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Establishment of a steady-state nucleation regime. Theory and comparison with experimental data for glasses

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The paper considers the homogeneous formation of new-phase nuclei after instantaneous formation of the initial meta-stable state. Expressions which are asymptotically accurate in the magnitude of the nucleation barrier are obtained for the nucleation "incubation time" and for the number of nuclei formed as a function of time. The results are supported by the numerical solution of the fundamental kinetic equation and are compared with known experimental data on the crystallization of glasses.

INTRODUCTION

The fundamental kinetic equation of nucleation theory, according to Zel'dovich,[1] is

$$\frac{\partial f}{\partial t} = -\frac{\partial j}{\partial g}, \quad j = -DN\frac{\partial}{\partial g}\frac{f}{N} = -D\frac{\partial f}{\partial g} + \dot{g}f. \tag{1}$$

Here g is the number of molecules in a nucleus ("size"), j is the flux in the size space, D(g) is the diffusion coefficient, $\dot{g}$ is the macroscopic growth rate of the nucleus, and f(g, t) and N(g) are respectively the kinetic and equilibrium distribution functions [the latter is proportional to the thermodynamic probability for the fluctuational formation of a nucleus N(g) ~ exp {−W(g)/kT}, where W(g) is the minimum work required to form a nucleus of the prescribed size].

The nucleation rate **I** is usually defined as the steady-state value of the flux j_{st}. A steady-state nucleation regime, however, is preceded by a transient period. The existence of such a period can have observable consequences in diverse physical situations, e.g., in the case of nucleation near the critical temperature[2] or in the case of condensation of an electron-hole liquid.[3] Evidently, the most important questions of this kind arise in connection with the quenching of metastable states, in particular, in the description of the crystallization of glasses.[4-10] The latter is due primarily to the fact that usually in the production of glass an attempt is made to shorten the time of intensive nucleation as much as possible and it is comparable to the transient period. On the other hand, experiments with glasses provide a unique opportunity to "freeze" the size distribution of the nuclei and to directly count the number of nuclei formed. This makes it possible to compare the theoretical and experimental data, which is done in this paper.

In Sec. 1 we discuss the various approximations in the theory of time-dependent nucleation. Expressions which are asymptotically accurate in the value of the barrier W_{max}/kT are obtained for the "incubation time" of nucleation and the time dependence of the number of nuclei formed. These results are compared with data from numerical simulation.

In Sec. 2 we compare the theoretical results wity experimental data.[6-8]

1. THEORY OF TIME-VARYING NUCLEATION

The establishment of a steady-state regime of nucleation was first considered by Zel'dovich,[1] who obtained an estimate of the characteristic time $\tau' = (g_* - g_1)^2/4D(g_*)$. Here g_* is the number of molecules in a critical nucleus, and g_1 is the size at which a steady distribution is established "instantaneously." Subsequently, numerous attempts were made to refine these estimates and to determine the analytical dependence I(t) (see the reviews in Refs. 4, 5, 11, 12 and also Refs. 2, 3, 13, and 14). The great diversity of the results obtained in studies of the same equation (1) is due mainly to the following reasons.

In general, Eq. (1) does not have a closed analytical solution, and the difference in the results is due to the particular approximations that were used, in particular, the methods used to separate the "diffusion" region from the "drift" region in the initial equation or the parabolic approximation of the barrier W(g)/kT, which is used in many studies (e.g., Refs. 2, 3, and 15). When in Ref. 16, for example, the diffusion region was extended to cover all the subcritical sizes, the result was a substantial overestimation of the transient period.

In the case of a parabolic approximation of the barrier and on the assumption that the value D_* of the diffusion coefficient does not depend on the size, Eq. (1) reduces to the Ornstein–Uhlenbeck equation, which admits an exact solution (e.g., Refs. 17 and 18). In this case we have

$$\tau = \frac{\Delta^2}{2D_*} = \left\{\frac{\partial\dot{g}}{\partial g}\Big|_*\right\}^{-1}, \quad \Delta = \left\{-\frac{1}{2kT}\frac{\partial^2 W}{\partial g^2}\Big|_*\right\}^{-1/2}, \tag{2}$$

where τ is the maximum relaxation time: the reciprocal of the first nonzero eigenvalue of the differential operator, $D_*\frac{\partial^2}{\partial g^2} - \frac{\partial\dot{g}}{\partial g}\Big|_*(g - g_*)\frac{\partial}{\partial g}$.

