

Supplemental Material for “Scaling law for the onset of solidification at extreme undercooling”

Philip C. Myint,^{1,2} Dane M. Sterbentz,^{1,2,3} Justin L. Brown,⁴ Brian S. Stoltzfus,⁴
Jean-Pierre R. Delplanque,³ and Jonathan L. Belof^{2, a)}

¹⁾ *These authors contributed equally to this work*

²⁾ *Lawrence Livermore National Laboratory, Livermore, CA 94550,
USA*

³⁾ *Department of Mechanical & Aerospace Engineering, University of California,
Davis, CA 95616, USA*

⁴⁾ *Sandia National Laboratories, Albuquerque, NM 87185,
USA*

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^{a)}Electronic mail: belof1@llnl.gov

I. DERIVING THE SCALING LAW WITH DIMENSIONAL ANALYSIS

This section provides more details on the dimensional analysis. For concreteness, we explain the procedure used to derive the scaling law in Equation (1) of the main paper for the transition pressure P_{trans} , which we repeat here for convenience:

$$\tilde{P}_{\text{trans}} = \tilde{P}_{\text{melt}} + 7.06298 (\Pi)^{0.06976} (\tilde{P}_{\text{melt}})^{0.56842} (\tilde{T}_{\text{init}})^{-2.97690}. \quad (1)$$

A similar procedure may be followed to obtain scaling laws for the particle speed $u_{p,\text{trans}}$ (which is shown in the insets to Figure 2 of the main paper) and other quantities. The first step in the dimensional analysis^{1,2} is to determine the full functional dependence of P_{trans} . We do this with models based on classical nucleation theory (CNT) and growth³⁻⁷ because they have proven to be successful in describing a wide variety of phase transitions, including those in our past theoretical/computational work on solidification kinetics under quasi-isentropic compression⁸⁻¹⁴. According to CNT, the phase transition is associated with a nucleation energy barrier ΔG^* given by

$$\Delta G^* = \frac{16\pi}{3} \frac{\sigma^3}{(\rho\Delta\mu)^2}, \quad (2)$$

where σ is the solid-liquid interfacial free energy, ρ is the density of the liquid, and $\Delta\mu$ is the bulk chemical potential difference between liquid and solid. The energy barrier arises because although $\Delta\mu$ favors the formation of solid (since we are considering metastable liquid phases), the very formation of this solid requires the creation of an interface between the liquid and solid, which is energetically unfavorable. With this CNT-based physical picture in mind, we postulate that

$$P_{\text{trans}} = f\left(\frac{dP}{dt}, \Delta\mu, P_{\text{melt}}, T, \rho, \sigma, R\right). \quad (3)$$

That is, we assume that P_{trans} is a function of seven dimensional parameters, including the compression rate dP/dt , which is our main independent variable of interest and is defined as the average compression rate between the time t_{trans} corresponding to P_{trans} and the time t_{melt} when the isentrope corresponding to the loading path crosses the melt curve (i.e., the earliest instance when solidification becomes theoretically possible):

$$\frac{dP}{dt} = \frac{P_{\text{trans}} - P_{\text{melt}}}{t_{\text{trans}} - t_{\text{melt}}}. \quad (4)$$

The temperature T and specific gas constant R also appear in Equation (3) because CNT-based nucleation and growth occur through thermally driven fluctuations and atomic-scale attachment processes.

