## 2D Monte Carlo simulations of radiation effects on non-equilibrium colloid growth

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**Abstract.** The formation of colloids in alkali halides induced by ionizing radiation is studied by the Monte Carlo simulation of aggregation of F centres. The model consists of the following mechanisms.

(i) A two-dimensional lattice-gas Ising model is defined by a Hamiltonian that describes the thermally driven transition from gas (F centres) into the solid phase (condensed colloids).

In addition two mechanisms specific for the irradiation of alkali halides are introduced.

(ii) Highly mobile recombining centres (H centres) diffuse to the colloid with a flux approximately equal to the flux of F centres. After reaching the surface the H centre and one surface alkali-metal atom of the colloid recombine.

(iii) The lattice is subject to a continuous generation of radiation-induced lattice excitations. At the surface of the colloid the excitations result in displacements, which distort the surface of the growing cluster.

The morphology and growth kinetics are studied. It appears that the F centres do not condense into one compact cluster. Instead they are spread over a confined region containing small subclusters. The spread increases considerably if the annihilation fraction is increased above 80%. The surface displacements prevent the cluster from relaxing to its equilibrium shape, causing a roughening of the cluster surface.

## 1. Introduction

During the last decade an increasing interest has developed in the field of diffusion-limited aggregation (DLA). The varying morphology of the clusters as well as the anomalous growth kinetics [1,2] are studied. Starting with models in which single particles execute a random walk on a lattice until they stick to the aggregate [3], the effort in this field had lead to a large variety of models, differing in complexity from a single-particle model with varying sticking probabilities to multiparticle models, in which the lattice is occupied by random walkers corresponding to an initial density $\rho_0$, which ultimately stick to the growing cluster [4–8].

During the past few years multiparticle models of the gas/fluid–solid phase transition [9–13] have been developed, in which the transition probabilities are based on local changes in the free energy of the system. The effect of the surface energy is incorporated in the models to investigate the restructuring and crossover from fractal growth to equilibrium compact growth as the system relaxes to a state of lower energy.

We have adopted the models of Sørensen and co-worker [10] and Saito and Ueta [11], which are based on a Hamiltonian formalism that permits the definition of a thermodynamic temperature. At low particle densities these models are closely related to the DLA models.

These models are particularly appropriate for the simulation of the aggregation of F centres in irradiated alkali halides. The irradiation of alkali halides leads to the formation of two primary defects, the F (an electron trapped in a vacancy) and the H centre (a halogen interstitial). Whereas the H centres are trapped by impurities and dislocations, the F centres aggregate to metallic colloids.

Our modelling efforts are motivated by difficulties in interpreting the results from a variety of experimental methods (electron spin resonance, Raman scattering, differential scanning calorimetry, optical absorption spectrometry) [14–16] on irradiated NaCl. Anomalous phenomena such as the observation of three melting peaks for the Na colloids and a double and/or wide colloid band in the optical absorption spectrum have been observed, which indicate that we are dealing with complex colloidal structures. Since the size and shape of the colloid strongly determines the response to these experimental methods, a good understanding of the colloid morphology is required.

Colloids in irradiated alkali halides are the net product of simultaneous aggregation of F centres and annihilation of colloidal centres at the surface by H centres. From the kinetics [17, 18] it is clear that the fraction of aggregated F centres that will be annihilated again by H centres is very large (typically > 90%). Such a large annihilation fraction will strongly affect the morphology of the colloids. On the other hand, the surface of the colloid will continuously be affected by radiation-induced lattice excitations. These excitations lead to displacements of atoms at the surface, which affects the surface structure. Preliminary work on the effect of both reactions was based on a single-particle DLA type of model [19], which showed clear differences as compared with the usual DLA structures.

The aim of this paper is to investigate the effects on the growth of colloids of these mechanisms, which are typical for the irradiation of alkali halides. We propose a model of the growth of a colloid in a diffusional field, in which the effects of annihilation by H centres and transformation by irradiation-induced excitations at the surface are taken into account. Our main concern is the cluster morphology as a function of the temperature and the evolution of the colloid growth as a function of the time.

We emphasize the general nature of the model. The aggregation of one type of reactant, undergoing a transition from the gas phase (the F centre) into the solid phase (the colloidal centre) is simulated, which process competes with the simultaneous annihilation with a second type of reactant (the H centre). Meanwhile, the growing cluster is continuously distorted by a random process, which changes the structure of the surface.

