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Explanation of the barrier-depression effect in ceramics undergoing microwave heating

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Thermal runaway observations on a variety of materials including strontium titanate, zinc oxide, iron oxide, and alumina, have been explained successfully in terms of a recent theory, which involves absorbing entities such as vacancies, bivacancies, or interstitials, which have to overcome an energy barrier in order to absorb microwaves to a significant extent. However, there is a tendency, unexplained so far, for the energy-barrier values extracted in this fashion to be slightly smaller than those obtained from diffusion measurements. The source of this systematic discrepancy is discovered through the application of a stochastic analysis of the mobility of the ceramic materials. New barrier values are obtained from the thermal runaway observations and found to be in better agreement with diffusion experiments.

I. INTRODUCTION

Sintering of materials while under the action of microwaves has been reported by several investigators to lower the sintering temperature by several hundreds of degrees and shorten the sintering time by several hours.[1-3] This has understandably led to a lot of activity[4-9] in the study of the effects of microwave sintering on ceramics. Not only are microwaves expected to increase the heating efficiency by concentrating the heating process within the material rather than in the furnace in which the material is placed, but they are also suspected to have rather fundamental consequences such as more efficient atomic diffusion within the material. Experiment[8] as well as theoretical[9] investigations of these fundamental issues have ben undertaken, and a general picture[7,9] has begun to emerge.

A recent theory[7,9] of some of these phenomena has provided what appears to be a satisfactory description of thermal-runaway measurements in a large number of materials and dielectric-loss observations in some of them. The positive aspects of the theory have been its simple explanation of the qualitative features of the runaway phenomenon, the diversity of time-temperature curves, and the variety of materials, viz., silica, zinc oxide, ferric oxide, alumina, and strontium titanate, that it has been able to address and the generally reasonable values of the theoretical parameters that have come out of the analysis. An interesting negative aspect has been the fact that the values of the energy barriers, which the absorbing entities (such as vacancies, bivacancies, or interstitials) have to surmount in order to absorb microwaves to an appreciable extent, have turned out to be always smaller by a small factor (such as 3) relative to barrier values extracted from diffusion measurements. While not large, the discrepancy is systematic. The source of the discrepancy is, therefore, of definite interest and constitutes the subject of the present paper.

The theory of Refs. 7 and 9 is based on the idea that the potential to which some of the absorbing entities within the ceramic material are subjected possesses a nonlinear nature which divides the phase space of the absorbing entities into two regions. One is a bound region which contributes negligibly to microwave absorption. The other is a free region whose contribution to absorption is substantial. The absorbers must cross an energy barrier to pass from the bound region to the free region. Thus, a nonlinear feedback is possible. Heating can provide the necessary energy to free the absorbing entities, and the freeing of the absorbers can then result in enhanced absorption and, consequently, in enhanced heating. The ensuing temperature-time equation is given by[7,9]

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

where P is the incident power, σ_1 is proportional to the Stephan-Boltzmann constant, and k_A and k_M are related to the absorption coefficients of two kinds of species of absorbers present in the material. The first kind, labeled by A, does not have the two-region phase space described above. The second kind, labeled by M, does. As stated above, when bound, the latter absorb negligibly. When free, they make a contribution to the heating rate. The fraction of the M absorbers which are free is itself dependent on temperature, and is denoted by $f(T)$. Details of (1.1) and its application may be found in Ref. 7.

