Hopping transport in the presence of site-energy disorder: Temperature and concentration scaling of conductivity spectra

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(Received 27 August 1999)

Recent measurements on ion-conducting glasses have revealed that conductivity spectra for various temperatures T and ionic concentrations n can be superimposed onto a common master curve by an appropriate rescaling of the conductivity and frequency. In order to understand the origin of the observed scaling behavior, we investigate by Monte Carlo simulations the diffusion of particles in a lattice with site energy disorder for a wide range of both T and n values. While the model can account for the changes in ionic activation energies upon changing n, it in general yields conductivity spectra that exhibit no scaling behavior. However, for typical n and sufficiently low T values, a fairly good data collapse is obtained analogous to that found in experiment.

I. INTRODUCTION

Electrical conduction processes in disordered solids show a remarkable similiarity with respect to their low-frequency dielectric loss behavior.¹ For frequencies ω smaller than some crossover frequency τ^{-1} , the real part $\sigma'(\omega)$ of the complex conductivity $\hat{\sigma}(\omega) = \sigma'(\omega) + i\sigma''(\omega)$ is constant $[\sigma'(\omega) = \sigma(0) = \sigma_{dc}$ for $\omega \tau \ll 1$], while for higher frequencies $\omega \tau > 1$, $\sigma'(\omega)$ increases monotonously with ω , until at typical phonon frequencies above 100-GHz vibrational contributions become dominant. The dispersive part can be characterized by *approximate* power laws, $\sigma'(\omega) \sim \omega^s$, with exponents $s \approx 0.6 - 0.8$, where s tends to increase weakly with increasing frequency (at fixed temperature T) and decreasing temperature (at fixed frequency ω). In fact, at low temperatures or high frequencies the exponents s becomes close to one. Alternatively, this general behavior may be discussed in terms of the complex dielectric function $\epsilon(\omega) = \epsilon'(\omega)$ $+i\epsilon''(\omega) = \epsilon_{\infty} + i\hat{\sigma}(\omega)/\epsilon_0\epsilon_{\infty}$, where ϵ_0 is the vacuum permittivity and ϵ_{∞} the high-frequency dielectric constant.

For ionically conducting glasses in particular, it was pointed out by Isard² already in 1962 that τ^{-1} and $\sigma(0)$ are proportional with an almost universal constant of proportionality. More precisely, it was found that the peak frequency ω_p in the "dielectric polarization part" $[\sigma'(\omega) - \sigma(0)]/\omega$ of the loss spectrum is proportional to $\sigma(0)$. However, since any reasonable definition of τ^{-1} yields values comparable to ω_p , we do not distinguish between ω_p and τ^{-1} here. A closer inspection of the relation between τ^{-1} and $\sigma(0)$ was done later independently by Barton, Nakajima, and Namikawa³ (BNN) on a variety of ionic and transition metal oxide glasses with the result

$$\sigma(0)\tau = p\epsilon_0 \Delta \epsilon', \qquad (1)$$

where *p* is a material-dependent constant of order one and $\Delta \epsilon' = \epsilon'(0) - \epsilon_{\infty} = -\epsilon_0^{-1} \lim_{\omega \to 0} \sigma''(\omega)/\omega$ is the dielectric loss strength. Equation (1) is commonly referred to as the BNN relation.

The question if and to what extent these common features of conductivity spectra in disordered solids can be cast into a universal scaling form has been debated for a long time. In fact, if τ is the only relevant time scale in the system and $\Delta \epsilon' \propto n/T$ with *n* being the charge-carrier concentration (this is the simplest behavior one can expect based on the fluctuation-dissipation theorem), the BNN relation suggests a scaling form

$$\frac{\hat{\sigma}(\omega)}{\sigma(0)} = F(\omega\tau), \quad \tau = \frac{\alpha n}{\sigma(0)T}, \quad (2)$$

where α is a constant of order unity. This type of scaling behavior will henceforth be referred to as the BNN scaling.

