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KINETICS OF THREE-DIMENSIONAL NORMAL GRAIN GROWTH*

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Kinetics of three-dimensional normal grain growth and related processes (*e.g.*, soap froth evolutions) described by the Mulheran–Harding model is studied. The model is represented by a diffusion equation with the grain–size–dependent diffusion coefficient. The equation is solved for an arbitrary initial distribution of grain sizes. It is proved that asymptotic kinetics do not depend on the initial state.

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1. Introduction

Throughout this study, we wish to reveal kinetic effects characteristic of the so-called normal grain growth [1, 2]. One may ask: Which are the basic (kinetic) signatures of the Normal Grain Growth (NGG) and why is the process of a certain importance? To answer at least the first question, one may start from a set of simple observations of the process of interest looking carefully at its temporal behavior. First, one can observe that the

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number of grains constituting the system decreases with time. Second, the average size of the grain increases with time. Another qualitative observation assigned to the NGG states that it passes quite smoothly over all stages that it meets on its way. One could say that the evolution is realized in a uniform manner. Moreover, it can naturally be described by zero or Dirichlet boundary conditions. Mathematically speaking, such conditions give no tolerance for existence of grains of zero as well as of infinite sizes. They assure the process as being realized in the so-called normal regime, with no re-nucleation as well as giants' domination events. Physically speaking, in turn, one may expect some abnormalities while more closely inspecting the growing conditions. On the level of a statistical-mechanical description of the phenomenon they are mostly subjected to some violation of either boundary conditions or when one is capable of keeping the system in the regime of preservation of its total volume [3,4].

To understand the driving mechanism(s) of the NGG, one is kindly sent to have a look at [5]. For the purpose of our description we would like to state that there are a few types of forces driving the system and/or damping its evolution. Among several types of forces driving the system let us mention a deterministic one, very much related to the surface tension effect (for bubbles, with a pressure difference) and named the capillary force. It is known [6] that this force may readily be accompanied by material density fluctuations present in the system. The forces that try to damp the system evolution are called the drag ("damping") forces, among which the Zener force, due to pinning, as well as the Mullins force, likely emerging in the high-temperature limit, can be mentioned [7]. They are, however, not taken into consideration in the present study. Therefore the presented study can be anticipated as some simplification of a real problem due to polycrystal evolution; for realizing the simplified mechanism of the growing process, see Fig. 1 and have also a look at a mini-review [8].

It is worth listing main types of a theoretical description of the threedimensional NGG kinetics, which is here of major interest. According to [2], these are the following:

- (i) the topological approach [9];
- (*ii*) the metric and combined approach, which may split into three subapproaches:
 - the grain growth as a deterministic process, described by a continuity equation [10];
 - the grain-diffusion approximation [11];
 - the stochastic description [12].



Fig. 1. Two snapshots representing a possible local scenario of the Normal Grain Growth taken at two successive time instants t_1 and t_2 , in which the content of the grain 1 has been absorbed by the grain 2, presumed that the grain 3 had to survive under such growing circumstances ($t_2 > t_1$). Some basic signatures of the process can be underscored: (*i*) neighboring crystallites 1, 2 and 3 pack entirely the available physical space; (*ii*) the process seems to be efficient (close-packing, but not of the Apollonian type, *cf.* Appendix) and its principal driving force appears to be the curvature-dependent capillary force; (*iii*) the grain boundaries undergo a strong fluctuation effect (see the corresponding double-arrows); (*iv*) the centers of inertia of the crystallites seem to undergo at most a short-range diffusional motion and the process keeps on growing toward some increase of the average grain radius.

There is a variety of papers that try to solve the basic kinetic problems related to the NGG phenomenon by means of computer simulation techniques, like [13,14], but their connections to the grain growth or evolution of polycrystals (bubbles) are unfortunately not very much established yet [15].

As seems to be known, the NGG processes can be described by their kinetics, which can be best seen while examining their asymptotic behavior though the obtained solutions are valid for the entire time domain. Thus, the goal of this paper is to reveal some characteristic kinetic features of the NGG, mostly by inspecting the temporal behavior of its main physical quantities like the number of grains constituting the system as well as their average radius [16].

