THERMAL SPIKES AND SPUTTERING YIELDS

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Recent experiments on the erosion of condensed gases and alkali halides by incident ions have renewed interest in the description of thermal spikes. In such spikes a localized, transiently heated region produced by incident radiation may induce activated processes like evaporation of atoms or molecules from a surface. In this paper are presented useful expressions to describe the thermal sputtering for materials having a temperature dependent thermal diffusivity, using a heat capacity and thermal conductivity which vary as $C = C_0 T^{-1}$ and $K = K_0 T^{-1}$, respectively, and assigning a width to the initial temperature distribution.

INTRODUCTION

There has been continuing interest in thermal spikes as a model describing the dispersal of energy deposited in a solid by radiation.\textsuperscript{1,2} Such spikes have been used to describe various thermally activated processes,\textsuperscript{2,3} such as the sputtering (or erosion) of particles from the transiently heated surface of irradiated materials. Recent experimental work on the sputtering of condensed gases\textsuperscript{4,5} and alkali halides\textsuperscript{6} suggests that thermal sputtering, resulting from the conversion of the deposited electronic energy into heat, may occur in these systems. Therefore it is useful to have simple analytical estimates of the sputtering yield for comparison with data.

Models of thermal spikes have been given by Vineyard\textsuperscript{2} and others.\textsuperscript{1} The applicability of these results to particular materials is limited by the assumption that the thermal diffusivity is temperature independent and often by the further assumption that the initial heat distribution is narrow and may be represented adequately by a delta function. It is straightforward, however, to generalize the results of Vineyard to allow for a temperature independent thermal diffusivity and an initial heat distribution with a finite width.

The details of the transfer of the deposited energy into heat are not well known. Here we assume that only a fraction of the total deposited energy initially contributes to the thermal spike which subsequently cools by conduction and that the resulting temperature rise activates certain processes. Assuming the classical heat conduction laws of the continuum for an isotropic, homogeneous medium are valid, the non-linear heat conduction equation is solved for cylindrical and spherical sources in a material having a thermal conductivity and heat capacity with different temperature dependences.

Once the temperature of the material is obtained as a function of position and time, the expression for the activated process, here thermal sputtering, may be integrated to obtain the total yield. The results for the cylindrical source are presented in terms of tabulated integrals which reduce to simple analytic expressions, such as those of Vineyard,\textsuperscript{2} when the initial distribution is a delta function. These expressions, we expect, will be useful for examining models of sputtering and for comparison with experimental data. They can also be used easily to describe other activated processes such as atom migration.

SOLUTION OF THE HEAT CONDUCTION EQUATION

In an isotropic, homogeneous medium with thermal conductivity $K$ and specific heat $C$ the equation for heat conduction is

$$\nabla (K \nabla T) = \frac{C}{\rho} \frac{\partial T}{\partial t}$$

(1)

We assume the temperature dependence of $K$ and $C$ over a reasonably wide temperature range can
be represented by
\[ K = K_0 T^{n-1} \]
\[ C = C_0 T^{n-1} \]
where \( K_0, C_0, m \) and \( n \) are constants for a given material. Making the substitution suggested by Vineyard,
\[ U = T^n \]
Equation (1) is non-linear in \( U \), having the form
\[ \nabla^2 U^{1+1/\delta} = \left( \frac{C_0}{(1 + \delta)K_0} \right)^{\delta} \partial \partial \]
where \( n, m = 1 + \delta, \delta \) being a measure of the non-linearity of the equation.

Making the further assumptions of a negligible background temperature and an infinite medium, the solution of (5) in a cylindrical geometry with radius \( \rho \) is
\[ U = \frac{\partial}{\partial z} \left[ 1 + \frac{\delta \rho^2}{\Delta_0^2} \right]^{1+1/\delta} \]
where \( \Delta_0^2 = \partial, 4K_0(T + t_0) C_0 \rho^2 \) and \( \partial \) is a normalization constant; \( \delta > -\frac{1}{2} \).

