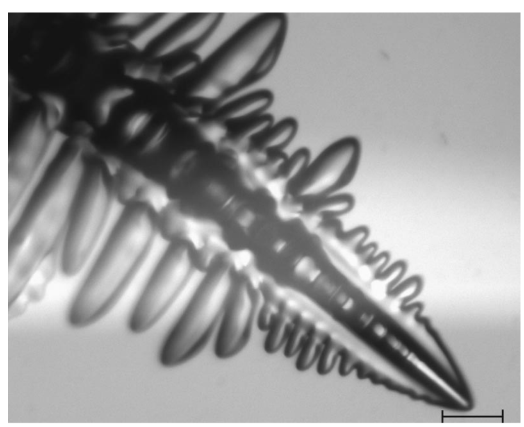

FIG. 1. Typical dendritic growth of a xenon crystal. The crystal is oriented in such a way that the maximal projection area is visible. The other two fins developed by the fourfold symmetry grow perpendicular to the focusing plane. The scale bar corresponds to 250 μm.

and a fourth-order polynomial could fit the tip of xenon dendrites equally well. However, for $y > 2R$ the power law has been found to fit the shape of the dendrite over a range of more than 600 μm, much better than a parabola or a fourth-order polynomial. The calculations of Brener and Temkin have been performed for a nonaxisymmetric dendrite. In Fig. 1 a dendrite is shown where the fins reach down to the dendrite tip as an example of a nonaxisymmetric dendrite tip. For dendrites with a somewhat more axisymmetric tip such as the ones found for succinonitrile (SCN) this fit may be less appropriate. Such dendrites have been studied in Refs. [5,6].

First attempts to characterize dendritic structures were based on the studies of Papapetrou [7] and Ivantsov [8], who found a rotational paraboloid to be a steady state solution of the diffusion problem of the latent heat. Fitting the two-dimensional projection of a dendrite with a parabola $y = ax^2$, Hürlimann *et al.* [9] realized that the fitting parameter a depends on the fitting height h and used an extrapolation to the fitting height $h \to 0$ to define the tip radius $R_0 = \lim_{h\to 0} R_{parab}(h)$. Dougherty and Gollub [10], on the other hand, used a fixed, arbitrary height $y = 3R$ for fitting. LaCombe *et al.* [5] used fourth-order polynomials to describe the shape of a dendrite. Finally Bisang [2] fitted the shape of a dendrite by a $y \propto x^{1.67}$ power law and found gratifying agreement with the findings of Brener [4]. This short historical overview shows that the experimental determination of the tip radius differs significantly. A similar overview can be found in Ref. [6].

It should be noted that depending on the underlying analytical assumption of the shape of the tip an exact determination of the tip radius is not possible (as shown above). The exception is the parabola, and at first sight the approach of Hürlimann *et al.* [9] seems quite reasonable to reduce arbitrariness. However, we will show that depending on the choice of the extrapolation a huge regime of different R_0 can be obtained. Let us assume that Bisang's finding of a description of the tip radius with Brener's model holds even for heights very close to the dendrite tip: $f(x) = ax^{5/3}$. For a given height h we will now fit this function with a parabola $f_2(x) = cx^2$:

$$\int_0^l (ax^{5/3} - cx^2)^2 \mathrm{d}x \overset{!}{=} \min, \tag{6}$$

where l is defined by $h = f(l) = al^{5/3}$. After a short calculation it is found that the tip radius follows the relation

$$R(h) = \frac{7}{15} \frac{\left(\frac{h}{a}\right)^{1/5}}{a}, \tag{7}$$

thus leading to an extrapolation $\lim_{h\to 0} R(h) = 0$ for an analytical function. As limits are never reached when dealing with experimental data usually an interval Δh is chosen where the slope of $R(h)$ does not change "significantly" and then is fitted with a straight line which will in 0 determine the approximated extrapolated value of the tip radius R_{0Exp}.

