Specific heat anomaly in a supercooled liquid with amorphous boundary conditions

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We study the specific heat of a model supercooled liquid confined in a spherical cavity with amorphous boundary conditions. We find the equilibrium specific heat has a cavity-size-dependent peak as a function of temperature. The cavity allows us to perform a finite-size scaling (FSS) analysis, which indicates the peak persists at a finite temperature in the thermodynamic limit. We attempt to collapse the data onto a FSS curve according to different theoretical scenaroi, obtaining reasonable results in two cases: a “not-so-simple” liquid with nonstandard values of the exponents $\alpha$ and $\nu$, and random first-order theory (RFOT), with two different length scales.

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In fragile glassformers, the relaxation time increases faster than the Arrhenius law as temperature is lowered [1]. This implies that the effective barrier to relaxation grows on cooling, which leads to expect concomitant structural, and perhaps thermodynamic, changes. Though not universally accepted, the idea that a thermodynamic transition may underlie the dynamic glass transition is old and at the core of random first-order theory (RFOT) and other theoretical approaches [2]. The question of the existence of a transition is open; in fact structural changes accompanying the slowdown have been found only recently [3–11], after more than a decade of study of dynamic correlations [12, 13].

The most general tools for probing structural correlations are the “order-agnostic” methods —which include patch correlations [7, 14], finite-size scaling (FSS) [15, 16], point-to-set (PTS) [17] and its related correlations—which do not need knowledge of the order parameter. Calculation of PTS correlations involves the study of confined systems, and in part for this reason a growing number of studies of liquids under various confined geometries have been reported, mainly cavities with amorphous boundary conditions (explained below) [3, 10, 18–21], “cavities” with open directions [21, 22] and systems with pinned particles [9, 21, 23]. These investigations have focused mostly on density correlations, from which a correlation length can be extracted.

Here we report numerical results on the specific heat $C_V$ of a system confined under amorphous boundary conditions (ABCs), therefore combining the ABCs and standard FSS approaches [24]. We find an anomalous peak as a function of temperature. The algorithm we use (swap Monte Carlo (MC) [25]) provides a complete sampling of configuration space at all the temperatures we report, so the peak is completely unrelated to the usual anomalies caused by the system falling out of equilibrium. We use FSS to study the changes of this thermodynamic anomaly as the cavity is enlarged, and our results indicate that it remains at a finite temperature in the thermodynamic limit. This is further evidence of the structural changes happening in supercooled liquids, and supports the existence of a thermodynamic transition.

We study the soft-sphere binary mixture of ref. [26] with size ratio 1.2 and unit density. To confine with ABCs, a spherical cavity of radius $R$ is created in an equilibrium configuration from a periodic boundary conditions (PBCs) system at temperature $T$, introducing a hard wall that conserves density and composition inside the cavity [3, 20, 27]. Inside particles evolve with swap MC [25] at the same temperature, while outside particles are held fixed. The specific heat is computed through energy fluctuations, $C_V = \frac{\langle E^2 \rangle - \langle E \rangle^2}{(MT^2)}$, where $E$ is the energy, $M$ the number of cavity (free) particles, and the overline means average over different realizations of the BCs. All results correspond to the (meta)equilibrium supercooled liquid. We used the energy time correlation function (checking for aging and finite-time effects) to estimate a correlation time and ensure that all relevant quantities were computed using runs lasting more than 100 relaxation times (including, self-consistently, the energy correlation). We used the bond orientation order parameter $Q_6$ [28] to exclude samples that showed signs of crystallization and could give a spurious contribution to the liquid $C_V$. Note that equilibration of small cavities is not problematic since with swap Monte Carlo smaller cavities are faster (not slower) than larger ones [29]. For detailed description of simulation and equilibration procedures and crystallization checks, see SM [30].

Fig. 1 shows the specific heat per mobile particle for ABCs for several cavity sizes (from 28 to 2000 mobile
have assumed no particular theory, nor any divergence of
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\( \lambda \)

hand, is stuck. Hence, the mere presence of this peak does not
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anomalous peak can be explained if we accept that the
served peak is an anomaly. A similar anomaly has been
the phonon theory of liquids [35]): in this sense, the ob-
T

increasing with temperature (as has been shown for a
For classical liquids, displaying a peak. The peak is not due to the
particles), displaying a peak. The peak is not due to the
system going out of equilibrium, or to crystal formation.
For classical liquids, \( C_V \) is expected to be monotonically
decreasing with temperature (as has been shown for a
large number of liquid models [33, 34] and follows from
the phonon theory of liquids [35]): in this sense, the ob-
served peak is an anomaly. A similar anomaly has been
reported before [25, 36], although in rather small systems
and without a FSS analysis.

