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An analytical model for nanoscale electrothermal probe recording on phase-change media

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Scanning probe memories are now emerging as a means of achieving nanoscale resolution data storage. The use of microscopic conductive tips in contact with a phase-change material to record data as amorphous and crystalline marks is one such approach, making use of the large difference in electrical conductivity between the two phases to distinguish between two binary states on replay and hence provide a memory function. The writing process is complex and involves electronic, thermal, and phase-change processes that are difficult to model and study except using numerical techniques. A simplified analytical model of electrothermal writing by probe on a basic two-layer phase-change structure is developed here, and used to predict the required voltage levels for recording and the expected diameters of recorded crystalline and amorphous marks. A simplified model of cooling and solidification was also developed to study the cooling rates during amorphization. The predictions are shown to be in agreement with published experimental measurements and numerical simulations. The developed analytical models were extended to investigate the effects of introducing coating layers on recording voltage levels, to study the depth profiles of recorded marks, and to derive expressions for the capacitance and resistance of the phase-change layer that contribute to the transient behavior of the recording system. © 2006 American Institute of Physics. [DOI: [10.1063/1.2165408](https://doi.org/10.1063/1.2165408)]

I. INTRODUCTION

The nanoscale resolution of scanning probe technologies is now being exploited to circumvent the limitations of conventional data storage techniques and attain ultrahigh areal storage densities.¹ Phase-change material, in particular, is an attractive storage medium when used with electrical scanning probes, mainly due to the large difference in electrical conductivity between the amorphous and crystalline phases that can be used to represent the two binary states for information storage. In electrical probe storage, the temperature in the phase-change layer is raised, through Joule heating, by the application of a suitably high voltage either to the transition temperature to induce irreversible amorphous to crystalline transformations and write crystalline marks in an amorphous material or to the melting temperature followed by fast cooling and quenching of the molten material to produce amorphous marks. Replay is achieved by applying a lower voltage across the storage layer and using the sense current from the scanning tip to detect, according to Ohm's law, changes in electrical conductivity between the crystalline and amorphous marks to recover data.

Raising the temperature through Joule heating to induce irreversible phase transformation has been demonstrated using scanning probes either in contact with or in close proximity to the phase-change layer. In the former, conductive atomic force microscope (AFM) tips or thin conductive wires were used to write crystalline marks in an initially amorphous material^{2–5} achieving crystalline dots less than 50 nm in diameter. The latter approach involved using scanning

tunneling probes to inject current into the phase-change material to induce phase transformation.^{3,6–9} Another technique for nanoscale recording on phase-change media uses microscopic heating filaments (heated by passing current through them) in contact with the phase-change layer to induce the phase transformation and record data.^{10,11} This paper is concerned with writing by electrical probe in contact with the storage medium.

The temperature distribution in the phase-change layer produced by Joule heating (which is a function of the geometry and electronic and thermal properties of the material structure) in addition to the kinetics and thermodynamic properties of the material determine the size and shape of the final recorded marks during writing. Thus any modeling or simulation of this writing process must take into account the electronic, thermal, and kinetic processes involved, which are complex. Dynamic numerical simulations of the writing process using the finite element technique were made incorporating all the above-mentioned processes,¹² and facilitated the determination of the ultimate sizes and shapes of the recorded marks.¹³

The purpose of this paper is to use analytical means to study the electrothermal writing process by electrical probes on phase-change media, to determine the conditions for writing crystalline and amorphous marks, and to predict their corresponding sizes as functions of the geometry of the system and material properties. These material properties are those applicable to the transition and melting temperatures. To keep the theory analytical, a simple two-layer structure consisting of a phase-change layer and an underlayer is modeled. This basic structure was used in writing experiments,^{3,4}

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FIG. 1. Geometry of tip and material structure used to model the recording system. The center of the coordinate system is in the middle of the tip with contact length L .

allowing direct comparison of the outcomes of this theory with the published measurements. Section II of this paper introduces the thermal and electrical models of the basic two-layer structure including the tip. These models are then used in Sec. III to derive an analytical expression for the temperature distribution in the phase-change layer during recording. The voltage requirements for irreversible amorphous-to-crystalline transition and for melting are determined in Sec. IV, using the derived temperature distributions, and compared with the reported measurements. In Sec. V, an approximate expression for the diameters of crystalline and amorphous marks is derived including an approximate model of cooling and solidification. A discussion is included in Sec. VI on the consequences of introducing a coating layer into the material structure on the voltage requirements and their dependence on phase-change layer thickness, the effect of the capacitance created by the tip/underlayer arrangement, and the role of the latent heat of fusion during cooling.

II. GEOMETRY OF RECORDING SYSTEM

Figure 1 illustrates the semi-infinite, two-dimensional geometry of the tip and material structure used to produce the thermal and electrical models in this paper. This arrangement makes use of the symmetry of the tip and material structure about the y axis. This system consists of a highly conductive tip making contact with a semi-infinite structure, with a contact length L . The thicknesses of the phase-change layer and underlayer are δ_p and δ_u , respectively. The thickness of the substrate is assumed to be much larger than the phase-change layer or underlayer. A positive voltage is normally applied to the tip with respect to the bottom electrode (underlayer) which is held at zero potential.

A. Thermal model

The temperature distribution in a material structure can be determined from solution of the heat conduction equation

$$\rho c_p \frac{\partial T}{\partial t} = k \nabla^2 T + P, \quad (1)$$

where ρ is the density of the material, c_p is the specific heat at constant pressure, k is the thermal conductivity of the material, and P is the power density of the heat source. For a two-layer structure, Eq. (1) can be solved subject to the thermal boundary conditions illustrated in Fig. 2 which provide

FIG. 2. Thermal model of medium. The top and bottom boundaries exhibit linear heat-flow conditions. The temperature is assumed continuous through the interface between the phase-change layer and underlayer.

for linear transfer of heat at the top and bottom surfaces and given by

$$-k_p \frac{\partial T_p}{\partial y} + H_t(T_p - T_o) = 0 \quad \text{at } y = 0,$$

$$k_u \frac{\partial T_u}{\partial y} + H_b(T_u - T_o) = 0 \quad \text{at } y = y_2,$$

where k_p and k_u are the thermal conductivities of the phase-change layer and underlayer, respectively, H_t and H_b are the thermal coefficients at the top and bottom surfaces, respectively, and T_o is the surrounding temperature (equal to 293 K here). The thermal coefficients are used to simulate different boundary conditions and substrate material in the thermal model. The temperature is taken to be continuous at the interface between the phase-change layer and underlayer thus satisfying the conditions

$$k_p \frac{\partial T_p}{\partial y} = k_u \frac{\partial T_u}{\partial y}, \quad T_p = T_u \quad \text{at } y = y_1.$$

