

Phase-change materials: Empowered by an unconventional bonding mechanism

J. Pries, O. Cojocaru-Mirédin, and M. Wuttig

Phase-change materials (PCMs) have demonstrated a wide range of potential applications ranging from electronic memories to photonic devices. These applications are enabled by the unconventional portfolio of properties that characterizes crystalline PCMs. Here, we address the origin of these unusual properties and how they are related to the application potential of these materials. Evidence will be presented that the properties are related to an unconventional bonding mechanism. Employing a novel map, which separates solids according to the number of electrons transferred and shared between adjacent atoms, it is shown that PCMs occupy a well-defined region. Depicting physical properties such as the optical dielectric constant as the third dimension in the map reveals systematic property trends. Such trends can be utilized to unravel the origins of the unconventional materials properties or alternatively, as a means to optimize them.

Introduction

Phase-change materials (PCMs) have attracted significant interest in the last few years. There are two major reasons for this attention. PCMs have a wide range of present and potential applications, ranging from rewriteable optical data storage to novel electronic memories and switchable photonic devices.[1–3] For these applications, it is crucial to understand the source of the unconventional properties of PCMs,[4,5] and how the properties can be tailored. Yet, PCMs are also stimulating from the perspective of fundamental sciences. What are the origins of the unconventional properties that characterize these remarkable solids? In this article, we attempt to address both perspectives.

We first summarize the properties portfolio that is desirable for different applications of PCMs, in particular, where improvements of materials properties promise significant gains in the application arena. Subsequently, we briefly address how material properties can be explained utilizing fundamental concepts of the relationship between bonding, structure, and properties. The emerging insights are then employed to address an aspect of significant application interest—the origin of the high switching speeds.[6,7] Finally, the potential of the approach presented is critically evaluated.

PCMs and switching speeds

PCMs are presently employed for rewritable optical and electronic data storage. Both means of storage utilize the fact that PCMs can exist at room temperature in both a metastable amorphous phase and a (more stable) crystalline state.[8] The coexistence of two such states can be realized in many materials, including Si and SiO_2 (glass and crystal). What makes PCMs stand out are the potentially high switching speeds from the amorphous to the crystalline state at elevated temperatures, which can be as short as 1 ns, or less,[9–11] while the amorphous phase can be stable for more than 10 years at room temperature. The switching speed exhibits an extraordinary temperature dependence in these materials, with an increase in temperature of about 300 K leading to an increase in switching speed by 16–17 orders of magnitude.

The second highly remarkable feature of this material class is the pronounced difference in properties between the amorphous and the crystalline states. While the optical properties of SiO_2 such as the optical reflectivity hardly differ in the amorphous (glassy) and crystalline states. This is very different for PCMs, where the amorphous and crystalline phase show a pronounced difference in refractive index and optical polarizability ε_∞, leading to applications such as their use in Blu-ray discs.[2]

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The requirements listed in **Table I** for PCMs in data-storage applications are closely interwoven with material properties, such as the atomic arrangement and its stability, the speed of crystallization and atomic relaxation processes, as well as the consequences of differences in atomic arrangement between the amorphous and crystalline states on switching speed and the contrast in their properties.

A number of approaches have been conducted over the last decade to explain the unusual properties of PCMs. One line of inquiry has focused on the atomic arrangement in the amorphous phase. The rationale behind this approach is that both the switching speed and the property contrast can potentially be attributed to differences in the atomic arrangements in the two phases. Indeed, these studies have provided interesting insights, addressing for example, the role of Ge in the structure of amorphous $Ge_2Sb_2Te_5$[12] and the importance of the distortion away from a perfectly octahedral atomic arrangement.[13] Yet, it has been difficult to derive insights into how novel PCMs could be found or how property trends can be predicted. Which prompted the question: is there an alternative approach?

Table I. Requirements for phase-change materials in data-storage applications.

High signal-to-noise ratio	• Large amorphous-crystalline property contrast
	• Small property variation with time
High switching speeds	• Very fast atomic rearrangement at elevated temperature
	• Small reduced glass-transition temperature T_g/T_m
Good stability of amorphous phase	• No major rearrangement at room temperature
	• High T_g
Multilevel memory	• Stable atomic arrangements at room temperature
Low power consumptions	• Low melting temperature
	• Low thermal conductivity
	• High switching speed
	• High fragility m, low T_g

