Stochastic derivation of the switching function in the theory of microwave heating of ceramic materials

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(Received 16 December 1991)

Stochastic arguments are presented to provide a fundamental derivation of a switching function introduced in a recent theory of microwave heating of ceramic materials. The phenomenon under study is thermal runaway wherein the temperature of a ceramic material increases suddenly after being heated under microwaves for a certain time. Observations in a wide variety of materials have been explained successfully via a recently constructed theory of the pheonomenon, which suggests that the mobility of absorbing entities, such as vacancies, bivacancies, or interstitials, has a temperature dependence that follows a switching behavior: The mobility, equivalently the absorption coefficient, starts at small values for small temperatures but rises quickly to high values as the temperature is increased. Phase-space considerations and static arguments have been given earlier to support the idea, and to calculate the detail, of the switching function. Here we provide a basic justification to the static arguments through a consideration of the dynamics. Although the evolution of the absorbing entities is governed by a nonlinear Langevin equation, which cannot be solved exactly, natural approximations are shown to lead to a Smoluchowski equation for the probability distribution, which can be solved. General results and specific calculational prescriptions are provided for a variety of potential shapes.

I. INTRODUCTION

Recent observations of beneficial effects of microwaves on the sintering of some materials, as reported, e.g., by Sheppard,[1] Bruce,[2] and Katz *et al.*,[3] have led to a revival of interest in the general subject of the interaction of microwaves with ceramics. One of the relevant phenomena under investigation consists of a sudden rise in the temperature of a material undergoing microwave heating, and is termed "thermal runaway." A theory of this phenomenon has been presented recently by Kenkre *et al.*[4,5] on the basis of a few simple concepts. The theory has addressed, with considerable success, a variety of time-temperature curve shapes in several different materials, such as strontium titanate, zinc oxide, iron oxide, and alumina. The primary purpose of the present paper is to provide a basic justification for a central conjecture made in that theory. However, our procedure and results turn out to have relevance also to the general problem of the calculation of transport coefficients of classical particles subjected to nonlinear potentials.

Underlying the theory in Refs. 4 and 5, which motivated this work, is the idea of the existence of absorbing entities in the material, such as vacancies, bivacancies, or interstitials, whose number available for absorption switches from a small value at small temperatures to a large value at higher temperatures, the resulting temperature-time relation being

$$\frac{dT}{dt}=[k_A+k_M f(T)]P-\sigma_1 T^4 . \tag{1.1}$$

Here k_A and k_M are related to the absorption coefficients of two different species of absorbers, P is the incident microwave power, and σ_1 is proportional to the Stephan-Boltzmann constant. The fraction of the absorbers of the M kind available for absorption is denoted by $f(T)$. The peculiar runaway nature of the heating arises generally from the "switching" nature of $f(T)$, which, in its simplest form, may be written as[4]

$$f(T)=2\frac{e^{-\Delta/kT}}{1+e^{-\Delta/kT}} , \tag{1.2}$$

Δ being an energy barrier and k the Boltzmann constant. The factor 2 shown explicitly in (1.2) represents a normalization such that $f(T)$ changes from the value 0 to the value 1 as the temperature increases from zero to infinity.

The argument provided in Refs. 4 and 5 to arrive at (1.1) and (1.2) is as follows. If the M absorbers each have mass m and electric charge q, their dynamics under an applied microwave field of amplitude E and angular frequency ω are governed by[4,5]

$$m\frac{d^2x}{dt^2}+m\gamma\frac{dx}{dt}+\frac{dU(x)}{dx}=qE\cos(\omega t)+R(t) , \tag{1.3}$$

where x is the coordinate of the particle and γ the effective damping constant. The important characteristic of the evolution equation (1.3) is the presence of two forces to which the absorber is subjected as a result of interactions with the rest of the lattice: the systematic force due to the *nonlinear* potential $U(x)$ and the stochastic force due to thermal fluctuations represented by $R(t)$. A typical form of $U(x)$ is sinusoidal, the characteristic feature being a periodic repetition of a region of space in which the absorber is bound and a region in which it is free, the amplitude of the potential being Δ. Absorption is significantly higher in the free region relative to the bound region. As a result, the absorption coefficient is proportional to the fraction of the absorbers which have energy greater than Δ. Equation (1.2) for $f(T)$ is then a consequence of the simple assumption that

the regions accessible to the absorbers may be regarded as just two states. Those in the higher-energy state are free and contribute to the absorption, whereas the rest, being in the bound state, do not, or do so negligibly. A further assumption, that standard Boltzmann reasoning regarding the relative population in the two states is applicable, then leads to (1.2).

More elaborate forms of $f(T)$ have also been provided.[6] The application of phase-space considerations without the simplifying assumption that the accessible states are only two, leads to

$$f(T)=\frac{\int_{\text{free region}} d\mathbf{x}\,d\mathbf{p}\exp\{-[\mathbf{p}\cdot\mathbf{p}/2m+U(\mathbf{x})]/kT\}}{\int_{\text{entire region}} d\mathbf{x}\,d\mathbf{p}\exp\{-[\mathbf{p}\cdot\mathbf{p}/2m+U(\mathbf{x})]/kT\}}\ , \tag{1.4}$$

where $\mathbf{p}$ is the momentum variable, $\mathbf{x}$ is the position variable, and by "free region" we understand the portion of the phase space corresponding to the total energy being greater than the maximum of the potential energy, viz., Δ. The integrations over $\mathbf{p}$ can be carried out easily to write $f(T)$ in terms of $\mathbf{x}$ integrals involving complementary error functions. Thus, for a one-dimensional system, (1.4) can be shown to reduce to[5,6]

$$f(T)=\frac{\int_{\text{entire region}} dx\, e^{-U(x)/kT}\operatorname{erfc}\sqrt{[-U(x)/kT]}}{\int_{\text{entire region}} dx\, e^{-U(x)/kT}}\ . \tag{1.5}$$

The forms of the switching function $f(T)$, given in the theory of Refs. 4 and 5, and shown above in (1.2), (1.4), and (1.5), are surely reasonable. Given the physical arguments[4,5] which support them and also the success of the theory in explaining observations on a wide variety of materials, there is no doubt about the validity of their qualitative T dependence. However, they must be regarded as the result of a conjecture with *static* considerations as their support. In the following we present a derivation of the switching function on the basis of the dynamics of the absorbers by attempting a direct solution of (1.3). The rest of the paper is laid out as follows. In Sec. II we set out the general framework of our approach and obtain the Fokker-Planck equation relevant to (1.3), as well as the Smoluchowski equation appropriate in the large damping limit. In Sec. III we derive expressions for the mobility and the switching function for constant fields but arbitrary potentials, and study specific cases of physical interest. An examination of some general questions constitutes Sec. IV, and conclusions form Sec. V.