To be able to find an analytical solution for the temperature distribution in (1) and to simplify the analyses, a number of assumptions are made. The first of these is that the phase-change layer is thin such that there is no temperature gradient in the y direction and hence only the thickness-averaged temperature is evaluated.¹⁴ Joule heating, due to current flow, is assumed to occur only in the phase-change layer thus defining the source of heat energy in the heat conduction equation. Further, it is taken that the thermal resistances between the various layers are negligible. Moreover, no lateral heat flow is assumed in the underlayer or in the substrate. Finally, the thermal conductivities of all the layers are taken to be constant and temperature independent.¹⁵ Using these assumptions and considering only the steady-state solution, the two-dimensional heat conduction equation in the phase-change layer in (1) becomes

$$\frac{\partial^2 T_p}{\partial x^2} + \frac{\partial^2 T_p}{\partial y^2} + \frac{P}{k_p} = 0, \quad (2)$$

where P is the power per unit volume generated in the phase-change layer due to Joule heating and is given by

$$P = \sigma_p |\mathbf{E}|^2, \quad (3)$$

where σ_p is the conductivity of the phase-change layer and $\mathbf{E}$ is the vector electric field developed in this layer. Integrating (2) through the thickness of the phase-change layer yields

$$\left. \frac{\partial T_p}{\partial y} \right|_{y=y_1} - \left. \frac{\partial T_p}{\partial y} \right|_{y=0} + \frac{\partial^2}{\partial x^2} \int_0^{y_1} T_p dy + \delta_p \frac{P}{k_p} = 0, \quad (4)$$

where $\delta_p = y_1$. Through solution of the steady-state, one-dimensional heat conduction equation for the underlayer (with no heat source) and application of the boundary and interface conditions described previously into (4) the heat equation in the phase-change layer becomes

$$\frac{\partial^2 \bar{T}_p}{\partial x^2} - G(\bar{T}_p - T_0) = \frac{-P}{k_p}, \quad (5)$$

where the average temperature is defined by $\bar{T}_p = \int_0^{y_1} T_p dy / \delta_p$ and the coefficient G is given by

$$G = \frac{1}{k_p \delta_p} \left[H_t + \frac{H_b}{1 + H_b(\delta_u/k_u)} \right]. \quad (6)$$

It can be seen from (5) and (6) that the effects of the thermal conductivities and thicknesses of the medium structure are all conveniently contained within the coefficient G in (6) which has dimensions of m^{-2} . The thermal model described in this section can easily be extended to more than two layers and to include thermal boundary resistances. This will not alter the form of Eq. (5) as only the coefficient G will have to be changed to include the thermal conductivities and thicknesses of the additional layers as will be discussed later.

B. Electrical model

The source of heat energy [Eq. (3)] in the heat conduction equation is Joule heating resulting from the passage of current through the finite conductivity phase-change layer. Thus the magnitude and distribution of the electric field in the phase-change layer will determine the maximum temperature developed and temperature distribution in the heated region. This will ultimately decide the size and shape of the recorded crystalline or amorphous marks. The electric field is produced by placing the material between a highly conductive tip and an electrode positioned underneath the phase-change layer, as shown in Fig. 1. In this work, the tip is modeled as a highly conductive semi-infinite rectangle biased at potential $\varphi = +V$, and the return electrode (or underlayer) is represented by an infinitely conducting sheet held at ground potential ($\varphi = 0$ V), as shown in Fig. 3. The tip and electrode are separated by a constant gap u .

In the absence of volume charges in the phase-change material, the electric field may be evaluated through solution of the time-independent current continuity equation $\nabla \cdot \mathbf{J} = 0$,

FIG. 3. Electrical model of the tip/electrode structure (symmetrical about $y=0$), with boundary conditions indicated. Also shown is the equivalent image model of the tip/electrode. Also shown are the continuous interface conditions required for evaluating the thickness-averaged temperature distribution in the phase-change layer using the determined y component of the tip/electrode electric field in (11).

where $\mathbf{J} = \sigma_p \mathbf{E}$ is the current density in the phase-change layer. When the phase-change layer is thin, current can be assumed to flow only in the y direction between the tip and the underlayer. It is further assumed that the electrical conductivity does not vary through the thickness of the phase-change layer. Expanding the dot product in the continuity equation and using the above assumptions lead to Laplace's equation $\nabla \cdot (\sigma \mathbf{E}) = \sigma \nabla \cdot \mathbf{E} = 0$ which may be solved to obtain the potential and hence electric fields developed in the phase-change layer.

With the underlayer being infinitely conductive, the tip/electrode arrangement in Fig. 1 may equivalently be represented by two equipotential, semi-infinite rectangles biased at equal but opposite voltages (images) and equidistant from the surface $\varphi = 0$ V, as shown in Fig. 3. Exact solutions to Laplace's equation for this geometry are available¹⁶ but, however, are in the form of infinite series. An approximation that yields closed-form expressions for the potentials and fields for this geometry will be derived next.

The geometry in Fig. 3 is divided into two regions: underneath the tip, where $|x| \leq L/2$, and beyond the tip corners, where $|x| > L/2$. The two-dimensional Laplace's equation for the potential φ in this case is

$$\frac{\partial^2 \varphi}{\partial x^2} + \frac{\partial^2 \varphi}{\partial y^2} = 0. \quad (7)$$

Inside the gap region ($|x| \leq L/2$), the potential varies only in the y direction, leading to the linear solution

$$\varphi = \frac{-V}{u} y + V, \quad |x| \leq L/2. \quad (8)$$

In the region $|x| > L/2$, the solution to (7) subject to a vanishing potential at $x \rightarrow \infty$ and a prescribed potential $\varphi(L/2, y)$ along boundary $x = L/2$ can be shown to be

FIG. 4. The y component of the electric field for the structure shown in Fig. 3 normalized by E_0 , where $E_0 = V/u$. The solid line is the field calculated using (10), and the dashed line is calculated using the first-order Padé approximation near the corner $x=L/2$ in (11). The plots are for $y=L/2$ and $u=2L$.

