Kinetics of reversible thermal breakdown in thin films

A. V. Subashiev and I. M. Fishman

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR
(Submitted 26 March 1987)
Zh. Eksp. Teor. Fiz. **93**, 2264–2266 (December 1987)

Reversible thermal breakdown in thin films is shown to resemble a first-order phase transition. The probability, size, and growth time of a critical nucleus are calculated.

Reversible thermal breakdown is a typical example of bistable behavior in nonequilibrium systems.[1] If a conducting specimen is placed in a (specified) external magnetic field, its temperature will depend on the relative rates of heat evolution and dissipation. If the heat evolution is nonlinear

$$P(T)=\sigma(T)E^2V$$

and exceeds the heat transfer $Q(T)$ for a certain range of temperatures (Fig. 1), reversible thermal breakdown, accompanied by a rapid temperature change with hysteresis, may occur in the system [here $\sigma(T)$ and V are the conductivity and volume of the specimen, and E is the electric field].

We will show below that reversible thermal breakdown in thin films is completely analogous to a first-order phase transition: at a certain "supersaturation," a macroscopic nucleus of hot "phase" forms in unstable equilibrium with the surrounding cold film. The thermal fluctuations are responsible for critical nucleation. Macroscopic critical nuclei do not occur in one- or three-dimensional systems.

The kinetics of thermal breakdown in a thin film of thickness d is described by the heat equation, averaged over the thickness of the film:

$$c\frac{\partial T}{\partial t}=\varkappa_d\Delta T(\mathbf{r},t)+P(T)-Q(T). \tag{1}$$

Here $\mathbf{r}$ is a point in the plane of the film, $\varkappa_d$ is the thermal conductivity, and c, $P(T)$, $Q(T)$ are the specific heat, heat evolution, and heat dissipation per unit film area. The homogeneous steady states of the film are found from the equation $P(T)=Q(T)$, which for a range of external fields E has three solutions T_l, T_c, and T_h, of which T_l and T_h are stable (Fig. 1). If the temperature distribution is nonuniform, the characteristic width of the interface (wall) between the hot and cold phases is

$$r_\varkappa\sim[\varkappa(d(P-Q)/dT)^{-1}]^{1/2}|_{T=T_l,\,T_h}$$

($r_\varkappa\gg d$). The width $r_\varkappa$ may vary widely from $\sim10^{-2}$ to $\sim10^{-6}$ cm, depending on the experimental conditions.

Let us first consider time-independent, axisymmetric solutions of (1), $T=T_0(r)$; these correspond to round nuclei of radius $R\gg r_\varkappa$. Multiplying (1) by dT_0/dr and integrating over r, we get

$$\varkappa\int_0^\infty\left(\frac{dT_0}{dr}\right)^2\frac{dr}{r}-\int_{T_l}^{T_h}(P-Q)\,dT=0. \tag{2}$$

Most of the contribution to the first term in (2) comes from the region $R_c-r_\varkappa\lesssim r\lesssim R_c+r_\varkappa$, where R_c is the radius of the nucleus. A steady state with $R_c\to\infty$ (a straight-line interface) is therefore possible if

$$J=\int_{T_l}^{T_h}(P-Q)\,dT=0. \tag{3}$$

The quantity J may be regarded as a "supersaturation," while Eq. (3) is analogous to Maxwell's rule.[2,3] If the supersaturation is small ($R_c\gg r_\varkappa$), Eq. (2) gives

$$R_c=\frac{\varkappa}{J}\int_0^\infty\left(\frac{dT_0}{dr}\right)^2dr. \tag{4}$$

The nucleus of new phase in the film is thus macroscopic, as in the capillary theory of first-order phase transitions.[3] Thermal conduction across the wall of the nucleus plays the role of surface tension.[1)]

As usual, nuclei with $R=R_c$ are unstable; they grow (decay) for $R>R_c$ ($R<R_c$). We will find the growth rate by exploiting the fact that for nuclei with small curvatures ($R\gg R_c$), Eq. (1) has a solution that describes a moving wall (a switching wave[1,4]), and the thickness of the wall remains comparable to $r_\varkappa\ll R$. We can therefore use the approximate relation $T=T_0[r-v(R)t]$ to find the average growth rate. Proceeding as in the derivation of (4), we find from (1) that

$$v(R)=dR/dt=v_\infty(1-R_c/R), \tag{5}$$

where the quantity

$$v_\infty=J\left[c\int_0^\infty(dT_0/dr)^2\,dr\right]^{-1}$$

can be regarded as the velocity of the moving planar interface (wall) at a given supersaturation J.

