

ON A STATISTICAL THEORY OF CRITICAL EVENTS IN MICROSTRUCTURAL EVOLUTION

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Abstract. One of the most challenging aspects of the microstructural evolution in polycrystalline materials is to understand the role of topological reconfigurations during coarsening. In this paper, we study these critical events in a one-dimensional grain-boundary system and a stochastic framework for modeling texture evolution. The model is based on a master equation derived from numerically determined statistical properties of the system.

Keywords. Grain boundary character, Coarsening, Texture, Continuous time random walk.

1. Introduction. Most technologically useful materials arise as polycrystalline microstructures, composed of a myriad of small crystallites, termed grains, separated by interfaces, called grain boundaries. The energetics and connectivity of the network of boundaries are implicated in many properties across wide scales, for example, functional properties, like conductivity in microprocessors, and lifetime properties, like fracture toughness in structures.

A central problem is to engineer a microstructure to attain a desired set of material characteristics. It likewise presents many challenges for mathematical modeling, simulation, and analysis. Historical emphasis here has been on the geometry, or more exactly, on statistics of simple geometric features of experimental and simulated polycrystalline networks, like grain area. We are now turning our attention to texture, the mesoscopic description of arrangement and properties of the network described in terms of both crystallography and geometry.

In recent years, we have witnessed a changing paradigm in the materials laboratory with the introduction of automated data acquisition technologies. This has permitted the collection of statistics on a vast scale and its use to optimize aspects of material behavior. There are situations, for example, where it is possible to quantify the amount of alignment or misalignment sufficient to produce a corrosion resistant microstructure [1]. To rise beyond this level of anecdotal observation, the thermodynamics of the material system must be related to texture and texture related properties. Said in a different way, are there any texture related distributions which are material properties? Some geometric features of the configuration, like relative area statistics have these properties in the sense that they are robust but they are not strongly related to energetics. Recent work has provided us with a new statistic, the grain boundary character distribution, which has enormous promise in this direction. The grain boundary character distribution is a measure of relative amount of grain boundaries with a given net misorientation. Owing to our new ability to simulate the

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evolution of large scale systems, we have been able to show that this statistic is robust and, in elementary cases, easily correlated to the grain boundary energy [2]–[4].

However, the mechanisms by which the robust distributions develop from an initial population are not yet understood. As a polycrystalline configuration coarsens, facets are interchanged, some grains grow larger, and other grains disappear. We refer to these topological rearrangements as critical events. Such critical events contribute to the evolution of both the relative area and the grain boundary character distribution via the motion of the triple junctions, with low energy boundaries sweeping out those with higher energy. Further, when triple junctions collide, new boundaries are created.

The regular evolution of the network of grain boundaries in two dimensions is governed by the Mullins equations of curvature-driven growth, supplemented by the Herring condition of force balance at triple junctions—a system of parabolic equations with natural boundary conditions [5]–[9]. (For the higher dimensional formulation of capillary driven growth, see [10].) A main feature, first observed in [9] is that the nonlinear system and boundary conditions satisfy complementing conditions [8]. When applied to a single evolving n -sided grain with constant grain boundary energy, this mechanism leads to a Mullins-von Neumann $n - 6$ rule [11]—the rate of change of the area of the grain is proportional to $n - 6$, i.e.,

$$\frac{dA_n}{dt} = \gamma(n - 6) \text{ where } A_n \text{ is the area of an } n\text{-sided grain,} \quad (1.1)$$

and $\gamma > 0$ is some material constant (MacPherson and Srolovitz [6] have given, very recently, higher dimensional generalizations of the $n - 6$ rule). In particular, grains with 3, 4 or 5 sides decrease in area. When averaged over a population of grains, equation (1.1) results in

$$\frac{d\bar{A}_n}{dt} = \gamma(n - 6) \text{ where } \bar{A}_n \text{ is the average area of } n\text{-sided grains.} \quad (1.2)$$

Inspection of Fig. 1.1 shows that, contrary to (1.2), the average area of five-sided grains in a columnar aluminum structure increases several fold over the course of an annealing experiment. The $n - 6$ -rule does not fail for the continuous changes of boundary positions, but most of the five-sided grains we observe at time $t = 2$ hours had 6, 7, 8, ... sides at some earlier time $t < 2$ hours. Thus in the network setting, the critical events grain deletion and side interchange play a major role.