In a previous paper[19] we demonstrated that this result also holds for arbitrary functions W(g) and D(g), provided that the barrier W_*/kT is high. It should be borne in mind, however, that the relaxation is preceded by a fairly prolonged "incubation period," during which the flux is virtually zero. We note that the first eigenfunction of the operator indicated above has a zero derivative at $g = g_*$, so that the establishment of the flux $j_*(t)$
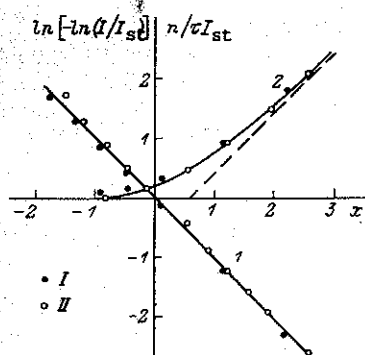
FIG. 1. 1, 2) Comparison of Eqs. (4) and (14) with the numerical solution of Eq. (1): $x = (t - t_{ind})/\tau + \gamma$. $W^*/kT = 20$ (I), 30 (II).
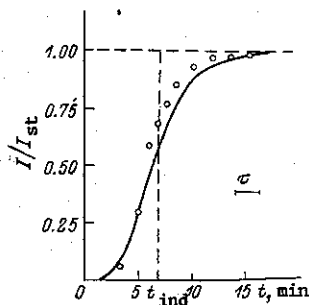

FIG. 2

is characterized by the second eigenvalue and the relaxation time $\tau_* = \tau/2$ (Ref. 19).

The transient period depends on the size g. Most studies involved the estimate of $g = g_*$. These estimates generally insufficient since the distribution of nuclei near g_*, because of their slow growth, is distorted considerably by diffusion in the transition to the region of large sizes. For such sizes j(t) does not correspond to the function $j_*(t)$ which is displaced in time. The transient process in the above-critical region was considered in Refs. 3, 14, and 20, but the quesiton of what size g_0 can be assigned initially to a nascent nucleus remained unanswered. This led, in particular, to an ambiguous interpretation of the results of numerical simulations.[5]

Among the analytical results for I(t), we can single out the simple "relaxation" relation

$$I(t) = I_{st} \left\{ 1 - \exp \frac{t_u - t}{\tau} \right\} \qquad (3)$$

and "diffusion" relation

$$I(t) = I_{st} \exp\left(-\frac{\tau'}{t} \right) \qquad (3')$$

of the types given in Refs. 4, 5, and 12.

Relations of the form (3) are unquestionably valid for along times but become meaningless for times comparable to the "incubation time" t_i. The applicability of (3') is limited to short times, as is evident from the fact that at $t \gg \tau'$ the linear dependence $n = I_{st}(t -$ constant) of the total number of nuclei on time does not follow from (3'). In Ref. 15, which has been used often in the interpretation of real experiments[7] and numerical simulations,[5,9] the function I(t) is represented by a

series which corresponds to (3) and (3') for long and short times; the relaxation time in Ref. 15 is $\tau_K = 8\pi^{-2}\tau$. The solution in Ref. 15, based on broad assumptions, is an approximation; this solution is discussed in Sec. 2.

The most natural approach to the study of non-stationary nucleation is provided by the solution of Eq. (1) on the assumption of a high barrier, $W_*/kT > 1$. This is one of the fundamental conditions in nucleation theory (it is closely linked with the condition $g_* \gg 1$, which determines the Fokker–Planck form of the initial kinetic equation, allows a steady-state solution to be obtained in closed form, etc.) and is satisfied satisfactorily in typical experimental situations. The first asymptotically rigorous analysis of the establishment of a steady-state regime of nucleation was carried out by Wakeshima.[21] His analysis, however, contained an additional assumption that the flux varies smoothly in comparison with the function N(g), but this assumption proved to be incorrect. The result was an underevaluation of the relaxation time, $\tau_W = \tau/4$, and an analytical form of I(t), which is different from (3), could not be determined.