## 2. The model

### 2.1. The basic dynamics

The simulations were carried out on a two-dimensional $256 \times 256$ square lattice subject to periodic boundary conditions. At the start of a simulation a single colloidal centre (the colloid nucleus) is introduced at the centre of the lattice. A number of F centres corresponding to a density $\rho_0$ of 0.005 is placed at random at unoccupied sites of the lattice. It is not allowed to occupy a single lattice site more than once. During one Monte Carlo step (MCS) each particle is (on average) examined in random order. With probabilities given by the characteristic frequencies $\tau_D^{-1}$ or $\tau_s^{-1}$ either diffusion or phase transition is selected.

In the case of diffusion there are two possibilities. If the selected particle is colloidal the trial is unsuccessful and a new cycle is started (i.e. the colloidal centres are not allowed to move). If the particle is an F centre one of its neighbour sites is selected randomly as a

possible next position. If unoccupied, the particle moves to this new site. If occupied, the particle remains on its site and a new cycle is started.

In the case of phase transition the F-centre–colloid transition is executed with a probability obtained from the following Hamiltonian:

$$\mathcal{H} = \mu_s \sum_i C_i + 2J \sum_{\langle ij \rangle} [C_i(1 - C_j) + (1 - C_i)C_j] \tag{1}$$

where the symbol $\langle i, j \rangle$ means that all nearest-neighbour pairs $i, j$ are summed once, and the crystallization order parameter $C_i$ on the $i$th lattice site is unity if it is occupied by a colloidal centre and vanishes otherwise. The first summation calculates the bulk free energy of the colloid and the summation in the second term accounts for the binding energy $2J$ for each missing bond. The Hamiltonians can be compared with the classical expression for the Gibbs free energy:

$$G = i\mu_\infty + \gamma A \tag{2}$$

where $\mu_\infty$ is the chemical potential of the bulk metal, $i$ the number of particles in the cluster, $\gamma$ the surface energy and $A$ the surface area. The transition probabilities are determined by the usual Metropolis Monte Carlo criterion [20] $w = \min[1, \exp(-\Delta\mathcal{H}/kT)]$ where $\Delta\mathcal{H}$ is the change in energy and $T$ is the temperature. Varying the number of nearest neighbours (NNs) between zero and four leads to 10 different transition classes as listed in table 1.

Table 1. The energy difference for all possible transitions.

| Class | Transition | NNs | $\Delta\mathcal{H}$ |
|-------|-----------|-----|---------------------|
| 1 | C→F | 0 | $-\mu_s - 8J$ |
| 2 | C→F | 1 | $-\mu_s - 4J$ |
| 3 | C→F | 2 | $-\mu_s$ |
| 4 | C→F | 3 | $-\mu_s + 4J$ |
| 5 | C→F | 4 | $-\mu_s + 8J$ |
| 6 | F→C | 0 | $\mu_s + 8J$ |
| 7 | F→C | 1 | $\mu_s + 4J$ |
| 8 | F→C | 2 | $\mu_s$ |
| 9 | F→C | 3 | $\mu_s - 4J$ |
| 10 | F→C | 4 | $\mu_s - 8J$ |

In close analogy to the model of Sørensen and co-workers we include an entropy term in the chemical potential and express it as $-\mu_s = E - TS$ (see below). Up to this point the modelling of the aggregation is similar to the approach of Sørensen and co-workers [10] and Saito and Ueta [11]. In figure 1 the processes are represented schematically. The dynamics governed by Monte Carlo sampling are denoted by *process 1*. We will now describe how we implemented the reactions associated with the radiation.

## 2.2. The radiation effects

In correspondence with the model of Saito the concentration of F centres at distances far from the cluster is kept at its initial value. This has been done by monitoring the number of F centres in a zone at the edge with a width of five lattice points and adding or removing F centres if necessary. In this way we have fulfilled the condition that the number of F centres in irradiated alkali halides reaches the saturation value, determined by the steady-state value at which the production by the irradiation source is equal to the loss to sinks and recombination.