$$\varphi(x, y) = \frac{(|x| - L/2)}{\pi} \int_{-\infty}^{\infty} \frac{\varphi(L/2, y')}{(y' - y)^2 + (|x| - L/2)^2} dy', \quad (9)$$

$$|x| > L/2.$$

The potential at the boundary $x=L/2$ can be approximated by the linear potential in (8) over $0 < y < 2u$ and equal to $\pm V$ along the top and (imaginary) bottom surfaces of the tip, respectively. This provides a first-order approximation for the potential in the gap region and yields closed-form expressions for the electric fields that are not far from exact and more accurate solutions.¹⁷ Substituting this prescribed potential (8) at the boundary $x=L/2$ into (9) and integrating produce the potential everywhere beyond $x=L/2$. The corresponding y components of the electric fields in each region are evaluated from

$$E_y = \frac{-\partial\varphi}{\partial y} = \frac{V}{u}, \quad |x| \leq L/2, \quad (10a)$$

$$E_y = \frac{-\partial\varphi}{\partial y} = \frac{V}{\pi u} \left[\tan^{-1} \left(\frac{2u-y}{|x|-L/2} \right) + \tan^{-1} \left(\frac{y}{|x|-L/2} \right) \right], \quad (10b)$$

$$|x| > L/2.$$

Equation (10b) is further simplified to yield an analytical solution for the heat conduction equation, by applying a first-order Padé approximation near the tip corner ($x=L/2$), yielding

$$E_y = \begin{cases} E_0, & -L/2 \leq x \leq L/2 \\ E_0 \frac{d}{d+2u(|x|-L/2)}, & |x| > L/2, \end{cases} \quad (11)$$

where $d = \pi y(2u-y)$ and $E_0 = V/u$. Equation (11) shows that the y component of the electric field in this case varies as x^{-1} beyond the tip corner.¹⁸ Figure 4 shows a plot of the y component of the electric field using the approximation in (11) and compares it with the more accurate version in (10). It can be seen that there is a close agreement between the two par-

ticularly near the tip corner $x=L/2$ which is of most interest in this work.

III. TEMPERATURE PROFILE IN PHASE-CHANGE LAYER

The approximate electric field in (11) represents the source of heat energy in the material structure and is now used to determine the resulting temperature distribution in the phase-change layer. From this temperature distribution, the required voltages to heat the phase-change layer to the transition and melting temperatures can be predicted in addition to written mark sizes as functions of the geometry and thermal properties of the material structure.

Solution of the heat conduction equation in (5) requires dividing the phase-change layer into two regions according to the electric fields defined in (11) with temperatures $\bar{T}_{p1}$ and $\bar{T}_{p2}$, as shown in Fig. 3. The boundary conditions for this problem are that $\bar{T}_{p2}$ tends to the ambient temperature T_0 as $|x| \rightarrow \infty$, and that the temperature is continuous at the interface $|x|=L/2$. Subject to these symmetry and boundary conditions, the solution to Eq. (5) in the region directly underneath the tip is

$$\bar{T}_{p1} - T_0 = \frac{P_0}{k_p G} [A \cosh(\sqrt{G}x) + 1], \quad -L/2 \leq x \leq L/2, \quad (12)$$

where the constant A is given by

$$A = -e^{-\sqrt{G}L/2} \times \left\{ 1 - \frac{d\sqrt{G}}{2u} \left[1 + \frac{d\sqrt{G}}{2u} e^{d\sqrt{G}/2u} \text{Ei} \left(\frac{-d\sqrt{G}}{2u} \right) \right] \right\}. \quad (13)$$

In (12) $P_0 = \sigma_p E_0^2$ is the power density in the phase-change layer directly underneath the tip, and $\text{Ei}(\cdot)$ in (13) is the exponential integral. In the region beyond the tip corner, the solution to (5) can be shown to be

$$\bar{T}_{p2} - T_0 = \frac{P_0}{k_p G} [B e^{-\sqrt{G}x} + S(x)], \quad x > L/2, \quad (14)$$

with the particular solution

$$S(x) = \left(\frac{d\sqrt{G}}{2u} \right)^2 \left[\frac{e^\eta \text{Ei}(-\eta) + e^{-\eta} \text{Ei}(\eta)}{2} \right], \quad (15)$$

where $\eta = \sqrt{G}(x-L/2+d/2u)$. The constant B satisfying the boundary and continuity conditions in (14) is

$$B = \frac{1}{2} \left(\left(\frac{d\sqrt{G}}{2u} \right)^2 \left\{ e^{\sqrt{G}[(d/2u)-(L/2)]} \text{Ei} \left(\frac{-d\sqrt{G}}{2u} \right) - e^{-\sqrt{G}[(d/2u)-(L/2)]} \text{Ei} \left(\frac{d\sqrt{G}}{2u} \right) \right\} + e^{\sqrt{G}L/2} \left(1 + \frac{d\sqrt{G}}{2u} \right) - e^{-\sqrt{G}L/2} \left(1 - \frac{d\sqrt{G}}{2u} \right) \right). \quad (16)$$

Equations (12) and (14) are plotted in Fig. 5 for a 20 nm phase-change film on top of a 20 nm metal alloy underlayer with a thermal conductivity of 50 W/m K. The properties of

FIG. 5. Temperature profile in the phase-change layer for different bottom layer boundary conditions simulating different substrate material. For the solid line, the thermal characteristic length $1/\sqrt{G}=5$ nm, and for the dashed line, $1/\sqrt{G}=49$ nm. The calculation parameters are $\delta_p=20$ nm, $u=\delta_p$, $y=\delta_p/2$, $L=20$ nm, $k_p=0.24$ W/m K, $k_u=50$ W/m K, $\sigma_p=0.4$ Ω^{-1} m $^{-1}$, $H_t=0$ W/m 2 K, and the tip applied voltage is 10 V.

the phase-change layer are chosen for an amorphous $\text{Ge}_2\text{Sb}_2\text{Te}_5$ chalcogenide and are listed in Table I. In these calculations, the top surface layer is thermally insulated with $H_t=0$. A potential of 10 V is applied between the tip and underlayer, and the tip contact length L is 20 nm.