Unconventional properties of PCMs

If the chalcogenides used as PCMs such as GeTe or $Ge_2Sb_2Te_5$ have so many interesting applications, this might be attributed to the fact that they possess a combination of unconventional properties.[5] This is indeed the case, as summarized in **Table II**, which compares the characteristic property portfolio of materials employing ionic, metallic, or covalent bonding and relates these properties to the property portfolio characteristics for chalcogenides such as GeTe, $Ge_2Sb_2Te_5$, PbTe, or Bi_2Se_3. This comparison reveals that these chalcogenides show rather unconventional properties. They have electrical conductivities between those of metals and covalently bonded solids. In addition, crystalline PCMs such as SnTe, GeTe, or $Ge_2Sb_2Te_5$ are characterized by high values of the optical dielectric constant ε_∞.[5] Their effective coordination number (ECoN) is incompatible with the 8-N rule, where N is the number of outer shell electrons or group number, which is usually obeyed for covalently bonded materials (e.g., Group IV element Si features four nearest neighbors). Hence, in SnTe, the Sn and Te atoms should on average have three nearest neighbors. Instead, the effective coordination number is closer to six, signaling a significant deviation from ordinary covalent bonding. Furthermore, these crystalline phases possess high values of the Born effective charge Z*, a measure of the chemical bond polarizability.

Finally, these materials are characterized by high values of the Grüneisen parameter for the transverse optical modes, γ_{TO}; a sign of anharmonic behavior, which describes how much phonon frequencies change with changing volume. Thus, a linear combination of properties typical for other bonding mechanisms (metallic and covalent) is unable to describe the

Table II. Property-based "fingerprints" to define bonding in inorganic materials.

	Ionic (e.g., NaCl, MgO)	Covalent (e.g., Si, GaAs)	Metavalent (incipient metals, e.g., GeTe, PbTe)	Metallic (e.g., Cu, NiAl)
Electronic conductivity (**electrical identifier**)	Very low ($<10^{-8}$ S cm^{-1})	Low to moderate (10^{-8}–10^{2} S cm^{-1})	Moderate (10^{2}–10^{4} S cm^{-1})	High ($>10^{5}$ S cm^{-1})
Effective Coordination number[a] (**structural identifier**)	4(ZnS), 6(NaCl), 8(CsCl)	8-N rule Typically satisfied	8-N rule not satisfied	8 (bcc), 12 (hcp/fcc)
Optical dielectric constant ε_∞ (**optical identifier**)	Low (≈2–3)	Moderate (≈5–15)	High (>15)	N/A[b]
Born effective charges Z* (**chemical bond polarizability**)	Low (1–2)	Moderate (2–3)	High (4–6)	Vanishes (0)
Mode specific Grüneisen parameters (**anharmonicity**)	Moderate (2–3)	Low (0–2)	High (>3)	Low (0–2)

The fingerprint for metavalent solids is a combination of five different identifiers, all of which need to be present in a given material (e.g., NaCl and PbTe have the same structural identifier, but the electronic conductivity in NaCl is extremely low).[5]

[a] For ionic and metallic systems, representative structure types are given, but there are many others especially for multinary systems (e.g., in Zintl phases).

[b] This indicator is not normally applicable to the metallic state.

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properties for PCMs such as GeTe, PbTe, or others. This can be seen, for example, by a comparison of the typical values of the Grüneisen parameter, γ_{TO}. It is high for materials such as GeTe or PbTe, while metallic and covalently bonded materials both generically show low values of γ_{TO}. Therefore, bonding in GeTe cannot be considered a bonding just at the boundary between covalent and metallic bonding. Instead, the bonding in GeTe or PbTe should be considered as a unique and novel bonding mechanism, distinctively different from covalent and metallic bonding. Nevertheless, this bonding mechanism seems to be related to both metallic and covalent bonding and has been coined metavalent; the materials that utilize this bonding mechanism have been named incipient metals.[5]

Bonding and material properties

It might seem bold to link a unique property portfolio to a unique bonding mechanism, but chemical bonding is at its best if it is employed to explain material properties. Nevertheless, it would be highly desirable to obtain further support and confirmation for the notion that there is a novel bonding mechanism in solids besides ionic, metallic, and covalent bonding, yet different from the weaker hydrogen and van der Waals bonding.

Indeed, such support was obtained by studies of various solids employing atom probe tomography (APT), which has been recently used to analyze PCMs.[14] These studies have helped not only to detect the exact composition at the nanometer level[15,16] in the bulk and grain boundaries (a signature for layer intermixing in the case of interfacial phase-change materials), but also to discover surprising behavior in bond rupture during the APT experiments. This was observed when various classes of materials, including covalent, ionic, metallic, and metavalent materials were investigated by APT. The difference in bond rupture is reflected in the probability of multiple events (PMEs) in APT experiments, as summarized in **Figure 1**.[14] The PME is defined as the probability of dislodging more than one ion when a laser pulse is applied to the apex of a needle-shape specimen, and expected to be generally very low for most materials.