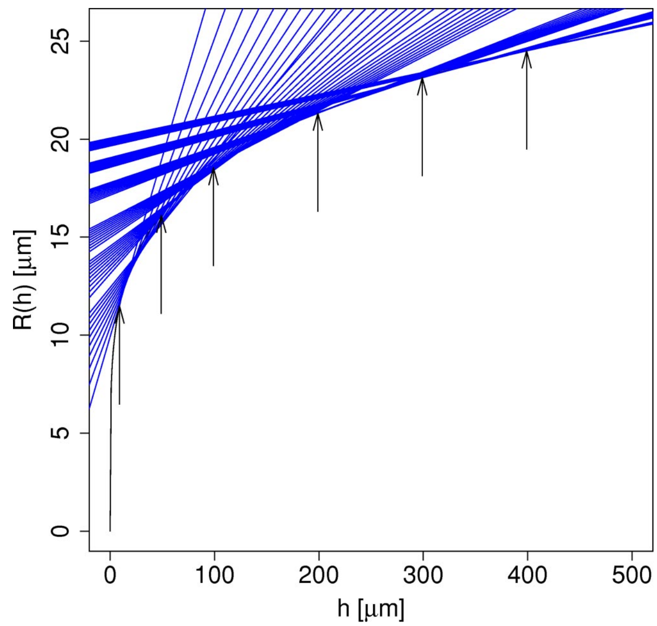

FIG. 2. Experimental determination of the tip radius by extrapolating the value R_0. Different fits for several Δh (starting at every arrow) are plotted. A huge domain of different R_0 can be found.

This is depicted in Fig. 2. Different fits for several Δh starting at each arrow are plotted. As can be easily seen, a huge domain of different extrapolated values for R_0 can be found. Therefore, this way of determination is not favorable.

Karma *et al.* [6], noticing the problem of the determination of the tip radius as well, presented an integral method of finding R_{tip} of three-dimensional simulated dendrites by integrating cross sections perpendicular to the axis of growth very close to the tip and determining the tip radius by the calculated areas. However, it should not be neglected that at least in certain cases integral methods might hide relevant and system dependent information, for example in the temporal evolution of the tip radius, as integral methods tend to smooth out noise or possible small oscillations.

We think that the tip radius, being such an important value, should be determined in a way that is independent of analytical assumptions of the shape of the tip and is only based on geometrical properties. Moreover the method should be resistant against pixelization errors and should not change significantly on successive images unless there is a physical reason for this behavior (such as tip splitting for example, where the tip radius increases before splitting [11]).

In Ref. [2] it was shown, at least for xenon dendrites, that although higher order polynomials ($n = 3, \ldots, 7$) approximate the tip shape better than a parabola, a dependence on the fitting height is found which cannot be neglected. For polynomials with $n > 7$ the fits became numerically unstable. Additionally the tip radius shows a dependence on the order of the fitting polynomial and no order of the polynomials that would best fit the contour could be found. Therefore it was concluded that the tip shape of xenon dendrites could not be described by a parabola or low-order polynomials in a well-defined and reproducible way. In Ref. [5], on the other hand, it was found that a fourth-order polynomial $y = -r^2/2 - Q(\phi) r^4$, where ϕ corresponds to the deviation from the

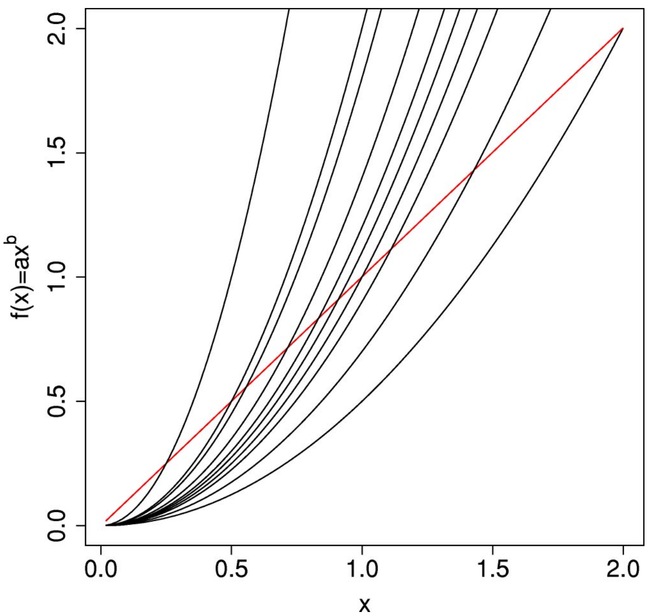

FIG. 3. Generalized length scale defined as half of the distance of the point of intersection between the curve [in this case $y=f(x)=ax^b$] with the line $y=x$. Here the parameter b goes from 0.7 to 4.0. This scale can be applied to any experimental data independently of an underlying assumption of the analytical shape.