In the simplest scenario, the qualitative origin of this
anomalous peak can be explained if we accept that the
effect of the border penetrates into the cavity as far as a
length-scale \( \lambda(T) \), and that this penetration length
increases for lower \( T \) [20]. At very high \( T \) the effect of the
boundary is weak (\( \lambda(T) \) is very small) and the \( C_V \) of
the liquid inside the cavity follows its bulk behavior (in
our case well described at high \( T \) by the Rosenfeld-Tarazona
law \( C_V \sim C_V^{(b)} = AT^{-2/5} \)). For low \( T \), on the other
hand, \( \lambda(T) \) will be large compared to the size \( R \), so that
the cavity will be almost frozen. The crossover from an
increase to constant gives rise to the peak. Notice we
have assumed no particular theory, nor any divergence of
\( \lambda(T) \) here, only that a very small cavity (relative to \( \lambda \))
is stuck. Hence, the mere presence of this peak does not
allow us to discriminate among theoretical frameworks.
We need to be more quantitative.

The penetration length \( \lambda \) is conceptually different from
the correlation length \( \xi \) [20, 22, 37, 38]. The correlation
length is a measure of the distance that two points must be
separated form each other so that the local state is
mutually independent (or in a cavity, how large must the
cavity be so that the state at the center is independent
from the state at the walls). The penetration length is
only meaningful in the presence of domains, and is a mea-
sure of the width of the domain walls (in a cavity, how
far from the wall a point must be to be independent from
the state outside). \( \xi \) can also be thought of as a measure
of the size of the domains (or cooperatively rearranging
regions), and \( \lambda \) as a measure of the interface width, or
the extent of the spatial fluctuations of the walls separating
such domains or regions. The lengths can be coincide in
simple cases (like the Ising model), but in principle they
measure two different phenomena.

There are thus two sides in our finite-size story: a finite
size is needed for the ABCs border to have an effect, but
the converse is not true: with PBCs, for instance, we can
have finite size but no border effects. Hence we must try
to include, but separate, both effects: that of the ABCs
border (related to \( \lambda \)) and that of the finite size (related
to \( \xi \)). We believe a reasonable way to do this, at least
near the peak, is to write

\[
C_V(R, T) = R^{\alpha/\nu} \tilde{c}(y)[1 - f(R/\lambda)].
\]

The factor \( R^{\alpha/\nu} \tilde{c}(y) \) is the border-free, FSS form of
the specific heat, and it would be a safe bet in most finite-size
systems with PBCs [39]. Through the scaling variable
\( y \equiv R^{1/\nu}(T - T_c)/T_c \), the finite-size term contains all
the information about the possible existence of a finite-
temperature transition, \( T_c \), and about the correlation
length \( \xi \sim (T - T_c)^{-\nu/\nu}; \) hence \( \tilde{c}(y) \) is a function of \( \xi/R \).
The second factor on the r.h.s. is meant to take care of
the border: for large \( R/\lambda \) the border function \( f \sim 0 \) and
the effect of the border is negligible (but not necessarily
that of finite size). But we need to be more specific as
regards \( f(x) \) to test our scaling ansatz Eq. 1, so we make
the simplest assumption: that the specific heat is zero
exactly at the border and relaxes exponentially to the
(finite-size) PBCs value. This results in [20]

\[
f(x) = 3 \left[ x^{-1} - 2x^{-2} + 2x^{-3}(1 - e^{-x}) \right],
\]

with \( x = R/\lambda \).

This form of \( f(x) \) is certainly an approximation; to
understand its significance we must first make three re-
marks. First, Eq. 1 is qualitatively different from stan-
dard FSS only if \( \lambda \) and \( \xi \) are two different length scales,
otherwise the effect of the border is a mere decoration
of the scaling function and standard FSS remains
unchanged. Second, in the critical region, \( R \sim \xi \), which
follows because the peak position is given by the position \( y_0 \)
of the maximum of \( \tilde{c}(y) \). This follows immediately in the
usual FSS case; in the general case the analysis is slightly
more complicated (see SM [30]), but it remains true that
the peak position and its temperature shift are given by
\( R^{1/\nu}(T - T_c) \approx y_0 \). Third, (at least in the scenarios we
consider), \( \lambda \) diverges at \( T_c \), but not as fast as \( \xi \).