The solid line in Fig. 5 represents the case of controlled heat flow at the underlayer/substrate interface with thermal coefficient $H_b=2 \times 10^8$ W/m 2 K [typical value within the range of $(0.3-7) \times 10^8$ W/m 2 K of measured thermal boundary conductances between metals and dielectrics reported in the literature at room temperature^{22,24,25}]. Reducing heat flow across the underlayer/substrate interface, by reducing the thermal coefficient, increases the temperature in the phase-change layer and forces heat to flow laterally, leading to broadening of the temperature distribution around the tip re-

gion, as illustrated by the dashed line in Fig. 5. It is ultimately desirable to make the temperature distribution in the phase-change layer confined to the tip contact region and follow closely the localized electric field to limit the extent of the heated region and write small marks. Figure 5 caption indicates that this can be achieved by maximizing the factor G (or minimizing the thermal characteristic length $1/\sqrt{G}$) through modifying the thicknesses and thermal properties of the adjacent layers.

The peak temperature in the phase-change layer, from Fig. 5, occurs at the center of the tip ($x=0$). Using the approximation $1 + \zeta e^\zeta \text{Ei}(-\zeta) \approx 1/(1+\zeta)$ in (13), the peak temperature in the phase-change layer can be written using (12) as

$$\bar{T}_p^m \approx T_0 + \frac{P_0}{k_p G} \left[\frac{1 + (1 - e^{-\sqrt{G}L/2})d\sqrt{G}/2u}{1 + d\sqrt{G}/2u} \right], \quad (17)$$

which illustrates the dependence of the maximum temperature in the phase-change layer on the applied voltage through P_0 , and on the factor G that encompasses the thicknesses and thermal properties of the two-layer structure. Equation (17) can be used to determine the power level required to heat the phase-change layer to a desired temperature, and is used next to determine the voltages required to achieve crystallization and melting.

IV. WRITING VOLTAGES

One of the most important factors that contribute to the performance of a recording system is the writing condition. In electrical probe recording, this represents the voltage necessary to write stable and small crystalline and amorphous marks. Sections IV A and IV B are dedicated for the derivation of simple expressions for this voltage.

A. Crystallization threshold voltage

The flow of current in an initially amorphous phase-change layer increases the temperature in the vicinity of the tip contact area due to Joule heating. Provided that sufficient time has passed for the formation of stable nuclei in the amorphous material to initiate the crystal growth process, and that the temperature reaches or exceeds the transition temperature T_t , irreversible phase transformation into the crystalline state occurs. This results in the formation of crystalline marks after the heat source is removed and the material cools to room temperature. The amorphous-to-crystalline phase transformation is accompanied by a significant increase in electrical conductivity, with the electrical conductivity of the crystalline phase being almost 10 000 times greater than that of the amorphous phase for $\text{Ge}_2\text{Sb}_2\text{Te}_5$ (Ref. 23).

The equations for the temperature distribution developed so far can be utilized to determine the voltage level that is required to raise the temperature in phase-change layer to the transition temperature. This voltage, referred to in this work as the threshold voltage, represents the minimum voltage necessary to produce an irreversible amorphous-to-crystalline phase transition, and yields the smallest possible recorded crystalline mark. Taking the region directly under-

TABLE I. Thermal and electrical properties of $\text{Ge}_2\text{Sb}_2\text{Te}_5$ used in the calculations. Subscripts c , a , and t denote values at the crystalline phase, amorphous phase, and transition temperature, respectively. T_L is the liquid temperature, T_M is the melting temperature, T_t is the transition (or peak crystallization) temperature, ρ is the density, L_f is the latent heat of fusion, c_p is the specific heat, k is the thermal conductivity, and σ is the electrical conductivity.

Symbol	Value	Units
T_L	911 ^a	K
T_M	894 ^a	K
T_t (1.6 K/min)	428 ^a	K
ρ_c	6130 ^b	kg/m 3
ρ_a	5860 ^b	kg/m 3
L_f	14.3 ^a	kJ/kg
c_p	208 ^c	J/(kg K)
k_c (300 K)	0.53 ^d	W/(m K)
k_a (300 K)	0.24 ^d	W/(m K)
σ_c (300 K)	3250 ^e	(Ω m) $^{-1}$
σ_a (300 K)	0.4 ^e	(Ω m) $^{-1}$
σ_t (428 K)	310 ^e	(Ω m) $^{-1}$

^aReference 19.

^bReference 20.

^cReference 21.

^dReference 22.

^eReference 23.

FIG. 6. Comparison of measured (from Ref. 4) and calculated values of threshold voltages. The parameters for the calculations are $\delta_p = 20$ nm, $u = \delta_p$, $y = \delta_p/2$, $k_t = 0.4$ W/m K, $k_u = 50$ W/m K, $\sigma_t = 310 \Omega^{-1} \text{m}^{-1}$ (at $T_t = 428$ K), $H_t = 0$ W/m² K, and $H_b = 2 \times 10^8$ W/m² K. The dashed lines show the calculated threshold voltage for two different values of H_b within the reported range in the literature.

neath the tip where the tip-medium contact length is largest to reach the transition temperature first, then substituting $x = 0$ and $\bar{T}_{p1} = T_t$ in (12), and solving for the voltage, making use of the approximation leading to (17), yield the threshold voltage:

$$V_t \approx u \sqrt{\frac{k_t G (T_t - T_0)}{\sigma_t}} \cdot \frac{(1 + d\sqrt{G/2u})}{[1 + (1 - e^{-\sqrt{G}L/2})d\sqrt{G/2u}]}, \quad (18)$$

where k_t is the thermal conductivity of the phase-change layer at the transition temperature. When the coefficient G is large, the threshold voltage reduces to the simple form

$$V_t \approx u \sqrt{\frac{k_t G (T_t - T_0)}{\sigma_t}}. \quad (19)$$

When G is large, moreover, the heat conduction equation in (5), for constant electrical conductivity, may well be approximated by

$$\bar{T}_p - T_0 \approx \frac{P}{k_p G}. \quad (20)$$

Hence when G is very large, corresponding to the case where most of the heat generated in the phase-change layer is dissipated vertically through the adjacent layers, then the temperature profile in the phase-change layer follows the applied power-density distribution. At the transition temperature T_t , the power density P_t can be written in terms of the threshold voltage V_t as $P_t = \sigma_t V_t^2 / u^2$ which, upon substitution in (20) and solving for V_t , yields Eq. (19) as expected.