Of special relevance to the understanding of thermal runaway observations via (1.1) is the dependence of $f(T)$ on the temperature. At small temperatures, this quantity is negligible. Therefore, the contribution made to the heating rate is also negligible. As the temperature rises, $f(T)$, and with it the heating rate, increases. This increase is sharp at first, but tends to saturate: Eventually, $f(T)$ attains the value 1. The physics behind this "switching" behavior of $f(T)$ is the existence of the potential barrier mentioned above, which the absorbing charges have to surmount to make a significant contribution to microwave absorption. The theory of Refs. 7 and 9 uses two forms of $f(T)$: a simple expression based on a two-state description of the system, viz.,

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

Δ being the energy barrier and k the Boltzmann constant, and a more complex expression based on phase-space considerations, viz.,

$$f(T)=\frac{\int_{\text{free region}} d\mathbf{x}\,d\mathbf{p}\,e^{-[\mathbf{p}\cdot\mathbf{p}/2m+U(\mathbf{x})]/kT}}{\int_{\text{entire region}} d\mathbf{x}\,d\mathbf{p}\,e^{-[\mathbf{p}\cdot\mathbf{p}/2m+U(\mathbf{x})]/kT}}=\frac{\int_{\text{entire region}} dx\,e^{-U(x)/kT}\text{erfc}\{[U(x)/kT]^{1/2}\}}{\int_{\text{entire region}} dx\,e^{-U(x)/kT}}\ , \tag{1.3}$$

where $\mathbf{p}$ and $\mathbf{x}$ denote the momentum and position coordinates, respectively, $U(x)$ is the potential to which the surrounding lattice subjects the absorber, and the last expression is valid for a representative one-dimensional system. Both forms (1.2) and (1.3) were based on a static analysis of the problem. A theory of $f(T)$ based on dynamic, in particular stochastic, considerations is now available.[10] The expression for $f(T)$ provided by this dynamic theory, and its comparison with the two-state expression used in Ref. 7 for extracting energy-barrier values, will form our point of departure in the present paper. In Sec. II we discuss the physics underlying the older forms (1.2) and (1.3) of $f(T)$ as well as the general expression (valid for arbitrary potentials) provided by the recent analysis of Ref. 10, along with arguments supporting that analysis. In Sec. III we compare the new result with the older ones and thereby provide an explanation of the barrier-depression effect. Concluding comments including the statement of explicit relations between the parameters k_A, k_M of (1.1) and microscopic quantities form Sec. IV.

II. EXPRESSIONS FOR THE SWITCHING FUNCTION $f(T)$

Figure 1 shows the bare essentials of the potential to which absorbing entities such as vacancies and interstitials are subjected within a ceramic material. *Ab initio* calculations of such potentials have been performed recently[11] for materials such as MgO. The basic feature of the potential is that it is spatially periodic and possesses the characteristic nonlinearity which separates the phase space of the absorber into two regions. For energies which are low enough, the absorber is trapped within one of the wells and, as can be shown,[7–9] absorbs negligibly. For energies which are large enough, the absorber can overcome the barrier between wells and absorb significantly. The switching function $f(T)$ can then be said to be proportional to the relevant Boltzmann factor and is at once given by (1.2) if no further attention is given to the differences in the *amount* of phase space associated with the two regions. If, on the other hand, the amount of phase space is calculated by working out integrals such as (1.3), a different expression results. As two examples of such an expression, we cite the cases of a rectangular potential and a sinusoidal potential. Clearly, the former is one of the simplest representations of the situation encountered in Fig. 1. One characterizes the potential in terms of an energy barrier, a length, and a period:

$$U(x)=\begin{cases}0 & \text{for } |x|\geq(a/2)\xi\ ,\\ -\Delta & \text{for } |x|\leq(a/2)\xi\ ,\end{cases}$$
$$U(x+a)=U(x)\ . \tag{2.1}$$

The switching function is then given by[9,10]

$$f(T)=\frac{re^{-\Delta/kT}+\text{erfc}(\sqrt{\Delta/kT})}{re^{-\Delta/kT}+1}\ , \tag{2.2}$$

where the so-called free/bound ratio is $r=(1-\xi)/\xi$. A more realistic representation of potentials such as the one in Fig. 1 is given by the sinusoidal form

$$U(x)=-\frac{\Delta}{2}\left[1+\cos\left[\frac{2\pi x}{a}\right]\right]\ . \tag{2.3}$$

In this case, (1.3) can be shown[9,12] to lead to the following $f(T)$:

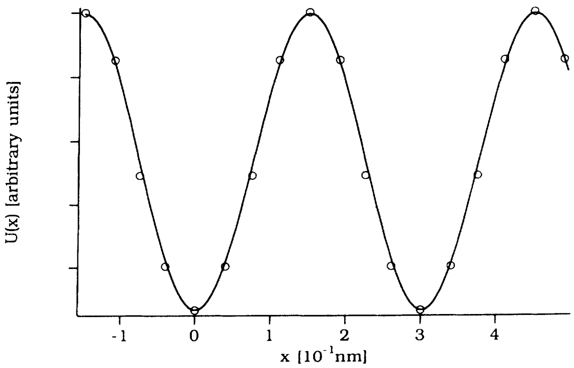

FIG. 1. Essential features of the potential seen by the absorbers in a ceramic material as exemplified by the result of *ab initio* calculations carried out in Ref. 11 for MgO. Because the calculational procedure in Ref. 11 produces reliable relative (but not absolute) values of the ordinate, we display the latter here in arbitrary units. The values of the distance x and the shape of the potential $U(x)$ are reliable and are seen to correspond very closely to the sinusoidal potential of Eq. (2.3). The latter is represented by the solid line, and the circles depict calculated values.

$$f(T)=\frac{\int_{-\pi}^{\pi} dx \exp[(\Delta/2kT)\cos x]\,\mathrm{erfc}[\sqrt{(\Delta/2kT)(1+\cos x)}]}{2\pi I_0(\Delta/2kT)}\,, \tag{2.4}$$

where I_0 denotes the modified Bessel function and erfc is the complementary error function. Further simplifications of (2.4) are presented in the Appendix.

All of the above results for $f(T)$, whether the simple two-state expression (1.2) or the more detailed forms (2.2) or (2.4), are obtained from *static* considerations. As stated in the Introduction, their application to experiments seems to suffer from the fact that the extracted values of the barrier energy Δ are consistently smaller than the corresponding values obtained from diffusion measurements. The point of departure in the present paper is our recently developed dynamic theory of the switching function $f(T)$. The basic idea of the theory[10] is to consider the full evolution equation for the absorbing particle,[9]

$$m\frac{d^2x}{dt^2}+m\gamma\frac{dx}{dt}+\frac{dU(x)}{dx}=qE\cos\omega t+R(t)\,, \tag{2.5}$$

where m and q are, respectively, the mass and charge of the particle moving in an applied microwave field $E\cos\omega t$, x being the coordinate of the particle. Absorber particles interact with the rest of the lattice via the systematic force corresponding to the potential $U(x)$, and via the stochastic force $R(t)$ which stems from thermal fluctuations. The strength of $R(t)$ is connected with the effective damping constant γ through the fluctuation-dissipation theorem. Since the instantaneous power absorbed is proportional to the product of the velocity $v=dx/dt$ of the particle and the applied electric field E, our aim is to calculate the mean velocity $\langle v(t)\rangle$ of the absorber particle. The exact solution of this problem is not known. Fortunately, typical values of the parameters involved allow reasonable approximations to be made, which reduce the problem to a soluble one. One of these helpful features is that the friction coefficient γ is typically much larger than the characteristic frequency of the potential, i.e.,

$$\gamma \gg \omega_0=\left[\frac{1}{m}\frac{d^2U}{dx^2}\right]^{1/2}.$$

This allows the reduction of the Fokker-Planck equation corresponding to (2.5) to a Smoluchowsky form.[10] Also, the frequency of the microwave field ω is rather small compared with other characteristic frequencies in the system. This allows the solution of the Smoluchowsky equation in the time-independent form. Thus we can reduce the problem to that of the calculation of the dc mobility of a particle in a spatially periodic potential and solve it explicitly.[10] Similar problems have arisen and been solved in various other physical contexts.[13] The result for the switching function for an arbitrary periodic potential $U(x)=U(x+a)$ with a period a is

$$f(T)=a^2\left[\int_0^a dx\, e^{-(1/kT)U(x)}\int_0^a dx\, e^{(1/kT)U(x)}\right]^{-1}. \tag{2.6}$$