An important part of the dimensional analysis is to choose characteristic values of $\Delta\mu$, T , ρ , and σ to substitute into Equation (3). A clear choice is to use the values of these quantities evaluated at the equilibrium melt curve (the solid–liquid equilibrium phase boundary). That is, for a particular quasi-isentropic experiment with an initial temperature T_{init} and pressure P_{init} (all of the experiments considered in this study are initiated at atmospheric pressure so that $P_{\text{init}} = 1$ bar), the characteristic values of $\Delta\mu$, T , ρ , and σ are evaluated at the point where the isentrope initiated from T_{init} and P_{init} intersects the melt curve. For the generic phase-diagram example depicted in Figure 1(a) of the main paper, this intersection point is labeled as P_{melt} . The characteristic values $\Delta\mu_{\text{melt}}$, T_{melt} , ρ_{melt} , and σ_{melt} evaluated at this intersection point not only define the physical scale of these quantities, they are also practical choices because they are purely equilibrium properties, meaning that they can be computed in a standalone manner from a given equation of state (EOS) and interfacial free-energy model without requiring any hydrodynamics or kinetics code. With this choice, however, the driving force $\Delta\mu$ becomes trivial because $\Delta\mu_{\text{melt}}$ is identically zero. In order to preserve in some form the information content that $\Delta\mu$ is intended to convey, we substitute T_{init} in place of $\Delta\mu_{\text{melt}}$, because when T_{init} is nondimensionalized by T_{melt} (as we will do in the next paragraph), this dimensionless temperature ratio $\tilde{T}_{\text{init}} = T_{\text{init}}/T_{\text{melt}}$ gives a sense of the driving force. Temperature differences have been used in previous CNT analyses to estimate $\Delta\mu$ of nonequilibrium phase transitions⁵, and the nondimensionalized initial temperature $\tilde{T}_{\text{init}} = T_{\text{init}}/T_{\text{melt}}$ that appears in Equation (1) is essentially a relative difference between T_{init} and T_{melt} . Larger values of $\tilde{T}_{\text{init}} = T_{\text{init}}/T_{\text{melt}}$ (i.e., values of $\tilde{T}_{\text{init}}$ that are close to unity) mean that T_{init} is close to T_{melt} , implying that small changes in T or P lead to large changes in $\Delta\mu$. For a material where $\Delta\mu$ is strongly sensitive to T or P , one would not expect to be able to drive the system very far past equilibrium (the material is difficult to undercool), and so large values of $\tilde{T}_{\text{init}}$ correlate with small values of $\Delta\mu$. As a result, $\tilde{T}_{\text{init}}$ may be used as a surrogate for $\Delta\mu$ in the dimensional analysis. Designating the characteristic values $\Delta\mu_{\text{melt}}$, T_{melt} , ρ_{melt} , and σ_{melt} in Equation (3) and also substituting T_{init}

66 for $\Delta\mu_{\text{melt}}$ in that equation, we have

$$P_{\text{trans}} = f\left(\frac{dP}{dt}, T_{\text{init}}, P_{\text{melt}}, T_{\text{melt}}, \rho_{\text{melt}}, \sigma_{\text{melt}}, R\right). \quad (5)$$

67 We now choose a subset of the seven parameters on the right-hand side of Equation (5)
 68 to carry out the crucial step of nondimensionalization. This subset should span the four
 69 physical dimensions that appear in our problem, which are mass, length, time, and temper-
 70 ature. For this purpose, we choose the subset T_{melt} , ρ_{melt} , σ_{melt} , and R . We use these four
 71 “repeating parameters” to nondimensionalize P_{trans} and the three remaining parameters in
 72 Equation (5): dP/dt , T_{init} , and P_{melt} . This involves finding the exponents a , b , c , and d in
 73 the following equation that produce a dimensionless transition pressure $\tilde{P}_{\text{trans}}$:

$$\tilde{P}_{\text{trans}} = \frac{P_{\text{trans}}}{(\rho_{\text{melt}})^a (\sigma_{\text{melt}})^b (R)^c (T_{\text{melt}})^d}. \quad (6)$$

74 We do the same to produce dimensionless versions of dP/dt , T_{init} , and P_{melt} . This process
 75 of determining the exponents may be posed as a matrix equation, and solving that equation
 76 yields

$$\tilde{P}_{\text{trans}} = \frac{P_{\text{trans}}}{\rho_{\text{melt}} R T_{\text{melt}}}, \quad (7)$$

$$\Pi = \frac{dP/dt}{\rho_{\text{melt}}^2 \sigma_{\text{melt}}^{-1} (R T_{\text{melt}})^{5/2}}, \quad (8)$$

$$\tilde{T}_{\text{init}} = \frac{T_{\text{init}}}{T_{\text{melt}}}, \quad (9)$$

$$\tilde{P}_{\text{melt}} = \frac{P_{\text{melt}}}{\rho_{\text{melt}} R T_{\text{melt}}}. \quad (10)$$

80 Here, Π is the dimensionless compression rate, $\tilde{T}_{\text{init}}$ is the dimensionless initial tempera-
 81 ture (which is a surrogate for the driving force $\Delta\mu$, as mentioned above), and $\tilde{P}_{\text{melt}}$ is the
 82 dimensionless melt pressure.