As the heat added per unit volume is \( \int_0^1 C \, dT = (C_0 n) T^2 = (C_0 n) U \), the factor \( \partial \) is obtained by requiring
\[ \int_0^1 \frac{dE}{d\Delta} \, d\partial = f \, \frac{dE}{dx} \]
Here \( f \) is the portion of the deposited energy converted to heat, \( dE \, dx \) is the material stopping power, and, using Eq. (6), \( f = (\pi C_0) \int f \, d(\partial E \, dx) \).
Finally the width of the distribution at \( t = 0 \), \( \Delta_0 = \partial, \) is parametrized by the quantity \( t_0 \). For the linear case, where \( n = m \) (i.e., \( \delta = 0 \)), Eq. (6) reduces to the usual Gaussian solution in \( \rho \) discussed by Vineyard and others. For \( -\frac{1}{2} < \delta < 0 \) the solutions are zero beyond \( \rho^2 = \Delta_0^2 \).

With the same assumptions, the solution of (5) in a spherical geometry with radius \( \rho \) is
\[ U = \frac{\partial}{\partial z} \left[ 1 + \frac{\delta \rho^2}{\Delta_0^2} \right]^{1+1/\delta} \]
where now,
\[ \Delta_0^2 = \partial, 4K_0 C_0 \rho^2 \] and
\[ A = 2(2 - \delta) K_0 C_0 \rho^2 \]
The normalizing factor \( \partial \) is obtained by requiring that \( \int (C_0 n) U \, d\partial = E \), where \( E \) is the energy deposited in the spike. This gives
\[ \partial = n E_c \pi^{3/2} C_0 N, \]
with
\[ N = \Gamma(1-\delta-1) \delta^{1/2} \Gamma(1-\delta) \quad \text{for} \quad 0 \leq \delta < 2, \]
and
\[ N = \Gamma(1-\delta-1) \delta^{3/2} \Gamma(3/2-\delta) \]
for \( 0 > \delta > -1/2 \), where \( \Gamma(x) \) is the tabulated gamma function. In the limit that \( \delta \to 0 \), \( N \to 1 \) and \( U \) is a gaussian. The above expressions for \( U \), hence temperature, are used in the following section to evaluate the spattering yields due to thermal spikes.

**SPUTTERING YIELD**

Describing the sputtering as due to evaporation from the transitory heated surface, the vaporization flux \( \phi \) is given by
\[ \phi = \frac{cP}{(2\pi MKT)^{1/2}} \]
where \( P \) is the equilibrium vapor pressure, \( M \) is the mass of the vaporizing species and \( c \) is the evaporation coefficient. A simple modification is necessary if the vapor species is not identical to the solid species. The equilibrium vapor pressure may be approximated reasonably well as
\[ P = P_0 \exp\left( -\frac{L}{T} \right) \]
where \( P_0 \) and \( L \) are slowly varying functions of temperature which are assumed, initially, to be constants.

With these definitions, the sputtering yield (particles removed per incident ion) from a plane surface is
\[ S_{\text{thermal}} = \int_0^{\infty} dt \int_0^{\infty} 2\pi \rho \, d\rho \, d\phi = \int_0^{\infty} dt \int_0^{\infty} 2\pi \rho \, d\rho \]
\[ \times P_0 \exp\left( -\frac{L}{T} \right) \]
Taking \( T = U^{1/\delta} \) for the cylindrical geometry, Eq. (6), and using the substitution \( T = 1/\sigma \) suggested by Vineyard, where in this case \( \tau = \partial, \Delta_0^2 \) and \( \sigma_0 = (1 + \delta \rho^2 \Delta_0^2)^{-1/2} \) we obtain
\[ S_{\text{v}} = n^2 m^2 P_0 \int f \, d(\partial E \, dx) \]
\[ \times \left( \frac{2\pi MKT}{(2\pi MKT)^{1/2}} \Delta_0 \right) \]
\[ \times \int_{\partial} \, d\tau \int_{\sigma_0}^{\infty} d\sigma \, \tau^{m+1-\delta/2} \sigma^{n-1-\delta/2} \exp(-L/\tau). \]
Here \( \tau_0 \) is the inverse of the maximum temperature of the spike, \( \tau_0^{-1} = T_0 = T(\mu = 0, t = 0) \). The double integral in Eq. (11) may be reduced to two single, tabulated integrals, yielding