The specific cases mentioned above, viz., those of the rectangular potential (2.1) and sinusoidal potential (2.3), can be shown to result[10] in the following explicit forms of $f(T)$, respectively:

$$f(T)=\left[1+4\xi(1-\xi)\sinh^2\left[\frac{\Delta}{2kT}\right]\right]^{-1} \tag{2.7}$$

and

$$f(T)=\frac{1}{I_0^2(\Delta/2kT)}\,. \tag{2.8}$$

In Sec. III we compare these forms to the respective phase-space counterparts (2.2) and (2.4) and to the two-state expression (1.2), and show how the differences can account for the barrier-depression effect.

III. COMPARISON WITH PREVIOUS THEORETICAL RESULTS AND WITH EXPERIMENT

The comparison of the two-state, phase-space, and stochastic switching functions for the above considered potentials is shown in Fig. 2. By the phrase "stochastic switching function" we mean here (2.7) and (2.8), which are calculated analytically from the fully stochastic point of departure (2.5). While the qualitative behavior is the same for all three curves, we note that the actual switching function as given by our recent theory[10] is steeper than the two-state or phase-space approximations. Herein lies the source of the depression effect which is the subject of the present paper. We will restrict our discussion to the comparison of the exact $f(T)$ with the two-state result (1.2) in the following because the latter is the basis of previous analyses[7,9] of experimental data. It is possible to prove explicitly[10] that, for an arbitrary potential $U(x)$, we always have

$$f(T)\geq 2\frac{e^{-\Delta/kT}}{1+e^{-\Delta/kT}}\,. \tag{3.1}$$

Thus, the actual $f(T)$ is always steeper than the two-state approximation. While we are certainly in possession of the mathematical proof[10] of this statement, it is not easy to provide a simple physical interpretation for it. One could argue that a heat reservoir performs two tasks: (i) providing phase-space (Boltzmann) weights according to the energies of different states and (ii) allowing fluctuations which could, for instance, cause escape from lower-energy states. Static arguments such as those leading to the two-state $f(T)$ take only (i) into account, whereas the dynamic derivation caters also to (ii). As the carrier escapes to the high-energy high-mobility states, it contributes more to the mobility. At any given temperature, the mobility or $f(T)$ is thus larger (steeper) than that calculated from the static approximation.

In Fig. 3 we take the actual switching functions, as given by (2.7) and (2.8), as data, and fit them with the