When \( R/\lambda \to \infty \), Eq. 2 gives correctly \( f \to 0 \). When
\( R/\lambda \to 0 \), \( f(x) \) tends unphysically to 1, but in the critical
region \( \lambda \) is large but \( R \sim \xi \), so that \( R/\lambda \ll 1 \). This
means the \( R/\lambda \to 0 \) limit of \( f(R/\lambda) \) is irrelevant in the
critical region. Very near \( T_c \), though, (which is outside

![Graph showing specific heat vs. temperature for amorphous boundary conditions (ABCs). Error bars omitted for clarity; the absolute error is bounded by 0.03. PBCs data are for a system of \( N = 8192 \) particles, where \( C_V \) can be measured down to \( T \approx 0.2 \) (see SM for details).]
In the simplest possible physical scenario we have no transition ($T_c = 0$) and only one length scale, $\lambda \sim \xi$ (i.e. $f \equiv 0$). In this case we are scaling the data as $C_V R^{-\alpha/\nu}$ vs $R^{1/\nu}$. We then need $\alpha \neq 0$ to account for the growth of the peak. Moreover, it seems reasonable to assume that the standard RG scaling relation (Josephson scaling) $\nu d = 2 - \alpha$ [40] holds in this simplest case. We are therefore left with just one parameter, $\alpha$. The result is quite bad and no reasonable collapse is obtained for any value of $\alpha$ (not shown).

Not-so-simple liquid (NSSL). What really seems to resist the scaling of the data in the simplest case is the assumption $T_c = 0$. We therefore relax this hypothesis, assuming that a standard phase transition exists at a finite temperature (standard meaning there is only one length scale, so that $\lambda \sim \xi$, and normal FSS ($f \equiv 0$) applies). One such case is that invoked by Tanaka et al. [6], with Ising-like critical exponents (which thus satisfy the relation $\nu d = 2 - \alpha$). This proposal does not achieve a reasonable collapse, irrespective of the value of $T_c$ (Fig. 2a). Fernández et al. [15] have studied the specific heat of our same system under PBCs and seemed to find a divergence at temperature $T_c = 0.195$, with $\alpha = 0.9$. With these values, however, we fail to obtain a collapse (violating Josephson scaling does not help much either). If, however we leave all three parameters $\alpha$, $\nu$ and $T_c$ free, we get a reasonable collapse for the data above $T_c$ (Fig. 2b).

Mosaic liquid. Now we assume that the penetration length, $\lambda$, and correlation length, $\xi$, grow differently. The increase of the peak for increasing $R$ implies that $\lambda \ll \xi$. This is the only case in which ABCs really have some nontrivial qualitative effect, because this is the only way in which we can achieve a growth of the peak with $\alpha = 0$: in this case, the specific heat has a kink in the bulk limit, rather than a divergence. This is exactly what is supposed to happen in the random first-order theory (RFOT), as well as in some mean-field spin-glass models, in particular the p-spin [18, 41–43]. The RFOT transition is first order in the sense that it has a discontinuous order parameter, but second order in the Ehrenfest sense [42, 43]. Quite generally $\alpha = 0$ in RFOT, as a consequence of the fact that the configurational entropy vanishes at $T_c$, giving a discontinuity of the derivative of the total entropy at the transition [42–44]. There is theoretical [38] as well as numerical [20, 22] evidence that within the RFOT scenario indeed penetration and correlation length are different things and that $\lambda \ll \xi$.

In this scenario both $\xi$ and $\lambda$ diverge at $T_c$, but with different critical exponents. For $\lambda(T)$, the prediction is that $\lambda \sim (T - T_c)^{-1/2}$ in three dimensions [38]. The exponent ruling the $\xi$ divergence is $\nu = 1/(d - \theta)$ [45], where $\theta$ is the stiffness exponent, for which different values have been predicted. Some approximations [45] give $\theta = d/2$, corresponding to $\nu = 2/d$, while others [22, 46] give $\theta = d - 1$, which yields instead $\nu = 1$. In three dimensions, both predictions imply $\nu > 1/2$ and thus $\xi \gg \lambda$ near $T_c$.
To perform the $\lambda \neq \xi$ scaling we need to plot $C_V R^{-\alpha/\nu} |1 - f(\lambda/R)|^{-1}$ vs. $R^{1/\nu}(T - T_c)/T_c$. We use Eq. 2 as an approximation for $f(x)$, but we need also $\lambda(T)$. For this we have taken the data of ref. [20] and fitted them to a power law $a|T - T_c|^{1/2}$ [38], leaving $a$ as fitting parameter but fixing $T_c$ self-consistently to the value that gives the best collapse of $C_V$. The resulting scalings are shown in Fig. 2 (panels c and d). Both have $\alpha = 0$ in accordance to RFOT predictions, with $\lambda(T)$ for $T > T_c$ taken from the power law fit as explained above. $T_c$ is a free parameter, while $\nu$ is fixed to the values 2/3 and 1 according to the different predictions. The RFOT scaling with $\nu = 2/3$ (Fig. 2c) does not give a good collapse of the data, while using $\nu = 1$ does a rather good job (Fig. 2d) for $T > T_c$. For $T < T_c$ we do not have data to fit $\lambda(T)$, hence we have used the same power law with a prefactor $a'$ chosen to give the best collapse. So the $T < T_c$ branch has a better-looking collapse but with two free parameters instead of one.