II. STOCHASTIC CALCULATIONS: FOKKER-PLANCK AND SMOLUCHOWSKI EQUATIONS

Following standard procedure, we assume that the random force $R(t)$ is a Gaussian, stationary stochastic process with zero mean,

$$\langle R(t)\rangle=0 \tag{2.1}$$

and that its correlation function is given by

$$\langle R(t)R(t')\rangle=\Gamma\delta(t-t')\ . \tag{2.2}$$

Angular brackets represent the average over the ensemble of realizations of the stochastic process $R(t)$. The above assumptions about the $R(t)$ process are in accordance with our understanding of the random force as resulting from many independent and fast interactions with the surroundings. The strength of the correlation Γ and the damping constant γ are connected via the fluctuation-dissipation theorem

$$\Gamma=2m\gamma kT\ . \tag{2.3}$$

Relation (2.3) follows from the fact that, in the absence of any deterministic external force, such as the microwave field force in (1.3), the system should come to rest in a state of thermal equilibrium and fulfill the principle of equipartition of energy, i.e.,

$$\frac{m\langle v^2\rangle}{2}=\frac{kT}{2}\ . \tag{2.4}$$

On rewriting the equation of motion (1.2) as a system of two differential equations of the first order,

$$\frac{dx}{dt}=v\ , \tag{2.5}$$

$$\frac{dv}{dt}+\gamma v=-\frac{1}{m}\frac{dU}{dx}+\frac{qE}{m}\cos(\omega t)+\frac{1}{m}R(t)\ , \tag{2.6}$$

it is straightforward[7] to obtain the corresponding Fokker-Planck equation for the time evolution of the probability distribution $P(x,v,t)$ of the phase-space variables x and v:

$$\frac{\partial P}{\partial t}=-v\frac{\partial P}{\partial x}+\left[\frac{dU}{dx}-qE\cos(\omega t)\right]\frac{\partial P}{\partial v}+\gamma\frac{\partial}{\partial v}(vP)+kT\frac{\partial^2 P}{\partial v^2}\ . \tag{2.7}$$

The multidimensional partial differential equation (2.7) is extremely hard to solve in the general case. Fortunately, the characteristics of the system under consideration allow several natural simplifications. The first of these characteristics is that the damping constant γ in (1.3), (2.6), and (2.7) is very large. Thus, the natural assumption to make is to neglect the time derivative of v in comparison with the term γv in (2.6) and obtain

$$v(x,t)=-\frac{1}{m\gamma}\frac{dU(x)}{dx}+\frac{qE}{m\gamma}\cos(\omega t)+\frac{1}{m\gamma}R(t)\ . \tag{2.8}$$

When substituted in (2.5), the expression (2.8) yields a single Langevin equation for the evolution of the variable x:

$$\frac{dx(t)}{dt}+\frac{1}{m\gamma}\frac{dU(x)}{dx}-\frac{qE}{m\gamma}\cos(\omega t)=\zeta(t) \tag{2.9}$$

with

$$\langle\zeta(t)\rangle=0,\quad \langle\zeta(t)\zeta(t')\rangle=\frac{2kT}{m\gamma}\delta(t-t') . \tag{2.10}$$

The corresponding Fokker-Planck equation for the probability distribution $P(x,t)$ of the position variable x reads

$$\frac{\partial P}{\partial t}=\frac{1}{m\gamma}\frac{\partial}{\partial x}\left[\left[\frac{dU}{dx}-qE\cos(\omega t)\right]P+kT\frac{\partial P}{\partial x}\right] \tag{2.11}$$

and is known in the theory of Brownian motion as the Smoluchowski equation.[7] We take (2.11) as the point of departure for the rest of the analysis in the present paper.

In order to calculate the switching function from (2.11), we make the following observations. The average power absorbed from the microwave field is proportional to the factor $k_M f(T)P$ appearing in (1.1). The calculation of the switching function is thus equivalent to the calculation of the average power absorbed. The latter is proportional to the field amplitude and to the average of the velocity of the absorber over the stochastic process. Calculating this average, multiplying it by the field amplitude, obtaining the time average over the microwave period, and dividing the result by the square of the electric field, yields the switching function unambiguously. The average of the velocity, viz., $\langle v(t)\rangle$, is given in terms of $P(x,t)$ as

$$\langle v(t)\rangle=\int dx\, vP(x,t) , \tag{2.12}$$

where the velocity v in the integrand is expressed through its dependence on x given in (2.8). Equations (2.11) and (2.12) give $\langle v(t)\rangle$ as depending parametrically on the magnitude of the electric field:

$$\langle v(t)\rangle=\langle v(t;E)\rangle . \tag{2.13}$$

The program to be followed is thus to solve (2.11), obtain the velocity from (2.13), specifically as

$$\begin{aligned}\langle v(t)\rangle&=-\frac{1}{m\gamma}\left\langle\frac{dU(x)}{dx}\right\rangle+\frac{qE}{m}\cos(\omega t)\\&=-\frac{1}{m\gamma}\int dx\, P(x,t)\frac{dU(x)}{dx}+\frac{qE}{m}\cos(\omega t) ,\end{aligned} \tag{2.14}$$

and to calculate the switching function from the result in the manner described. The solution of the time-dependent Smoluchowski equation (2.11) requires nontrivial labor which we postpone to a forthcoming paper. Here, we will make use of another simplification natural to the system under analysis. The applied field lies in a frequency range which is low enough to justify the neglect of its time dependence in our calculation. We shall, therefore, consider the simpler problem of a dc field, i.e., assume that the field period $2\pi/\omega$ is larger then all other characteristic times involved in the evolution (1.3). We can then obtain the switching function from the *steady-state* solution of the $\omega=0$ counterparts of (2.12):

$$\frac{\partial P}{\partial t}=0=\frac{1}{m\gamma}\frac{\partial}{\partial x}\left[\left[\frac{dU}{dx}-qE\right]P+kT\frac{\partial P}{\partial x}\right] . \tag{2.15}$$