Various theories of charge-carrier transport in disordered systems have been developed over the past 30 years, and already within the classical pair approximation⁴ and the continuous-time random-walk (CTRW) model⁵ the possibil-

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ity of scaling was considered. The problem has been investigated in more detail then by a number of authors. Summerfield suggested,⁶ based on the extended pair approximation (EPA),⁷ that the real part $\sigma'(\omega)$ in doped crystalline and amorphous semiconductors should follow Eq. (2) (but without considering the dependence of τ on the charge-carrier concentration n). Tests of this scaling ansatz for data on evaporated amorphous germanium (for 51 K<T<102 K) and doped crystalline silicon (for 1 K<T<10 K) in a frequency range 10^{-3} – 10^{5} kHz, showed a good data collapse for various temperatures and indeed almost the same scaling function $F(\omega \tau)$ for doped crystalline silicon and amorphous germanium.⁸ Hunt⁹ suggested a scaling behavior for the real part $\sigma'(\omega)$ using an "augmented pair approximation" at intermediate frequencies and a "cluster theory" at low frequencies, and discussed the degree of universality of the BNN relation.

Dyre¹⁰ studied a special electric-circuit model resembling a discretized form of Maxwell's equations for a medium with spatially fluctuating dielectric function. By treating this model in the effective-medium approximation (EMA), he showed that at low temperatures the complex conductivity $\hat{\sigma}(\omega)$ can be written as

$$\frac{\hat{\sigma}(\omega)}{\sigma(0)}\log\left(\frac{\hat{\sigma}(\omega)}{\sigma(0)}\right) = i\,\omega\,\tau,\tag{3}$$

where $\tau \propto 1/T \sigma(0)$. This low-temperature limit of the EMA was first derived by Bryksin¹¹ and verified to hold true for various distributions of the impedances in the electric circuit model,¹⁰ thus providing a possible explanation for the experimentally observed "quasiuniversality." Bryksin's equation (3) also results from an EMA treatment of a microscopic hopping model^{12,13} with uniformly distributed random freeenergy barriers at low *T* (see also Ref. 14), and it predicts a unique scaling function F(z) being determined by $F(z)\log F(z)=iz$.

The Bryksin scaling function was shown to describe the normalized real part $\sigma'(\omega)/\sigma(0)$ of various materials fairly well,¹⁵ both for electronic conductors (e.g., *n*-doped crystalline silicon or amorphous germanium) and ionic conductors (e.g., sodium silicate glass or the glass former 0.4CaNO₃.0.6KNO₃). It must be noted, however, that the EMA fails to predict the correct dc conductivity $\sigma(0)$. The low-temperature activation energy $E_a \propto -\log[T\sigma(0)]$ resulting from the EMA is not in accordance with the value obtained from a critical percolation-path analysis,¹⁶ which is known to become exact at low temperatures¹⁷ (see also the results in Sec. III below). Only with a rescaled dc conductivity may the EMA be regarded as a valuable concept to describe conductivity spectra in strongly disordered systems.

For a long time the BNN scaling (2) has never been tested with respect to changes in charge-carrier concentration *n*, until Roling *et al.* recently studied $\hat{\sigma}(\omega)$ for various ionically conducting glasses^{18,19} (see also the recent study of Sidebottom²⁰). In these materials, in contrast to most other systems, the concentration of charge carriers (mobile ions) can be varied to a large extent. Roling *et al.* found that the *real parts* of conductivity spectra in sodium borate glasses with compositions xNa₂O(1-x)B₂O₃ (and also other types of ionic glasses¹⁹) can be superimposed fairly well onto a single curve²¹ not only for various temperatures, but also for various modifier contents $x \sim n$. However, it was found also¹⁹ that the dielectric strength scales as $\Delta \epsilon' \propto x^{1/3} \sim n^{1/3}$, in contrast to the assumption $\Delta \epsilon' \propto n/T$ made when deriving the BNN scaling (2). This implies that the imaginary part cannot obey the BNN scaling because of causality requirements (Kramers-Kronig relations). Moreover, in Ref. 20 alkali germanate glasses have been studied over *n* values varying by a factor of ten, and it was found that the BNN scaling has to be modified in order to obtain a data collapse for this large concentration range. We therefore have to consider the observed data collapse in $\sigma(\omega)$ as an effective one.

The effective scaling behavior is nevertheless quite remarkable, since the activation energy E_a of the dc conductivity $\sigma(0)$ [and hence the activation energy for $\tau^{-1} \propto \sigma(0)$] usually decreases logarithmically^{22,23} with ionic concentration n, $E_a = A - B \log n$, for typical $n \approx 10^{21} - 10^{22}$ cm⁻³. Accordingly, one finds a quite complicated explicit dependence of the crossover time τ on n, $\tau \sim n^{1-B/k_{\rm B}T}$.