The calculated threshold voltage from (19) is now compared, in Fig. 6, with published recording measurements on a simple structure consisting of a thin film of amorphous $\text{Ge}_2\text{Sb}_2\text{Te}_5$ with various thicknesses between 1 and 900 nm on top of 20 nm Au–Ni underlayer, and finally deposited on a 1 mm oxide-glass substrate.⁴ Writing was performed by applying, through a series resistor, a constant voltage between either a Au–Ni-coated Si_3N_4 tip or a sharpened Au wire in contact with the phase-change material and the Au–Ni underlayer. The surface of the phase-change layer in these experiments was covered with an inert liquid having

very small electrical and thermal conductivities (compared to the amorphous material) to prevent oxidation. In the calculated curves in Fig. 6, the thermal conductivity of the phase-change layer at the transition temperature was taken to be 0.4 W/m K (lying between the amorphous and crystalline thermal conductivities^{26,27} in Table I). The thermal conductivity of the gold alloy underlayer was taken to be 50 W/m K, and a thermal coefficient value of 2×10^8 W/m² K was assumed at the Au–Ni/glass interface (solid line).

In spite of using approximate models and estimated material values, it can be seen from Fig. 6 that there is a good agreement between the theoretical and measured threshold voltage values. This is also true for the range of measured values of T_t for $\text{Ge}_2\text{Sb}_2\text{Te}_5$ reported in the literature at different heating rates^{19,23} (413–446 K at 0.5–80 K/min), giving a maximum variation of 6% in the threshold voltage. Even using much higher transition temperatures that may be applicable to very high heating rates, such as 700 K reported from pulsed laser heating investigations on phase-change media,^{21,28} the predicted threshold voltage still remains within the boundaries of the measurements presented in Fig. 6. Varying H_b within the range of published measured values also keeps the agreement of the predicted voltages within the boundaries of the measurements, as indicated by the dashed lines in Fig. 6. Hence it is reasonable to use the value of 2×10^8 W/m² K for H_b in the remainder of this paper.

To explore more closely the dependence of the threshold voltage on the phase-change layer thickness for this particular structure, the coefficient G in (6) is substituted in (19), noting that $u = \delta_p$, to yield

$$V_t = \sqrt{\delta_p} \sqrt{\frac{H_b (T_t - T_0)}{\sigma_t [1 + H_b (\delta_u / k_u)]}}. \quad (21)$$

This clearly shows that the threshold voltage is proportional to the square root of the phase-change layer thickness, as observed experimentally in Ref. 4.

B. Amorphization melting voltage

Creating an amorphous mark in an initially crystalline material involves melting the phase-change material followed by fast cooling and quenching of the molten material. The previous analyses used for crystallization can be applied here to determine the voltage that is required to be applied between the tip and underlayer to raise the temperature in the phase-change layer to the melting point. It is assumed here that the heat of melting is negligible compared to the heat required to raise the temperature of the phase-change layer to the melting temperature. This is reasonable since the liquids and melting temperatures for $\text{Ge}_2\text{Sb}_2\text{Te}_5$ are close, as seen in Table I, and such an assumption simplifies the solidification analyses presented later. Hence, this voltage is determined by replacing the transition temperature in (19) with the melting temperature T_M and using the electrical and thermal conductivities of the crystalline phase. The results are shown in Fig. 10(b), where it can be seen that for the basic two-layer structure the calculated melting voltage is less than the crystallization threshold voltage [shown in Fig. 10(a)]. This is since

although the melting temperature is significantly higher than the crystallization temperature, this is more than offset by a lower heat generation rate in the crystalline phase due to its higher electrical conductivity.

V. DOT DIAMETERS

Evaluating theoretically the size of recorded marks is important for producing estimates of the achievable linear and areal densities in electrical probe recording, and for identifying the factors that determine the mark size.

A. Crystalline marks

The location of the transition temperature isotherm (along the x direction at which $\bar{T}_p = T_t$) can be used to estimate the diameter of recorded marks. For voltages that are greater than or equal to the threshold value, the location of the transition temperature isotherm in the region $|x| \geq L/2$, where $\bar{T}_p = T_t$, written here as x_0 , may be found by solving (14) for x numerically. An approximate analytical solution can be obtained by substituting the electric-field distribution in the region $|x| \geq L/2$ defined in (11) into (20) and solving for x , yielding

$$x_0 = \frac{L}{2} + \frac{d}{2u} \left\{ \sqrt{\frac{P}{k_t G(T_t - T_0)}} - 1 \right\}. \quad (22)$$

Equation (22) shows that the size of the crystalline mark scales with the tip contact length, and depends on the power supplied, the thicknesses, and thermal properties of the phase-change and adjacent layers. The properties of the phase-change material enter implicitly through the transition temperature T_t , which is a function of the heating rate, the activation energy of crystallization, and the kinetics of phase transformation.^{15,29}

At the threshold voltage, where $P_t = k_t G(T_t - T_0)$, the position of the transition temperature isotherm is at the corner of the tip ($x_0 = L/2$), thus defining the minimum size of a recorded crystalline mark. For values of the threshold voltage $F = V/V_t \geq 1$, the power density becomes $P = F^2 k_t G(T_t - T_0)$ and substitution into (22) yields the simplified form

$$x_0 = \frac{L}{2} + \frac{d}{2u} (F - 1), \quad F \geq 1. \quad (23)$$

The diameter (assuming circular marks) of the crystalline region is simply $2x_0$ and is shown in Fig. 7 as a function of phase-change layer thickness for two applied writing voltages corresponding to 1.2 and 2 times the threshold value. The tip contact lengths in these calculations were chosen to be $L = 10$ and 20 nm. It is not possible to compare directly the outcome of Eq. (23) with the experimental dot diameters published in Ref. 4 since the authors applied the writing voltages through series resistors, so the actual potential between the tip and underlayer would vary with the change in electrical conductivity of the phase-change layer during crystallization and is not known. However, as shown in Fig. 7, even with these unknowns, the theoretical results follow the experimental trend of increasing dot size with phase-change layer thickness. Also shown in Fig. 7 is the calculated power

FIG. 7. Calculated diameters of recorded crystalline marks compared with measurements in Ref. 4. The applied voltage is 1.2 and 2 times the threshold value for each respective layer thickness and for two tip contact lengths $L = 10$ and 20 nm. Also shown is the calculated supplied power for $L = 20$ nm and $F = 1.2$ times threshold voltage assuming a cylindrical depth profile for the crystalline dot. The parameters for the calculations are $\delta_p = 20$ nm, $u = \delta_p/2$, $L = 10$ and 20 nm, $k_p = 0.24$ W/m K, $k_u = 50$ W/m K, $\sigma_t = 310 \Omega^{-1} m^{-1}$ (at $T_t = 428$ K), $H_t = 0$ W/m² K, and $H_b = 2 \times 10^8$ W/m² K.