We thus have to find the solution of an *ordinary* differential equation, which we do in a straightforward manner. Similar analyses can be found in the literature in connection with studies of voltage-current characteristics of Josephson junctions and conductance of superionic crystals.[8,9] Equation (2.15) leads to

$$\begin{aligned}P(x)=&A\exp\left[-\frac{U(x)-qEx}{kT}\right]\\&+B\exp\left[-\frac{U(x)-qEx}{kT}\right]\\&\times\int_0^x\exp\left[\frac{U(z)-qEz}{kT}\right]dz .\end{aligned} \tag{2.16}$$

The integration constants A and B are determined from the normalization condition

$$\int_{-a/2}^{a/2}P(x)dx=1 \tag{2.17}$$

and from a boundary condition on $P(x)$. We shall now restrict our analysis explicitly to potentials which are periodic in space with period a. This results in

$$P(x+a)=P(x) , \tag{2.18}$$

the periodicity of $P(x)$ being an obvious consequence of the periodicity of the potential $U(x)$. The substitution of (2.17) and (2.18) in (2.16) leads to two simultaneous equations for the unknowns A and B appearing in (2.16):

$$a_1A+b_1B=1,\quad a_2A+b_2B=0 , \tag{2.19}$$

where the quantities a_1, b_1, a_2, and b_2 are given by

$$a_1=\int_{-a/2}^{a/2}dx\exp\left[-\frac{1}{kT}[U(x)-qEx]\right] , \tag{2.20}$$

$$\begin{aligned}b_1=&\int_{-a/2}^{a/2}dx\exp\left[-\frac{1}{kT}[U(x)-qEx]\right]\\&\times\int_0^x dy\exp\left[\frac{1}{kT}[U(y)-qEy]\right] ,\end{aligned} \tag{2.21}$$

$$a_2=\exp\left[\frac{1}{kT}qEa\right]-1 . \tag{2.22}$$

$$b_2=\int_0^a dx\exp\left[\frac{1}{kT}[U(x)-qE(x-a)]\right] . \tag{2.23}$$

The steady-state version of (2.14) yields, for the mean velocity $\langle v\rangle$,

$$\langle v\rangle=-\frac{akT}{m\gamma}B=\frac{akT}{m\gamma}\frac{a_2}{a_1b_2-a_2b_1} . \tag{2.24}$$

Equation (2.24) describes the full nonlinear dependence of the steady-state velocity on the electric field when the moving particle is under the simultaneous action of a dc field of arbitrary magnitude (which appears in the quantities a_1, b_1, a_2, and b_2), a nonlinear periodic potential of otherwise arbitrary spatial dependence, and normal stochastic interactions characterized by (2.1) and (2.2). In the following analysis we will use (2.24) to obtain expressions for the particle mobility and the switching function.

III. MOBILITY AND THE SWITCHING FUNCTION

While it is certainly possible to evaluate the full expression (2.24) obtained above, microwave field amplitudes applied lie within breakdown limits and are therefore small enough to justify invoking Ohm's law. One may, therefore, expand (2.24) as a function of E and keep only linear terms:

$$\langle v(E)\rangle=\langle v(0)\rangle+\left.\frac{d\langle v(E)\rangle}{dE}\right|_{E=0}E+\cdots=0+\mu E\ . \tag{3.1}$$

The constant term obviously vanishes in equilibrium and the coefficient of the linear term is the mobility μ, an expression for which can be derived immediately from (2.24):

$$\mu=\frac{qa^2}{m\gamma}\left[\int_{-a/2}^{a/2}dx\,\exp\left[-\frac{1}{kT}U(x)\right]\int_0^a dx\,\exp\left[\frac{1}{kT}U(x)\right]\right]^{-1}\ . \tag{3.2}$$

On comparing the result we have derived for the mobility, viz., (3.2), to

$$\mu=\frac{q}{m\gamma}\ , \tag{3.3}$$

which is the well-known Drude result, we observe that, in the presence of stochastic interactions, the simple Drude result merely gets multiplied by a dimensionless temperature-dependent factor. Equation (3.3) is recovered in the limit of infinite temperature or when there is no potential.

Equation (3.2) can now be used to calculate the power absorbed from the microwave field. In the linear approximation, which, as we have stated above, is amply valid in systems of interset to us, the instantaneous power absorbed in one period is given by $\langle v(t)\rangle qE\cos(\omega t)$. The time average over one period of the microwave field is

$$R=\frac{q^2E^2a^2}{2m\gamma}\left[\int_{-a/2}^{a/2}dx\,\exp\left[-\frac{1}{kT}U(x)\right]\int_0^a dx\,\exp\left[\frac{1}{kT}U(x)\right]\right]^{-1}\ . \tag{3.4}$$

Except for proportionality constants such as the specific heat, this average power absorbed is identical to the heating rate, i.e., to the rate of temperature increase arising from the field. A comparison of (3.4) to (1.1) shows then that the temperature-dependent factor appearing in (3.2) and (3.4) is nothing other than the switching function $f(T)$, which is the object of this paper:

$$f(T)=a^2\left[\int_{-a/2}^{a/2}dx\,\exp\left[-\frac{1}{kT}U(x)\right]\int_0^a dx\,\exp\left[\frac{1}{kT}U(x)\right]\right]^{-1}\ . \tag{3.5}$$