A significant difficulty in developing a theory of the grain boundary character distribution lies in the lack of understanding of these stochastic events associated with coarsening. As a motivation for such theory consider the following, highly over-simplified example. Let α_t be a stochastic process in which low energy “boundaries” grow at the expense of those with higher energy and new boundaries are created and disappear at random

$$d\alpha = -\gamma'(\alpha)dt + \epsilon dB_t. \quad (1.3)$$

Here α represents the difference of “orientations” on either side of the grain boundary, $\gamma(\alpha)$ represents the “energy” of the boundary, and ϵ is related to the rate at which the new grain boundaries enter the system. The stationary distribution for α is given by the Boltzmann-like expression

$$\rho(\alpha) = \frac{1}{Z} e^{-\frac{2\gamma(\alpha)}{\epsilon^2}},$$

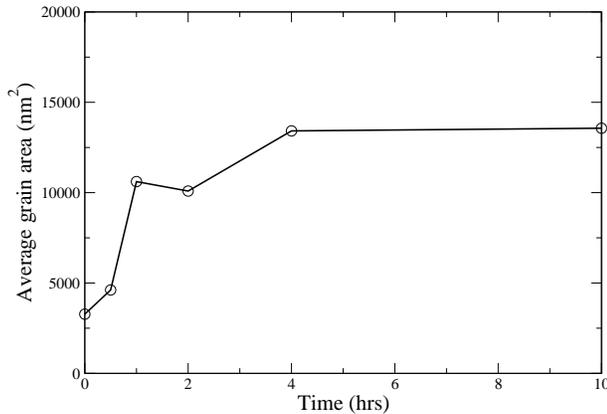


FIG. 1.1. *Average area of five-sided grains in an Al columnar structure.*

that can be found by solving the stationary Fokker-Planck equation for (1.3). Note that the maxima of this distribution function correspond to the minima of the energy—this is an essential feature of a grain boundary character distribution [2]-[4].

There are several stochastic approaches one might adopt to develop a more realistic model of the grain boundary character distribution. A framework based on statistical mechanics is adopted in [12], where we constructed a Boltzmann-type equation modeling grain interaction that successfully reproduces simulation data on a long timescale and has a good potential for generalization to higher dimensions.

In [13]-[14] we conjectured the fractional nature of the grain boundary kinetics and proposed a unified approach to model it in terms of a fractional master equation. We tested our theoretical ideas on the one-dimensional model by identifying the set of stable statistics and confirming the subdiffusive nature of the underlying kinetics for a prolonged transient regime exhibited by the system. Further, we demonstrated a reasonable agreement between the statistics obtained by direct simulation and the solution of the generalized master equation developed through the continuous time random walk (CTRW) theory.

Through our investigations, however, it became apparent that, while some stages of the evolution of one-dimensional system can be described using the formalism of the fractional CTRW theory, a complete understanding of the observed dynamics may require a new, more general stochastic framework. The main difficulty here is that which framework is appropriate is not known in advance and, in fact, it may not be available altogether.

In order to either develop a new or to select an existing theory, the statistical characteristics of the process need to be determined through an experiment. Given a deterministic mathematical model, the experiments can be conducted numerically with an advantage over physical experiments being that a large number of trials can be conducted in a relatively short time. Here we demonstrate the applicability of this approach to grain boundary evolution by developing a stochastic model for the times of grain boundary disappearance events. A generalization of our ideas to the entire grain boundary kinetics is a subject of a forthcoming publication.