\[
S_e = \frac{m^2}{8\pi(K_0 L^m)(C_0 L^m)(2\pi MkL)^{1/2}} I_e
\]

where \( I_e \) is a dimensionless quantity,

\[
I_e = \Gamma \left( n + m + \frac{1}{2}, \frac{L}{T_0} \right) - \left( \frac{L}{T_0} \right)^{2\alpha} \times \Gamma \left( n - m + \frac{1}{2}, \frac{L}{T_0} \right)
\]

The function, \( \Gamma(a, n) = \int_0^\infty t^{a-1} e^{-t} \, dt \) is the incomplete gamma function which is tabulated or may be evaluated numerically. For a narrow line source as an initial condition, \( T_0 \to \infty \) and \( I_e = \Gamma(n + m + \frac{1}{2}) \). If, in addition, \( n = m \), the result of Vineyard is recovered, however the more general expression (12) is almost as simple to use. In the other limit, \( L \gg T_0 \), \( I_e \to 2nL(T_0)^{n+1} \times \exp(-L/T_0) \).

Adopting the solution, \( \psi \), in Eq. (7), for an infinite medium and applying it to a semi-infinite medium, the usual procedure the sputtering yield for a spherical source at a depth \( x_0 \) is

\[
S_e = \frac{m^2}{4\pi(3m-n)(nL)^{1/3}} \left( \frac{L}{T_0} \right)^{2\alpha} \frac{1}{(2\pi MkL)^{1/2}} I_e
\]

where the double integral may be written

\[
I_e = \frac{P_0}{(2\pi MkL)^{1/2}} \int \tau^{m+2} \exp(-L\tau) \left( \frac{L}{T_0} \right)^{2\alpha} \frac{1}{T_0} \times \Gamma(n-m+1, \frac{L}{T_0})
\]

where \( \tau = \Delta_t \Delta_s \), \( \Delta_t(t) \) is the temperature of the spike at \( t = 0 \), and \( T_0 \) is the maximum temperature of the spike from the surface. The expression for \( \Delta_t \) is approximated and \( T_0 \) is the maximum temperature of the spike at \( t = 0 \). For this case the first integral must be approximated or numerically evaluated. The above expressions are simply modified if \( P_0 \) and \( L \) are also given a power-law, temperature dependence.

CONCLUSIONS

Results for the thermal sputtering yield given in Eqs. (12) and (13) are useful for comparison with experiment without the rather restrictive assumption of a temperature independent thermal diffusivity. Such a comparison has been made in Ref. (4) for ions incident on ice. The expressions for the temperature and the yield are not significantly complicated by using a temperature dependent thermal diffusivity. However, the expressions in Eqs. (12) and (13) are quite sensitive to the choice of \( L \). In the cylindrical case. In the limit that the initial widths are narrow (i.e., \( L \), \( T_0 \) small) simple, analytic expressions for sputtering yield result whether or not the thermal diffusivity is temperature dependent, which is quite useful. In this limit, for example, \( S_e = (dE/dx)^n \), but for a finite width, the yield becomes a rapidly varying function of \( dE/dx \) only approaching the narrow width limit when \( dE/dx \) is large. Using the above expressions, large \( dE/dx \) means \( \int dE/dx \), etc.

The validity of the thermal spike model of particle erosion described here has the same deficiencies discussed by previous authors (e.g., validity of the classical heat conduction equation, the use of a solution based on an infinite medium, background temperature, change of phase, etc.) In addition, if the time for transfer of deposited energy into heat energy is comparable to the characteristic heat conduction times, which may be the case for sputtering due to electronic processes, then a source term should be included in the conduction equation, Eq. (1). However, the thermal sputtering model does provide a useful starting point for interpreting experimental data. This is true both for experiments measuring total yield, using Eqs. (12) and (13) for comparison, as well as recent experiments on the velocity distribution of sputtered products. Employing the expression for particle flux, \( \psi \), in Eq. (8).

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REFERENCES