Equation (3.5) is the central result of this paper. It gives a prescription to calculate the switching function of the theory of thermal runaway[4,5] from dynamic considerations rather than static phase space arguments.[4–6] Explicit quantities entering the prescription are the temperature, the shape of the potential, and its period. Most situations involve symmetric potentials $U(x)=U(-x)$. For these, the limits of integration in (3.5) can be readjusted:

$$f(T)=a^2\left[4\int_0^{a/2}dx\,\exp\left[-\frac{1}{kT}U(x)\right]\int_0^{a/2}dx\,\exp\left[\frac{1}{kT}U(x)\right]\right]^{-1}\ . \tag{3.6}$$

If the difference between the maximum and minimum values the potential attains is Δ, one may reexpress the potential as

$$U(x)=-\Delta V(2x/a)\ , \tag{3.7}$$

where $V(y)$, which we call the reduced potential, is a periodic function which varies between the values 0 and 1 as its argument y runs over its period $[-1,1]$. The switching function can then be written in the compact form

$$f(T)=\left[\int_0^1 dy\,\exp\left[-\frac{\Delta}{kT}V(y)\right]\int_0^1 dy\,\exp\left[\frac{\Delta}{kT}V(y)\right]\right]^{-1}\ . \tag{3.8}$$

We end this section by considering three physically motivated examples of the potential $U(x)$, and presenting the corresponding forms of the switching function obtained by applying (3.8).

(a) Sinusoidal potential. The potential is given by

$$U(x)=-\frac{\Delta}{2}\left[1+\cos\left[\frac{2\pi x}{a}\right]\right] \tag{3.9}$$

and corresponds, via (3.7), to the reduced potential

$$V(y)=\tfrac{1}{2}[1+\cos(\pi y)] \ . \tag{3.10}$$

Substitution of (3.10) in (3.8) shows that the switching function is the reciprocal of the square of the modified Bessel function of order 0, and of argument $\Delta/2kT$:

$$f(T)=\frac{1}{I_0^2(\Delta/2kT)} \ . \tag{3.11}$$

(b) Square potential. The potential is given by

$$U(x)=\begin{cases}0 & \text{for } a/2\geq|x|\geq a/4\\ -\Delta & \text{for } 0\leq|x|\leq a/4\end{cases}, \quad U(x+a)=U(x) \ . \tag{3.12}$$

Equation (3.8) shows, in this case, that the switching function is the reciprocal of the square of the hyperbolic cosine of Δ/kT:

$$f(T)=\frac{1}{\cosh^2(\Delta/2kT)} \ . \tag{3.13}$$

(c) Triangular potential. The potential is given by

$$U(x)=-\Delta\left[1-\frac{2|x|}{a}\right]$$
$$\text{for } 0\leq|x|\leq a/2, \quad U(x+a)=U(x) \ . \tag{3.14}$$

The integrations in (3.8) yield the switching function to be

$$f(T)=\frac{(\Delta/2kT)^2}{\sinh^2(\Delta/2kT)} \ . \tag{3.15}$$

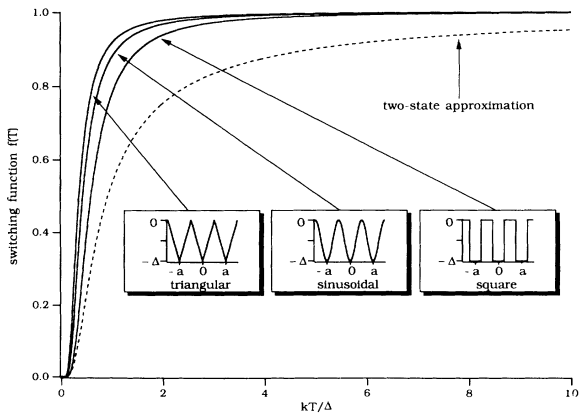

FIG. 1. Exact temperature dependence of the switching function or mobility for the sinusoidal, square, and triangular potentials, along with the two-state approximation (1.2). Here and in Figs. 2–4 below, the insets depict the potentials.

The potentials in the above three cases and the corresponding switching functions are shown in Fig. 1. In all three cases the switching character is clearly visible: all three functions interpolate between the values 0 and 1 for small and large temperatures, respectively. This behavior of $f(T)$ is crucial in the explanation of the thermal runaway phenomenon.[4,5] We also see that, while the $f(T)$'s obtained through our prescription (3.8) are seen to have the general behavior displayed by the two-state expression of (1.2), shown in Fig. 1 for comparison, they all rise faster than the latter. The source of this behavior will be clear in Sec. IV below.

IV. GENERAL COMMENTS ON THE SWITCHING FUNCTION

The expressions derived for the switching function for the three cases discussed in Sec. III show a peculiar resemblance to expressions for the *partition function* describing the equilibrium behavior of three magnetic systems, which we will denote by (a), (b), and (c). Each of the magnetic systems is a collection of a large number of noninteracting spins. In case (a) any spin can exist in one of two states, in case (b) it is free to rotate continuously in a plane, and in case (c) it may rotate freely in three dimensions. As is well known from texts on statistical mechanics,[10] the squares of the partition functions of systems (a), (b), and (c) are identical to the reciprocals of the right hand sides of (3.11), (3.13), and (3.15), respectively. Thus, they correspond exactly to the respective potentials (a), (b), and (c) discussed in Sec. III. Given that the calculations in Sec. III are not aimed at the equilibrium behavior of any magnetic system, this apparent coincidence is intriguing. What is the source of this storage connection? It is easy to answer this question when we realize that, in all the three cases considered, the potential $U(x)$ obeys the condition

$$U\left[\frac{a}{2}-x\right]+U(x)=\text{const} \ . \tag{4.1}$$

Loosely stated, this means that the shape of the potential in the inner region of each confining well is the same as in the outer region. Equation (3.7) translates (4.1) into the simpler condition

$$V(1-y)+V(y)=\text{const} \tag{4.2}$$

for the reduced potential, and allows us, through (3.8), to write at once that

$$f(T)=\left[\int_0^1 dx\,\exp\left[-\frac{\Delta}{kT}V(x)\right]\right]^{-2} \ . \tag{4.3}$$

Equation (4.3) shows that the switching function is indeed the reciprocal of the square of a partition function of a system possessing no kinetic energy and the potential $U(x)$. Comparing the physics underlying the magnetic systems with that behind our moving particle in the nonlinear potential, we see that it is trivial to appreciate the connection between the two-state spin and our system with a (two-state) square-well potential. It is also not difficult to see the correspondence between the rotation of

a spin in a plane and the motion of our system under the action of a sinusoidal potential as soon as the identification of the angular variable in the former case is made with the translational variable in the latter. It is perhaps a bit more surprising to see that the three-dimensional spin corresponds to the triangular potential in our system.

