

Formation of fractal structures in an explosion

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In an explosion of condensed explosives free carbon is typically released. The chemical reaction can proceed under conditions where diamond is stable, and some results on the production of the diamond phase are described in Refs. 1 and 2. This circumstance explains the interest in the particle-growth process. The characteristic size of a single crystal is about 40 Å.^{2,3} Larger aggregates form from such particles. It is reasonable to conjecture that a fractal structure arises.

In this work we measured the fractal dimension of clusters in powder of ultradisperse diamonds produced in an explosion. A two-step model of growth (atoms—compact particles—clusters), which agrees qualitatively with experiment, is introduced.

1. The experiments were performed by the method of small-angle x-ray scattering for angles in the range $7' < 2\theta < 7^\circ$ at the wavelength $\lambda = 1.54 \text{ \AA}$. The results for two samples of diamond powder, prepared in different experiments, are presented in Fig. 1. The intensities were recalculated in order to correct for the aperture averaging of the device.

The sections of the curves whose slope is close to -4 correspond to scattering by separate particles. For values of the transferred momentum $q = (4\pi/\lambda) \cdot \sin(\theta/2)$ ranging from $5 \cdot 10^{-3}$ to $3 \cdot 10^{-2} \text{ \AA}^{-1}$ the slope gives the fractal dimension $D \approx 1.9$ characteristic for cluster-cluster aggregation.⁴ The boundaries of the fractal interval make it possible to estimate the characteristic sizes of the particles and aggregates (30 and 200 Å).

2. During an explosion free carbon is liberated over a time $\approx 0.3 \text{ \mu s}$ — the chemical reaction time. The characteristic hydrodynamic expansion time t_H is several microseconds. The quantity of hydrogen can be $\geq 10\%$ of the mass of the charge and the number density of atoms $n_C \sim 10^{22} \text{ cm}^{-3}$. In the case of strong nonequilibrium, particle growth should start with fast coagulation. However, the assumption that the coagulation is unbounded results in particle sizes which are too large. The average mass of a particle (in units of the mass of the C atom) increases linearly with time⁵:

$$\langle m \rangle \sim K n_C t; \quad K = 4kT/3q.$$

For $T = 3 \cdot 10^3 \text{ K}$, $\eta = 10^{-2} \text{ P}$ (Ref. 6), $kn_C \sim 10^{12} \text{ s}^{-1}$. Within 1 μsec particles are formed which consist of, on the average, 10^6 atoms and are about 200 Å in size. In addition, the experiment of Ref. 1 did not show any dependence of the particle size on t_H (proportional to the size of the charge). Therefore the growth of compact particles must be bounded.

Next, we shall assume that when two particles encounter one another they coalesce if at least one of them contains fewer than $m_0 \sim 10^3$ atoms; i.e., if it is less than $\sim 20 \text{ \AA}$ in size. This assumption is supported by several arguments,² in particular, the fact that smaller particles have a sharply lower melting point.⁷ During the reaction time t_r , intro-

ducing the unit of time $\sqrt{t_r/kn_C}$ and the unit of concentration $\sqrt{n_C/kt_r}$, we obtain the following modified system of Smoluchowski's equations:

$$\frac{dn_m}{dt} = 2 \sum_{j=1}^{M_+} F_{j,m-j} n_j n_{m-j} - 2n_m \sum_{j=1}^{M_-} F_{j,m} n_j + \delta_{m,1}, \quad (1)$$

where n_m is the concentration of particles containing m atoms, $M_+ = \min[m/2, m_0]$, $M_- = m_0$ for $m < m_0$; $\delta_{m,1}$ describes uniform arrival of single atoms; the factor $F_{j,m} = (r_j + r_m)^2/4r_j r_m$ takes into account the difference in the sizes r of the particles with masses j and m .

Over a time $t \sim 5$ the distribution $n_m \sim m^{-3/2}$ (Ref. 8), which is cut off at the limit $m \sim t^2$, is generated. The limiting mass m_0 appears at $t \sim \sqrt{m_0}$. Small particles ($m < m_0$) continue to stick to large particles. For $m \gg m_0$, expanding $n_{m-j} = n_m - j \frac{\partial n_m}{\partial m}$ and setting $F_{j,m-j} = F_{j,m}$, we obtain the wave equation

$$\frac{\partial n}{\partial t} + c \frac{\partial n}{\partial m} = 0, \quad c = 2 \sum_{j=1}^{m_0} j n_j F_{j,m}.$$

The numerical solution of the system (1) demonstrates that a wave is generated at the boundary and propagates (Fig. 2a). The velocity of the wave is close to the velocity computed according to a distribution n_j which is not disturbed by the boundary.

When the reaction ends, over a time $\Delta t \sim \sqrt{m_0}$, the small particles die away and a frozen distribution (Fig. 2b) with a quite narrow range of sizes is generated. The times $t = 31.6, 100,$ and 316 correspond to chemical reaction times of 1, 10, and 100 ns. The true reaction time t_r can be shorter than the gross time of 0.3 μs , determined according to the end of the reaction as a whole (if the reaction proceeds by flare-up from hot points).