Our main goal is to determine the equation governing the evolution of the arrival rate—probability that a critical event will be observed at a given time—based on numerically determined statistical properties of a large deterministic system. We study a simplified one-dimensional model (Section 2) that we introduced in [12] exhibiting the main features of the interacting grain boundary network. In particular, the model incorporates boundaries and junctions between boundaries moving under a form of gradient flow.

In Section 4 we develop a generalized arrivals master equation for independent, but not identically distributed time increments. We show that the independence assumption holds for the model one-dimensional system and find the probability density functions (pdfs) for waiting times between critical events. The waiting times are not identically distributed but rather are time-dependent scalings of an exponential distribution; this reflects the fact that the coarsening process slows down with time.

The arrival times master equation can be explicitly solved for the model system (Section 5) and we have found that arrival rates obtained both analytically and via the numerical experiment are in a close agreement. The form of the pdfs for waiting times suggests that, initially, the arrival rate corresponds to a stationary Poisson process while it can be approximated by a solution of a fractional ODE at later times.

2. Simplified model. Here we report on a model which emphasizes the role of critical events. For a precise description, fix $L > 0$ and consider the intervals $[x_i, x_{i+1}]$, $i = 0, \dots, n-1$ on the real line where $x_i \leq x_{i+1}$, $i = 0, \dots, n-1$ and $x_n = x_0 + L$. The locations of the endpoints x_i , $i = 0, \dots, n$ may vary in time and the total length L of all intervals remains fixed. For each interval $[x_i, x_{i+1}]$, $i = 0, \dots, n-1$, choose a number α_i from the set $\{\alpha_j\}_{j=1, \dots, n}$. The intervals $[x_i, x_{i+1}]$ correspond to grain boundaries and the points x_i represent to the triple junctions. The parameters $\{\alpha_i\}_{i=1, \dots, n}$ can be viewed as representing crystallographic orientations. The length of the i^{th} grain boundary is given by $l_i = x_{i+1} - x_i$.

For a non-negative energy density $\gamma(\alpha)$, we define the energy

$$E(t) = \sum \gamma(\alpha_i)(x_{i+1}(t) - x_i(t)) \quad (2.1)$$

and consider gradient flow dynamics characterized by the system of ordinary differential equations

$$\dot{x}_i = \gamma(\alpha_i) - \gamma(\alpha_{i-1}), \quad i = 1, \dots, n. \quad (2.2)$$

An important feature of the thermodynamics of grain growth is that it is dissipative for the energy during normal grain growth. The dynamics (2.2) has this property [13].

The parameter α_i is randomly prescribed for each grain boundary and does not change during its lifetime. The velocities of the grain boundaries can be computed from the relation

$$v_i = \dot{x}_{i+1} - \dot{x}_i = \gamma(\alpha_{i+1}) + \gamma(\alpha_{i-1}) - 2\gamma(\alpha_i) \quad (2.3)$$

Note that the velocities in the system remain constant between critical events corresponding to disappearances of individual grain boundaries. Every such critical event changes the statistical state of the model through its effect on the grain boundary velocities and therefore affects further evolution of the grains.

3. Stochastic properties of the system. The first step toward a mesoscopic model is the identification of stable statistics. The stable statistics determined from numerical experiments for a system of grain boundaries can be found in [13]. In particular, the analog of the grain boundary character approaches a stable distribution related to the energetics of the system (Fig. 3.1).

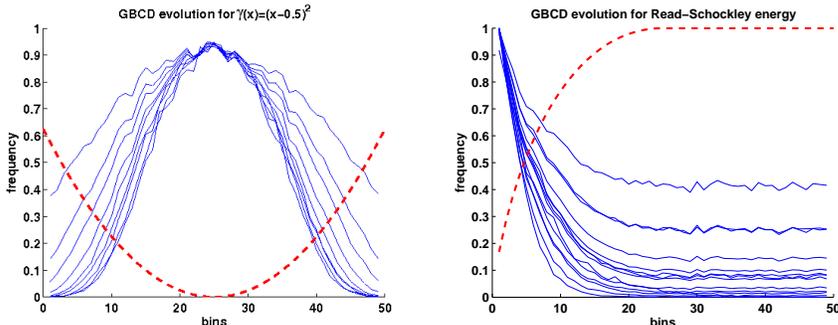


FIG. 3.1. Evolution of the grain boundary character distribution for various choices of γ .