(dash-dot line) supplied to the phase-change layer calculated from

$$P_s = F^2 k_t G(T_t - T_0) v_p, \quad (24)$$

using $F = 1.2$, 20 nm contact length, and assuming a cylindrical depth profile for the crystalline dot with volume $v_p = \pi x_0^2 \delta_p$.

B. Amorphous marks

Recording an amorphous mark in crystalline material involves heating the phase-change layer to the melting temperature followed by fast cooling, with rates of the order of a few tens of K/ns (Refs. 21 and 30), and quenching processes that ultimately define the final size or diameter of the amorphous mark.

When heating to the melting temperature, the “initial” size or diameter of the recorded (melted) mark may be evaluated from the melting temperature isotherm³⁰ using Eq. (22) by replacing the transition temperature with the melting temperature, and using the electrical and thermal conductivities of the crystalline starting phase. The “final” size of the recorded amorphous mark is then determined by cooling of the molten material, in particular, whether or not the cooling rate at a given location exceeds the critical rate required to prevent recrystallization. Hence an estimation of the cooling rate would give insight into the regions of the melted phase-change layer that would amorphize upon cooling and help determine the final size of the amorphous mark.

To find the cooling rate at a given location along the phase-change layer, the heat conduction equation in (5) is modified to include solidification and movement of the solidification front upon cooling, as demonstrated in Fig. 8. The time-dependent heat equation in the phase-change layer after removing the heat source now becomes

FIG. 8. Geometry of phase-change layer used to determine the final size of an amorphous mark written in a crystalline material during cooling and quenching.

$$\frac{\rho c_p}{k_p} \frac{\partial \bar{T}_p}{\partial t} = \frac{\partial^2 \bar{T}_p}{\partial x^2} - G(\bar{T}_p - T_0). \quad (25)$$

In this solidification problem, the liquid is initially at the melting temperature T_M and contained in the region $x < x_0$. The liquid then solidifies as a result of cooling through the adjacent vertical layers, and laterally through the boundary $x = x_0$ where $\bar{T}_p = T_0$ (liquid is assumed to extend to infinity to the left in Fig. 8). As latent heat of fusion is removed from the liquid by cooling, it changes to the solid phase and the solid/liquid interface or solidification front moves in the negative x direction, as indicated in Fig. 8. The boundary conditions at the moving interface, assuming that there is no temperature gradient in the liquid phase with constant temperature T_M , are given by³¹

$$\bar{T}_p = T_M, \quad k_p \frac{\partial \bar{T}_p}{\partial x} = -\rho L_f \frac{df}{dt} \quad \text{at } x = x_0 - f(t),$$

where $f(t)$ is the displacement of the solidification front from the initial location x_0 , L_f is the latent heat of fusion, and ρ is the density of the solid region. The initial condition is that $f(t) = 0$ at $t = 0$, where t is the time from cooling and quenching.

The heat conduction equation in this case subject to the boundary and initial conditions cannot be solved analytically, and therefore the heat-balance integral method³² is used in the Appendix to yield the approximate linear temperature distribution in the region $f(t) < x < x_0$ given by

$$\bar{T}_p = \begin{cases} T_M, & x < x_0 - f(t) \\ T_0 + (T_M - T_0) \frac{(x_0 - x)}{f(t)}, & x_0 - f(t) < x < x_0 \\ T_0, & x > x_0 \end{cases}. \quad (26)$$

The displacement of the solidification front is

$$f(t) = \sqrt{\frac{2}{G} \left[\exp\left(\frac{\alpha t}{\beta}\right) - 1 \right]}, \quad (27)$$

where $\alpha = 2k_p G(T_M - T_0)/(\rho c_p)$ and $\beta = (2L_f/c_p) + (T_M - T_0)$. The cooling rate can now be determined by differentiating (26) to give

FIG. 9. Plots of the (a) temperature and (b) cooling rate upon cooling and quenching at two locations in a 20 nm phase-change layer. The initial location x_0 was determined by the melting temperature isotherm for an applied voltage that is 1.2 times the melting voltage ($=0.84$ V). The tip contact length is 20 nm, and the underlayer thickness is 20 nm. The electrical and thermal parameters of the crystalline phase were used in the calculations.

$$\frac{\partial \bar{T}_p}{\partial t} = \begin{cases} 0, & x < x_0 - f(t) \\ -(T_M - T_0) \frac{(x_0 - x)}{f(t)^2} \frac{df(t)}{dt}, & x_0 - f(t) < x < x_0 \\ 0, & x > x_0 \end{cases}, \quad (28)$$

where the velocity of the solidification front is

$$\frac{df(t)}{dt} = \frac{1}{f(t)} \frac{\alpha}{\beta G} \exp\left(\frac{\alpha t}{\beta}\right). \quad (29)$$

Figure 9 illustrates the calculated temperature and the corresponding cooling rate, as functions of time, after removing the heat source at two different locations in a 20-nm-thick phase-change layer. In these calculations, the initial size of the recorded mark x_0 , when cooling starts, was determined from the location of the melting temperature isotherm and was found to be 14.7 nm for an applied voltage that is 1.2 times the melting voltage (equal to 0.84 V). It can be seen from this figure that the predicted cooling rates over the length of the cooled region are in excess of the reported values required for amorphization. This indicates that the entire melted region, for this particular structure, will cool and quench to the amorphous phase and hence the size of the final amorphous mark can be determined approximately from the location of the melting temperature isotherm using (22). The dot diameters in this case will follow Fig. 7 for voltages that are in excess of the melting voltage.