The asymptotically exact analysis of Eq. (1), carried out in Ref. 19, enabled us to unambiguously single out the diffusion and drift regions in the initial kinetic equation and to obtain the analytical relation

$$j(g_0, t) = I_{st} \exp\{-\exp(-x)\}, \quad x = \frac{t - t_i(g_0)}{\tau}. \qquad (4)$$

The initial nucleus size g_0 can be chosen in the interval $\Delta \ll g_0 - g_* \ll g_*$. The flux $j(g_0, t)$ is carried by drift into the region of large sizes g, where the analytical form of its dependence on time is preserved with allowance for the lag

$$t_i(g) = t_i(g_0) + t'(g, g_0), \quad t'(g, |g_0) = \int_{g_0}^{g} \frac{dg}{\dot{g}}, \qquad (5)$$

i.e., the flux $j(g_0, t)$ determines the nucleation rate I(t).

To calculate the incubation time $t_i(g)$, we use the result[19]

$$t_i(g_0) = \tau \left\{ \ln \frac{2g_*(g_0 - g_*)}{\Delta^2} - C \right\}, \quad C = \int_0^{g_*} dg \left(\frac{1}{\dot{g}} - \frac{1}{g - g_*} \right). \qquad (6)$$

Taking in (5) and (6) the limit $g_0 = g_*$, we find

$$t_i(g) = \tau \left(\ln \frac{2g_*^2}{\Delta^2} - 2C \right) + \int_0^{g} \frac{dg}{\dot{g}}, \qquad (7)$$

where the integral is evaluated in the sense of the principal value. As expected, the final results do not contain the "initial size" of the nucleus, which can be chosen arbitrarily. The last expression can be rewritten more clearly by introducing, in addition to the growth time t', the positive decay time of a nucleus with $g < g_*$,

$$t''(g) = \int_0^{g} \frac{dg}{-\dot{g}}. \qquad (8)$$

Taking into account the asymptotic smallness of the ratio Δ/g_*, we find from (6) and (7)

$$t_i(g) = t'\left(g, \; g_* + \frac{\Delta}{\sqrt{2}}\right) + t''\left(g_* - \frac{\Delta}{\sqrt{2}}\right). \tag{9}$$

Expression (9) is more convenient than (7) for specific calculations (see below) when the integral $\int dg/\dot{g}$ is expressed in terms of elementary functions.

To determine $t_i(g)$ explicitly, it is necessary to make the expression for the macroscopic growth rate more specific. In many physical situations the rate of change of the radius R of the nucleus is described by an equation of the type

$$\dot{R} = \frac{R_*}{\tau}\left(\frac{R_*}{R}\right)^{\vartheta}\left(1 - \frac{R_*}{R}\right), \tag{10}$$

where the exponent ϑ is determined by the manner in which the material is introduced to the nucleus (Ref. 22).[1]

A more general expression is obtained when allowance is made for the discrete nature of the molecules in a nucleus (but the macroscopic expression for the work required to form the nucleus is retained). In this case the factor $1 - R_*/R$ in Eq. (10) is replaced by $a^{-1}\{1 - \exp[-a(1 - R_*/R)]\}$, where $a = \delta\mu/kT$, and $\delta\mu$ is the difference in the chemical potentials of the metastable and stable phases. The discreteness reduces the decay time t'' and increases the growth time t'. It can be expected that for reasonably large values of R the relative role of the discreteness is small even in those cases in which the parameter a is of the order of unity. This assumption is confirmed indirectly by the results of Ref. 19 and by numerical simulations (see the discussion below).

To determine the incubation time $t_i(R)$, let us consider several of the most frequently encountered integer values of ϑ in the growth law (10); the general results are given in the Appendix.

Case $\vartheta = 1$ applies when the material is introduced to the nucleus by diffusion. From (9) we find

$$\frac{1}{\tau}t_i(R) = \frac{1}{2}\left(\frac{R}{R_*} - 1\right)^2 + 2\left(\frac{R}{R_*} - 1\right) \tag{11}$$
$$- \frac{3}{2} + \ln\left(\frac{R}{R_*} - 1\right) + \ln\frac{6W_*}{kT}.$$

For $\vartheta = 0$, which corresponds to free-molecular ("ballistic") delivery of material to the nucleus, we obtain

$$\frac{1}{\tau}t_{ii}(R) = \frac{R}{R_*} - 2 + \ln\left(\frac{R}{R_*} - 1\right) + \ln\frac{6W_*}{kT}. \tag{12}$$