83 The great benefit of the nondimensionalization is that it reduces the seven-parameter
 84 space in Equation (5) to a much more manageable three-parameter space involving the
 85 dimensionless quantities:

$$\tilde{P}_{\text{trans}} = f(\Pi, \tilde{T}_{\text{init}}, \tilde{P}_{\text{melt}}). \quad (11)$$

86 For each quasi-isentropic compression experiment, we employ an appropriate EOS and in-
 87 terfacial free-energy model to determine $\tilde{T}_{\text{init}}$ and $\tilde{P}_{\text{melt}}$. We use our two-phase EOS⁸ in the

case of water and ice VII, while we use a recent multiphase gallium EOS¹⁵ for the liquid
 and Ga-III phases of gallium (Ga), which are the two phases that are relevant to our experi-
 ments on Ga. The interfacial free energy of both materials is obtained from a general model,
 published in a recent paper of ours¹⁴, that we have developed to be applicable to metallic
 and nonmetallic systems alike. The transition pressure P_{trans} and compression rate dP/dt
 can, in principle, be extracted from experimental wave profiles obtained through velocimetry
 (e.g., VISAR¹⁶). But we have chosen instead to extract them from wave profiles generated
 by our simulations, which we have shown in our previous studies^{10,13,14} to yield good agree-
 ment with experimental wave profiles; Section III of this Supplemental Material (SM) also
 shows comparisons between our Ga experiments and simulations. The simulations addition-
 ally provide insight into quantities like the nucleation rate J and energy barrier $\Delta G^*/k_{\text{B}}T$,
 which are illustrated in Figure 3 of the main paper, that are not available experimentally.
 Specifically, we run our phase-transition kinetics code Samsa^{9,10,13}, which is coupled to the
 multiphysics continuum-scale code Ares^{17,18} (see our 2020 paper¹³ for numerical details on
 how this coupling is implemented), to simulate the experiments. The input drives for many of
 the experiments involving water are obtained through application of our drive-optimization
 code¹². We run Samsa/Ares to simulate each experiment, and we use the aforementioned
 EOS, interfacial free-energy, and CNT models (which are all implemented in Samsa) to com-
 pute $\tilde{P}_{\text{trans}}$, Π , $\tilde{T}_{\text{init}}$, and $\tilde{P}_{\text{melt}}$ in each simulation. This gives us a set of “data points” in the
 dimensionless three-parameter space indicated by Equation (11).

We then apply a multivariable regression analysis on these data points to obtain the
 scaling law in Equation (1). We have employed methods from the `scikit-learn` Python
 library¹⁹ to perform the regression. As shown in the main paper, our study considers a total
 of 18 experiments: 14 on water and our four experiments on Ga, the latter of which are
 described further in Section III of this SM. We split up these 18 experiments into a test set
 composed of five experiments (four on water and one on gallium) and a training set composed
 of the remaining 13 experiments. To combat overfitting in the regression, we have employed
`ElasticNet` regularization¹⁹ (which utilizes a combination of L_1 and L_2 regularization) in
`scikit-learn`, with the `GridSearchCV` method used to perform a grid search over the α
 and L_1/L_2 ratio hyperparameters involved in the `ElasticNet` regularization. We have ex-

118 amined α values ranging from 10^{-6} to 1.0 and L_1/L_2 ratios ranging from 10^{-6} to 0.99. The
 119 **GridSearchCV** method found that the best set of hyperparameters yield an average deviation
 120 (here, the deviation is defined as the difference between the target value of $\tilde{P}_{\text{trans}}$ and the
 121 value of $\tilde{P}_{\text{trans}}$ predicted by the regression with Π , $\tilde{T}_{\text{init}}$, and $\tilde{P}_{\text{melt}}$ serving as the predictors)
 122 of less than 3% for both the training set and the test set. The resulting regression coefficients
 123 produced by this regularized fit are displayed in Equation (1).