Our goal in this paper is to describe the dynamics of critical events by understanding the stochastic properties of waiting times between these events. Fig. 3.3 shows typical distributions for waiting times. Note that, although the waiting times are close to being exponentially distributed, their means increase with time.

In what follows, we sometimes refer to each critical event as a simulation "step". Hence, unless there are coincident events, n boundaries disappear exactly after n steps.

Waiting times between successive critical events can be treated as random variables. To formulate a probabilistic theory, we need to establish the properties of these variables, e.g. their joint probability distribution functions (pdf's) etc. This task would be significantly simpler if we could assume that the waiting times are mutually independent so that it would be sufficient to determine marginal i -th waiting time distribution $w_i(t)$. In all tests that follow, we use data collected from 15000 runs with $n = 5000$ grain boundaries each. The runs are initialized with a random configuration of orientations and lengths and utilize the same energy functional $\gamma(x) = (x - 0.5)^2$.

Fig. 3.2 below provides the justification for the independence assumption. Fig. 3.2(a) shows the correlation coefficient between two successive waiting times T_i and T_{i-1} , where $1 \leq i \leq 4500$. Observe that this correlation does not grow with time. Fig. 3.2(b) depicts the 2000-th row of the correlation matrix. This picture is generic in the sense that an analogous plot is observed for an arbitrary $j \neq 2000$. Therefore, the matrix is close to being diagonal, and there is no significant correlation between the j -th and $k \neq j$ -th waiting times.

Next, we identify the pdfs $w_i(t)$ for the waiting times. The histograms of $w_i(t)$ for each 100-th arrival are plotted in Fig. 3.3 in log-log scale. The trend is clearly linear, showing that each probability density exhibits an exponential behavior. However, as shown in Fig. 3.3(b), the means of the exponential distributions increase with time, due to the slowing effect of the grain growth dynamics attributed to increase in the average grain boundary length. Hence we conclude that the waiting times distributions depend on the step number and are given by

$$w_i(t) = r_i \exp(-r_i t),$$

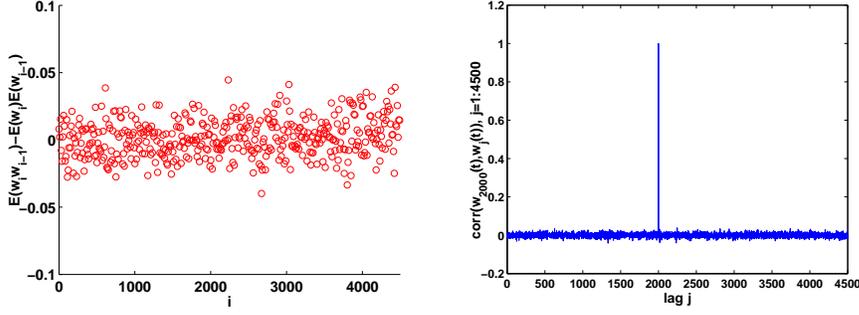


FIG. 3.2. (a) Correlation between successive waiting times as a function of jump number, showing no growth of dependence over time. (b) A j -th row of the correlation matrix with $j = 2000$, showing the absence of long-order correlations.