It is important to note that these experimental findings for ionically conducting glasses cannot be explained by the random free-energy barrier model, since in this model the conductivity is proportional to n but does not steeply increase with *n* as $\sigma(0) \sim n^{B/k_{\rm B}T}$. The perhaps simplest approach that allows one to account for a concentration dependence of E_{a} is a model of particles hopping in a lattice with site energy disorder (see Refs. 24-26 and below), where only one particle can occupy a given lattice site (Fermi statistics). In order to see if such a model also yields the BNN scaling (2), we perform Monte Carlo simulations for Gaussiandistributed site energies in a range of concentrations n and temperatures T both extending over almost two orders of magnitude. We find that the model can account for the logarithmic dependence of E_a on n, but in general exhibits no effective BNN scaling behavior corresponding to Eq. (2). However, for typical ionic concentrations and sufficiently low temperatures an effective BNN scaling behavior is found in agreement with experiment.

II. HOPPING MOTION AND TRANSPORT QUANTITIES

The jump motion of particles in a lattice with site-energy disorder is a standard model for describing hopping transport in disordered systems.²⁷ We choose here a simple cubic lattice with spacing a and assign to each lattice site i independently a random energy ε_i drawn from a Gaussian distribution $\psi(\varepsilon)$ with zero mean and variance σ_{ε}^2 . The particles jump among nearest-neighbor sites and the jump rate from site *i* to a vacant neighboring site *j* is $W(\varepsilon_i, \varepsilon_j)$ $= \nu \min(1, \exp[-(\varepsilon_i - \varepsilon_i)/k_{\rm B}T])$ with ν being an attempt frequency (Metropolis transition rates). If the neighboring site *j* is occupied, the jump rate is zero. The size of the lattice is L^3 and periodic boundary conditions are used. At equilibrium, the probability for a site with energy ε to be occupied is given by the Fermi distribution $f(\varepsilon) = [1 + \exp[(\varepsilon) +$ $-\mu/k_{\rm B}T$]⁻¹, where the concentration $c = na^3$ per lattice site determines the chemical potential through c $=(a/L)^3 \Sigma_i f(\varepsilon_i) \cong \int d\varepsilon \, \psi(\varepsilon) f(\varepsilon)$. For $T \to 0$, $f(\varepsilon)$ approaches the step function $f(\varepsilon) = \theta [\varepsilon_f(c) - \varepsilon] [\theta(x) = 1$ for $x \ge 0$ and zero else] with the Fermi energy $\varepsilon_{\rm f}(c)$ given by



FIG. 1. Plot of (a),(d) $D(t)/D_0$ vs νt , (b),(e) $\sigma'(\omega)/\sigma_0$ vs ω/ν , and (c),(f) $\sigma''(\omega)/\sigma_0$ vs ω/ν ($\sigma_0 = ne^2 D_0/k_B T$). In (a)–(c) the concentration is fixed, c = 0.02, and the temperature is varied, $\sigma_{\varepsilon}/k_B T = 2$ (full squares), 3 (open diamonds), 4 (full triangle), 5 (open stars), 6 (full circles), and 7 (open squares), while in (d)–(f) the temperature is fixed, $\sigma_{\varepsilon}/k_B T = 6$, and the concentration is varied, c = 0.005 (full stars), 0.01 (open circles), 0.02 (full squares), 0.04 (open diamonds), 0.08 (full triangles), and 0.16 (open stars). The solid lines in (a) and (d) represent the fits used for the Laplace transform.

$$\operatorname{erf}[\varepsilon_{\mathrm{f}}(c)/2^{1/2}\sigma_{\varepsilon}] = 2c - 1, \qquad (4)$$

where $erf(\cdot)$ denotes the error function.