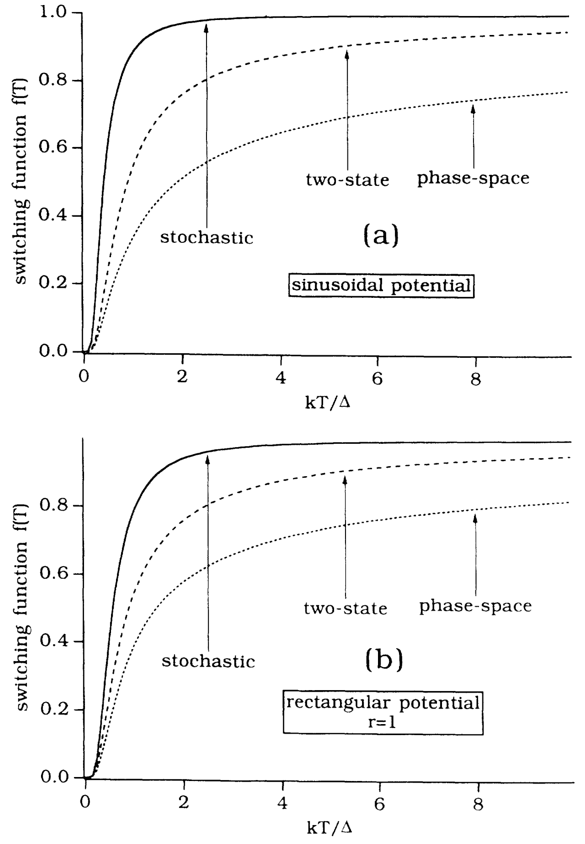

FIG. 2. Comparison of the new *stochastic* switching function $f(T)$ [Eq. (2.6)] used in the present paper with the two-state approximation [Eq. (1.2)] and the phase-space approximation [Eq. (1.3)] used in earlier work (Refs. 7 and 12). The potential is sinusoidal [Eq. (2.3)] in (a) and rectangular [Eq. (2.1)] in (b). In both cases the stochastic $f(T)$ is seen to be the steepest.

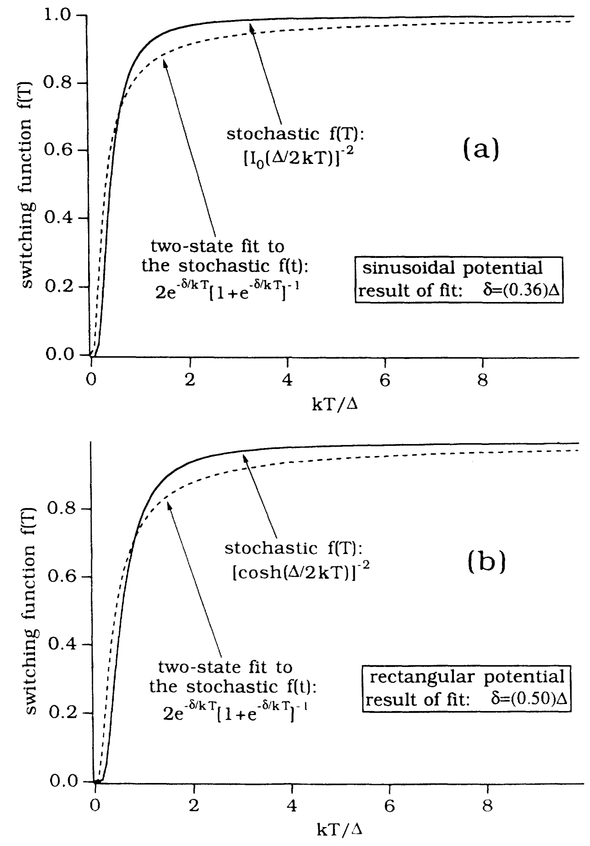

FIG. 3. Explanation of the barrier-depression effect: The actual switching functions for the rectangular and sinusoidal potentials, as given by (2.7) and (2.8), respectively, are fitted with the help of the two-state approximation (1.2) by varying the energy-barrier parameter of (1.2) to get a best fit. The extracted values of the barrier are seen to be smaller than the actual value. This explains the systematic discrepancy observed between the interpreted values of the energy barrier and values obtained from diffusion experiments.

help of the two-state approximation (1.2) by varying the energy barrier. In other words, we fit (2.7) and (2.8) with the help of

$$f(T)=2\frac{e^{-\delta/kT}}{1+e^{-\delta/kT}} \tag{3.2}$$

and vary δ to get a best fit. As Fig. 3 shows clearly, the extracted value of the barrier is 0.36 times the actual value in the sinusoidal case and 0.5 times the actual value in the rectangular case. We have here a clear source of the systematic discrepancy observed between the interpreted values of the energy barrier[7] and values obtained from diffusion experiments.[14,15]