In addition, we have implemented two extra mechanisms, which are directly connected to the irradiation (see figure 1).
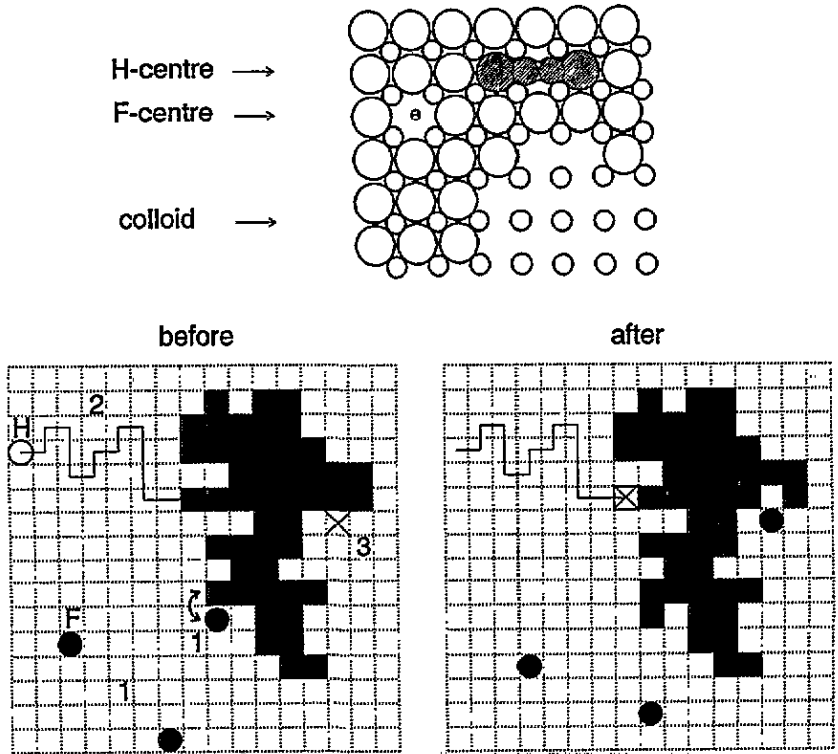
Figure 1. A schematic illustration of the defects in the lattice and the three processes incorporated in the model. Note that each intersection of the dotted lines represent one alkali-metal atom. *Process 1*, the model dynamics governed by Monte Carlo sampling, i.e. the diffusion and phase transition (solid circles, F centres; solid squares, colloidal centres). *Process 2*, the random walk of an H centre (open circle), ultimately annihilating a colloidal centre. *Process 3*, the displacement at the surface due to lattice excitations resulting in the annihilation of a colloidal centre at the surface and the creation of an F centre.

*2.2.1. Process 2, annihilation.* Highly mobile H centres diffuse to the colloid, annihilating colloidal centres at the cluster surface. The H centres have a mobility a factor of about $10^{10}$ higher than the mobility of the aggregating particles, the F centres. Thus we are allowed to keep the positions of the F centres fixed, while the H centre carries out a random walk until it reaches a lattice site occupied by a colloidal centre. This colloidal centre is then removed from the system. To enhance the efficiency of the program the direct recombination of an H and an F centre is forbidden.

The H centres are released randomly from the edge, with a rate proportional to the number of F centres added to the system. From the kinetics of growth of the colloids in irradiated NaCl [17, 18] it is known that the flux of H centres towards the colloids is approximately equal to the flux of F centres. Therefore, the annihilation fraction is set to a relatively high value, $f_a = 0.9$, i.e. only 10% of the aggregated F centres effectively contributes to the growing cluster. In order to avoid annihilation of all colloidal centres the minimum number of colloidal centres in the lattice is set to unity, the colloid nucleus.

*2.2.2. Process 3, surface displacements.* The second reaction was implemented in the following way. A lattice site is randomly chosen and checked to see whether it is unoccupied and next to a colloidal centre. If so, (one of) the bordering colloidal centre(s) is removed

from the lattice and an F centre is released at the original coordinates. This procedure will be repeated at fixed time intervals, corresponding to a total number of trials per lattice point (LP) $d$ during one simulation. According to this definition the parameter $d$ is proportional to the irradiation dose.

## 3. Parametrization

Unless stated otherwise the standard simulation parameter values are $\rho_0 = 0.005$, $\tau_S/\tau_D = 1$, $f_a = 0.9$, $d = 0.5$ trials/LP, $J/kT = 6.22$ ($kT/J = 0.161$), $E = 5.5J$ and $S = k$. The time duration of a complete simulation was set to 2.5 million MCSs.