In order to analyze the mobility of the particles for various concentrations c and temperatures T we determine the mean-square displacement $\langle r^2(t) \rangle$ of a tracer particle by means of Monte Carlo simulations,²⁸ and calculate the time-dependent diffusion coefficient $D(t) = \langle r^2(t) \rangle / 6t$. D(t) approaches the short-time diffusion constant $D_{\rm st}$ for $t \to 0$, and the long-time diffusion constant D_{∞} for $t \to \infty$. The frequency-dependent conductivity $\hat{\sigma}(\omega)$ is, to a good approximation,²⁹ given by

$$\hat{\sigma}(\omega) = -\frac{ne^2}{k_{\rm B}T} \frac{\omega^2}{6} \lim_{\epsilon \to 0^+} \int_0^\infty \langle r^2(t) \rangle \exp(i\omega t - \epsilon t) dt, \quad (5)$$

and approaches $\sigma(0) = (ne^2/k_{\rm B}T)D_{\infty}$ and $\sigma(\infty) = (ne^2/k_{\rm B}T)D_{\rm st}$ in the low- and high-frequency limits, respectively.

The short-time or high-frequency dynamics is governed by the mean jump rate of a particle,

$$\langle W \rangle = \int d\varepsilon \, \frac{\psi(\varepsilon)f(\varepsilon)}{c} \int d\varepsilon' \, \psi(\varepsilon') \left[1 - f(\varepsilon')\right] W(\varepsilon, \varepsilon'), \tag{6}$$

which determines $D_{st} = \langle W \rangle a^2/6$. For $k_B T \gg |\varepsilon_f(c)|$ we can use the high-temperature expansion of the chemical potential, $\mu = k_B T \{ \log[c/(1-c)] + (c-1/2)(\sigma_{\varepsilon}/k_B T)^2 + O(\sigma_{\varepsilon}/k_B T)^3 \}$ to evaluate Eq. (6) and obtain

$$\langle W \rangle \simeq \nu (1-c) \left(1 - \frac{\sigma_{\varepsilon}}{\pi^{1/2} k_{\rm B} T} \right).$$
 (7)



FIG. 2. (a) Plot of cD_{∞}/D_0 vs $\sigma_{\varepsilon}/k_BT$ for concentrations c = 0.001 (open triangles) 0.005 (full stars), 0.01 (open circles), 0.02 (full squares), 0.04 (open diamonds), 0.08 (full triangles), and 0.16 (open stars). The solid line indicates the high-temperature activation energy (Ref. 33) $E_0 = \sigma_{\varepsilon}/\pi^{1/2}$. The dashed lines are the fits used to determine the activation energy E_a . (b) Plot of the activation energy E_a/σ_{ε} vs c. The open circles represent the values obtained from the Monte Carlo simulation shown in (a), while the solid line marks the result from the critical path analysis. The dashed line is a fit with respect to a logarithmic dependence of E_a on c, $E_a = A - B \log(c)$ with $A \cong 0.23$ and $B \cong 0.37$.



FIG. 3. Scaling plot of (a),(d) $D(t)/D_{\infty}$ vs t/τ , (b),(e) $\sigma'(\omega)/\sigma_{dc}$ vs $\omega\tau$, and (c),(f) $\sigma''(\omega)/\sigma_{dc}$ vs $\omega\tau$, for the data presented in Fig. 1.

At low temperatures $k_{\rm B}T \leq |\varepsilon_{\rm f}(c)|$ we can replace $f(\varepsilon)$ by the step function in Eq. (6) and it follows that $\langle W \rangle = (4\nu/c) \exp[(\sigma_{\varepsilon}/k_{\rm B}T)^2] \operatorname{erfc} E_{-}\operatorname{erfc} E_{+}$ with $E_{\pm} = 2^{-1/2} (\sigma_{\varepsilon}/k_{\rm B}T \pm \varepsilon_{\rm f}(c)/\sigma_{\varepsilon})$ [erfc(·)=1-erf(·) denotes the complementary error function]. For $k_{\rm B}T \leq \min(\sigma_{\varepsilon}^2/|\varepsilon_{\rm f}(c)|, |\varepsilon_{\rm f}(c)|)$, in particular, we then find, by using $\operatorname{erfc}(x) \sim \exp(-x^2)/\pi^{1/2}x$ for $x \to +\infty$,

$$\langle W \rangle \simeq \frac{\nu}{2 \pi c} \left(\frac{k_{\rm B} T}{\sigma_{\varepsilon}} \right)^2 \exp[-\varepsilon_{\rm f}(c)^2 / \sigma_{\varepsilon}^2].$$
 (8)

Note that $\langle W \rangle$ is not thermally activated in this lowtemperature limit. This is a desirable feature of the model, since the high-frequency conductivity is indeed only weakly temperature dependent in ionically conducting glasses (and other disordered hopping systems).