maximal projection area, was sufficient to describe the tip shape of SCN dendrites. Indeed, the coefficients of such a presumed polynomial have a physical meaning. However, in order to introduce a certain uniformity into the measurements of different substances we propose a more model independent determination of the tip shape.

It is our aim to motivate the generalization by another geometrical interpretation of the tip radius $R=1/2a$ in the parabolic case. Let us consider the function $y=f(x)=ax^2$, having its tip in the origin. The length $R=1/2a=\frac{1}{2}(1/a)$ is exactly $\frac{1}{2}$ of the distance x, where $f(x)$ intersects the function $y=x$:

$$ax^2=x \Rightarrow x=\frac{1}{a}. \tag{8}$$

We therefore define the generalized scaling length as $\frac{1}{2}$ of the distance where a convex function [for example $y=f(x)=ax^b$ for Brener's model] meets $y=x$:

$$ax^b=x \Rightarrow x=\left(\frac{1}{a}\right)^{1/(b-1)} \tag{9}$$

and

$$L=\frac{1}{2}\left(\frac{1}{a}\right)^{1/(b-1)}, \tag{10}$$

recovering in the case of $b=2$ the parabolic tip radius. The behavior for different parameters b is shown in Fig. 3. In the case of dendritic growth with $b=\frac{5}{3}$ we find $L=\frac{1}{2}(1/a)^{3/2}$.

Scaling dendrites by this length scale ($\tilde{x}=x/L$, $\tilde{y}=y/L$) will result in

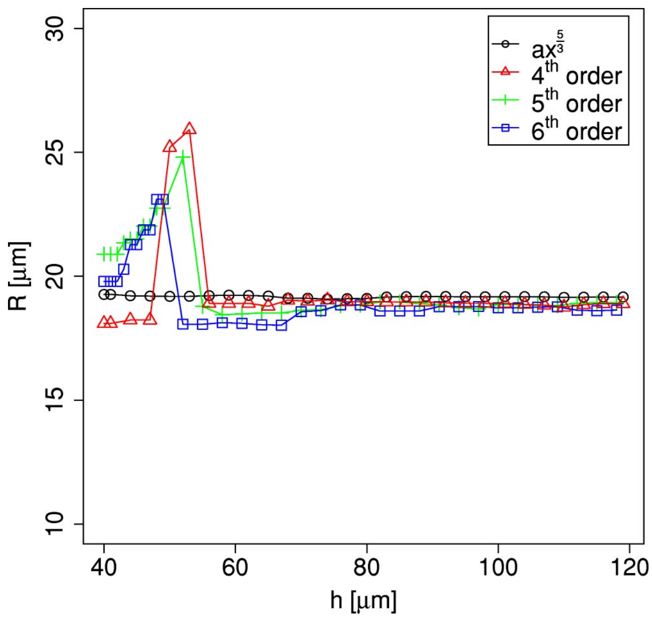

FIG. 4. Generalized tip radius for different fitting heights and different fitting methods $f_1 \ldots f_4$. The variance of the tip radius is less than 1.5% of the average tip radius.

$$L\tilde{y}=a(L\tilde{x})^b \tag{11}$$

$$\tilde{y}=L^{b-1}a\tilde{x}^b=a\left(\frac{1}{2}\right)^{b-1}\left[\left(\frac{1}{a}\right)^{1/(b-1)}\right]^{b-1}\tilde{x}^b=\left(\frac{1}{2}\right)^{b-1}\tilde{x}^b. \tag{12}$$

The prefactor $c=(\frac{1}{2})^{b-1}$ depends only on b and is therefore universal for all shapes with the same b. By introduction of the length scale L it is thus possible to define a valid scale for all $b>1$.