The paper is organized as follows. In the next section we briefly recall the Mulheran–Harding (M–H) model¹, which stands for a random walk approximation of the NGG process, with some emphasis that the random walk is realized in the space of grain sizes. In Section 3, we provide a sketch of the method of a formal solution of the M–H model generally based upon the method of separation of variables. We also present explicitly the propagator of the system evolution, which contains a possibility of including arbitrary prescribed initial state f(v, 0). Then, in Section 4 we arrive at the most interesting asymptotics of the model which appears to be consistent with

¹ It would perhaps be equally well named the Mulheran–Harding–Louat model because Louat contributed first to this issue [10], and solved the problem in a one-dimensional case.

the M–H asymptotics, though they do not mention explicitly how do the asymptotics depend upon f(v, 0), if this is the case. We finish the paper by sending a final address and preparing a quite recent list of literature, also incorporating some links to applicational aspects of our study, *cf.* Appendix for having a wider outlook.

2. Mulheran–Harding model

In the M–H model it has been argued [10,11,16] that the grain size- and time-dependent evolution of the system satisfies the continuity equation of diffusion type, namely,

$$\frac{\partial}{\partial t}f(v,t) = -\frac{\partial}{\partial v}j(v,t) = D_0 \frac{\partial^2}{\partial v^2} v^{2/3} f(v,t), \quad v \in [0,\infty),$$
(1)

where v is a volume of a grain, D_0 is a constant representing a random walk behavior in grain growth (we choose $D_0 \equiv 1$ because it enters only into the product $D_0 t$ leading to the rescaling of time t), f(v, t) is the distribution function of grains (bubbles) at time t, *i.e.*, f(v, t)dv is a relative number of grains of sizes in the volume range [v, v + dv]. The flux j(v, t) takes the form

$$j(v,t) = -D_0 \frac{\partial}{\partial v} v^{2/3} f(v,t).$$
(2)

In this model it is assumed that the flux j(v, t) is proportional to the change of number of particles on the surface $v^{2/3}$ of the grain. Unfortunately, the microscopical justification of the M–H model does not exist. Nevertheless, it describes quite well kinetics of a certain class of grain-growth processes.

Eq. (1) is completed by the following conditions [10, 16]:

(a) The initial distribution of grains,

$$f(v,0) = f_0(v),$$
(3)

where $f_0(v)$ is a given function.

(b) The boundary conditions,

$$f(0,t) = f(\infty,t) = 0.$$
 (4)

They signify that the number of grains of zero volume as well as of infinite volume at arbitrary time is zero. The former means that the boundary v = 0 is absorbing. The latter is natural and minimal for existing statistical moments of the distribution f(v, t).

3. Propagator of evolution

A fundamental problem concerning the grain growth process is to determine the solution f(v, t) of (1) with the conditions (3) and (4). The relation between the distribution f(v, t) at the instant t and the distribution f(v, s)at the earlier instant s defines an evolution operator $\hat{U}(t, s)$ via the formal equality

$$f(v,t) = \hat{U}(t,s)f(v,s), \quad t \ge s.$$
(5)

The evolution operator $\hat{U}(t,s)$ is an integral operator given by the relation

$$f(v,t) = \int_{0}^{\infty} U(v,t|w,s)f(w,s)dw,$$
(6)

where U(v, t|w, s) is an integral kernel of the operator $\hat{U}(t, s)$. The function (or more precisely the distribution) U(v, t|w, s) is called a propagator and has the following properties

$$U(v,t|w,s) = \int_{0}^{\infty} U(v,t|z,\tau)U(z,\tau|w,s)dz, \quad t \ge \tau \ge s,$$
(7)

$$\lim_{t \to s} U(v, t|w, s) = \delta(v - w) \tag{8}$$

which follow from the equality (6). What we need is in fact the propagator for the particular time s, namely, for the initial time s = 0. Then (6) is given by

$$f(v,t) = \int_{0}^{\infty} U(v,t|w,0) f(w,0) dw$$
(9)

and it is sufficient for solving Eq. (1) with the initial condition (3).

To find the propagator we assume that

$$f(v,t) = \int_{0}^{\infty} e^{-\lambda t} \mathcal{G}_{\lambda}(v) d\lambda, \qquad (10)$$

where $\mathcal{G}_{\lambda}(v)$ is an unknown function of two variables λ and v. The representation (10) is the version of the method of separation of variables in the case when the spectrum λ of the diffusion operator in (1) is continuous.