By contrast, the long-time diffusion constant or dc conductivity is thermally activated at low temperatues, $D_{\infty} \propto \sigma(0) \propto \exp(-E_a/k_BT)$. The activation energy E_a can, at sufficiently low $c(\{\psi[\varepsilon_f(c)]k_BT\}^{1/3} \ll 1)$, be calculated from the critical path analysis¹⁶ as the difference between the critical energy ε_c and the Fermi energy $\varepsilon_f(c)$, i.e., $E_a = \varepsilon_c$ $-\varepsilon_{\rm f}(c)$. Here, $\varepsilon_{\rm c}$ is the highest energy, which at least has to be encountered by a particle when it wants to move between two (far distant) sites close to the Fermi level. According to percolation theory^{30,31} $\int_{-\infty}^{\varepsilon_{\rm c}} \psi(\varepsilon) d\varepsilon = [1 + {\rm erf}(\varepsilon_{\rm c})]/2 = p_{\rm c}$, where $p_{\rm c} \approx 0.3117$ denotes the percolation threshold in the simple cubic lattice. Solving numerically for ε_c we obtain $\varepsilon_c \approx -0.491\sigma_{\varepsilon}$ and hence

$$E_{\rm a} \simeq -0.491 \sigma_{\varepsilon} - \varepsilon_{\rm f}(c). \tag{9}$$

For $c \to 0$, $\varepsilon_{\rm f}(c) \to -\infty$, such that the implicit equation $\operatorname{erf}[\varepsilon_{\rm f}(c)/2^{1/2}\sigma_{\varepsilon}] = 2c-1$ for $\varepsilon_{\rm f}(c)$ can be simplified to $\exp\{-[\varepsilon_{\rm f}(c)/(2^{1/2}\sigma_{\varepsilon})]^2\} \simeq -2\pi^{1/2}c[\varepsilon_{\rm f}(c)/(2^{1/2}\sigma_{\varepsilon})]$, which yields

$$\varepsilon_{\rm f}(c)/\sigma_{\varepsilon} \simeq -2^{1/2} [-\log(2\pi^{1/2}c) - \log(2\pi^{1/2}c)]^{1/2}]^{1/2}$$

III. RESULTS AND DISCUSSION

Figure 1 shows the Monte Carlo results for D(t) (in units of $D_0 \equiv \nu a^2$) and the real and imaginary parts $\sigma'(\omega)$ and $\sigma''(\omega)$ of the conductivity (in units of $\sigma_0 \equiv ne^2 D_0 / k_{\rm B}T$) be-



FIG. 4. Scaling plot of (a) $D(t)/D_{\infty}$ vs t/τ , (b) $\sigma'(\omega)/\sigma_{dc}$ vs $\omega\tau$, and (c) $\sigma''(\omega)/\sigma_{dc}$ vs $\omega\tau$, for $(c,\sigma_{\varepsilon}/k_{B}T) = (0.02,6)$ (open triangles), (0.02,7) (open stars), (0.04,6) (open circles), (0.04,7) (open squares), (0.08,7) (open diamonds), and (0.08,8) (open upside-down triangles).

ing calculated from D(t) by numerical a Laplace transformation according to Eq. (5). In the upper figures [Figs. 1(a)-1(c)], the curves refer to a fixed concentration c = 0.02 and various (inverse) temperatures ranging from $\sigma_{e}/k_{\rm B}T=2$ to 7, while in the lower figures [Figs. 1(d)-1(f)] the temperature is held fixed, $\sigma_{\varepsilon}/k_{\rm B}T=6$, and the concentration is varied between c = 0.005 and c = 0.16. The overall behavior of the complex conductivity $\hat{\sigma}(\omega)$ compares fairly well with that found in the experiments. There exists a crossover time τ , such that the real part $\sigma'(\omega)$ is almost constant for $\omega \tau \ll 1$, $\sigma'(\omega) \simeq \sigma(0)$. For $\omega \tau > 1$, $\sigma'(\omega)$ increases monotonously with ω until for $\omega \simeq \omega_{\rm hf}$ it reaches the high-frequency plateau being determined by $\langle W \rangle$ (see Sec. II). In real materials this high-frequency plateau is usually masked by phonon excitations, but may be resolved by substracting the vibrational contributions.³² The dispersive part of the spectrum for τ^{-1} $\ll \omega \ll \omega_{\rm hf}$ can be described by an approximate power law at sufficiently low temperatures $[\sigma_{\epsilon}/k_{\rm B}T \lesssim 4, \text{ see Fig. 1(b)}]$ with typical exponents s in the range 0.5–0.8. The imaginary part $\sigma''(\omega)$ behaves similar to $\sigma'(\omega)$ in the dispersive regime $\tau^{-1} \ll \omega \ll \omega_{\rm hf}$, i.e., $\sigma''(\omega) \sim \omega^s$, while in the dc- and high-frequency limits it rapidly approaches zero, since the phase shift between current and electric field approaches rapidly 0 and π , respectively.

