Effect of Material Parameters on the Attenuation and Amplification of an Incident Laser Beam

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Abstract Laser processing is being increasingly used for treatment and processing of various materials. Etching, computer board manufacture, rapid prototyping and NC Machine tool technology have all adapted to laser use. Laser heating supplements traditional methods like nitriding and carburizing. The type of laser and its power output can vary significantly along with the depth of affected material. The moving heat source in laser melting is modeled by transformations together with a decoupling for the heat and mass transfer terms. The scale for heat diffusion is different from conduction and when small times are involved as in rapid solidification, the effect may be pronounced. Very little published work has appeared on the stability of the solid-liquid interface. A few solutions are known for certain geometries for the moving heat source. Approximate solutions incorporating the convective surface flux are obtained. It is shown that under certain conditions, a high Stefan number can attenuate an impinging laser beam and sustain thermal oscillations in the substrate. Application to thin films and amorphous material formation give criteria derived for stability in terms of surface parameters. The analysis does not include quantum effects likely in the nano region. Additionally, the surface reflectivity would influence the attenuations of the incident beam.

1 Introduction

Moving heat sources of laser melting are treated with various transformations along with a decoupling for the heat and mass transfer terms. The scale for heat diffusion is different from that of conduction and when small intervals of time are involved as in rapid solidification, the effect may be pronounced. Very little published work

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Metal	Cp (J/kg K)	θ	L(kJ)	Ste	$1/Ste(\mu)$	α
Cast iron	544	1810	272	0.33	3.03	0.134
Ni	456	1728	297	0.45	2.25	0.155
Ag	235	1235	111	0.49	1.11	1.7
Al	904	933	398	0.67	1.5	0.86
Cu	385	1358	205	0.49	2	1.14

Table 1 Important thermal and non dimensional parameters for some engineering materials

exists on the stability of the solid-liquid interface. It is essential to understand this because instabilities in solidification processes will affect the final finish of the surface. Novel transforms specific to melting with convective surface conditions are applied to this analysis. Phase field theory notions are applied to specific cases for amorphous film transformation. The effect of material parameters to attenuate or even amplify input frequencies to nano or tetra levels is outlined.

1.1 Theoretical Considerations and Review of Past Work

Laser surface modification is through a phase change. More recently, laser processing has found application in computer controlled machine tools and Rapid prototyping, 3D printing and 3D sintering. 3D printing was described in USP 5398193 [\[1](#page-6-0)]. Sahoo et al. [\[2](#page-6-0)] describes pulsed laser treatment for TiC coating on Al. The depth of affected materials can range from nanometers to millimeters depending on types and power outputs (Table 1). Laser processing has been used to form p-n junctions and semiconductor annealing. The application of a pulsed laser gives interesting possibilities for further experimental verification due to the precision involved in focusing and controlling the intensity and time. The subsequent theoretical considerations assume that heat is not dissipated either by ablation or pore formation. Apart from the heat input, the frequency of the input laser also may affect the results. The properties of the underlying material attenuate the pulse, and it is subsequently shown that in certain cases the pulse can be sustained or even amplified.

The problem of phase change with a moving interface has been described by Carslaw and Jaeger [\[3](#page-6-0)], Stefan [\[4](#page-6-0)], Ingersoll et al. [\[5](#page-6-0)], as well as Crank [[6\]](#page-6-0), among others. A comprehensive study by Langer [\[7](#page-6-0)] appeared dealing with many core issues that occurred during solidification and growth of solid phases where thermal and concentration effects were combined. Many important issues are addressed with the linearization used by Mullins and Sekerka [[8\]](#page-6-0). In the present work, the coupled heat diffusion and mass equations are also solved using a decoupling technique (Table [2\)](#page-2-0).

Laser	Wavelength	Pulse length	$(Dt)^{0.5}$ Si	$(Dt)^{0.5}$ GaAs	Optical depth Si	Optical depth GaAs
XeCl excimer	308	30	1660	973	6.8	12.8
KrF excimer	248	30	1660	973	5.5	4.8
ArF excimer	192	30	1660	973	5.6	10.8
Nd; Y AG	1060	₍	743	435	1000	N/A

Table 2 Characteristics of some laser types

Source Pulsed laser heating and melting, Sands [[10](#page-6-0)]