From (1) and (10) it follows that $\mathcal{G}_{\lambda}(v)$ is a solution to the ordinary differential equation of the second order, namely,

$$v^{2/3}\mathcal{G}_{\lambda}''(v) + \frac{4}{3}v^{-1/3}\mathcal{G}_{\lambda}'(v) - \frac{2}{9}v^{-4/3}\mathcal{G}_{\lambda}(v) = -\lambda\mathcal{G}_{\lambda}(v), \qquad (11)$$

where the prime denotes a derivative with respect to v. Eq. (11) can be reduced to a simpler form by introducing the new variable

$$y = v^{2/3}$$
. (12)

Let us define a new function $F_{\lambda}(y)$ by the relation

$$F_{\lambda}(y) = \mathcal{G}_{\lambda}(v). \tag{13}$$

In consequence, Eq. (11) can be rewritten in terms of the new function $F_{\lambda}(y)$ as follows

$$y^{2}F_{\lambda}''(y) + \frac{3}{2}yF_{\lambda}'(y) + \left(\frac{9}{4}\lambda y^{2} - \frac{1}{2}\right)F_{\lambda}(y) = 0.$$
 (14)

It is the Bessel equation [17], the solutions of which are well known. In our case, the solution has the form [17]

$$F_{\lambda}(y) = \mathcal{G}_{\lambda}(v) = v^{-1/6}C(\lambda) J_{3/4}\left(\frac{3}{2}\sqrt{\lambda} v^{2/3}\right), \qquad (15)$$

where $J_{\nu}(x)$ stands for the Bessel function [18]. The 'constant' of integration $C(\lambda)$ is determined by the initial condition (3). Now, using the same method as in [4], one readily finds for the propagator the explicit result

$$U(v,t|w,0) = \frac{3v^{-1/6}w^{1/2}}{4t} \exp\left[-\frac{9(v^{4/3}+w^{4/3})}{16t}\right] I_{3/4}\left(\frac{9(vw)^{2/3}}{8t}\right), \ (16)$$

where $I_{\nu}(x)$ is the modified Bessel function [18]. Hence, Eq. (9) with the propagator (16) is a solution of (1) for an arbitrary initial distribution f(v, 0) of the grains. It is worth to note that the propagator (16) is not symmetric under the transformation $v \leftrightarrow w$. It is in clear contrast to the case considered in [4], where a slightly different model describing the phase transformation has been analyzed.

4. Analysis of solution

Some general properties of the grain growth process described by (1) can be obtained from the representation (9) of the solution of (1). Firstly, let us consider the zero-order moment, designated by N(t), namely

$$N(t) = \int_{0}^{\infty} f(v, t) dv$$
(17)

which is the relative number of grains at the instant t, cf. statements beneath Eq. (1). Inserting (9), completed by (16), into (17) and integrating it over the variable v yields

$$N(t) = \frac{4\beta^{3/4}(t)}{3\Gamma(3/4)} \int_{0}^{\infty} F(w,t)dw,$$
(18)

where

$$F(w,t) = w e^{-\beta(t)w^{4/3}} M\left(1;7/4;\beta(t)w^{4/3}\right) f(w,0)$$
(19)

and

$$\beta(t) = \frac{9}{16t}.\tag{20}$$

The function M(a; b; z) is the confluent hypergeometric (Kummer) function and $\Gamma(z)$ is the Euler gamma function [18].

Now, we prove that for long time, $t \gg 1$, the asymptotics

$$N(t) \sim t^{-3/4}$$
 (21)

holds, independently on the initial distribution f(v, 0) of the grains, which means, that some algebraic fall of the total number of grains (bubbles) is observed. Indeed, from the definition of the Kummer function [18] it follows that

$$M(1;7/4;z) \le M(1;1;z) \equiv e^z$$
 (22)

and therefore

$$F(w,t) \le w f(w,0) \tag{23}$$

for any time t. It enables us to exploit the Lebesgue's dominated convergence theorem [19], namely,

$$\lim_{t \to \infty} \left[\int_{0}^{\infty} F(w, t) dw \right] = \int_{0}^{\infty} \left[\lim_{t \to \infty} F(w, t) \right] dw.$$
(24)

Because $\lim_{t\to\infty} F(w,t)$ exists, the function N(t) behaves asymptotically as $N(t) \sim \beta^{3/4}(t) \sim t^{-3/4}$. It completes the proof.