Yet, Figure 1 shows that for those materials characterized by an unconventional combination of properties (PCMs), such as high dielectric constant ε_∞, large Born effective charge Z*, high effective coordination number ECoN, and large Grüneisen parameter γ_{TO}, a high PME (>60%) is observed. On the contrary, for all other materials that utilize covalent, ionic, or metallic bonding, a low PME (<33%) was detected, in agreement with the value expected and observed when a standard coevaporation phenomenon takes place in APT.[17] The exceptionally high PMEs observed in the former are hence an intrinsic property of crystalline PCMs. The mechanism responsible for this behavior was termed enhanced correlated field evaporation,[14] and should not be mistaken for the high PME of approximatively 40–45% detected when either field dissociation of molecular ions[18,19] or known measurement artifacts[20,21] occur.

Bonding in solids

So far, we have characterized materials and the bonding mechanism they utilize by their properties and their bond-breaking behavior. At the same time, other bond descriptors have been developed based on the quantum theory of atoms in molecules and crystals.[22] Two quantities are frequently utilized to characterize the nature of the chemical bond, the number of electrons transferred to/from an atom and the number of electrons shared between neighboring atoms. The amount of electrons shared between adjacent atoms (ES) and the number of electrons transferred to an atom (ET) can be used to distinguish between ionic, metallic, and covalent bonds[4] (e.g., purely covalent bonding features only shared electrons and no transferred electrons, while the opposite is true for the ionic bond). The ES and ET values are calculated from the difference in the electron wave function between the single atoms and the compound, as described in Reference 4. Employing ES and ET values as coordinates in a plot, metals, ionic, and covalent

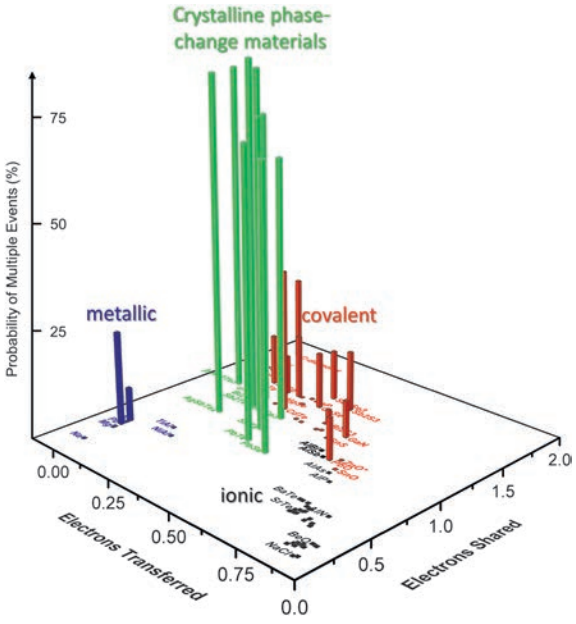


Figure 1. A 3D map using the basal plane of electrons transferred and electrons shared between atoms in various materials and, in the vertical axis, the "probability of multiple events" (PMEs) measured by laser-assisted atom probe tomography. The coordinates of the basal plane are the degree of electron transfer and electron pair formation between atoms in each material. All compounds with a high probability of generating multiple ionization events upon exposure to a single laser pulse are located in the green area of this plot, which is characterized by unconventional physical properties. In the green region, the PMEs range from about 60% to more than 80%, while the highest value observed outside this region is about 33%. This is thus clear experimental proof that the bond-breaking behavior of the crystalline phase-change materials differs from materials employing conventional bonding mechanisms (covalent, metallic, and ionic).[14] The number of electrons transferred is adapted from Reference 4, but renormalized by the formal oxidation state.

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compounds as well as incipient metals cluster in groups (i.e., occupy different regions) in this plot (see **Figure 2**). In this map, crystalline PCMs such as GeTe or Sb_2Te_3 are situated in between typical metallic and covalent materials. Yet, their properties are different (Table II) and feature a very different bond-breaking behavior (Figure 1).

We can use this insight in a number of ways. First, it helps us to identify other materials that should have the desired properties of PCMs. Similar maps can also be utilized to understand property trends, such as trends for ε_∞ or Z*. This is demonstrated in **Figure 3**, where both quantities are shown.