2 Theoretical Considerations and Stability

The linearization $T = T_0 + Gz$ for the solubility phase boundary, and the similarity transformation, moving with the interface at velocity V give for a dimensionless "diffusion field" u: $\nabla^2 U + 2I\delta u/\delta z = 0$ with a thermal parameter $u = \Delta T/(L/cp)$ and the corresponding chemical potential $u = \mu/\Delta c$ ($\delta u/\delta c$), where $l = 2D/V$ (diffusive length) and $L =$ latent heat, c_p the specific heat. The diffusion equations for both heat and mass are similar. A similar linearization was used by Davis and Schulze [\[9](#page-6-0)] for studying morphological instability for small Stefan numbers. Li and Beckermann [[10\]](#page-6-0) related growth to the critical wave number found by Mullins and Sekerka [[8\]](#page-6-0). Pulsed laser heating used for surface modification was described in detail by Sands [\[10](#page-6-0)]. Ragas-Trigos and Calderon [\[11](#page-6-0)] have analysed the effects of a periodic heat source with sinusoidal modulation along with various boundary conditions by Greens function method. Kim et al. [[12\]](#page-7-0) have given predictions of solidification velocity in ion implanted GaAs (Gallium Arsenide). Laser spot welding with enthalpy method analysis has been described in Duggen et al. [[13\]](#page-7-0). A 3-D finite element analysis for laser melting has been described by Contuzzi et al. [\[14](#page-7-0)]. Following Carslaw and Jaeger [\[3](#page-6-0)], the point source is practically modelled as a rectangular, disc shaped or strip source, for which solutions are available. All these solutions are of the form $\exp(-x^2)/(\alpha t)$, with α being diffusivity. Specific solutions for a moving source are also given in Paterson $[15]$ $[15]$. For metals, the transients are damped out very quickly, corresponding to the Fourier number $2\pi/k$. So for one wavelength, the attenuation goes as $\exp(-2\pi) = 0.0019$. Hence the solid of more than one wavelength depth may be regarded as infinite for all practical purposes. Now consider a 1-dimensional geometry for simplicity. This could also correspond to spherical symmetry after applying certain simple transformations. Applying a similarity transformation to the thermal diffusion equation with re-melting terms, results in (after suitable normalized variables are applied);

$$
T'' + \eta/2T' + (\varepsilon L/\rho c_p)dc/dt = 0,
$$
\n(1)

T is temperature, η similarity parameter, α diffusivity, ε porosity, c concentration, t time, ρ Density, ∇ nabla.

The transformation of the concentration derivative to non dimensional similarity parameter η, a characteristic length may be extracted from the Fourier number after knowing the characteristic time related to the frequency of the pulsed laser source. By examination of the coefficients of the equation and introducing a small parameter $\mu = g \theta c_p/(gL) = g/(gSte)$, one obtains

$$
\mu\theta'' + 0.5 \,\mu\text{Fo}\theta' + 0.5 \,\text{Bi}\theta = 0,\tag{2}
$$

(where g is the thermal gradient, Ste Stefan number, Bi Biot number, and θ non dim temperature). The above equation can be solved exactly, with the inner and outer solutions easily obtained by letting $\mu \ge 0$ and $\text{Bi} \ge 0$ respectively, (assuming μ Fo > 0). Accordingly, θ '/ θ = $-(BiSte/\epsilon)(gFo)$, θ ''/ θ = $-Fo/2$ which have immediate solutions. For small μ , the Eq. (2) can be written without the second derivative terms, thus becoming one dimensional in θ . On the other hand, if Bi is very small, the reverse holds true, and the higher terms appear. If F_0 is very small a third option is available leading to hyperbolic or sinusoidal solutions depending on the sign of Bi. The equations can still be solved exactly using a similarity transformation. The small parameter μ is calculated for some materials in Table [1](#page-1-0). The parameter Fo²(gK)/(eSten) can be recognized as (Fo²/Bi)Cp θ , and can be extracted from columns 1 and 2 of Table [1](#page-1-0). (Note: this is effectively a modification of the Fourier length given by Carslaw and Jaeger [[3\]](#page-6-0), through the Stefan number due to the interface effect. It is seen to be a scaling through an effect on time.)

2.1 Computational Results

Two time scales which can be introduced are due to Fourier conduction and phase front motion. The scales are quite different in most cases, especially if the freezing is slow. In effect, the temperature sensed due to heat conduction of the normal boundary value problem has a superimposition of a "wave" of temperature caused by the motion of the phase front. The Dirichlet solution is modified for convective conditions by the approximations:

$$
h(TS - T0) = k dT/dx
$$
 (3)

where

$$
dT/dx = (TS - Tm)/1
$$
 (4)

Fig. 1 Dirichlet simulation for phase change

Fig. 2 Convective effects due to Bi number

Giving

$$
hl/k(Ts - T_0) = (Ts - T_m)
$$
\n⁽⁵⁾

Normalizing to Tm = 0, Ts = $\frac{BiT_0}{1 + Bi}$ where Bi = hl/k, h the convective film coefficient, l characteristic length. The Dirichlet solution is used to obtain the convective solution after replacing the boundary temperature by the modified temperature due to Bi. Simulations using WOLFRAM for the spherical case are shown in Fig. 1 along with the modification for convection in Fig. 2.