The next important characteristics of the process is the average value of volume of the single grain. It is defined by the relation

$$\langle v(t) \rangle = \frac{\int_{0}^{\infty} v f(v,t) dv}{\int_{0}^{\infty} f(v,t) dv}.$$
(25)

Using (9) and (16), one can show by an explicit evaluation of the integrals that the *average* total volume V(t) of the system is preserved in the course of time, *i.e.*

$$V(t) = \int_{0}^{\infty} v f(v, t) dv = \int_{0}^{\infty} v f(v, 0) dv = V(0).$$
 (26)

In consequence, the average grain volume is closely related to evolution of the average number of grains,

$$\langle v(t) \rangle = V(0)N^{-1}(t), \qquad (27)$$

cf. (17) and (26), so that a quantitative volumetric "fragmentation" relation can be obtained this way. Because the average volume $\langle v(t) \rangle$ of the single grain is proportional to the third power² of its average radius $\langle r(t) \rangle$, therefore

$$\langle r(t) \rangle \sim \langle v(t) \rangle^{1/3} \sim N^{-1/3}(t).$$
 (28)

Independently of the initial conditions, the average radius of the grains displays at large times the power-law dependence, namely

$$\langle r(t) \rangle \sim t^{\mu}$$
 (29)

with the exponent $\mu = 1/4$.

From (18) and more explicitly from (21) it is seen that the average number of grains drops with time. We observe that, by integrating (1) over the phase space $v \in [0, \infty)$, the change of the grain number is related to the flux across the absorbing boundary at v = 0,

$$\frac{dN(t)}{dt} = j(0,t) < 0$$
(30)

 $^{^2}$ A discussion of considering the (average) grain as a non-Euclidean object of a certain fractal dimension d_f is left for the Appendix.

(the total flux at infinity $j(\infty, t) = 0$). So, there are less and less grains since infinitesimally small grains disappear attaching some bigger ones.

Now, let us present examples of the grain distributions. *E.g.*, let us assume that initially at t = 0 there are N_0 grains each of non-zero volume v_0 . Hence, the total volume of the system is $V(0) = v_0 N_0$ (see (26)) and the initial distribution f(v, 0) reads

$$f(v,0) = N_0 \delta(v - v_0) = \frac{V(0)}{v_0} \delta(v - v_0), \qquad (31)$$

where $\delta(x)$ denotes the Dirac delta distribution. In this case one gets

$$f(v,t) = f_1(v,t) = N_0 U(v,t|v_0,0)$$
(32)

and the solution of the problem (1)-(4) is expressed by the well-known functions.

In the papers [10, 11], only the particular solution of (1) has been presented. It reads

$$f(v,t) = f_2(v,t) \sim t^{-7/4} v^{1/3} \exp\left[\frac{-9v^{4/3}}{16t}\right].$$
 (33)

This solution can be obtained for a special initial distribution. We have guessed the function f(v, 0) which reads

$$f(v,0) = \frac{V(0)}{v}\delta(v), \qquad (34)$$

where V(0) is the average total volume of the system, cf. (26). Indeed. let us insert this distribution to (9), represent the modified Bessel function $I_{3/4}(z)$ in (16) as a power series with respect to the argument z [18] and use the definition of the Dirac delta distribution. Then one obtains (33). The initial condition (34) is rather a non-typical distribution. For it, the initial mean number of grains N(0) is infinite and the initial mean volume of the single grain $\langle v(0) \rangle$ is zero. On the other hand, the initial distribution (31) gives as a result a finite N(0) and a non-zero value of $\langle v(0) \rangle$. Moreover, the boundary condition (4) is fulfilled for (31) but not for (34). It implies that the value of the function $f_2(v,t)$ for v=0 suddenly jumps from infinity to zero at infinitesimally small time, *i.e.*, $f_2(v = 0, t = 0) = \infty$ and $f_2(v = 0, t = 0) = \infty$ (0, t > 0) = 0. It is inconsistent and unphysical. Nevertheless, from the mathematical point of view, the solution (33) is correct. The comparison of the distributions (32) and (33) is shown in Figs 2 and 3 at two various instants. The normalization constants of two distributions have been chosen in such a way that the average total volume (26) is the same in both cases. One can notice that in the case of the M–H initial condition, there are a greater number of smaller grains than for (32) and a smaller number of greater grains than for (32). It seems to be obvious because the mean volume of both systems is the same.