124 **II. TWO APPLICATIONS OF THE SCALING LAW IN HIGH-PRESSURE** 125 **SCIENCE**

126 **A. Effect of compression rate on homogeneous vs. heterogeneous nucleation**

127 By following the same arguments used to analyze Figure 3 in the main paper, it is straight-
 128 forward to see why heterogeneous nucleation (i.e., nucleation due to impurities dissolved in
 129 the metastable liquid) becomes less important relative to homogeneous nucleation with in-
 130 creasing compression rates dP/dt . Recall from our discussion of Figure 3 that CNT gives the
 131 (steady-state) homogeneous nucleation rate as $J = \mathcal{A} \exp(-\Delta G^*/k_B T)$, where the preexpo-
 132 nential factor $\mathcal{A}$ largely reflects the availability of nucleation sites, and $\Delta G^*/k_B T$ is a nondi-
 133 mensionalized version of the nucleation energy barrier ΔG^* defined in Equation (2). The het-
 134 erogeneous nucleation rate³ may be similarly expressed as $J_{\text{hetero}} = x \mathcal{A} \exp(-\Delta G_{\text{hetero}}^*/k_B T)$,
 135 in which x is the impurity mole fraction and $\Delta G_{\text{hetero}}^* = \psi \Delta G^*$, where $0 < \psi \leq 1$ is a barrier-
 136 reduction factor that accounts for atomic-scale interactions facilitated by the impurities. At
 137 ambient conditions, the ratio

$$J_{\text{hetero}}/J_{\text{homo}} = x \exp[(1 - \psi)\Delta G^*/k_B T], \quad (12)$$

138 tends to be greater than one, meaning that heterogeneous nucleation is the dominant mech-
 139 anism for the phase transition at ambient conditions, because the barrier reduction factor ψ
 140 tends to be sufficiently small that it can compensate for the impurity fraction x being perhaps
 141 much less than 1. However, as discussed in our previous work^{9,10}, homogeneous nucleation
 142 can overtake heterogeneous nucleation as the primary mechanism if the liquid becomes un-
 143 dercooled to high-pressure metastable states that lie well past the equilibrium melt point.

Such highly undercooled states are possible in the quasi-isentropic platforms considered in this study, making these platforms unique in being able to probe these kinds of states. At these high-pressure conditions, the barrier $\Delta G^*/k_B T$, which appears in an exponential in Equation (12), becomes small enough that the ratio $J_{\text{hetero}}/J_{\text{homo}}$ remains less than unity. In other words, as the liquid gets increasingly undercooled past the equilibrium melt boundary to higher-pressure metastable states, the effect of the barrier reduction ψ , which favors heterogeneous nucleation, diminishes so that it becomes too small to compensate for the fact that $x \ll 1$. Since the transition pressure P_{trans} increases with dP/dt (which is a reflection of the fact that it is easier to overdrive a liquid if it is compressed/undercooled more quickly), $\Delta G^*/k_B T$ also decreases with dP/dt , as illustrated in Figure 3(a) of the main paper. Therefore, the ratio $J_{\text{hetero}}/J_{\text{homo}}$ diminishes with increasing compression rates. We repeat here some implications of this fundamental result: even if heterogeneous nucleation is found to be negligible at high compression rates, say in a laser-drive experiment, one cannot necessarily conclude that it would also be unimportant at the lower compression rates of a gas-gun experiment. Conversely, if heterogeneous nucleation is found to be important in a gas-gun experiment, the same may not be true in a laser-drive experiment.

B. A novel means to determine equilibrium melt temperatures at high pressures

The scaling law in Equation (1) is predicated on the idea that by applying dimensional analysis on a well-chosen set of characteristic values which define the physical scale of the problem, one can obtain a general relation that enables prediction of the observed, nonequilibrium transition pressure P_{trans} as a function of the compression rate dP/dt . The characteristic values chosen in our case are the equilibrium melt properties P_{melt} , σ_{melt} , ρ_{melt} , and T_{melt} . As stated in the main paper and elsewhere in this SM, these characteristic values are properties evaluated at the point where the corresponding isentrope intersects the melt curve (the solid-liquid equilibrium phase boundary), which is the point labeled as P_{melt} in the example shown in Figure 1(a) of the main paper. Thus, the scaling law is a relation that allows one to predict the nonequilibrium transition pressure P_{trans} as a function

of the compression rate dP/dt from knowledge of the material’s equilibrium melt properties P_{melt} , σ_{melt} , ρ_{melt} , and T_{melt} . This equilibrium $\rightarrow$ nonequilibrium prediction is the intended “forward” direction of the scaling law that is demonstrated in the main paper. Here, in this section of the SM, we consider whether the scaling law can also be applied in the “reverse” direction of nonequilibrium $\rightarrow$ equilibrium. That is, if one can experimentally obtain the nonequilibrium quantity P_{trans} for some material whose melt properties are not well known, can P_{trans} (as well as the compression rate dP/dt) be used in conjunction with the scaling law to back out P_{melt} , σ_{melt} , ρ_{melt} , and T_{melt} of that material?