In the light of this curious connection with the calculation of textbook partition functions, it might be interesting to evaluate the switching function for another potential arising through an analogy with yet another magnetic system,[10] viz., a spin with quantum number greater than $\frac{1}{2}$.

(d) Multistep potential. With M,N as integers, and $M=0,1,\ldots,N-1$, the potential is given by

$$U(x)=-\Delta\left[1-\frac{M}{N-1}\right], \tag{4.4}$$

$$(a/2)\frac{M}{N}<x<(a/2)\frac{M+1}{N}\ ,$$

$$U(x+a)=U(x)\ .$$

The switching function $f(T)$ takes the form

$$f(T)=\frac{N^2\sinh^2(\Delta/2(N-1)kT)}{\sinh^2(N\Delta/2(N-1)kT)} \tag{4.5}$$

and is once again the reciprocal of the square of the partition function of the corresponding magnetic system.[10] The results (3.13) and (3.15) for the square and triangular potentials can be recovered from the multistep result (4.5) by the respective limits $N\to 2$ and $N\to\infty$. This is shown in Fig. 2.

When the potential in our problem does not obey (4.1) or (4.2), the switching function does *not* equal the reciprocal of the square of a partition function. Two cases of such potentials, the general rectangular potential and the general piecewise linear potential, are treated below.

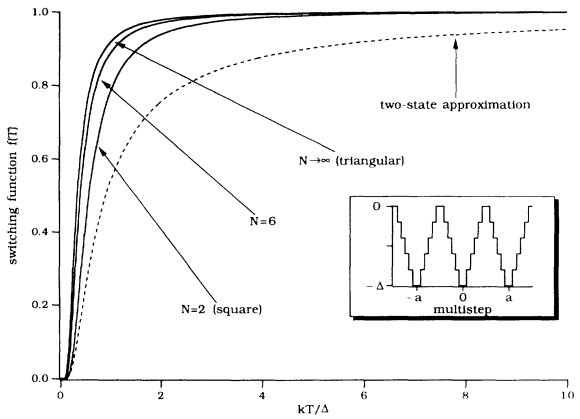

FIG. 2. Case of the multistep potential: exact temperature dependence of the mobility or switching function along with the two-state approximation. The extreme limits $N\to\infty$ and $N\to 2$, which yield the triangular and the square potential, respectively, are shown along with an intermediate case ($N=6$). The switching function for the multistep potential is related to the partition function of a spin system with quantum number greater than $\frac{1}{2}$.

(e) Rectangular potential. The potential is given by

$$U(x)=\begin{cases}0 & \text{for } |x|\geq(a/2)\xi\\ -\Delta & \text{for } |x|\leq(a/2)\xi\end{cases},\quad U(x+a)=U(x)\ , \tag{4.6}$$

where ξ is the fraction of the configuration space in which the particle is not free, i.e., ξ is the ratio of the length over which $U(x)$ equals $-\Delta$ to the total period a of $U(x)$. The switching function is given by

$$f(T)=\left[1+4\xi(1-\xi)\sinh^2\left[\frac{\Delta}{2kT}\right]\right]^{-1}. \tag{4.7}$$

In the special case of $\xi=\frac{1}{2}$, i.e., when the free and the bound regions are of equal extent, (4.7) reduces to the square potential result (3.13). This limit as well as other cases of Eqs. (4.6) and (4.7) are depicted in Fig. 3.

(f) Piecewise linear potential. The potential is given by

$$U(x)=\begin{cases}-\Delta\left[1-\left[\frac{\zeta}{\xi}\right]\frac{2x}{a}\right] & \text{for } 0\leq x\leq a\xi/2\\ -\Delta\left[\frac{1-\zeta}{1-\xi}\right]\left[1-\frac{2x}{a}\right] & \text{for } a\xi/2\leq x\leq a/2\end{cases},\quad U(-x)=U(x)=U(x+a) \tag{4.8}$$

and thus consists of two linear pieces. The fraction of the configuration space in which the particle is in the energetically lower of the two sections is denoted by ξ, in correspondence with case (e) above. The additional parameter ζ defined in the present case is such that $(1-\zeta)$ denotes the ratio of the depth of the potential at the point where the two linear pieces meet to its total depth. The switching function is given by

$$f(T)=\frac{(\Delta/2kT)^2\zeta(1-\zeta)}{\xi(1-\xi)\sinh^2(\Delta/2kT)+(1-\xi/1-\zeta)(\zeta-\xi)\sinh^2[(1-\zeta)\Delta/2kT]+(\xi/\zeta)(\xi-\zeta)\sinh^2(\zeta\Delta/2kT)}\ . \tag{4.9}$$

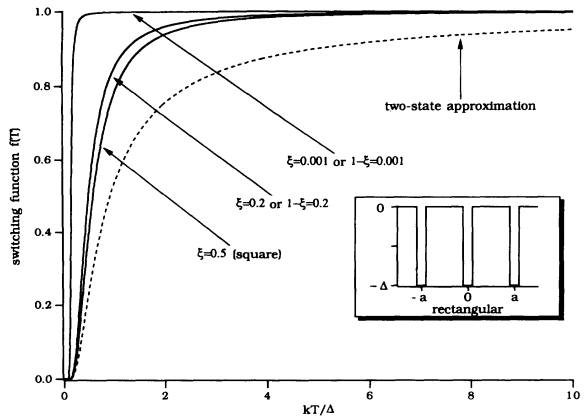

FIG. 3. Case of the rectangular potential: exact temperature dependence of the switching function or mobility, compared to the two-state approximation. Along with the square potential limit ($\xi=0.5$) and an intermediate case ($\xi=0.2$), the extreme limits of $\xi\to 0$ and $\xi\to 1$ are also shown such that ξ in one case equals $1-\xi$ in the other. The latter two limits result in different potential shapes (narrow vs wide wells) but in identical switching functions. For this case of very small or very large ξ (the value assumed is 0.001 or 0.999), the switching function is seen to rise very steeply to the value 1. This would correspond to extreme manifestation of thermal runaway.