VI. DISCUSSION AND CONCLUSIONS

An analytical, electrothermal model of probe recording on phase-change media has been produced. This model was used to predict the voltage levels that are required to reach the transition temperature for crystallization and the melting temperature for amorphization. These voltages were then used to derive analytical expressions for the expected sizes of crystalline and amorphous marks. This theory used a basic two-layer structure consisting of a phase-change layer along with a high electrical conductivity underlayer deposited on a substrate. The effect of the substrate material was taken into account through conductive boundary conditions at the underlayer/substrate interface. For this arrangement, the threshold voltage required to induce irreversible amorphous-to-crystalline transformation was found to depend on the square root of the phase-change layer thickness, as observed experimentally.⁴

In reality, the phase-change layer is usually coated with a thin protective layer to prevent oxidation and for tribology purposes. Moreover, further underlayers may be included for thermal optimization and for current limiting.⁵ In these cases, the dependence of the threshold voltage on the thickness of the phase-change layer may be different from the one predicted for the simple two-layer structure. The simple case of introducing a coating layer to the basic two-layer structure can be investigated here by solving Laplace's equation for the potential in the region directly underneath the tip, $-L/2 \leq x \leq L/2$. The corresponding electric field in the phase-change layer, subject to the boundary conditions shown in Fig. 3 and continuity of currents and voltages across the interfaces, can then be shown to be

$$E_y = \frac{V}{(\sigma_p/\sigma_{\text{coat}})\delta_{\text{coat}} + \delta_p},$$

where V is the applied voltage and σ_{coat} and δ_{coat} are the electrical conductivity and thickness of the coating layer, respectively. Substituting this electric field into (20) and solving for the voltage at the transition temperature yield the modified crystallization threshold voltage as

$$V_t = \left(\frac{\sigma_t}{\sigma_{\text{coat}}} \delta_{\text{coat}} + \delta_p \right) \sqrt{\frac{k_t G (T_t - T_0)}{\sigma_t}}. \quad (30)$$

Following the same analyses presented in Sec. II A, the factor G in (30) now becomes

$$G = \frac{1}{k_p \delta_p} \left[\frac{H_t}{1 + H_t(\delta_{\text{coat}}/k_{\text{coat}})} + \frac{H_b}{1 + H_b(\delta_u/k_u)} \right],$$

where k_{coat} is the thermal conductivity of the coating layer. From (30) it can be seen that the dependence of the threshold voltage on the phase-change layer thickness no longer follows a square root. Considering a 2 nm coating layer, for example, with an electrical conductivity of $50 \Omega^{-1} \text{ m}^{-1}$ (Ref. 12) (a value that is a compromise between the electrical conductivities of the amorphous and crystalline phases⁵), the threshold voltage from (30) as a function of phase-change layer thickness is plotted in Fig. 10(a) and compared with the previous calculations without a coating layer. In both calculations, the top boundary of the structure is assumed ther-

FIG. 10. Plots of (a) the threshold voltage for an initially amorphous material, and (b) the melting voltage for an initially crystalline material with and without a coating layer on top of the phase-change layer. The coating layer is 2 nm thick with electrical and thermal conductivities of $50 \Omega^{-1} \text{ m}^{-1}$ and $100 \text{ W/m} \cdot \text{K}^{-1}$. The top of the coating layer is thermally insulated with $H_t=0$.

mally insulated with $H_t \rightarrow 0$, making the value of the factor G the same for both cases. It can be seen from the figure that the threshold voltage no longer follows the square-root dependence on the thickness of the phase-change layer, is large at small thicknesses, has a minimum value at a specific thickness (13 nm), and tends to the limit of no coating when the thickness of the phase-change layer is much larger than the coating layer.

For an initially crystalline material, on the other hand, the voltage required for melting in the presence of a coating layer can also be computed using (30) by substituting the melting temperature and electrical and thermal conductivities of the crystalline phase in the equation. This melting voltage is plotted in Fig. 10(b) for the same coating layer parameters used in Fig. 10(a), where it can be seen that larger voltages are required to melt thin crystalline films compared to the threshold voltage, and that the minimum occurs at a larger phase-change layer thickness (130 nm).

Introducing coating and additional layers with different electrical and thermal properties and with different thicknesses is also expected to change the dependence of the mark size on the thickness of the phase-change layer from that shown in Fig. 7. This is due to the modification of the electric fields and hence temperature distributions throughout the multilayer structure. This will not be treated here. Note also that if there is appreciable heat flow through the tip during writing, Eqs. (12) and (14) can still be solved, with the same boundary conditions, using different thermal coefficients di-

FIG. 11. Power-density contours in 20-nm-thick amorphous phase-change layer, computed using the y component of the electric field in (10) for a 20 nm tip contact length and 10 V applied voltage.

rectly beneath the tip and beyond the tip corners (this was not done here since comparisons are made with experiments⁴ where a thermal insulating layer was placed on the top surface of the sample). It can be shown that this causes the temperature maxima to be at the tip corners and a reduced temperature beneath the tip. This in turn leads to an increased voltage requirement for crystallization and amorphization.

The analytical models developed in this work only considered the temperature distribution averaged over the thickness of the phase-change layer for simplicity, therefore not providing detail on the depth profile of the resulting recorded crystalline and amorphous marks. Nevertheless, the derived models of the electric fields can give insight into the distribution of the power density or heat source that determines the profile of recorded dots through the thickness of the phase-change layer. For an amorphous material in the basic two-layer structure (or including a coating layer whose electrical conductivity is larger than the amorphous material but less than the crystalline conductivity), the power-density contours through the phase-change layer can approximately be computed using the y component of the electric field in Eq. (10) and are shown in Fig. 11. The power density in this figure broadens with increased depth into the phase change layer beyond the tip corner. Therefore the temperature distribution and hence the recorded crystalline bit are expected to follow this profile with an increase of the size of the crystalline material through the thickness of the phase-change layer in agreement with the more detailed finite element simulations in Ref. 12. In the high electrical conductivity crystalline phase, on the other hand, the electric fields are small in the phase-change layer and a large proportion of the applied voltage drops across the small conductivity (compared to the crystalline conductivity) coating layer. The power density or heat source in this case is not shown here but would be expected to be confined in the coating layer near the tip contact region leading to a temperature distribution and hence bit shape that is located at the coating/phase-change layer interface and not distributed through the whole thick-

ness of the phase-change layer. The finite element simulations in Ref. 12 have shown hemispherical amorphous bit shapes in a crystalline material that are confined to the upper region of the phase-change layer interface during amorphization, confirming the above predictions.