Though ABCs differ from more usual BCs such as PBCs in that they bring forward the existence of two lengthscales, the critical temperature and exponents are independent of the boundary conditions, since in the $R \to \infty$ limit all observables are independent of the boundary conditions [47] unless control parameters are such that the system is below a thermodynamic transition [48]. It is not possible to perform the same analysis under PBCs in this system, because systems very small or below $T \approx 0.2$ crystallize before $C_V$ can be measured. The values we have been able to obtain are compatible with the NSSL scaling (Fig. 3c), but also with other values (see SM [30] for more details). It is not possible to collapse the PBCs data using Eqs. 1 and 2, since $f(x)$ is constructed specifically for cavities (i.e. frozen boundaries). We do not delve into how the existence of two lengthscales as proposed by RFOT should manifest itself under PBCs; we merely point out that these data do not contradict a scaling with a nonzero critical temperature.

Finally, we have tried a different BC on a cavity, repeating the analysis with random boundary conditions (RBCs). RBCs are the same as ABCs except that the outer (fixed) particles are at random positions. Fig. 3 shows the result of applying the two most successful scalings to the RBCs data. This figure introduces no new parameters: RBCs data are scaled using the same $T_c$, $\lambda(T)$ and exponents adjusted for the ABCs case. Above $T_c$ both sets of data can be scaled with the same parameters, and, at least under RFOT, with the same scaling function. Below $T_c$ (open symbols), the scaling function seems to depend on the boundaries; we note in particular that the RBCs data can be scaled in the RFOT scenario without adjusting the prefactor of the $\lambda(T)$ power law.

In summary, we have studied spherical cavities with amorphous and random BCs. Together with swap MC that can equilibrate small cavities, this has allowed us to do a FSS analysis of the specific heat, which is impossible under PBCs due to crystallization. We have found a peak in $C_V$, which can be scaled under two different scenarios, NSSL and RFOT. The first implies a divergence of $C_V$ in the thermodynamic limit, while the second predicts a discontinuity. Both are of comparable quality, but NSSL has three free parameters, compared to one (two below $T_c$) for RFOT. Within RFOT, only $\nu = 1$ gives a reasonable collapse, suggesting a stiffness exponent $\theta = d - 1$.

We finally emphasize that in all cases our collapse attempts yield a finite $T_c$, of around 0.17. This means that the peak survives the thermodynamic limit. Since in this limit observables must be independent of the boundary conditions (unless there is phase coexistence [47]), this result implies the existence of an anomaly or phase transition in the $R \to \infty$ limit, independently of the particular BCs we have employed. Investigation of more realistic and better glassforming liquids is needed. This will be a challenge, as swap MC is unsuitable for most systems. Nevertheless, these results seem a strong support for thermodynamic theories of the glass transition.

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FIG. 3. Scaling plots for $C_V$ data under random boundary conditions (RBCs) (red squares) and amorphous boundary conditions (ABCs) (black circles) according to the NSSL (panel a) and RFOT (panel b) scenarios. $T_c$, $\alpha$, $\nu$ and $\lambda(T)$ are the same for both sets of data and are those adjusted for ABCs and quoted in Fig. 2. The only difference is that in the RFOT case, we have set $a' = a$ in the $\lambda(T)$ expression to scale RBCs data. Panel c: PBCs data for cubic systems of side $L = 8$ (black crosses), 12.7 (red squares), 20.16 (blue circles), and 32 (purple triangles), scaled with the same exponents and $T_c$ of the NSSL case (panel a).
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