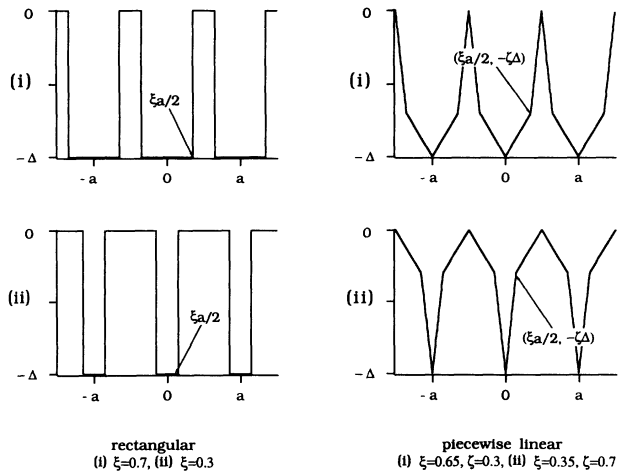

FIG. 5. Two cases of the rectangular potential of Fig. 3, and two cases of the piecewise linear potential of Fig. 4, shown to elucidate what potential features control the form of the switching function. For each kind of potential, case (i) represents wide wells ($\xi=0.7$ for the rectangular and $\xi=0.65$, $\zeta=0.3$ for the piecewise linear case) while case (ii) represents narrow wells ($\xi=0.3$ for the rectangular and $\xi=0.35$, $\zeta=0.7$ for the piecewise linear case). The relation between cases (i) and (ii) is that the values of ξ in the two cases add up to 1 and so do the values of ζ. Although potentials (i) and (ii) look quite different from each other, their switching functions are identical.

The special case $\xi=\zeta$, which reduces this result to the triangular potential result (3.15), as well as other cases of (4.9), are shown in Fig. 4.

An inspection of Figs. 3 and 4 as well as of expressions (4.7) and (4.9) reveals an additional invariance property of the switching function for potentials (e) and (f). In the case of the rectangular potential, the transformation

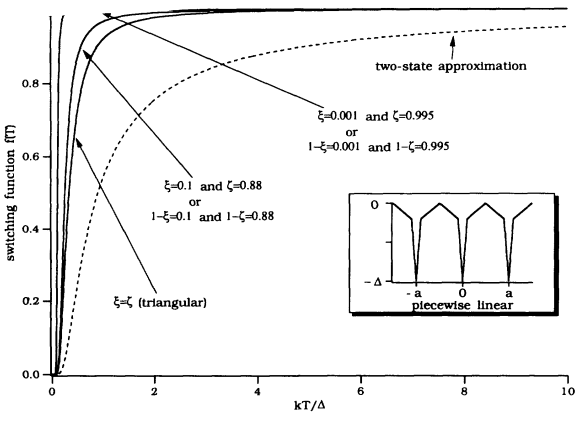

FIG. 4. Case of the piecewise linear potential: exact temperture dependence of the mobility or switching function, compared to the two-state approximation. Along with the limit $\xi=\zeta$, which yields the triangular potential, and an intermediate csa ($\xi=0.1$, $\zeta=0.88$), the extreme case of $\xi=0.001$ with $\zeta=0.995$ is also displayed. The latter choice of parameters is seen to result in a very steep switching function.

$\xi\to 1-\xi$ changes the potential shape (see Fig. 5) but not the switching function. Similarly, in the case of the piecewise linear potential, despite the change in the potential shape brought about by the transformation $\xi\to 1-\xi$, $\zeta\to 1-\zeta$ (see Fig. 5), there is no effect whatsoever on the switching function. This is remarkable in view of the fact that the ratio of the configuration space in which the particle is free (or less bound) to that in which the particle is bound (or more bound) changes drastically under these transformations. Thus, the potential shapes depicted in Fig. 5 for the rectangular case might tempt one to conclude that, in case (i), in which the particle appears to have more bound configuration space available to it, the mobility should be less than that in case (ii). We see, however, that such a conclusion would be erroneous and that phase-space arguments can be misleading. We will see below that the key property of the potential is its dispersion.

We will now address three general issues concerning the switching function. First, we will prove that $f(T)$ indeed possesses the switching characteristics displayed by the elementary two-state result (1.2), viz., that it equals 0 and 1 in the limits of vanishing and infinite temperature, respectively. Next, we will investigate what features of the potential control the degree of steepness of $f(T)$ and develop an approximate prescription to calculate $f(T)$ easily from global features of the potential. And last, we will establish an inequality which compares several expressions for the switching function including the exact result and the two-state result.