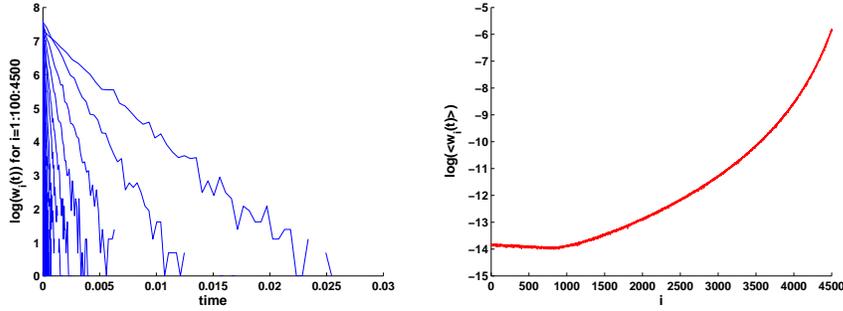


FIG. 3.3. (a) Double-log scale plot of the histograms for waiting times $w_i(t)$, revealing exponential behavior with decaying rates. (b) The mean of the waiting times distribution for the i -th waiting time $w_i(t)$ grows with time.

where r_i is a constant dependent on i . The dependence of r_i on i is essentially quartic, as demonstrated by the least-squares fit in Fig. 3.4. Hence $r_i \sim (N_\infty - i)^4$.

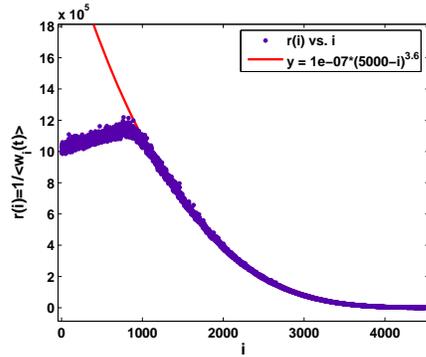


FIG. 3.4. Least squares fit for the exponential distribution parameters r_i .

4. Probabilistic derivation of the arrival rates master equation. We now derive the arrival rate master equation. Consider the sequence of times of critical

events during grain coarsening. These events can be described within the framework of a renewal process as long as times T_1, T_2, \dots between consecutive steps of the random walker are independent, identically distributed random variables. Here we consider a more general case, where T_i , $i = 1, 2, \dots$ are nonnegative, independent random variables that *may not be identically distributed*, that is each T_i is drawn from a distribution $w_i(t)$, $i = 1, 2, \dots$

Set $T(0) = 0$ and let

$$T(n) = \sum_{i=0}^n T_i \quad (4.1)$$

be the time of the n -th jump. Consider the random process

$$N(t) = \max\{n \geq 0 : T(n) \leq t\} \quad (4.2)$$

counting the number of jumps up to time t and denote

$$t_k = \sum_{i=1}^k T_i, \quad k \geq 1$$

to be renewal times of the process. Denote the probability that at least k arrivals have occurred prior to time t by

$$\Lambda_k(t) = \mathbb{P}(t_k \leq t), \quad (4.3)$$

and the corresponding density function by

$$\lambda_k(t) = \frac{d\Lambda_k(t)}{dt}. \quad (4.4)$$

Note that $\lambda_k(t)dt$ is the probability that the k -th arrival occurs during the time interval $[t, t + dt]$. The probability that an arbitrary arrival will be observed during the same time interval is given by

$$\lambda(t)dt = \mathbb{P}(\cup_k \{t_k \in [t, t + dt]\}) = \sum_{k=0}^{\infty} \lambda_k(t)dt, \quad (4.5)$$

where the last equality follows from independence of time increments. We call $\lambda(t)$ the arrival rate.

It is easy to see that while being a density of a sum of random variables T_i , $i = 1, \dots, k$, the density $\lambda_k(t)$ can be computed as a k -fold convolution of individual waiting time densities $w_i(t)$, $i = 1, \dots, k$, so that in the Laplace space

$$\hat{\lambda}_k(u) = \prod_{i=1}^k \hat{w}_i(u), \quad (4.6)$$

where by $\hat{f}(u)$ we denote a Laplace transform of a well-behaved function f .