3. We shall examine qualitatively the formation of aggregates under conditions of cluster association

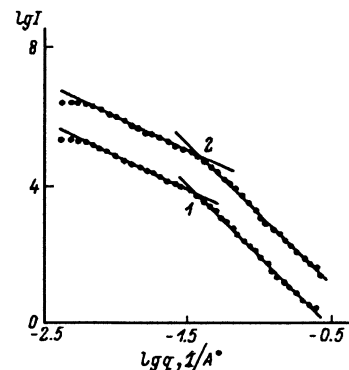


FIG. 1. The intensity of scattered radiation as a function of q . Slopes of the straight lines: for data group 1 (-1.94 and -4.18); for data group 2 (-1.89 and -4.10).

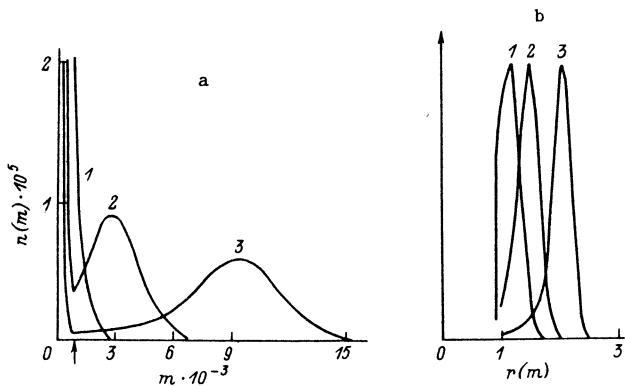


FIG. 2. a) Mass distribution of concentrations for the dimensionless time $t_1 = \sqrt{m_0} = 31.6$, $t_2 = 100$, $t_3 = 316$. b) Frozen size distributions obtained after the reaction ends at the moments t_1 , t_2 , and t_3 , scaled to the same amplitude. The limiting size $r(m_0)$ is taken to be unity.

Neglecting polydispersity of small clusters, we obtain for the concentration A of aggregates

$$\frac{dA}{dt} = \frac{dP}{dt} - A^2$$

(P is the particle concentration). At the end of the reaction $A \approx 1/t$, $P \sim m_0^{-1/2}$. The average number of particles in a cluster is $Z = P/A \sim t/\sqrt{m_0}$ or, in dimensional units is t/τ ; $\tau = \sqrt{t_r m_0 / kn_C}$. Taking hydrodynamic expansion into account the time t must be bounded by the time t_H , so that $Z \leq t_H/\tau \sim 100$ ns with $t_H = 1 \mu\text{s}$ and $t_r = 100$ ns ($\tau = 10$ ns). The cluster size $R \sim Z^{1/D} \approx \sqrt{Z} \sim 10$ particle sizes is

in agreement with experiment. When the true volume fraction of the solid phase is ~ 0.1 the looseness of the fractal structure makes gel formation probable (which limits the size of an aggregate approximately by the same value $R \sim 10$). As the medium expands the gel will rupture. In conserved samples structures of the next orders observable microscopically³ form from the initial aggregates.

4. Individual particles and clusters are "relics" of the chemical reaction. A more detailed study will make it possible to determine the true reaction time t_r . The interaction of the solid and gas phases is of interested. The processes studied can be affected by phase transitions and changes in the rheology of the medium.

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Phase and distance effects in the Mössbauer effect in the presence of ultrasonic excitation

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The study of the propagation of Mössbauer γ rays through matters exposed to radio-frequency fields is of great interest because of the possibility of studying both the dynamic processes occurring in the solid and the characteristics of the rf fields themselves.¹ The effects arising in systems containing more than one Mössbauer object (source or absorber), exposed to ultrasound have now been studied in detail theoretically,²⁻⁴ but they have not been adequately confirmed experimentally. The only experiment of which we know in this field is described in Ref. 5.

In this paper we report the results of the first experiments on how the intensity of Mössbauer γ rays that have passed through a system of two absorbers exposed to ultrasound depends on the difference of the phases of ultrasonic oscillations and on the distance between the absorbers (the so-called phase and distance effects).

Theoretical calculations based on the work of Refs. 2-4 show that when coherent ultrasonic excitation is present in thin absorbers and when there is no hyperfine splitting in the source and in the absorbers and the frequencies and amplitudes of the ultrasound in both modulators are equal then the frequency-modulated spectrum of coherent Mössbauer γ -rays will consist of a collection of equally spaced lines with the frequencies $\omega_n = \omega_0 + n\Omega$ ($n = 0, \pm 1, \pm 2, \dots$), where ω_0 is the frequency of the Mössbauer transition, Ω is the frequency of the ultrasound, the intensities of the time will be

$$I_n \sim J_n^2(2\bar{k}a \sin \frac{\theta}{2}), \quad \theta = \Delta\varphi + \frac{L\Omega}{c}, \quad (1)$$

where $J_n(z)$ is the Bessel function of the first kind, of order n , k is the wave vector of the γ -radiation.