Fig. 2. The distribution function f(v) = f(v, t = 1) of grains for two various initial conditions: for the Mulheran-Harding initial distribution (34) and for the Dirac-delta distribution (31) with $v_0 = 1$. For both systems the mean total volume V(t) = V(0) = 100.



Fig. 3. Same as in Fig. 2 but taken at the later moment t = 1.5.

It turns out that the asymptotic kinetics (like $\mu = 1/4$ in (29)) obtained above differs distinctly from the standard or normal case (for which $\langle r(t) \rangle \propto t^{1/2}$ and which has been also got by Louat [10] for one-dimensional systems). It is also in disagreement with findings of computer simulations [14] for two-dimensional systems. It is not a surprise because exponents like μ depend on the dimension of the system as well as on mechanisms taken (or not taken) into account in modelling of the growth phenomena.

5. Final remarks

We have solved the Mulheran-Harding model for an arbitrary initial distribution of the grain sizes, see (9), (16), (18) and (26)–(28). It allows, in fact, to investigate the influence of the initial preparation of the system on its later evolution, *cf.* [20]. Concerning the very basics of the NGG kinetics of the three-dimensional system as well as soap froth³ evolution, one may state the following:

- (1) The process under study can be treated as the random walk in the space of grain sizes. However, the distribution function is not normal-ized as it does in the case of diffusion motion of the Brownian particle. In the case considered, the first statistical moment is conserved in time, see (26).
- (2) In the asymptotic (late times) regime, the evolution does not depend upon the prescribed initial state. It resembles the ergodic behavior of a class of standard stochastic processes.
- (3) In the non-asymptotic regime, one may expect some differences between various prescribed initial conditions (in the early times limit, for example), but further the process goes smoothly towards some visible increase of the average grain size [12].
- (4) If some conditions of the NGG, *i.e.* the constancy of average total volume or the prescribed boundary (Dirichlet) condition(s), are violated, the system is supposed to enter the so-called abnormal regime, *cf.* [1,7]. It is also known that the abnormality may be caused by, *e.g.*, preparing a special initial state of the grain growth process in stainless steels, when (indirectly) post-recrystallization strains effect on the microstructure as well as the grain misorientation texture, so that some incorporation of such a behavior would certainly make the modelling more advantageous as well as applicable to polycrystals [21].

Why is the kinetic study of some importance? We prefer to make a response to the posed question just by recalling one example. The example concerns the bumpers of cars, and one can think of the process of casting the bumper structure as being of NGG type (metallic soap froths), and one might imagine that the evolution will go up to the large times regime. But it may also be stopped somehow artificially in some earlier time domain, so that

 $^{^3}$ Notice that in our model no explicit notion of texture and/or crystallographic (mis)orientations, very characteristic of polycrystals but clearly absent in soap froths, has appeared.

a fine-grained structure will be a certain result thereof. Fine-graining typically implies that the structure can be mechanically more robust, whereas a structure containing bigger grains (bubbles), though perhaps not very tough (but rather brittle), may also be of use in another maybe equally important context, *cf.* [22]. Moreover, the reader is also encouraged to see the book by Okatsaki [23], where grain growth processes in ceramics (BaTiO₃) have been analyzed and where the obtained asymptotics conform very much to ours (for them the exponent $\mu = 1/4$ is quite often fulfilled). Last but not least according to [13] one may think about possible applications in magnetic (recording) tapes, in which certain (cylindrical) microdomains emerge, *e.g.* in garnet or ferrite tapes typically about a few μm thick [24].

A.G. wishes to thank D.E. Czekaj and M. Cieplak for providing some useful informations about specific systems that may exemplify our study.