We have shown in Ref. 10 that a simple but astonishingly accurate approximation can be given for the switching function, which is highly useful for practical computations. Its usefulness stems from the characterization of $f(T)$ by a single parameter u^2:

$$\frac{u^2}{4}=\int_0^1 dx\, V^2(x)-\left[\int_0^1 dx\, V(x)\right]^2 , \tag{3.3}$$

where $V(x)=U(ax)/\Delta$ is the reduced form of the potential $U(x)$, Δ being the difference between the maximum and minimum values of $U(x)$ (i.e., the height of the energy barrier). The expression for $f(T)$ is exact for any rectangular potential, is extremely accurate for most other potentials,[10] and is given by

$$f(T)=\frac{1}{1+u^2\sinh^2(\Delta/2kT)} . \tag{3.4}$$

In the case of a rectangular potential, the quantity of u^2 of (3.4) is related to the free/bound ratio r through

$$r=\frac{1-(1-u^2)^{1/2}}{1+(1-u^2)^{1/2}} \quad\text{or}\quad r=\frac{1+(1-u^2)^{1/2}}{1-(1-u^2)^{1/2}} . \tag{3.5}$$

In order to compare the predictions of our improved theory with observations on thermal runaway, we carried out best fits of solutions of (1.1), with the new switching functions obtained in the present paper, to data pertaining to alumina,[4] strontium titanate,[4] iron oxide,[1] and zinc oxide.[1] We used the two forms of the stochastic $f(T)$: (2.7) corresponding to the sinusoidal potential (2.1) and the approximate (3.4) involving the global parameter u^2. The results are presented in Table I (see also Figs. 4 and 5) together with their previously obtained counterparts.[7]

TABLE I. Parameters of the temperature-time equation (1.1) deduced from the application of the present theory to thermal-runaway observations on several materials. Fits are shown in Figs. 4 and 5. Two forms of the switching function are used: (2.7) which corresponds to a sinusoidal potential and (3.4) which is a "universal" approximate form involving the parameter u defined in (2.11).

	Pk_A (K/s)	Pk_M (K/s)	Δ/k (K)	σ_1 ($K^{-3}s^{-1}$)	u^2
		Ferric oxide			
Two-state	13.30	8.0	0.0017	0.5×10^{-12}	
Sinusoidal	0.78	10.3	0.0020	5.2×10^{-12}	
Universal	1.28	16.8	0.0020	5.2×10^{-12}	0.60
		Strontium titanate			
Two-state	1.1×10^{-2}	230	3600	4.1×10^{-12}	
Sinusoidal	9.6×10^{-2}	37	4575	2.7×10^{-12}	
Universal	6.7×10^{-2}	23	4020	2.5×10^{-12}	0.30
		Alumina			
Two-state	0.08	72	2600	1.9×10^{-12}	
Sinusoidal	0.66	30	4350	2.3×10^{-12}	
Universal	0.63	18	3800	1.9×10^{-12}	0.32
		Zinc oxide			
Two-state	0.75	250	2800	0.46×10^{-12}	
Sinusoidal	2.07	220	2650	296.00×10^{-12}	
Universal	1.53	535	2835	157.00×10^{-12}	0.68

IV. CONCLUDING REMARKS

In Refs. 7, 9, and 12, we presented a successful theory of microwave interactions with ceramics. Using a fully stochastic development of the ideas introduced in that theory, we provided a justification for qualitative aspects of the central conjecture of the theory in Ref. 10. In the present paper, we have carried out a quantitative development of the formalism in Ref. 10 and have extended the previous work by applying the new results to resolve the energy-barrier-depression problem. In order to complete our presentation of these ideas, we give below an explicit prescription for an *ab initio* calculation of quantities such as k_A, k_M, and σ_1 appearing in the temperature-time equation (1.1), which we introduced in Ref. 7.