A. Proof of the switching behavior

The explanation of the source of the switching behavior becomes transparent if we rewrite $f(T)$ given by (3.8) in the form of a double integral

$$\frac{1}{f(T)} = \int_0^1 dx \exp\left[\frac{\Delta}{kT}\right] \int_0^1 dx \exp\left[-\frac{\Delta}{kT}\right] = \int_0^1 dx \int_0^1 dy \exp\left[\frac{\Delta}{kT}[V(x)-V(y)]\right], \quad (4.10)$$

which, upon renaming the variables, can also be written as

$$\frac{1}{f(T)} = \int_0^1 dx \int_0^1 dy \exp\left[\frac{\Delta}{kT}[V(y)-V(x)]\right], \quad (4.11)$$

On combining (4.10) and (4.11), one obtains

$$\frac{1}{f(T)} = \frac{1}{2}\left[\int_0^1 dx \int_0^1 dy \exp\left[\frac{\Delta}{kT}[V(x)-V(y)]\right] + \int_0^1 dx \int_0^1 dy \exp\left[\frac{\Delta}{kT}[V(y)-V(x)]\right]\right] = \int_0^1 dx \int_0^1 dy \cosh\left[\frac{\Delta}{kT}[V(x)-V(y)]\right]. \quad (4.12)$$

Since $\cosh(z) > 1+z^2/2$ for $z>0$, we have the inequality

$$f(T) < \frac{1}{1+C(\Delta/kT)^2}, \quad (4.13)$$

where

$$C = \int_0^1 dx \int_0^1 dy\, [V(x)-V(y)]^2 > 0$$

provided $V(x)$ is not identically a constant function [in which case, $f(T)$ is a temperature-independent constant]. The switching behavior is now completely clear from (4.13). The first of the switching characteristics, viz.,

$$\lim_{T\to 0} f(T) = 0, \quad (4.14)$$

follows from (4.13), while the second characteristic is obvious directly from (3.8):

$$\lim_{T\to\infty} f(T) = \int_0^1 dx \int_0^1 dy = 1. \quad (4.15)$$

Thus, we have given a general proof of the fact that, for any nonconstant potential, $f(T)$ switches between the values 0 and 1 as the temperature changes from 0 to infinity.

B. Source of steepness and an approximate prescription

The degree of severity of the thermal runaway phenomenon, i.e., the extent to which the temperature rise is sudden, will obviously depend on the degree of steepness of the switching function, which, in turn, will depend on the shape of the potential. It is of interest to know that features of the potential determine the steepness. To answer this question, as well as to develop reasonable *approximations* for $f(T)$ when one knows only some global characteristics of the potential rather than its complete detail, we present the following analysis. Equation (4.12) may be recast in the form

$$\frac{1}{f(T)} = \int_0^1 dx \int_0^1 dy \left[1+2\sinh^2\left[\frac{\Delta}{2kT}[V(x)-V(y)]\right]\right]. \quad (4.16)$$

From the definition (3.7) of $V(x)$, we know that $[V(x)-V(y)]$ is of the order of unity. The following approximation is, therefore, justified:

$$\sinh^2\left[\frac{\Delta}{2kT}[V(x)-V(y)]\right] \approx [V(x)-V(y)]^2 \sinh^2\left[\frac{\Delta}{2kT}\right]. \quad (4.17)$$

We now substitute (4.17) in (4.16), define a single global quantity u^2 which characterizes the potential through

$$u^2 = 4\left[\int_0^1 dx\, V^2(x) - \left[\int_0^1 dx\, V(x)\right]^2\right], \quad (4.18)$$

and obtain a simple new expression for the switching function for any potential:

$$f(T) = \frac{1}{1+u^2\sinh^2(\Delta/2kT)}. \quad (4.19)$$

Equation (4.19) is another important result in our analysis. While not exact, the expression provided for the switching function in (4.19) involves an excellent approximation. The high degree of accuracy with which (4.19) approximates exact expressions may be appreciated from Fig. 6. The prescription in (4.19) is, furthermore,

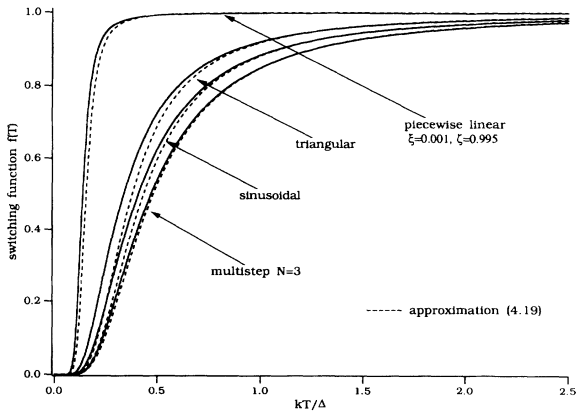

FIG. 6. The approximate prescription (4.19) (dashed lines) compared to the exact mobility or switching function (solid lines) for various potentials: the sinusoidal potential, the triangular potential, the multistep potential with $N=3$ and the piecewise linear potential with $\xi=0.001$ and $\zeta=0.995$. The approximate prescription has the following respective values of u^2: $\frac{1}{2}$, $\frac{1}{3}$, $\frac{2}{3}$, and 0.0017. The prescription is seen to provide an excellent approximation in every case. The case of the rectangular potential is not shown as it would be redundant: the prescription (4.19) is *identical* to the exact result in that case.

simple, easily usable, and extremely convenient for actual computations from potentials. The simplicity and ease of use are evident from the expression itself. The convenience stems from the fact that methods to obtain potentials in realistic cases, which involve *ab initio* computational procedures[11] or direct deduction from observations such as those in diffusion experiments, can provide global characteristics of the potential with reasonable confidence but not the detail.

What feature of the potential controls the steepness of the switching function is now clear. Equation (4.19) shows that the *dispersion* of the potential, viz., the quantity u^2, decides the steepness of $f(T)$. It is this dispersion that remains the same as one goes from case (i) to case (ii) of the potentials depicted in Fig. 5, although the potential shapes change. The lower the value of u^2, the more precipitous the switch of $f(T)$ from 0 to 1.