The initial density $\rho_0 = 0.005$ (in two dimensions) is chosen relatively low in correspondence with the low F-centre concentration in irradiated alkali halides, $c_F \simeq 10^{-4}$ (in three dimensions). With the characteristic time ratio set to $\tau_S/\tau_D = 1$, on average one MCS can be associated with one-half of the jump time, i.e. $\frac{1}{2}\tau_D$. The jump time $\tau_D$ is determined by the jump frequency of the diffusing F centre $1/\tau_D = \nu_0 \exp(-E_m/kT)$, where $\nu_0$ is the vibrational frequency and $E_m$ the migration energy. Thus at high temperatures one MCS represents a shorter time step than at low temperatures. With $\nu_0 = 10^{13}$ s$^{-1}$ and $E_m = 0.88$ eV, we obtain 1 MCS $= \frac{1}{2}\tau_D = 0.06$ s at 100 °C, i.e. one simulation corresponds to an irradiation time of about 40 h. Consequently, a dose $d = 0.5$/LP in 2.5 million MCSs leads to a dose rate of $3 \times 10^{-6}$ dpa s$^{-1}$, which is in the range of physical interesting values (100 Mrad h$^{-1} \equiv 1.65 \times 10^{-6}$ dpa s$^{-1}$).

The following considerations have led to the values of the Hamiltonian parameters $J$, $E$ and $S$. The transition of one particle from the solid into the gas phase is associated with a change in the chemical potential of $\Delta\mu = \mu_g - \mu_s = \Delta H_v - T\Delta S$, where $\Delta H_v$ is the enthalpy of formation of an F centre from the colloid and $\Delta S$ the change in entropy. Splitting the entropy term into the configurational entropy $\Delta S_c$ and the vibrational entropy $\Delta S_v$, where $\mu_g = -T\Delta S_c$ [21, 11], we obtain for the chemical potential the expression $\mu_s = -(\Delta H_v - T\Delta S_v)$. With $\Delta H_v = 1.1$ eV and $\Delta S_v = k$ [21], the complete expression for the chemical potential becomes $|\mu_s| = E - TS - 1.1 - kT$ in electronvolts.

The NN interaction energy $2J$ can be obtained from the interface energy $\gamma$ of the Na inside the NaCl matrix. Calculations of the energies arising from Coulombic and Van der Waals forces, the metal surface energy and the formation of image charges indicate that the interface energy of an Na colloid embedded in NaCl is about twice the surface energy of Na (0.2 J m$^{-2}$), i.e. 0.4 J m$^{-2}$ [22]. From the surface energy per atom for a flat $\gamma a^2 = 2J$, we find (where $a = 4 \times 10^{-8}$ cm is the lattice distance ($J = 0.2$ eV, i.e. $J/kT = 6.22$ at 100 °C. Consequently a variation in the temperature between $kT/J = 0.150$ and $kT/J = 0.215$ corresponds to a variation between 75 °C and 225 °C.

## 4. Results and discussion

### 4.1. General behaviour

Figure 2(a) shows the development in time of a growing cluster, which is subjected to all the processes. Due to annihilation and surface displacements the aggregate is fragmented into many small subclusters. The subclusters are scattered in a region close to the original nucleation site. If the cluster grows larger, the outer part is distorted continuously by the aggregating and annihilating random walkers, while the inner part remains relatively unaffected due to screening by the outer part.
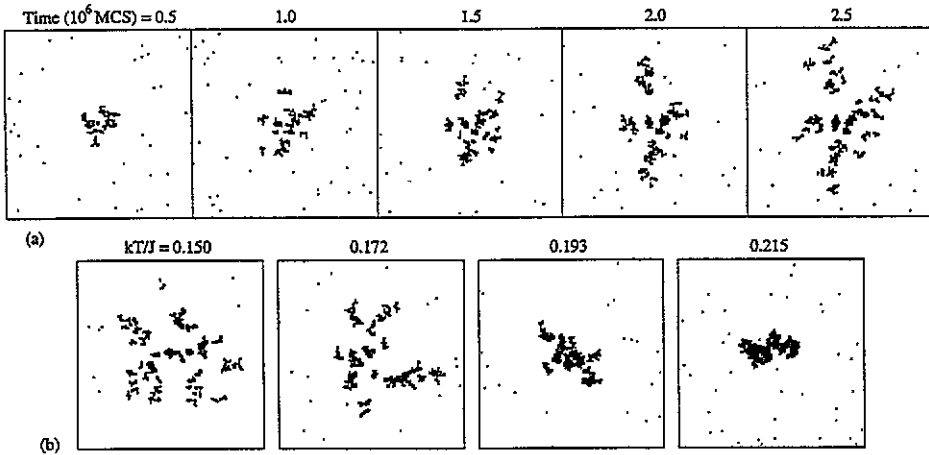
Figure 2. (a) A typical time sequence of a growing cluster. Shown is a window of $128 \times 128$ sites centred in the $256 \times 256$ lattice box. $kT/J = 0.161$. The increasing numbers of colloidal centres are respectively $N_c = 150, 237, 346, 452$ and $610$. (b) The influence of the temperature after 2.5 million MCSs, $kT/J$ varying from 0.150 to 0.215. From left to right $N_c = 648, 627, 495$ and $447$. The colloidal centres are denoted by black squares and the F centres are denoted by black dots.