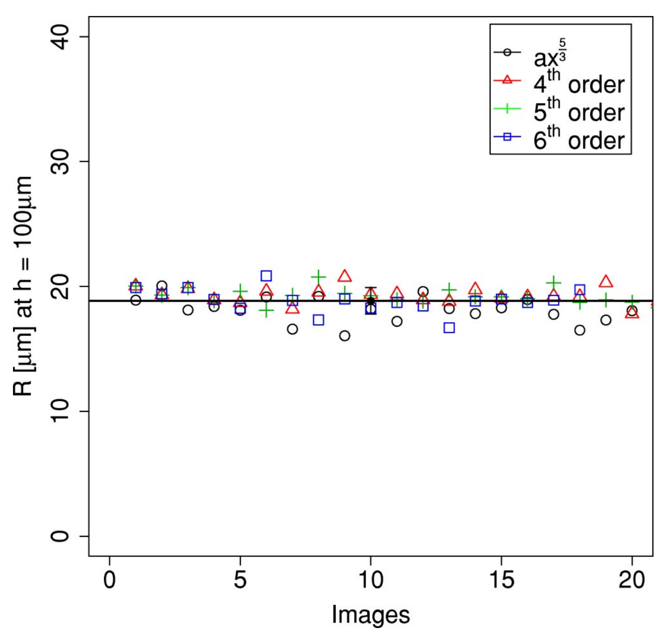

FIG. 5. Generalized tip radius for successive images at a given fitting height $h=5L$ for the different fitting functions $f_1 \ldots f_4$. The variance of the tip radius is less than 6% of the average tip radius.

Indeed the presented method is independent of any underlying analytical assumptions of the shape. The presented determination can be easily used for simulations and analytical functions as well as for experimental data. We would like to emphasize that this generalized tip radius is derived from the tip radius for a parabola and corresponds only in the parabolic case to the actual radius of curvature. A circle for example described by $y=r-\sqrt{r^2-x^2}$ having a radius of curvature of $R_{curv}=r$ would have a generalized tip radius of $R_{tip}=\frac{1}{2}r$.

It should be noted that experimental data are usually not smooth at all due to measurement errors or pixelization. If the method is applied directly to the pixels huge errors must be taken into account. We therefore suggest fitting the experimental data by a sufficiently high order polynomial (4–6) in order to receive a least square analytical description of the data. These coefficients of the polynomial as such are not physically relevant as they serve only to accurately describe the data. A subsequent intersection with $y=x$ is very easy to perform.

In order to test the independence of analytical assumptions we have used different fitting methods $f_1=ax^{5/3}$, $f_{2/3/4}=\mathrm{pol}(4,5,6)$, where $\mathrm{pol}(n)$ means nth order polynomial. However, we did not use a simple parabola fit as Bisang found very good agreement for Brener's $y=ax^{5/3}$ dependence and found that a parabola fit does not adequately describe the shape of our dendritic tips. We have found that independent of the functions $f_1 \ldots f_4$ the relative error for different heights $h=3 \ldots 5L$ is below 1.5% and for a fixed height $h=5L$ the relative error between two successive images is less than 6%. These errors are the worst case we have encountered. We have also found sequences showing 0.5% for different heights and also 0.5% for successive images. A plot for different heights is given in Fig. 4. The heights are plotted up to a critical height, where the side branches start to be visible. The behavior for successive images is shown in Fig. 5.

We have presented a method which determines the tip radius for any given experimental structure without any assumption about the analytical description of the data. It was shown that another reasonable scheme for determining the tip radius $R_0=\lim_{h\to 0}R_{parab}(h)$ is not converging and should therefore not be used. Fitting the dendrite tip by a fourth order polynomial does not eliminate the fitting height problem, although it approximates the tip shape much better than a parabola. We have proved that with the presented method the variance for the tip radius of fits with different fitting heights applied to one image is less than 1.5% and the relative error of the tip radius between successive images is less than 6%.

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