Relation (5) can be used to calculate the critical nucleation time (Zel'dovich time[5]): $\tau_n\approx R_c/v_\infty$.

An analysis shows that all the properties of a round nucleus apart from its radius are stable to small temperature fluctuations $\delta T=T-T_0(\mathbf{r})$, again as would be expected for a first-order phase transition.

A nucleus of critical size can form only due to fluctuations in P, Q, and T. We can estimate the probability for a nucleus of radius R_c to form spontaneously at a given supersaturation J. In contrast to the case of first-order phase transitions, here the standard thermodynamic formulas cannot

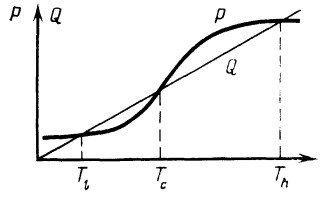

FIG. 1.

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be used and one must solve a kinetic problem. If $g(R)$ denotes the size distribution of the nuclei, then the flux of nuclei as a function of R is given by

$$j=v(R)g(R)-D(R)\,[dg(R)/dR], \qquad (6)$$

where the effective diffusion coefficient $D(R)$ (see, e.g., Ref. 6) is proportional to the correlation function $\delta v(R,T)$ for the fluctuations in nucleus growth rate caused by the fluctuations $\delta P, \delta Q, \delta T$. We will assume that the correlation radius for the fluctuations is small, $r_{\delta v} \ll r_\varkappa$. It is then easy to show that $D(R) = D_0 r_\varkappa / R$ holds. The steady-state distribution function for $R \leqslant R_c$ follows from the condition $j(R) = 0$:

$$\ln g(R_c) = -(v_\infty R_c/2D_0) R_c/r_\varkappa. \qquad (7)$$

Up to exponentially small terms, expression (7) gives the probability for the formation of a critical nucleus. If the magnitudes of the fluctuations δP and δQ are close to their equilibrium value, the probability (7) is nearly equal to the probability of a temperature fluctuation $\delta T = T_h - T_l$ in a volume $V = 2\pi R_c r_\varkappa d$ and is minute for any reasonable choice of parameter values. However, the size of the current fluctuations in the heat evolution may substantially exceed the equilibrium value if the power applied to the material is large and the temperature is low. For example, during avalanche breakdown in a p-n junction (Ref. 7), P can reach 10^{10} W/cm^3. The width of the strong-field region ($E \approx 10^6$ V/cm) is $d \approx r_\varkappa \approx 10^{-5}$ cm. Thus

$$\ln g(R_c) \approx c r_\varkappa^2 \Delta T/eEd \approx 10^2 - 10^3,$$

which is large enough so that the nucleation kinetics should be observable.

Large fluctuations may also be produced by fluctuations δP in films heated by optical radiation.[2,4] The problem considered above is also relevant to degradation processes in semiconductor lasers.

We are grateful to B. M. Ashkinadze for helpful discussions.

[1)] In a one-dimensional system (rod), the heat flux from the hot to the cold region is independent of the length of the former, apart from exponentially small terms; nuclei of new phase are therefore "microscopic," i.e., $R_c \approx r_\varkappa \ln J_0/J$ (Ref. 4). In a three-dimensional system, because heat transfer proceeds from the surface while heat is evolved throughout the interior, the size of a nucleus (inhomogeneity) may be either $r_\varkappa$ or else close to the length l of the specimen (when $r_\varkappa > l$). In the latter case, the entire specimen may be regarded as a critical nucleus.

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Translated by A. Mason