The effect of the temperature is illustrated in figure 2(b) by a series of 'snapshots' at 2.5 million MCSs for four different temperatures. At high temperatures the cluster becomes more compact. Although this trend is evident, the shape of the cluster is far from the square equilibrium shape. The formation of the equilibrium shape takes extremely long simulation times (up to $10^{12}$ MCS [10, 11]. Moreover, this relaxation is prevented by the continuous distortion of the cluster surface due to the annihilation and the surface displacements.

In figure 3 the results obtained by averaging over 20 independent runs with different random number sequences are presented for four different values of $kT/J$. In figure 3(a) the average number of colloidal centres $N_c$ is plotted as a function of time (MCSs). The growth is approximately linear for almost all times. In the early stages of the simulations there is a deviation of the relation $N(t) \sim t$. This deviation is caused by the initial random configuration of the F centres. The system needs a short time to obtain the steady-state concentration gradient. This leads to an increased growth rate in the first stage of the simulations.

The behaviour of $N_c$ can be understood in terms of usual growth kinetics, using the radius of gyration $R_g$ as a measure of the size of the cluster. We can derive the growth rate of the colloid from the concentration gradient using the steady-state diffusion equation in two dimensions:

$$(1/r)(d/dr)r(d/dr)\rho = 0. \qquad (3)$$

Using spherical symmetry with the boundary conditions $\rho = \rho_0$ at $r = R$ and $\rho = \rho_1$ at $r = r_1$, where $R$ is determined by the size of the $L \times L$ box, $R = \frac{1}{2}L$ and $r_1$ is the radius of the region containing the subclusters, we obtain the following equation for the growth rate:

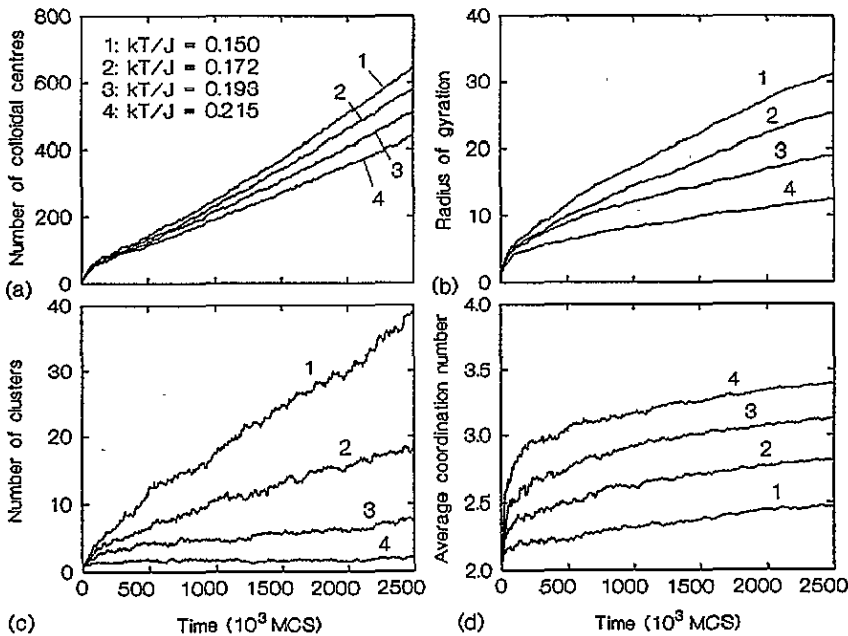$$dN/dt = (1 - f_a)2\pi D(\rho_0 - \rho_1)/\ln(R/r_1). \qquad (4)$$

**Figure 3.** Results of simulations at four different temperatures. (a) The number of colloidal centres $N_c$, (b) the radius of gyration of the colloid region $R_g$, (c) the number of subclusters $n_{cl}$ and (d) the average coordination number ACN versus time, $kT/J$ varying from 0.150 to 0.215.

Here $D = 1/\tau_D = 0.5$/MCS is the diffusion coefficient and $(1 - f_a) = 0.1$ is the correction for the annihilation. If $r_1 \sim R_g$ is relatively small the logarithmic factor will only change very slowly, such that we expect in the early-time regime a linear growth curve $N(t) \sim t$. The increase of the cluster region leads to an increase in the growth rate due to the smaller distance between the source (the edge of the simulation box) and the sink (the region with subclusters). This results in a deviation of the linear behaviour, which is accounted for by the logarithmic correction term $\ln(R/r_1)$.