The situation $\vartheta = -1$ arises if the increase in the nucleus size is not limited because of the introduction of the material, in the case of cavitation.[24] In this case we have

$$\frac{1}{\tau}t_i(R) = \ln\left(\frac{R}{R_*} - 1\right) + \ln\frac{6W_*}{kT}. \tag{13}$$

In typical situations we have $6W_*/kT \gtrsim 10^2$ and the constant $\ln 6W_*/kT$ in (11)–(13), which is asymptotic, is indeed large. It can be said, with sufficient accuracy, that besides $\ln 6W_*/kT$ the expressions for $\tau^{-1}t_i(r)$ contain only terms that increase without bound as $R \to \infty$, which allows the incubation time to be found approximately in those cases in which the corresponding integrals cannot be evaluated explicitly (see Appendix).

The experimentally observable quantity usually is not the nucleation rate I itself but the total number of nuclei, $n(t) = \int_{-\infty}^{t} I(t')dt'$. Integrating (4), we find

$$n(t) = \tau I_{st}E_1(e^{-x}), \tag{14}$$

where $E_1(z)$ is an exponential integral function.[25] At long times, (14) has the asymptotic form

$$n(t) \simeq I_{st}(t - t_{ind}), \; t_{ind} = t_i + \gamma\tau \tag{15}$$

($\gamma = 0.577\ldots$ is Euler's constant), which enables us to relate the incubation time t_i to the experimentally determined "induction" time t_{ind}.

To illustrate the accuracy of these analytical relations, we numerically solved Eq. (1) in the region of sizes $6 \leq g \leq 96$ for two values of the barrier $W_*/kT = 20$ ($g_* = 22.2$) and $W_*/kt = 30$ ($g_* = 45.9$). Virtually no data on $\dot{R}(R)$ are available for crystallites in glasses [although the experimentally observed slight effect of the size on the growth rate when $R \gg R_*$ does permit the assumption that this relation is similar to Eq. (10) with $\vartheta = 0$]. Because of this circumstance, we chose the physical parameters corresponding to a supercooled gas, where law governing the growth of a nucleus has the form (10), which was modified to allow for the discreteness at $\vartheta = 0$ (the scheme of the numerical simulation is similar to that described in Ref. 26). The specific form of $\dot{R}(R)$ for a given $D(g_*)$ can affect only the induction time t_{ind}, which therefore is a more convenient, experimentally measurable quantity. The use of Eq. (12) in this case gave a correct result for t_{ind} within 10%, although the "discreteness parameter" $a = \delta\mu/kT$ was not small: 1.8 for $W_*/kT = 20$ and 1.3 for $W_*/kT = 30$.

The results of the calculations are shown in Fig. 1 (I, II). The solid lines represent the theoretical relations; straight line 1 corresponds to taking the logarithm of (4) twice and curve 2
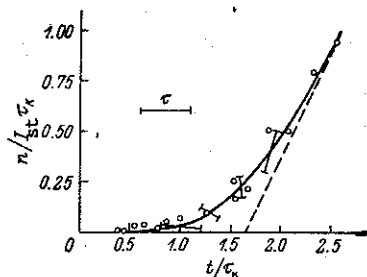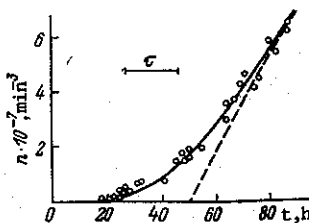

FIG. 3

FIG. 4

represents the function $E_1 \{\exp(-x)\}$, consistent with (14). The numerical and analytical results virtually coincide everywhere, except at the small initial stage where asymptotic methods are inapplicable because of the large gradients of the distribution function, and the dependence of the nucleation rate on time is evidently closer to (3').

2. COMPARISON WITH EXPERIMENTAL DATA

In accordance with the results of the preceding Section, we suggest a possible scheme for comparing the theory with experimental data on the functional dependence $n(t)$.

1) Isolating the linear part of $n(t)$, we determine I_{st} and t_{ind} in accordance with (15).

2) We find $n(t_{ind})$ and determine, in accordance with (14), that

$$\tau = \dot{n}(t_{ind})\{I_{CT}E_1(e^{-\gamma})\}^{-1} \simeq 2n(t_{ind})I_{st}^{-1}. \tag{16}$$

3) In accordance with (14), we plot $n_{theor}(t)$.