Figure 1 below demonstrates that it is indeed possible to reliably obtain the melt temperature T_{melt} in this manner, though not the other three melt properties (P_{melt} , σ_{melt} , and ρ_{melt}) for reasons that we will explain shortly. We have produced this figure by simultaneously adjusting the value of all four melt properties in each experiment of that figure and plugging these values into the scaling law in Equation (1) to try to produce a value for P_{trans} that closely matches the input, target value of P_{trans} . We then compare these optimized values for ρ_{melt} , P_{melt} , T_{melt} , and σ_{melt} against their actual values (which we know from our EOS and interfacial free-energy models) to see how well the scaling law can reproduce these melt properties. Practically any multivariable optimization technique could be suitable for this purpose, and in our case, we utilize particle swarm optimization (PSO), which is a now widely used stochastic method^{20,21}. Specifically, we perform 50 PSO runs for each experiment in Figure 1, and the T_{melt} values shown in that figure are averages over those 50 runs. In each run, we provide wide-ranging lower and upper bounds over which the melt properties are allowed to vary in order to simulate the situation where we are working with a material whose EOS and interfacial free energy are not known to a high degree of certainty: these bounds are $\pm 5\%$, $\pm 20\%$, $\pm 50\%$, $\pm 80\%$ from the actual values for ρ_{melt} , P_{melt} , T_{melt} , and σ_{melt} , respectively. Figure 1 shows that the initial $\pm 50\%$ uncertainty imposed on T_{melt} reduces down to $\pm 4\%$ (and often times within $\pm 2\%$) at the end of the optimization process. Thus, the scaling law can reliably reproduce the melt temperature T_{melt} given information on just the transition pressure P_{trans} and compression rate dP/dt .

Interestingly, while this optimization procedure works well for T_{melt} , it cannot also be used to reliably infer the other three melt properties ρ_{melt} , P_{melt} , and σ_{melt} , as we have

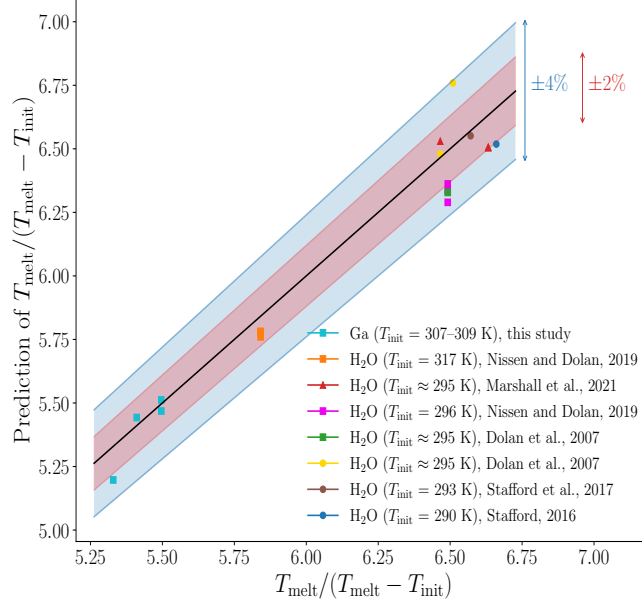


FIG. 1. The melt temperature T_{melt} (normalized by its difference with the initial temperature T_{init}) predicted by the scaling law for each experiment compared against the actual value of $T_{\text{melt}}/(T_{\text{melt}} - T_{\text{init}})$, with the black line indicating perfect agreement ($y = x$). As explained in the text, we have generated this figure by performing a multivariable optimization on the scaling law to infer T_{melt} from the corresponding values of the transition pressure P_{trans} . When applied in this reverse direction, the scaling law reproduces T_{melt} to within $\pm 4\%$ (and within $\pm 2\%$ for nearly all of the experiments), suggesting that it can be used to obtain a fairly precise estimate of T_{melt} for a material with an unknown melt curve from experimental measurements on P_{trans} . The melt temperature T_{melt} takes on a special status in this regard, as the three other melt properties (P_{melt} , σ_{melt} , and ρ_{melt}) that appear in the scaling law cannot be inferred in the same manner.