The solution in equation (4) is exact for compact spherical clusters. We can use the expression to evaluate our simulations; however, the exact numerical value of the growth rate depends on the definition of the effective radius of the cluster $r_1$ and the density $\rho_1$ at this radius. We choose $r_1 = R_g$, and assume that $\rho_0 \gg \rho_1 \simeq 0$. The condition $\rho_1 \simeq 0$ is doubtful; however, the exact value is difficult to obtain, since we are dealing with very low densities.

At $t = 0.5 \times 10^6$ MCSs we find for the highest temperature, i.e. the most compact cluster, $R_g = 6$ (figure 3(b)). Thus we derive a growth rate of $0.52 \times 10^{-3}$ colloidal centres per MCS (CC/MCS). This value can be compared with the actual simulation from which we obtain a growth rate of $0.13 \times 10^{-3}$ CC/MCS. Regarding the crudeness of our approximations the agreement between these values is good.

In figure 3(c) the number of subclusters $n_{cl}$ as a function of time shows an increase with decreasing temperature. This is as expected, because at low temperatures the probability of a transition from the gas phase to the condensed phase is larger than that at high temperatures. Therefore an F centre next to a colloidal centre will almost certainly undergo a phase transition at low temperatures, while at high temperatures this certainty is reached only if an F centre is neighboured by more than one colloidal centre. This results in a more

compact shape of the colloid with increasing temperature. This trend is also evident in figure 3(d), where the average coordination number (ACN) versus time is plotted. This number is defined as the average number of NN colloidal centres surrounding a colloidal centre, in other words, the average number of bonds of a colloidal centre.

On a $512 \times 512$ lattice with the standard simulation parameters and $kT/J = 0.150$, the morphology of the cluster is similar to the morphology of a cluster grown on $256 \times 256$ lattice, with a final number of colloidal centres of about 730 at $t = 2.5 \times 10^6$ MCSs. In the first stage of the simulation, the steep growth continues for four times as long, until about 300 000 MCSs, corresponding to the four times as large box area. If the steady state is reached, the growth of the number of colloidal centres is linear with time until the simulation is stopped afte 2.5 million MCSs. In order to study the effect of the relatively low mobility of the F centre on the cluster growth we have performed several simulations with a smaller ratio $\tau_S/\tau_D$, down to $10^{-3}$. The resulting relative decrease in the mobility of the F centres strongly delays the growth of the cluster, but does not change the cluster morphology.
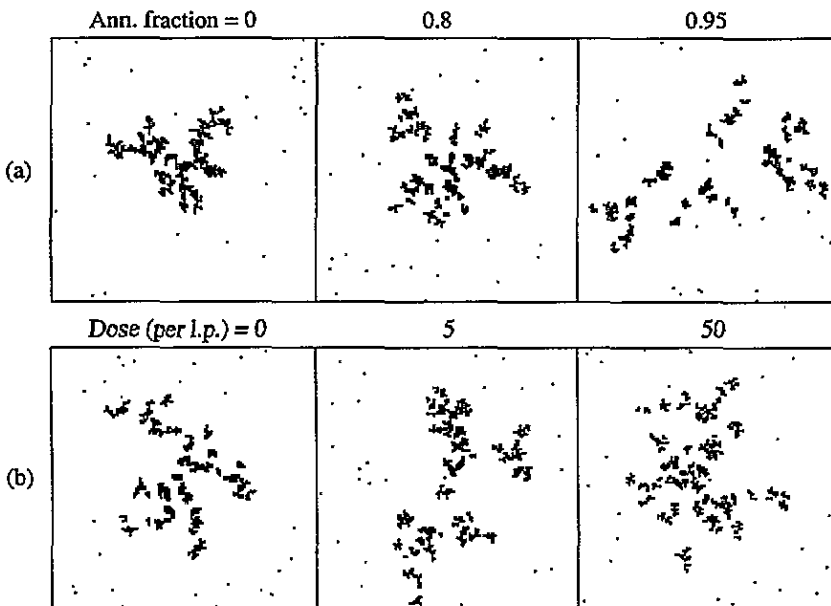


**Figure 4.** Typical clusters for three different values (a) of the annihilation fraction $f_a$ and (b) of the dose $d$. $kT/J = 0.161$. Characteristic numbers, from left to right, (a) $N_c = 542$, 564, 534, $R_g = 18.6$, 23.0, 38.6, $n_{cl} = 6$, 25, 27, ACN $= 2.66$, 2.59, 2.59, (b) $N_c = 583$, 663, 628, $R_g = 25.7$, 32.1, 27.0, $n_{cl} = 27$, 31, 49, ACN $= 2.65$, 2.54, 2.23.