Figure 2(a) displays Arrhenius plots of $\sigma_{dc}T \propto cD_{\infty}/D_0$ for various c. The linear behavior of the curves at low $k_{\rm B}T$ $\ll \sigma_{\varepsilon}$ (see the dashed lines in the figure) allows us to determine³³ the activation energy E_a whose dependence on cis shown in Fig. 2(b) (in a semilogarithmic plot). This dependence is in good agreement with the behavior predicted by the critical path analysis [see the solid line in Fig. 2(b)]. As indicated by the dashed line in Fig. 2(b), the data for c ≥ 0.01 can be approximated by a logarithmic dependence $E_a/\sigma_{\epsilon} = A - B \log c$, in accordance with the experimental findings. We also see that for low concentrations $c \leq 0.01$, the data start to fall off the straight dashed line. This is expected to occur in view of the low-c limit of $\varepsilon_{\rm f}(c)$ discussed after Eq. (9). The behavior is not in contradiction with the experimental situation, however, since in ionically conducting glasses the logarithmic c dependence of E_a is typically found at relatively high ionic concentrations (corresponding

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to network modifier contents of $5-40 \mod \%$ which are most interesting for applications).

Let us now discuss as to what extent the data can be superimposed by a BNN scaling ansatz as in Eq. (2). To this end the data in Fig. 1 are plotted in a scaled form in Fig. 3. As can be seen from Figs. 3(a)-3(c), the data for fixed *c* and various *T* do not collapse onto one master curve. The BNN scaling with respect to various concentrations at fixed (low) temperature *T* is significantly better [Figs. 3(d)-3(f)], but it is important to note that this approximate scaling behavior ceases to be present at higher *T*, $\sigma_{\varepsilon}/k_{\rm B}T \lesssim 5$. We conclude that the hard-core lattice gas with Gaussian site-energy disorder in general exhibits no BNN scaling.

However, the data collapse for the lowest temperatures $(\sigma_{\varepsilon}/k_{\rm B}T=6 \text{ and } 7)$ in Figs. 3(a)–3(c) is fairly good. In order to see if this holds true also for other c values, let us first estimate what is a typical range of ionic concentrations used in the experiments. For this purpose one might consider sodium silicate glasses $x \operatorname{Na}_2 O(1-x) \operatorname{SiO}_2$ with modifier contents x between 10% and 40%. By assuming about 10 sites per silicon to exist, we would have c = 2x/[10(1-x)] ≈ 0.01 and $c \approx 0.13$ for x = 0.1 and x = 0.4, respectively. For these c values we always found an approximate BNN scaling for $\sigma_{\varepsilon}/k_{\rm B}T \gtrsim 6$. A scaling plot combining various low T and typical c values is shown in Fig. 4. The quality of the approximate data collapse is not as good as the one found in experiment, but might become better at lower T. In fact, when considering typical activation energies for ionic conduction in glasses of 0.3 to 1 eV, one should assume similar values for σ_{ε} [see Fig. 2(b)], and accordingly $\sigma_{\varepsilon}/k_{\rm B}T \leq 10$ at room temperature (this, however, is true only when neglecting the Coulomb interaction between the mobile ions, see Ref. 34).

ACKNOWLEDGMENTS

We thank K. Funke, M. D. Ingram, and B. Roling for very helpful discussions. M.P. and P.M. gratefully acknowledge financial support from the Alexander-von-Humboldt Foundation (Feodor-Lynen Program) and the Deutsche Forschungsgemeinschaft (Ma 1636/2-1), respectively.

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