C. Inequality relating the exact and the two-state forms of $f(T)$

The reduced potential $V(y)$, defined by (3.7), is bounded by the values 0 and 1. It is clear then that

$$\int_0^1 dy\ V^2(y) \le \int_0^1 dy\ V(y)\ . \tag{4.20}$$

Since the square of $(\int_0^1 dy V(y) - \frac{1}{2})$ cannot be smaller than 0, we can write

$$\int_0^1 dy\ V(y) \le \left[\int_0^1 dy\ V(y)\right]^2 + \tfrac{1}{4}\ . \tag{4.21}$$

On combining (4.21) and (4.20), and recalling the definition (4.18) of u^2, we obtain the result that the dispersion can never exceed 1:

$$u^2 \ge 1\ . \tag{4.22}$$

Comparison of the right-hand sides of (1.2) and (3.13) along with a straightforward expansion shows that the square potential switching function (3.13) is always larger than the two-state expression (1.2):

$$\frac{1}{\cosh^2(\Delta/2kT)} \ge 2\frac{e^{-\Delta/kT}}{1+e^{-\Delta/kT}}\ . \tag{4.23}$$

Also, since $\sinh^2(cz) \ge z^2\sinh^2 c$ whenever $z^2 \le 1$, the passage from (4.16) to (4.19) makes it clear that the exact $f(T)$ is always larger than the approximate expression (4.19):

$$f(T) \ge \frac{1}{1+u^2\sinh^2(\Delta/2kT)}\ . \tag{4.24}$$

The combination of (4.26)–(4.28) yields the inequality

$$f(T) \ge \frac{1}{1+u^2\sinh^2(\Delta/2kT)} \ge \frac{1}{\cosh^2(\Delta/2kT)} \ge 2\frac{e^{-\Delta/kT}}{1+e^{-\Delta/kT}}\ , \tag{4.25}$$

which states that the exact switching function, the approximate prescription we have given in terms of the dispersion of the potential, the square potential case which contains no potential parameters other than Δ, and the two-state approximation given in Refs. 4 and 5 all lie in a strict order of decreasing magnitude.

V. CONCLUSIONS

The aim of the analysis presented in this paper has been to provide support to the form and detail of a conjecture made in a recent theory[4,5] of microwave heating of ceramic materials. Because the procedure we use involves the calculation of the mobility of a particle subjected simultaneously to a nonlinear periodic potential, an applied electric field, and stochastic interactions with its environment, our analysis is of interest not only to the microwave heating problem which motivated it, but also in the general context of transport calculations for nonlinear potentials. Our goal is the justification of the switching function (1.2) used in the temperature-time evolution given in (1.1). Our point of departure is the nonlinear Langevin equation (1.3) presented in Refs. 4 and 5. Our method consists of the conversion of (1.3) into the Fokker-Planck equation (2.7), the reduction of these two equations to (2.9) and (2.11), respectively, in the limit of large damping, the calculation of the steady-state mean velocity in the case of a time-independent electric field to give (2.24), and the simplification, in the linear field regime, to the mobility expression (3.2). The final step is the calculation of the switching function (3.5).

The central result of this paper is the mobility expression

$$\mu = \mu_0 f(T)\ , \tag{5.1}$$

where the mobility in the absence of stochastic interactions, viz., μ_0, is given by the Drude result (3.3), and the switching function $f(T)$ is given generally by (3.5). The switching function expression takes the form presented in (3.6) or (3.8) for symmetric potentials. We have explored a variety of potentials represented by (3.9), (3.12), (3.14), (4.4), (4.6), (4.8), and obtained the corresponding expressions for the switching function in (3.11), (3.13), (3.15), (4.5), (4.7), and (4.9), respectively.

We have noted the curious fact that the switching function for some of the potentials considered equals the square of the reciprocal of the partition functions of certain well-known magnetic systems, and have explained the source of the relation to be an additional symmetry condition, viz., (4.1), which those potentials possess.

In addition to the exact expressions discussed above, we have also presented a prescription to evaluate the switching function from a single global characteristic (rather than the detail) of the potential, viz., the quantity u^2 defined in (4.18). This prescription is (4.19) and it is exact for arbitrary rectangular potentials. While not exact, in general, it involves an excellent approximation as Fig. 6 shows, and it provides one with a measure of the steepness of the switching function as related to the features of the potential. We have also shown that, at all temperatures, the exact switching function (or mobility) for an arbitrary potential is always larger than that given by the approximation (4.19), that the latter is always larger than the square potential result (3.13), which, in turn, always exceeds the two-state result (1.2). In Table I,

TABLE I. Steepness measure $1/u^2$ of the mobility or of the switching function.

Kind of potential	Equations in text for potential and $f(T)$	Measure $1/u^2$ of the steepness of $f(T)$
Square	(3.12) and (3.13)	1
Sinusoidal	(3.9) and (3.11)	2
Triangular	(3.14) and (3.15)	3
Multistep	(4.4) and (4.5)	$3(N-1)/(N+1)$
Rectangular	(4.6) and (4.7)	$1/[4\xi(1-\xi)]$
Piecewise linear	(4.8) and (4.9)	$3/[4\xi(1-\xi)+(1-\xi-\zeta)^2]$

we provide the potentials studied in this paper along with the measure $1/u^2$ of the steepness of the resulting switching function.

The approximations that have gone into our analysis are (i) the limit of large damping that has allowed us to reduce the exact Fokker-Planck equation (2.7) to the Smoluchowski equation (2.11), (ii) the assumption of a dc field that has allowed us to solve (2.11) in the steady state, and (iii) the neglect of terms of order higher than linear in the electric field represented by (3.1). The justification of the three approximations is as follows. The ceramic systems which motivated our analysis are indeed believed to involve very large damping, compatible with Debye relaxation.[4,5] The period of the microwaves employed is large enough (relative to other time constants in the problem) to represent the electric field as independent of time, in the first approximation, and Ohm's law is certainly expected to be obeyed in the systems as the field magnitudes are limited by breakdown. Because our analysis could find easy application to systems other than ceramic materials, it is of interest to study the problem in cases wherein all these three assumptions must be dropped. We are in the process of carrying out such an investigation.

We have used the results of this paper to analyze several problems of direct interest to experimental observations in ceramic materials. One of these concerns the question of the disparity between values of the energy barrier, Δ in our analysis above, obtained from diffusion experiments on the one hand and from thermal runaway observations on the other. Another involves the effect of grain boundaries and confinement phenomena on microwave absorption in materials. These are being reported elsewhere.[12]

ACKNOWLEDGMENTS

This work was carried out in part with the support of the UNM-NSF Center for Microengineered Ceramics, a collaborative effort of the National Science Foundation (CDR-8800352), the Los Alamos and Sandia National Laboratories, the New Mexico Research and Development Institute, and the Ceramics Industry.

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