The energy $\Delta\varepsilon$ absorbed by the ceramic material as its temperature increases by ΔT is given by

$$\Delta\varepsilon=\rho VC\,\Delta T\ , \tag{4.1}$$

where V is the volume of the sample, ρ its density, and C the specific heat per unit mass. On dividing this absorbed energy by Δt and taking the limit as Δt tends to 0, we get the average power absorbed from the microwaves. According to the Stephan-Boltzmann law, radiative losses

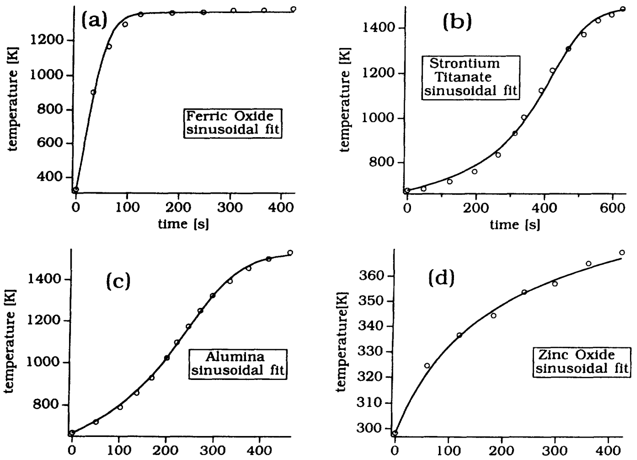

FIG. 4. Thermal-runaway observations in several ceramic materials fitted with the theory of Ref. 7 with the help of the stochastic switching function (2.6) rather than the two-state switching function (1.2) used in Ref. 7. The specific form of (2.6) corresponds to a sinusoidal potential and is given by (2.7). The data are from Refs. 1 and 4.

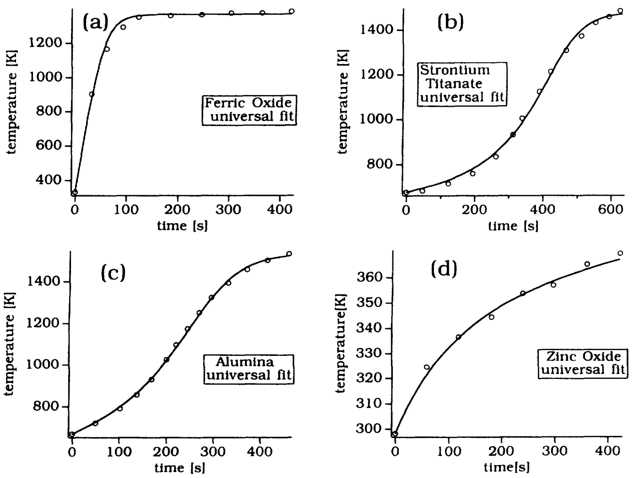

FIG. 5. Thermal-runaway observations fitted by our theory as in Fig. 4, but with the "universal" approximation to the switching function given by (3.4).

are proportional to the surface S of the sample, so that

$$\frac{d\varepsilon}{dt}=P_{\text{abs}}-S\sigma T^4 , \tag{4.2}$$

where σ is the Stephan-Boltzmann constant. The sample mass ρV does not change during heating. Nor is the value of C influenced considerably by changes of temperature. It is important to observe here that C is the specific heat of the lattice rather than of the absorber particles. The latter do change their specific heat during heating as they are freed form the confining wells. However, the process of heating involves energy being received from the microwave field by the absorbed particles and handed over to the "sink," viz., the lattice. On an average (over the microwave cycle), the absorber particle does not retain the energy absorbed. The energy poured into the lattice sink raises the temperature of the lattice through (4.1). Acting as a reservoir, through stochastic interactions described by the last term of (2.5), the lattice then raises the temperature of the entire sample including the absorber particles. On combining (4.1) and (4.2), we obtain

$$\frac{dT}{dt}=\frac{P_{\text{abs}}}{\rho VC}-\frac{S\sigma}{\rho VC}T^4 . \tag{4.3}$$