One can see that Z* is particularly large for materials that employ metavalent bonding and lie next to the region of metallic bonding. For ε_∞, high values are again found in the same region, but now for smaller values of electron transfer. It is quite interesting to observe such clear property trends as a function of changes in chemical bonding. However, regarding the requirements listed in Table I, neither ε_∞ nor Z* provide crucial guidance for property design. In this regard, a quantity such as γ_{TO} is more interesting, since it is closely related to the thermal conductivity of the lattice κ_{lat}. High values of γ_{TO} lead to low values of the thermal conductivity, which helps to ensure that the power consumption upon melting is reduced. The generally low thermal conductivities also help to understand why many materials in the region of metavalently bonded materials are also good thermoelectrics.

There are other properties that are potentially more important for PCMs. As can be seen from Table I, a crucial attribute of PCMs is their ability to be rapidly switched. Since crystallization of a PCM memory cell takes longer than melt-quenching,[9] the focus is on how to improve the crystallization speed and the material parameters involved.

Usually the regime of fast crystallization lies between the glass-transition temperature, T_g, and the temperature of melting, T_m. For PCM memory devices, it is necessary to enter this regime with minimal energy dissipation. In order to minimize energy dissipation, the absolute value of the glass-transition temperature, T_g, and the melting temperature, T_m, should be low. However, when the glass-transition temperature is too low (close to room or operating temperature), slight variations in temperature could lead to an unintended crystallization and reduced data retention times. On the other hand, when the glass-transition temperature is too high, too much energy will be required for achieving the temperatures needed for rapid crystallization. Additionally, the reduced glass transition, T_g/T_m, should be small for rapid crystallization.[23] When the glass transition and reduced glass-transition temperature meet these criteria, the fragility, m, of the material should be as high as possible. A high fragility of an undercooled liquid (UCL) is related to bad glass-forming ability and is characterized by a tremendous decrease in viscosity, η, upon the transition from a glass into the UCL, as depicted in **Figure 4** (see the Wei et al. article in this issue for more information).[24–26] Since the crystal growth velocity and nucleation rate are inversely related to viscosity η,[23–30] these increase hugely when the temperature exceeds the glass-transition temperature for high-fragility materials. Thereby, the necessary temperature increase for rapid crystallization and the energy required, as well as the time for complete crystallization, are reduced further by

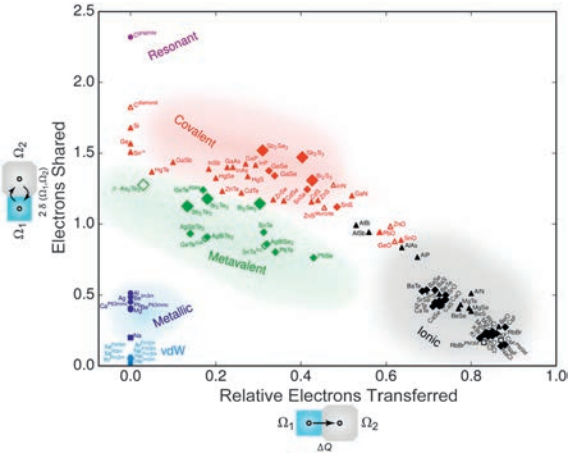


Figure 2. Two-dimensional map describing bonding in solids. The map is spanned by electron transfer (ET, ΔQ) between atomic basins (Ω_i), and the formation of electron pairs between them (ES), $\delta(\Omega_1,\Omega_2)$. Triangles, diamonds, squares, and circles denote tetrahedrally bonded solids, distorted and ideal rock-salt-type (octahedrally coordinated) structures, body-centered solids, and close-packed metals, respectively, while filled and open symbols represent thermodynamically stable and metastable phases (vdW, van der Waals).[4] Phase-change materials such as GeTe or Sb_2Te_3 all occupy a well-defined region of the map highlighted in green.

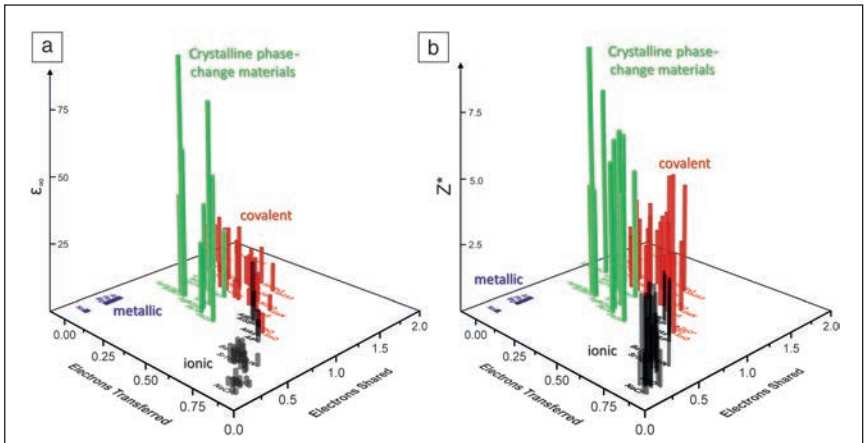


Figure 3. Three-dimensional maps showing unconventional properties of crystalline phase-change materials (in green) as compared with other classes of materials (covalent in red, metallic in blue, and ionic in black). (a) The optical dielectric constant, ε_∞, is plotted against ET and ES. ε_∞ is an optical identifier of chemical bonding.[4,5] (b) The Born effective charge, Z*, is plotted against electrons transferred (ET) and electrons shared (ES). Z* is a signature of the chemical bond polarizability.