found that PSO does not consistently further narrow down the bounds stated above for these properties. The reason why T_{melt} is unique in this regard can be understood from both a mathematical and physical perspective. Mathematically, this is because, as shown in Equation (9), T_{melt} appears by itself in the nondimensionalization of T_{init} , and the large magnitude of the exponent on $\tilde{T}_{\text{init}}$ [from Equation (1), this exponent is -2.97690 , as opposed to 0.06976 and 0.56842 for the exponents on Π and $\tilde{P}_{\text{melt}}$, respectively] means that the scaling law is very sensitive to even small changes in T_{melt} . This reduces the “degeneracy” of the

209 scaling law with respect to T_{melt} so that values of T_{melt} that are even somewhat off from the
 210 actual value (say more than 5%) will lead to wildly wrong predictions for P_{trans} . Physically,
 211 the reason why the exponent on $\tilde{T}_{\text{init}}$ is relatively large in magnitude is because $\tilde{T}_{\text{init}}$ is a
 212 surrogate for the driving force $\Delta\mu$, which has a powerful influence on the nucleation rate,
 213 solidification kinetics, and ultimately also P_{trans} . The exponent is negative because as we
 214 have explained earlier in Section I, large values of $\tilde{T}_{\text{init}} = T_{\text{init}}/T_{\text{melt}}$ (or equivalently, small
 215 differences between T_{init} and T_{melt}) are correlated with small values of $\Delta\mu$, meaning that it is
 216 difficult to undercool such a material far past equilibrium. It follows then that large values
 217 of $\tilde{T}_{\text{init}}$ are also correlated with small values of P_{trans} , hence a negative exponent on $\tilde{T}_{\text{init}}$.

218 It is encouraging that this ability of the scaling law to infer T_{melt} from knowledge of
 219 just two experimental observables (P_{trans} and dP/dt) appears to be equally applicable to
 220 water and Ga, despite the contrasting nature of the two materials (e.g., water, being a
 221 molecular substance, can exhibit a much larger entropy of fusion ΔS_{melt} than the monatomic
 222 metal Ga). This reflects the generality of the scaling law to a wide range of materials
 223 where solidification is well described by CNT. Obtaining reliable temperature measurements
 224 under dynamic compression, especially the melt temperature, is a long-standing area of
 225 interest (for example, see some recent studies on iron^{22–25}). The scaling law thus provides
 226 a way to determine T_{melt} of a material whose melt curve is not known to a high degree of
 227 precision, and interestingly, this equilibrium property can be inferred (by running the scaling
 228 law “in reverse”) from the nonequilibrium transition pressure P_{trans} , which is a seemingly
 229 unrelated quantity that is generally more accessible experimentally than T_{melt} . In fact, if
 230 one obtains P_{trans} measurements for several different quasi-isentropes with each originating
 231 from a different initial temperature, the scaling law could then potentially be applied to map
 232 out T_{melt} at several points along the melt curve.

233 **III. MAGNETIC-DRIVE EXPERIMENTS AND SIMULATIONS ON** 234 **GALLIUM**

235 A schematic of our four Ga magnetic-drive experiments is given in the inset in Figure 2(a),
 236 with details summarized in Table I. The 1-D configuration shown is a simplified version of

a typical experiment conducted on the Thor pulsed-power machine²⁶ and are largely an extension of previously published works on water^{27,28} but adapted for use with liquid Ga. The aluminum electrodes are separated by a 0.5 mm A-K gap formed by a solid Kapton dielectric. Thor produces a tailored time-dependent current that flows through the electrodes and induces a magnetic field within this A-K gap that then launches a stress wave into each electrode. One electrode is backed by a [100] lithium fluoride (LiF) crystal while the other side contains a thin liquid Ga sample sandwiched between the aluminum (Al) and LiF. This Al/LiF system was chosen because these materials have well-matched shock impedances, which minimize wave interactions and simplify interpretation of the experiment. Additionally, LiF is optically transparent, which enables the velocity to be measured through conventional velocimetry techniques; here, VISAR¹⁶ is used to measure Ga/LiF interface velocities. Both Al panels contain a type-II anodized coating (MIL-A-8625) on the surface opposite the A-K gap. This coating was found to be sufficient to prevent the well-known phenomena of Ga embrittlement²⁹ of the Al panels, which caused failures in preliminary testing of noncoated panels. The coatings are estimated to be $\sim 20 \mu\text{m}$ thick and are not believed to alter the interpretation of the data.