We can construct the master equation based on the last jump as follows:

$$\begin{aligned} \lambda_{k+1}(t) &= \int_0^t \lambda_k(s) w_{k+1}(t-s) ds \\ \lambda(t) = \lambda_1(t) + \sum_{k=1}^{\infty} \lambda_{k+1}(t) &= w_1(t) + \sum_{k=1}^{\infty} \int_0^t \lambda_k(s) w_{k+1}(t-s) ds \\ &= w_1(t) + \int_0^t \lambda(s) W(s, t-s) ds, \end{aligned}$$

where

$$W(s, t-s) := \sum_{k=1}^{\infty} \frac{\lambda_k(s)}{\lambda(s)} w_{k+1}(t-s) \quad (4.7)$$

is the kernel describing the probability of a single jump between times s and t . Hence the arrival rates satisfy

$$\lambda(t) = w_1(t) + \int_0^t \lambda(s)W(s, t-s)ds, \quad (4.8)$$

which will be referred to as a generalized renewal equation from now on. In the case of i.i.d waiting times distributed according to a common law $w(t)$, we obtain a standard renewal equation

$$\lambda(t) = w(t) + \int_0^t \lambda(s)w(t-s)ds, \quad (4.9)$$

which, in turn, yields $\lambda(t) = r = \text{const}$ in the case of a regular Poisson process with the waiting times $w(t) = re^{-rt}$. Notice, however, that the behavior of the arrival rate is more complicated in the case of non-identically distributed variables. For instance, if $w_i(t) = r_i e^{-r_i t}$, by a simple but tedious calculation of k -fold convolutions we obtain

$$\lambda(t) = \sum_{k=1}^{\infty} \sum_{i=1}^k r_i e^{-r_i t} \prod_{i=1, i \neq j}^k \frac{r_j}{r_i - r_j}. \quad (4.10)$$

In order to avoid possible convergence issues and to reflect the fact that the number of events in the model systems is finite we will assume that the first summation in (4.10) is performed up to a large $m < \infty$.

5. Comparison between theory and numerics. Our numerical experiments show that the cumulative arrivals do not depend linearly on time. In Fig. 5.1, we plot the cumulative number of arrivals $N(t)$ (the number of critical events) before time t , and again observe the slowing effect comparing to what we expect from regular Poisson-type process (where $N(t)$ grows linearly with time). What is even more intriguing is that the arrival times match those obtained via the regular renewal equation (4.9) for the choice of $w(t) = 0.051 t^{-1.3}$ for $3500 \leq i \leq 4500$, as shown in Fig. 5.1. This suggests existence of an intermediate fractional diffusive regime in our model [13]. To study this more precisely, we need to identify if the fractional exponent $\beta = 0.3$ conforms with the arrival rates given above. Fig. 5.2(a) shows the log-log graph of the number of intervals in the system surviving at time t , $N_{\infty} - N(t)$, for the period from 1000 to 4500-th arrivals. This behavior conforms well with $N(t) \sim t$ in the beginning. However the dependence of $N_{\infty} - N(t)$ on t for the period between 3000th and 5000th arrivals (Fig. 5.2(a)) is given by

$$N_{\infty} - N(t) \sim t^{-\beta}, \text{ where } \beta \sim 0.3.$$

The dynamics of the process experiences a transition from one mode to another at some critical point t_{cr} in the simulation. At the same critical time t_{cr} the stabilization of relative distributions is observed. Note that, although by the time $t = t_{cr}$, almost half of the boundaries have disappeared, the absolute time elapsed from the onset of

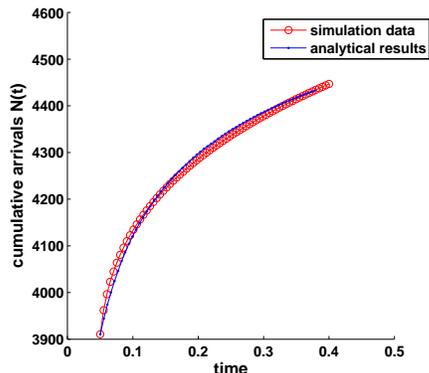


FIG. 5.1. Empirical cumulative arrivals at time t produced via simulation and analytically determined from the generalized fractional renewal equation (4.9) with $w(t) \sim t^{-1.3}$.