Comparison of (4.3) and (1.1) makes clear the meaning of the quantities k_A, k_M, and σ_1. The absorbed power P_{abs} is proportional to the absorption coefficient K'' (imaginary part of the dielectric constant), the volume V of the sample, the energy density of the electromagnetic field $u_{\text{EM}}=E^2/8\pi$, and the frequency ω of the microwave radiation:

$$P_{\text{abs}}=K''V\frac{E^2\omega}{8\pi} . \tag{4.4}$$

The power of the microwave field, $E^2\omega/8\pi$, is proportional to the incident power P of the source:

$$\frac{E^2\omega}{8\pi}=zP , \tag{4.5}$$

where z is a parameter determined by geometrical and propagational conditions of the experimental setup. On combining the formulas (4.1)–(4.5) and comparing with (1.1), we obtain the prescription for calculating the parameters of the temperature-time equation (1.1) from microscopic considerations:

$$k_A=\frac{n_AK_A''z}{\rho C} , \tag{4.6}$$

$$k_M=\frac{n_MK_M''z}{\rho C} , \tag{4.7}$$

$$\sigma_1=\frac{S\sigma}{\rho VC} . \tag{4.8}$$

Here n_A and n_M are the numbers of the two kinds of absorber particles A and M (as explained in the Introduction and in Ref. 7) and K_A'' and K_M'' are the respective absorption coefficients *per particle.* The quantities c_A and c_M introduced in Ref. 7 are, obviously, $zK_A''/\rho C$ and $zK_M''/\rho C$, respectively, and depend on the conditions of the experimental setup through z as well as on the properties of the ceramic material through K'', ρ, and C.

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APPENDIX

In the course of the application of the phase-space formulas for the switching function, we have found the following reduction of (2.4) which is valid for the sinusoidal potential (2.3). The complementary error function erfc can be approximated by[16]

$$\text{erfc}(x)=(a_1y+a_2y^2+a_3y^3)\exp(-x^2) , \tag{A1}$$

where

$$y=\frac{1}{1+px} \tag{A2}$$

and the numerical coefficients p, a_1, a_2, and a_3 have the values

$$\begin{aligned} &p=0.470\,47, \quad a_1=0.348\,024\,2 , \\ &a_2=-0.095\,879\,8, \quad a_3=0.747\,855\,6 . \end{aligned} \tag{A3}$$

With the substitution the integral in the numerator of the right-hand side of (A6) reduces to a sum of elementary integrals:

$$\int_{-\pi}^{\pi} dx \exp\left[\frac{\Delta}{2kT}\cos x\right]\text{erfc}\left[\left[\frac{\Delta}{2kT}(1+\cos x)\right]^{1/2}\right]=\exp\left[-\frac{\Delta}{2kT}\right]\sum_{n=1}^{3} a_n\int_{-\pi}^{\pi} dx \left[1+p\left[\frac{\Delta}{2kT}\right]^{1/2}\cos\frac{x}{2}\right]^{-n} .$$

On carrying out these integrals, one obtains

$$f(T)=\frac{2\exp(-\Delta/2kT)}{\pi I_0(\Delta/2kT)}F(a) , \tag{A4}$$

where

$$F(a)=\begin{cases}\left[a_1-\dfrac{a_2}{a^2-1}+a_3\dfrac{2+a^2}{2(a^2-1)^2}\right]G(a)+\left[a_2+a_3\dfrac{a^2-4}{2(a^2-1)^2}\right]\dfrac{a}{a^2-1} & \text{for } a\neq 1\ ,\\ a_1+\frac{2}{3}a_2+\frac{7}{15}a_3 & \text{for } a=1\end{cases} \tag{A5}$$

and

$$G(a)=\begin{cases}\dfrac{2}{(1-a^2)^{1/2}}\arctan\left[\left[\dfrac{1-a}{1+a}\right]^{1/2}\right] & \text{for } a<1\ ,\\ \dfrac{1}{(a^2-1)^{1/2}}\ln\left|\dfrac{\sqrt{a-1}+\sqrt{a+1}}{\sqrt{a-1}-\sqrt{a+1}}\right| & \text{for } a>1\ .\end{cases} \tag{A6}$$

In (A6), a is given as $a=p\sqrt{\Delta/kT}$. Equation (A4) is a useful representation of the switching function (2.4).

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