Elemental Ga becomes a liquid at 303 K. The samples were assembled by starting with solid high purity (99.99%) solid lump of Ga. The lump was placed in a furnace at ~ 320 K and was observed to liquify when it pooled and became specular in its reflectivity. A heated injector was then used to load the liquid Ga into the panel assembly, which was heated alongside the Ga in the furnace. The panel assembly was then loaded into the Thor machine. The central powerflow region of Thor is a stainless steel block that was heated using an engine block heater to a steady-state temperature near 308 K. This block effectively acts as a heat sink and the panel assembly was allowed to equilibrate with it over the several hours it took to attach the velocity diagnostics and make Thor ready to fire. K-type thermocouples located near the panel assembly were used to measure the equilibrium temperature and the liquid state of the Ga was verified optically through examination of the reflectivity.

The VISAR velocity measurements of the Ga/LiF interface for the four Ga experiments are given in Figure 2(a). The current pulses were intentionally varied to alter the applied loading rate and peak state, resulting in the differences in the measurements. This quasi-

TABLE I. Summary of our four magnetic-drive experiments on Ga; see Figure 2 for an illustration of the setup configuration and the velocimetry results. From left to right, the columns indicate the experiment number, thickness of the Al panel, thickness of the Ga sample, initial temperature, and values for the transition pressure P_{trans} and compression rate dP/dt from our Samsa/Ares simulations (see Section I of this SM and our earlier papers^{9,10,13,14} for more details) and our experiments. The thickness of the Ga samples were determined through liquid-only simulations, as there are large uncertainties in the measured pre-shot estimates of these initially liquid samples.

Experiment	Al thickness (mm)	Ga thickness (mm)	T_{init} (K)	P_{trans} sim (GPa)	P_{trans} exp (GPa)	dP/dt sim (GPa/ns)	dP/dt exp (GPa/ns)
Ga-1	0.548	0.15	308.5 ± 2	18.7	18.5 ± 0.4	0.77	0.85 ± 0.10
Ga-2	0.606	0.19	309.8 ± 2	18.2	18.1 ± 0.4	0.53	0.51 ± 0.07
Ga-3	0.581	0.25	307.2 ± 2	17.8	17.6 ± 0.4	0.39	0.40 ± 0.06
Ga-4	0.583	0.26	307.2 ± 2	17.5	17.3 ± 0.4	0.22	0.22 ± 0.05

isentropic loading couples with the Ga thickness to give the solidification signatures, which manifest as a pullback in the velocity near 1 km/s. The experiments were designed to quantify the Ga solidification kinetics based on this velocity signature. Since the Al and LiF are impedance matched and the Ga sample is significantly thinner, the spatial pressure gradient across the sample is minimized. Thus, the known properties of the LiF can be used to estimate the pressure history throughout the Ga thickness. The Sesame 7271 tabular EOS for LiF³⁰ was used to extract the principal isentrope and this was used to map the measured LiF velocities to pressures. Estimated uncertainties for the EOS are 2% in pressure and are added in quadrature to the velocity uncertainties to give the errors in the pressure histories. The results of this mapping for the four velocity profiles in Figure 2(a) are shown in Figure 2(b). The estimated Ga melt pressure along its isentrope is shown as a horizontal dashed line and labeled P_{melt} . The transition pressure, P_{trans} is taken to be the peak of the pressure loop associated with solidification of the initially liquid Ga to the Ga-III solid phase. As shown in Figure 2(b), these points define $\Delta P = P_{\text{trans}} - P_{\text{melt}}$ and $\Delta t = t_{\text{trans}} - t_{\text{melt}}$ and are used to estimate dP/dt through Equation (4), where the uncertainty in ΔP is combined