Appendix

It is not exclusively an intellectual puzzle, if one tried to argue, what would happen, when a formal substitution of d = 3 by $2 < d_f < 3$ would be realized, where d_f stands for a fractal dimension. We might even propose to choose $d_f \simeq 2.47$, which was pointed out as a very accurate numerical estimate by Borkovec et al. [25] as being very characteristic of some almost perfect close-packings of Apollonian type, generally called the obsculatory packings. The Apollonian sphere-packing could be a model of a porous fine-grained microstructure under evolution, or for an Apollonianlike soap froths, met sometimes in nature, cf. Ref. [1] by Mandelbrot in [25], or [26], and Ref. [32] therein. One can quite generally think about a defectscontaining partly ordered system, e.q. that of percolation-type, not forgetting, however, that this notion is preferred to be used for amorphous media; by the way, note that for 3D-percolation a typical value of $d_f \simeq 2.6$, cf. [27], and references therein. From the physical point of view, the problem of sphere-packing in a three-dimensional space was probably first studied by Lieb and Lebowitz [28] in a context of fundamental constitution of the matter organisation, being a thermodynamical system composed of electrons and nuclei (perhaps, if one wished to be in agreement with history of natural sciences, one would even be encouraged to move back to R. Descartes, who wanted to examine 'the disposition of matter in the solar system and its environs', *i.e.*, to propose a quasi-cellular as well as voids-containing model of the large-scale matter organisation in the universe [29]). If the above is taken into account, the relation (28) must be replaced by the similarity relation

with the Hausdorff–Besikovitch dimension $d_f \simeq 2.47$ [25], which yields a somewhat different value for the growth exponent μ , namely

$$\mu = \frac{3}{4d_f} \approx 0.3. \tag{36}$$

That means, that under some effective geometrical close-packing conditions the average radius (characteristic linear quantity) of the Apollonian gasket grows faster with time than in the afore presented standard case, cf. the relation (29), *i.e.*

$$\langle r(t) \rangle \sim t^{0.3},\tag{37}$$

roughly; here, the exponent $\mu = 0.3$ may probably be a first signature of the cellular system auto-separation or formation of (micro)voids, *cf.* [30] and references therein, that would facilitate the grain-growth or the evolution of bubbles-containing system. Notice, however, that the condition (26) has to be replaced by a weaker (limiting) one, namely

$$\lim_{t \to \infty} V(t) = V(0), \tag{38}$$

which by the way stands for completeness condition for the osculatory packings [31]. This remains an open question: Whether the natural close-packing concept could be proposed as a way of relaxing the condition (26), which in reality may easily be violated, *e.g.* by exerting, or even taking into account, a small mechanical stress on the system as a whole (or inside the system).

The argumentation may also be of use while studying statistical properties of focal conic textures of smectic liquid crystals of type A, cf. [32]; also soap froths belong to the same category of patterns. Some relation of the presented problem to Moon craters' formation as well as to the evolution of Earth-impacting meteorites would be anticipated [13]. For them some Apollonian-like structural arrangement appears to be always a key feature.

Some doubts may arise while asking whether an Apollonian grain, or simply the Apollonian gasket (maybe a "percolated grain" too?), even under growth, may solely be determined by a single fractal dimension, representing its volumetric (or mass) characteristics. Such a question can also be posed in our context, since after readily performing the differentiation in (2), one automatically shows up the two basic signatures of the mechanism of the evolution in question. After so doing, one immediately notices a curvaturedependent part of the mechanism proportional to $v^{-1/3}f(v,t)$, and because $v \propto r^3$, it is proportional to $(1/r) \times f(r^3, t)$, where certainly $r \equiv r(t)$ and $v \equiv v(t)$. For the 'fractal-case' one should formally replace 3 by d_f , but nevertheless the last equality holds also for this case. In consequence, one may recognize that every subtleness being assigned to a curvature change may also change the overall system's behavior. But Apollonian gaskets seem to have quite typical curvature behavior, they are mass fractals rather than surface fractals, so that we are likely to presume quite trivial surface characteristics of them, cf. discussions about fractal characteristics for soil (and, related) systems, or for some polymeric crystals, revealed mostly by the X-ray scattering method [33]. In short, the single fractal characteristics, d_f , would probably suffice at least in the first attempt towards modelling the evolution of self-similar close-packings.

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