The above discussion highlights the importance of the choice of the thickness and electrical conductivity of the coating layer for not only determining the voltage levels required for crystallization and amorphization, but also the shape of the recorded bit. This will in turn determine how close bits can be written next to each other (before they start to overlap), the shape of the replay signals from such bits,¹² and the storage densities that can be achieved.

Another important factor to consider in this storage technique is the writing speed. The placement of the phase-change layer between the tip and underlayer produces a finite capacitance that, along with the equivalent parallel resistance, contribute to the overall time constant of the system and therefore the writing speed. This capacitance also influences the magnitude and the rate of current flowing in the phase-change layer and tip during the application and removal of writing voltages. Estimating theoretically this capacitance together with the parallel equivalent resistance of the phase-change layer enables the evaluation of the transient behavior of a system including any additional external components. This capacitance can be determined from the charge accumulated per applied voltage according to $C_p = \int_v \nabla \cdot \mathbf{D} dv / \int_y \mathbf{E} dy$, where $\mathbf{D} = \epsilon \mathbf{E}$ is the electric-field density and ϵ is the dielectric constant of the phase-change layer. Using the y component of the electric field in (10) and integrating over the volume of the phase-change layer assuming a rectangular tip with a width W in the z direction and constant ϵ give the capacitance as

$$C_p = \frac{\epsilon L W}{u} \left[1 + \frac{2 \ln(4) u}{\pi L} \right]. \quad (31)$$

Equation (31) gives a close agreement with the more accurate approximation of Eq. (3) in Ref. 33 for the same geometry, particularly for small separations between the tip and bottom electrodes where the vertical component of the electric field dominates. Additional accuracy can be obtained by including the x component of the electric field in the evaluation of the capacitance. The corresponding parallel equivalent resistance of the phase-change layer then follows from Ohm's law and is $R_p = \epsilon / \sigma_p C_p$. For a 20-nm-thick amorphous material with a dielectric constant of 17 (Refs. 34 and 35) in the basic two-layer structure using a tip of a square cross section having $L = W = 20$ nm, the calculated capacitance using (31) is 5.7×10^{-18} F and the resistance is 66.4 M Ω . In the crystalline phase with a dielectric constant of 37 (Ref. 34) and the same tip cross section, the calculated capacitance is increased to 12.3×10^{-18} F, while the resistance is reduced to 8.2 k Ω . The overall time constant in a practical storage system following the application and removal of the writing voltages will be influenced by this intrinsic capacitance and parallel resistance of the phase-change layer, and will also of course depend on the rise and fall times of the voltage source and the values of external series resistors and stray capacitances in the network.

FIG. 12. The velocity of the solidification front as a function of time for different values of the latent heat of fusion. Time $t=0$ represents the start of the cooling time. The calculation parameters are the same as those for Fig. 9. The cooling rate was evaluated at $x=5$ nm.

An approximate model of cooling and solidification was developed to determine the final size of amorphous marks when the temperature in the phase-change layer falls below the melting point with cooling rates of few tens of K/ns. This model showed, for the simple two-layer structure, that the cooling rate over the melted region was in excess of the values required for amorphization, indicating that the whole melted region (determined by the melting temperature isotherm) will cool and quench to the amorphous phase. Cooling in this theory was assumed to occur through only one-half of the geometry shown in Fig. 8, while in reality the melted region would cool and quench through two solidification fronts on both halves of the geometry. Hence the calculated cooling rates from this theory represent the slowest cooling case. Even with this slow cooling condition, appropriate cooling rates were achieved to amorphize the whole melted region. This theory can also be used to investigate the effect of the latent heat of fusion on the velocity of the solidification front and on the cooling rate which ultimately determines the final size of the quenched mark. Figure 12 illustrates the velocity of the solidification front calculated using Eq. (29) for the two-layer structure (with or without coating) and the thermal parameters of the crystalline phase in Table I, showing that increasing the latent heat reduces the velocity of the solidification front. The cooling rate, calculated using (28), on the other hand, increases with increasing latent heat, as illustrated also in Fig. 12.

APPENDIX

In this appendix, the heat-balance integral method is used to find an approximation for the temperature distribution in the phase-change layer during cooling and quenching upon amorphization. Detailed description of this method and a review can be found in Ref. 31. Starting the solution by writing $R=T-T_0$, the heat conduction equation in the phase-change layer in (25) becomes

$$\frac{\rho c_p}{k_p} \frac{\partial R}{\partial t} = \frac{\partial^2 R}{\partial x^2} - GR$$

and the boundary conditions are transformed into

- (i) $R=0$ at $x=x_0$,
- (ii) $R=T_M-T_0$ at $x=x_0-f(t)$,
- (iii) $k_p \frac{\partial R}{\partial x} = -\rho L_f \frac{df}{dt}$ at $x=x_0-f(t)$.

The initial condition is that $f(t)=0$ at $t=0$.

Integrating the heat conduction equation over the thermal layer thickness $f(t)$ and applying boundary condition (iii) produce the heat-balance equation for this problem as

$$\left. \frac{\partial R}{\partial x} \right|_{x_0} + \frac{\rho L_f}{k_p} \frac{df(t)}{dt} = G\theta + \frac{\rho c_p}{k_p} \frac{d}{dt} [\theta - (T_M - T_0)f(t)], \quad (\text{A1})$$

where $\theta = \int_{x_0-f}^{x_0} R dx$.

The next stage of derivation involves approximating the temperature profile within the thermal layer using a mathematical function. Second- and higher-order polynomials are quite often used but would lead here to a nonlinear differential equation for $f(t)$ when substituted in the heat-balance equation (A1). Hence a first-order approximation is used which is defined as $R=a+bx$, where a and b are constants that are to be determined subject to the boundary conditions of this problem. Applying boundary conditions (i) and (ii) to the linear approximation yields

$$R = (T_M - T_0) \frac{(x_0 - x)}{f(t)}. \quad (\text{A2})$$

Substituting this temperature profile in the heat-balance equation (A1) produces a first-order differential equation for $f(t)$ whose solution (satisfying the initial condition) was found to be

$$f(t) = \sqrt{\frac{2}{G} \left[\exp\left(\frac{\alpha t}{\beta}\right) - 1 \right]}, \quad (\text{A3})$$

where $\alpha = 2k_p G(T_M - T_0) / (\rho c_p)$ and $\beta = 2L_f / c_p + (T_M - T_0)$. Substituting (A3) in (A2) gives an approximation for the temperature profile within the thermal layer upon solidification in the phase-change layer.

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