## 4.2. The radiation effects

The annihilation and surface displacements influence the appearance of the clusters. In figure 4(a) we see how the subclusters are spread out over a larger region if the annihilation fraction $f_a$ is increased. Existing bonds are broken by the H centres, resulting in a large number of separate subclusters. These subclusters are subject to continuous aggregation and
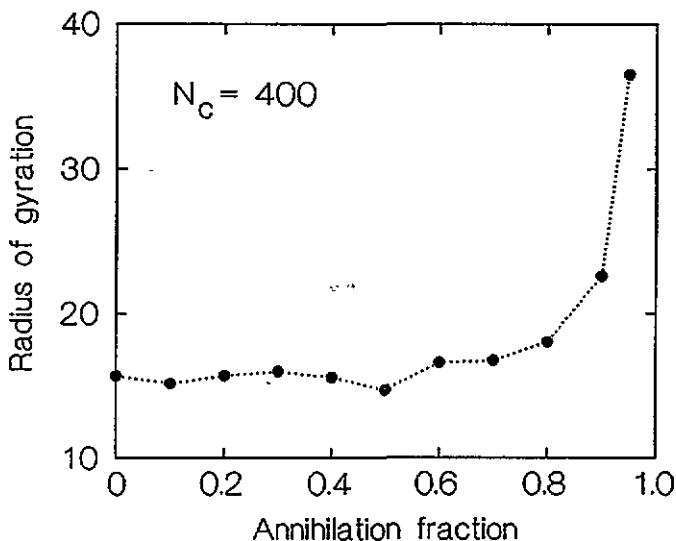
**Figure 5.** The radius of gyration versus the annihilation fraction $f_a$. The number of colloidal centres $N \simeq 400$. $kt/J = 0.161$.

annihilation, which results in a slow, semi-diffusional movement in a direction away from the centre of the cluster (see also figure 2).

To illustrate the effect of $f_a$ more clearly, the radius of gyration of the cluster is plotted versus $f_a$ in figure 5 for a total number of colloidal centres $N \simeq 400$. Up to $f_a = 0.5$ the radius of gyration remains approximately the same. If the annihilation fraction increases further, the radius of gyration increases gradually, while above $f_a = 0.8$ the spread of the subclusters increases very strongly.

The effect of the surface displacements is shown in figure 4(b). The cluster surface is continuously affected by the process, resulting in a fragmentation of the clusters into even smaller subclusters with rough surfaces. As a consequence the total number of subclusters increases and the average coordination number decreases.

### 4.3. The Hamiltonian parameter

The sensitivity of the aggregation processes with respect to the various Hamiltonian parameters is visualized in figure 6, where some typical clusters are shown. The value of $J/kT$ is varied between 4.66 and 7.77 (i.e. a variation of $J$ between 0.15 and 0.25 eV, at a temperature of 100 °C), while the other simulation parameters have the standard values. Due to the increase of the interaction parameter $J$, the clusters are more compact. A decrease of the energy parameter $E/kT$ from 40.4 to 28.0 (variation of $E$ between 0.9 and 1.3 eV, at 100 °C) has the same effect (not shown). For a relatively small $J/kT$ a change of the entropy term $S/k$ from one to three does not result in a significant change of the cluster morphology. If $J/kT$ is increased the influence of $S/k$ is more pronounced. The figure shows that the distortion of the cluster due to the radiation effects is relatively small if the interaction energy and the entropy are both relatively large.