In one of the first studies of time-varying nucleation in glass[6] the function $I(t)$ was determined from $n(t)$. Calculating τ from the foregoing scheme, we plot $I(t)$ in accordance with (4) (Fig. 2). Allowing for the small scale of the graphs in Ref. 6, which gives us a reasonably good accuracy in determining τ, we consider the agreement between the theoretical and experimental results to be satisfactory. The condition

$$I(t_{ind}) = I_{st}\exp(-\exp(-\gamma)) \simeq 0.57I_{st},$$

which is also satisfied reasonably well, does not depend on the choice of τ.

The results of the comparison of the theory with the experimental data[7] are shown in Fig. 3, where the times for various groups of experiments were reduced to one single scale by means of a multiplier which, in the opinion of ~~many~~ authors [*] coincides with τ_K. The possible spread of the theoretical results because of the limited accuracy in the determination of $n(t_{ind})$ is also shown; it was assumed that the asymptotic dependence $n(t)$, and hence I_{st} and t_{ind}, were determined accurately. Satisfactory agreement is observed between theory and experiment everywhere, except in the small initial stage where the experimental points lie slightly ~~lower~~ higher.

The approximate solution of Kashchiev,[15] which was used in Ref. 7 and which can be written as $I(t) = I_{st} \vartheta_4 [0, \exp(-t/\tau_K)]$ (ϑ_4 is an eliptical theta function[25]), has a number of drawbacks because of the absence of an auxiliary time parameter, in addition to the relaxation time τ_K. The last factor in this case turns out to be closely linked with the induction time $t_{ind} = (\pi^2/6)\tau_K$, which depends on the conditions of the experiment (minimum observable size). In accordance with Ref. 15, we have

$$n(t_{ind}) = 2I_{st}\tau_K \sum_{m=1}^{\infty} \frac{(-1)^{m+1}}{m^2} e^{-\frac{\pi^2}{6}m^2} \simeq 0.39I_{st}\tau_K.$$

$$\text{or } \tau_K$$

According to (16), this value is more than 50% higher than the relaxation time τ, which is calculated from the experimental values of $n(t_{ind})$ and I_{st}.

The establishment of a steady-state nucleation regime in glasses of various compositions was studied comprehensively by Fokin et al.[8] We have chosen their study because the linear portion of $n(t)$ given in it has the largest number of points. As follows from the graph of the function $E_1(e^{-x})$ in Fig. 1, the points with $t - t_{ind} > 2\tau$ [or, according to (16), with $t - t_{ind} > 4n(t_{ind})I_{st}^{-1}$] must be used to determine the asymptotic form of $n(t)$ with sufficient reliability. The results of the comparison are shown in Fig. 4. Satisfactory agreement is observed everywhere, except in the small initial stage.

The experimental results[7],[8] are higher than the theoretical values in the initial stage apparently because of either the presence of finite-sized nuclei in the initial state, which was studied in Ref. 27, or the existence of additional relaxation processes, which would lead to a time dependence of the barrier W_*/kT. The latter was considered theoretically in Ref. 19, but a comparison seems premature because of the large relative error in measurements in the region of extremely low nucleation rate. Of the other possible causes of this divergence, in addition to heterogeneity, we point out the multiparameter nature of the nucleation process. In this case, according to Ref. 24, at short times the formation of nuclei may proceed predominantly along energetically less advantageous trajectories, but with a shorter transient period.

CONCLUSION

1. Relations have been derived for determining the nucleation induction time from the macroscopic equations for nucleus growth.

2. The nucleation rate and the number of nuclei formed have been determined as analytical functions of time. These functions are nearly exact in the range of applicability of the initial kinetic equation.

3. The results which we have obtained are in satisfactory agreement with known experimental data (with the exception of the relatively small initial stage of nucleation). There results can be used to determine the relaxation time and the diffusion coefficient $D(g)$.

I wish to express my profound thanks to I. M. Fishman, whose comments stimulated the execution of this study.