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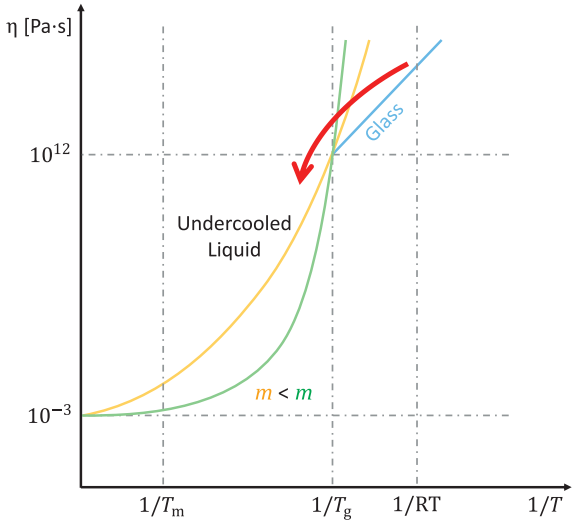


Figure 4. Schematic sketch of viscosity, η, as a function of inverse temperature (Angell plot). When a glass is heated from room temperature (RT) to above the glass-transition temperature, T_g, it enters the undercooled liquid phase. Upon glass transition, the temperature dependence of viscosity experiences a discontinuity.[24] When heated further to exceed the glass transition, the viscosity, η, decreases faster at higher fragility, m.[25] Since the crystal growth velocity and nucleation rate are both inversely proportional to viscosity, η, a more fragile material should crystallize faster than a less fragile one.

high fragility. Hence, it would be highly desirable to explore systematic trends for T_g, T_g/T_m, and fragility, m, as a function of chemical bonding in a map (Figure 3).

The glass-transition temperature is often determined in calorimetric experiments, where in the case of PCMs, crystallization initiates before the glass transition is revealed.[31] Because of this, T_g can only be estimated, which leads to a wide spread of reported T_g values ranging from 100 to 200°C in the case of the prominent PCM $Ge_2Sb_2Te_5$.[27,28,31–34] For another prominent PCM, AIST (AgInSbTe), values ranging from 105 to 185°C are reported.[31,32,35–37] However, in order to precisely predict crystallization kinetics of PCMs, accurate and physically meaningful values for T_g are mandatory. Until the development of suitable techniques to obtain these values experimentally, the range of possible T_g values can be restricted only due to the combined knowledge of its crystallization kinetics, glass dynamics, microscopic behavior, and by additional supportive simulations.[38]

A similar situation is present for assigning fragility values to PCMs. Since PCMs crystallize readily, the time scale to observe the UCL for measuring the viscosity is limited. For this reason, viscosity data, if any, only exist above or close to the melting temperature, T_m, but not close to the glass-transition temperature, T_g, where the fragility is usually determined. This lack of viscosity data in between T_g and T_m is truly a limitation for predicting the inversely proportional crystal growth velocity.

Summary

Besides being used in memory applications today, PCMs still offer the opportunity of further technological advances as well as exciting fundamental scientific insights. The unique bond breaking behavior observed in atom probe tomography is indicative of a novel bonding mechanism. This mechanism partly resembles metallic or covalent bonding, and yet PCMs feature a portfolio of properties which is not a linear superposition of metallic and covalent bonding. The maps described in this article, in which bonding character is clearly linked to trends in material properties, constitute a useful new route in material engineering. Unfortunately, so far it has not been possible to extend this map to crystallization kinetics, in order to tailor switching speeds of PCMs. The values reported so far for the glass-transition temperature of PCMs are inconclusive and contradictory. In addition, there are significant difficulties in determining the fragility of these materials. Hence, it has not yet been possible to produce a figure such as Figure 1 or Figure 3, where either T_g, T_g/T_m, or fragility values are depicted as a function of chemical bonding parameters (i.e., ES and ET). However, as such data become available, they will provide a framework to understand and optimize switching speeds and hence possibly provide an intriguing approach to tailor PCMs.

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