3 Discussion

According to Granasy and James [[16\]](#page-7-0), studies of laser treated materials began with amorphous layers in oxides, where it was found that some oxide systems with simple stoichiometric glass compositions had crystal phases with the same composition as the parent matrix. In certain cases, homogenous nucleation occurs without the need for nucleating agents aided by surface imperfections. Nucleation rates (excluding heterogeneous nucleation), were studied for several stoichiometric glasses by James [[17\]](#page-7-0), with steady state rates varying from 1.9×10^{12} m³ s⁻¹ for Ba_2SiO_2 glass to 1.7×10^6 m³ s⁻¹ for CaO–SiO₂ glass. Selective laser melting for processing metallic glasses is described by Pauly et al. [[18\]](#page-7-0). Surface modifications of CoCe alloys have been reported by Hoekstra et al. [\[19](#page-7-0)], Li et al. [[20\]](#page-7-0), using large area electron beam irradiation of microsecond duration. Interesting results for Pt foil are reported by Xraysweb [\[21](#page-7-0)]. Taking typical values for Co as $L = 66$ cal/gm, Cp as 0.42 cal/gK, the Stefan number for a temperature differential of 400 K comes to approximately 0.4. From (1) (1) the critical Stefan number is seen to be 0.025 g/ ε . For suitable values of the Stefan number, it is possible for the laser frequency to match the resonant frequency and allow for thermal oscillations to be maintained, thus preventing homogenous nucleation. The wide range of nucleation frequencies seen for glasses indicates that other mechanisms are involved in modifying the laser action, suggested here to be an attenuation mechanism. Apart from attenuation, the high under cooling required for amorphous alloy formation may be possible by using very short laser pulses, according to Pauly et al. $[18]$ $[18]$. The effect of under cooling on amorphous alloy formation is widely acknowledged, however, the separation of this from pulse duration and initiation of homogeneous nucleation, the shape of the amorphous phase field and other factors remain an area of further work. Recently, work on high-frequency laser pulses by Mankowsky and Subedi [[22\]](#page-7-0), shows that due to electronic effects of THz frequency laser pulses, superconductivity and transitions from insulator to metal have been observed. The effect has been attributed to phononics and staggered dilation and contraction of $Cu₂O$ interlayer distances in Y Ba CuO compounds. Gulian et al. [\[23](#page-7-0)] have also discussed evidence of superconductivity in laser processed $Sr₂RuO₄$ (triplet superconductor) using SQUID measurements even up to 200 K and beyond, in the top layer affected by the treatment (the "crust"). In both these examples, the presence of Oxygen and distortion of the lattice are contributing factors as are the phonon contributions. High temperature super conductors processed by a laser are described by Bauerle [\[24](#page-7-0)]. Laser pulses appear to shift the atoms in ceramic crystals and possible room temperature superconductivity for trillionths of a second has recently been reported.

4 Conclusion

It is found that for the usual metals, the second order terms cannot be neglected (inner solutions), and similarly for the Biot numbers encountered, the outer solution also needs to be used. The lengths involved may be seen to vary with conductivity and other parameters and thus for thin films and small dimensions the transient effects may become significant. Under certain conditions, the film can be made to sustain thermal oscillations, analogous to a mechanical spring damper system. The approximations for Fo and Bi around 0.2 and 0.1 reveal that Ste is around 0.025 g/e, and consequently the second order inertia terms varying as 1/Ste are one order of magnitude more than the damping and stiffness terms in the analogy discussed. Corrosion and oxidation due to the mass influx of foreign species from the surface layer are not considered, but would be an area of further interest in the future. Similarly the formation of an amorphous surface layer through rapid self-quenching with short heating times is an exciting possibility for development of new two phase film materials and has already been reported by several researchers. However, further work on separating the effect of the deep eutectic characteristic of amorphous glass formers from the phase field effect needs to be done. Extension of this work to the application of high temperature superconductivity seems a possibility, the data on femto-second results are however scarce and need corroboration.

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