APPENDIX

INCUBATION TIME WITH AN ARBITRARY NUCLEUS GROWTH LAW

Let us first consider the case of an arbitrary ϑ in the growth law (10). We rewrite Eq. (7) in the form

$$\frac{1}{\tau_1} t_1(R) = \ln \frac{2R_1^2}{\Delta_R^2} - 2C_R + \frac{1}{\tau}\int_0^R \frac{dR}{R}, \tag{A.1}$$

$$\Delta_R = \Delta \left.\frac{dR}{dg}\right|_*, \quad C_R = C + \ln 3.$$

We have

$$\frac{1}{\tau}\int_0^R \frac{dR}{R} = \int_0^{\frac{R}{R_*}} du \left(\frac{u^{\{\vartheta\}-1}}{u-1} + \sum_{m=0}^{[\vartheta]+1} u^{\vartheta-m}\right), \tag{A.2}$$

where $\{\vartheta\}$ is the fractional part of ϑ, and $[\vartheta]$ is its integer part.

Taking the infinite limit in the converging part of the integral (A.2), we find[28]

$$\frac{1}{\tau}\int_0^R \frac{dR}{R} \xrightarrow[a\to\infty]{} -\pi \cot \pi\{\vartheta\} + \sum_{m=0}^{[\vartheta]+1} (\{\vartheta\}+m)^{-1}(R/R_*)^{\{\vartheta\}+m}. \quad (A.3)$$

On the basis of (5) and (10) we represent C_R by the relation[28]

$$C_R = \psi(2+\vartheta) - \psi(1),$$

where $\psi(z)$ is the digamma function, or using the known relations for $\psi(z)$ (Ref. 25), we represent C_R by the relation

$$C_R = \psi(1-\{\vartheta\}) - \psi(1) - \pi\cot\pi\{\vartheta\} + \sum_{m=0}^{[\vartheta]+1} \frac{1}{\{\vartheta\}+m}. \quad (A.4)$$

From (A.1), (A.3), and (A.4) we find

$$\frac{1}{\tau}t_u(R) \underset{R\gg R_*}{\simeq} \ln\frac{6W_*}{kT} + 2(\psi(1)-\psi(1-\{\vartheta\}))$$

$$+\pi\cot\pi\{\vartheta\} - \frac{1}{\{\vartheta\}} + \frac{\left(\frac{R}{R_*}\right)^{\{\vartheta\}}-1}{\{\vartheta\}} + \sum_{m=1}^{[\vartheta]+1} \frac{\left(\frac{R}{R_*}\right)^{\{\vartheta\}+m}-2}{\{\vartheta\}+m}. \quad (A.5)$$

In the case of integer values of ϑ, from the last expression we have

$$\frac{1}{\tau}t_i(R) \underset{R\gg R_*}{\simeq} \ln\frac{6W_*}{kT} + \ln\frac{R}{R_*} + \sum_{m=1}^{\vartheta+1} \frac{\left(\frac{R}{R_*}\right)^m-2}{m}, \quad (A.6)$$

which corresponds to (11)-(13).

If the discrete nature of the moleculer in a nucleus is taken into account, we can write the growth law (10) in the form

$$R = \frac{R_*}{\tau_i}a^{-1}\left(1-\exp\left[-a\left(1-\frac{R_*}{R}\right)\right]\right) \quad (\vartheta=0). \quad (A.7)$$

Expanding the exponent for $R \gg R_*$ and retaining only the terms which make a divergent contribution to the integral, we find

$$\frac{1}{\tau}t_i'(R) \underset{R\gg R_*}{\simeq} \ln\frac{6W_*}{kT} + \frac{ae^a}{e^a-1}\frac{R}{R_*} + e^a\left(\frac{a}{e^a-1}\right)^2\ln\frac{R}{R_*}. \quad (A.8)$$

The generalization to the case of arbitrary ϑ is obvious.

In contrast with relations (A.5) and (A.6), which have an accuracy of $O(R_*/R)$, the accuracy of (A.8) is roughly $O(1)$ (we assume that $a \simeq 1$); the asymptotically large term $\ln 6W_*/kT$ is the only constant that has been retained.

A relation such as (A.8) can be used only when the condition $R \gg R_*$ is satisfied sufficiently well, e.g., if R is of the order of the size of the nuclei which greatly deplete the initial phase (t_i is of the order of the "lifetime" of the metastable state). The discrete nature of the molecules can

be taken into account through extremely crude approximation of the results for R_*/R. When this quantity is not two small (as was the case in the experiments under consideration), it is desirable to use relations of the type (11)-(13) or (A.5).

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[1])Such relations also arise in situations in which the nucleus can be described by a continuously varying scalar order parameter.[23a]

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