simulation remains minuscule (of the order of 10^{-4} sec for a 1 sec long simulation). The change in the behavior of the system can be attributed to "washing-out" of transients during the relaxation stage of coarsening. Note, however, that the "stable" regime corresponding to the stabilized distributions deviates significantly from regular diffusion, with $\beta = 0.3$, in contrast with the normal diffusion, where $\beta = 1$. Hence we have a case of an anomalous (sub)diffusion.

Further, note that the analytical solution of the generalized renewal equation (4.10) predicts both stationary Poisson and fractional behavior observed in our experiments. Indeed, there is a close match between the arrival rates at the intermediate stage of coarsening as can be seen in Fig. 5.2(b). Here, for the interval between $i = 3600$ and $i = 4500$, we compare the empirical arrival rate versus the solution of the renewal master equation (4.10). We approximate waiting times to match the least-squares fit in Fig. 3.4 by setting $w_i(t) = r_i \exp(-r_i t)$ with $r_i = 3.7 \cdot 10^{-9}(1400 - i)^4$. We compensate for the time elapsed before the 3600 arrival by translating the absolute time by $3.4 \cdot 10^{-2}$ —the mean of $\sum_{i=1}^{3600} T_i$. The plot shows a close agreement in the double-logarithmic scale, indicating that the generalized renewal equation captures the system dynamics in this interval.

If, on the other hand, we examine the expression $r_i \sim (N_\infty - i)^4$, we observe that $r_i \approx N_\infty^4$ for $i \ll N_\infty$. Hence, at its initial stages, the renewal process is close to a stationary Poisson process. The drawback of this is that the solution (4.10) of the generalized renewal equation is numerically ill-behaved for $i \ll 5000$. Having made these observations, it is clear that the jump process underlying the grain growth dynamics in the one-dimensional case is far from being simple. Not only it does not fit into the regular diffusion framework but, due to the decaying arrival rates, it also deviates from the more general framework of continuous time random walks. By means of the generalized renewal theory, we have been able to completely determine the arrival rates throughout the evolution. Moreover, we have shown that the model bears striking similarity to the fractional sub-diffusion in an intermediate regime where most relevant distributions stabilize. Similar observations can be made for the full process that takes into account jumps in orientation, as we will show in a forthcoming paper.

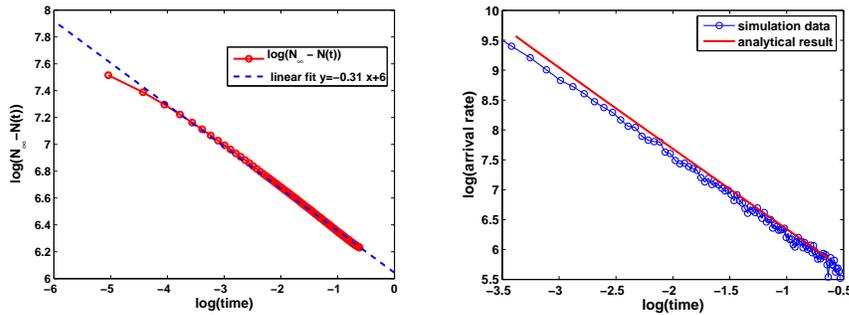


FIG. 5.2. (a) Least square power law fit for the number of surviving grain boundaries at time t . (b) Comparison of arrival rates in log-log scale obtained in simulations of the full system and analytically via equation (4.10).

6. Acknowledgments. The authors wish to thank their colleagues Eva Eggeling, Gregory Rohrer, and A. D. Rollett. Research supported by grants DMS 0405343 and DMR 0520425. DG acknowledges the support of DMS 0407361 and DK acknowledges the support of DMS 0305794.

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