### 5. Conclusions

The model presented here offers the possibility of studying the effects of specific mechanisms
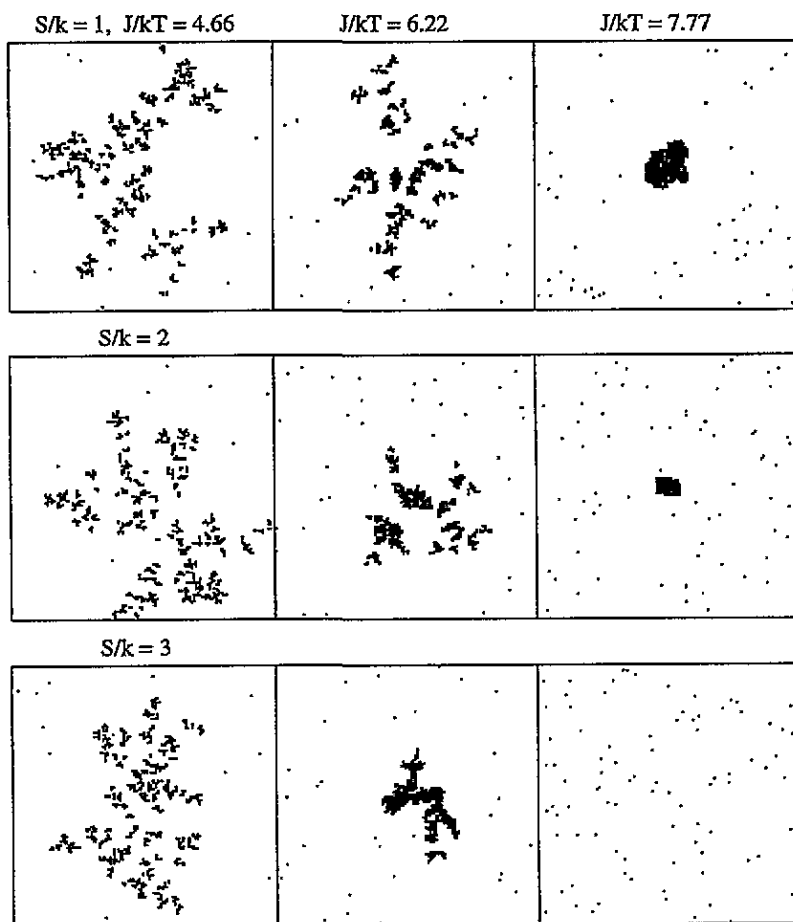
**Figure 6.** The influence of the Hamiltonian parameters on the cluster morphology. $S/k$ and $J/kT$ are varied, while $E/kT = 34.2$ is kept at its orignal value. From left to right, top, $N_c = 711$, 610, 335, $R_g = 36.3$, 28.0, 7.77, $n_{cl} = 77$, 31, 1, ACN $= 2.08$, 2.60, 3.64; middle, $N_c = 715$, 545, 97, $R_g = 35.5$, 20.4, 4.13, $n_{cl} = 84$, 17, 1, ACN $= 1.98$, 2.89, 3.53; bottom, $N_c = 683$, 489, 1, $R_g = 31.2$, 14.5, 0, $n_{cl} = 74$, 2, 1, ACN $= 2.04$, 3.26, 0.

that typically take place during the irradiation of alkali halides. The model shows that annihilation and displacements of colloidal centres at the surface as a result of the irradiation can seriously affect the aggregation of F centres. The colloidal centres are spread, forming small subclusters in a confined region surrounding the nucleation site. If the temperature is increased the colloid has a more compact morphology. The formation of small subclusters explains the experimental observations of quantum size effects, which indicate that the colloids formed during irradiation remain relatively small.

The growth of the number of colloidal centres can be described by usual growth kinetics, by applying the steady-state diffusion equation and defining an appropriate radius, e.g. the radius of gyration, for the size of the colloid. The exact rate can only be obtained from a careful investigation of the concentration of F centres in the vicinity of the subclusters. This concentration depends strongly on the shape and surface of the subclusters, and, therefore, on the radiation effects, which strongly affect the cluster morphology.

As a consequence, it is important to note that the rate constants in the kinetic models for the production of radiation damage [17, 18] need to be adjusted in order to take into account the larger effective radius of the colloid. Even more important for modelling the growth of colloids is the dependence of the evaporation of F centres on the cluster morphology. The strong dependence of the morphology on the radiation parameters and the temperature implies that the description of the F centre evaporation will become very complex in these models. Since the evaporation of F centres from the colloids limits the production of radiation damage at high temperatures, an exact knowledge of this quantity is of major importance.

In order to be able to make a comparison between the model and the experimental results the model should be extended to three dimensions. Such a model has to be based on a more detailed description of the interatomic potentials. In this respect a natural extension of our lattice-gas model will be the dynamical Monte Carlo method, first described by Voter [23]. This method takes into account the effect of the local environment by calculating the interatomic interactions for all possible configurations. These calculations are used to describe the Hamiltonian parameters in more detail. In particular the properties of the low-order aggregates containing only two, three or four F centres (M, R, N centres) are of major importance. These centres are the predecessors of the colloidal clusters and their properties will, if the cluster is fragmented into small irregular subclusters, affect the morphology of the colloid significantly.

## Acknowledgment

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