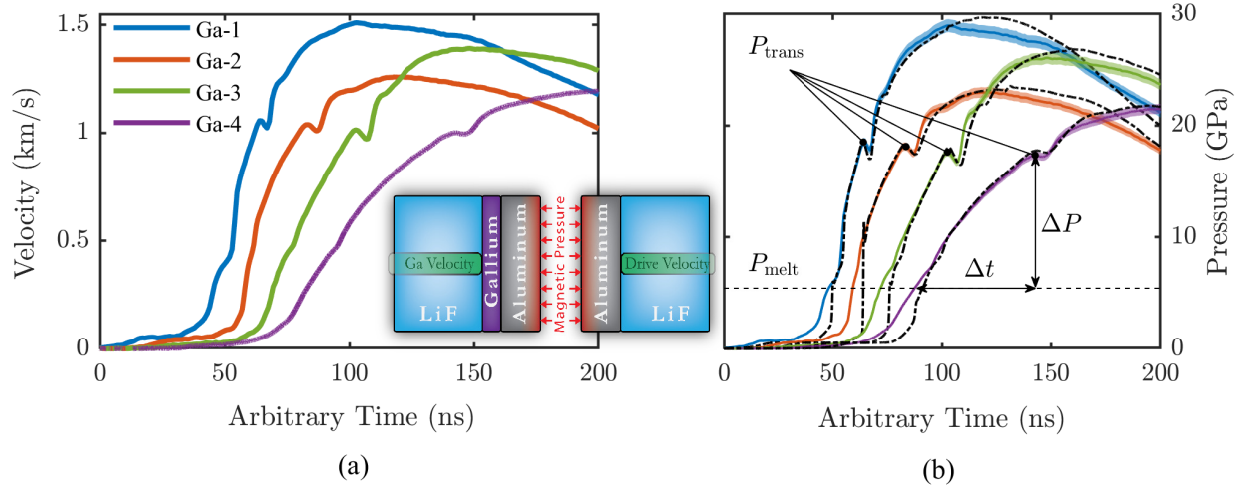


FIG. 2. Ga magnetic-drive experimental configuration and data. A shaped-current pulse is used to create a time-dependent magnetic pressure wave that propagates into the Al electrodes and eventually into the Ga. In (a), measured velocities at the Ga/LiF interface are shown for four different experiments utilizing different current pulses to alter dP/dt . Solidification of the Ga causes the distinct pullback feature near velocities of 1 km/s. In (b), the velocities are translated to pressures through a LiF EOS³⁰. Measurements are depicted as solid lines with $\pm 1\sigma$ uncertainty bands, and simulations are depicted as black dashed lines. The simulations agree well with the experiments for the pullback feature, including the pressure P_{trans} at the peak of this phase-transition loop. An example is shown for Ga-4 detailing how P_{trans} and P_{melt} determine ΔP and Δt for the calculation of dP/dt .

with the VISAR's 1 ns timing uncertainty to estimate the error in dP/dt . The results of this analysis are summarized in Table I, which show good agreement between our simulations and experiments for both dP/dt and P_{trans} .

Even more detailed information can be extracted through magnetohydrodynamic simulations of the experiments. The Al/LiF (non-Ga) measurement is referred to as the *drive* measurement because it allows for quantification of the time-dependent magnetic field within the A-K gap. Coupling simulations of the drive-side configuration with an assumption of known material properties of the Al and LiF leads to an inverse problem where the magnetic

field can be constructed through a least-squares fit of the simulated to experimental drive measurements; details for the method and material properties used are described in earlier references^{31,32}. Once the magnetic field is determined, the only remaining unknown is the response of the Ga. Towards this purpose, we employ Samsa/Ares simulations, and the physical models implemented into these codes, that are briefly described in Section I of this SM. Details on these models and codes are presented in our earlier papers^{9,10,13,14}. We feed in the drive profiles described earlier in this paragraph as inputs into Samsa/Ares to simulate each experiment. As illustrated in Figure 2(b), the simulations achieve agreement to within the estimated experimental uncertainties for the pullback loop feature that indicates solidification of Ga. This agreement gives confidence in other simulated kinetic quantities that provide further insight into the scaling law, like the nucleation rate J , energy barrier ΔG^* , and Kolmogorov time τ discussed in Figure 3 of the main paper.

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