Thermalization and possible signatures of quantum chaos in complex crystalline materials

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Contributed by Aharon Kapitulnik, August 21, 2019 (sent for review June 12, 2019; reviewed by Erez Berg and Subir Sachdev)

Analyses of thermal diffusivity data on complex insulators and on strongly correlated electron systems hosted in similar complex crystal structures suggest that quantum chaos is a good description for thermalization processes in these systems, particularly in the high-temperature regime where the many phonon bands and their interactions dominate the thermal transport. Here we observe that for these systems diffusive thermal transport is controlled by a universal Planckian timescale $\tau \sim h/k_B T$ and a unique velocity $v_F$. Specifically, $v_F \approx v_{ph}$ for complex insulators, and $v_F \lesssim v_p$ in the presence of strongly correlated itinerant electrons ($v_{ph}$ and $v_p$ are the phonon and electron velocities, respectively). For the complex correlated electron systems we further show that charge diffusivity, while also reaching the Planckian relaxation bound, is largely dominated by the Fermi velocity of the electrons, hence suggesting that it is only the thermal (energy) diffusivity that describes chaos diffusivity.

Significance

Quantum chaos has been suggested as a framework to understand transport of charge excitations in strongly correlated electron systems exhibiting the “strange metal” state without long-lived quasiparticle excitations. Identifying a characteristic local scrambling time and a velocity by which perturbation propagates into nonlocal degrees of freedom, a chaos diffusivity is identified and related to the measured charge and energy diffusivities. Here we scrutinize the underlying hypothesis that lattice dynamics can be ignored, particularly at high temperatures, where many phonon bands and their interactions dominate the thermal transport in complex materials. We argue that much of the chaotic behavior, which is also observed in complex insulators, originates from phonons, and in equivalent itinerant systems only the thermal (energy) diffusivity describes chaos diffusivity.

Author contributions: J.Z., E.D.K., K.B., and A.K. designed research; J.Z., E.D.K., K.B., and A.K. performed research; J.Z., E.D.K., K.B., and A.K. analyzed data; and K.B. and A.K. wrote the paper.

Reviewers: E.B., Weizmann Institute of Science; and S.S., Harvard University.

The authors declare no conflict of interest.

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First published September 12, 2019.
that it is the thermal (energy) diffusivity that describes chaos diffusivity.

Observations and Analyses

Overview. Recently, in a comprehensive study of low thermal conductivity insulators, Behnia and Kapitulnik (12) analyzed the magnitude of the thermal diffusion constant in the high-temperature regime where \( T \)-linear thermal resistivity is observed. Possibly the most striking result of this study has been the observation that complex oxides (such as cubic perovskites) exhibit a thermal relaxation time that is of order (but bound from below) of the Planckian relaxation time \( \tau_p = h/k_B T \). More structurally complex insulators such as glasses may come even closer to that bound. Thus, following these observations, we consider real complex material systems such as perovskites where at high temperatures (of order of room temperature or above) a large number of phonon bands are active within each Brillouin zone, many of them dispersive and hence able to carry entropy. Moreover, at the high-temperature range of the experiments anharmonic effects are appreciable, thus strongly influencing the many-body states of the phonon system. It is therefore reasonable to assume that the phonon system creates a chaotic environment, exhibiting levels of statistics that follow Wigner–Dyson statistics (15, 16). If in addition these systems are doped, only a very small number of electron bands (often only 1) cross the Fermi energy to become relevant. This large number of phonon modes in the presence of appreciable electron–phonon interaction causes the electrons to locally relax their energy and momentum.

Complex Insulators. As alluded to above, even without the contribution of electrons, insulating perovskites exhibit anomalous diffusivity that is likely to be described as chaotic. A ubiquitous \( D_{\text{ph}} \propto 1/T \) has been highlighted in recent studies of high-temperature thermal diffusivity of complex insulators, particularly perovskites (20, 24). In general, such a temperature dependence has been attributed to Umklapp scattering of phonons with a scattering rate that decreases as \( \theta_D/T \) (25), commencing at temperatures as low as \( \theta_D/5 \) [here \( \theta_D = \hbar \omega_c/k_B \) is the Debye temperature (26)]. However, for the complex oxides, a \( \sim 1/T \) behavior must have a different origin, since in the relevant temperature range, typically 250 K \( \lesssim T \lesssim 700 \) K, the phonon mean-free path is very small, of order of or even smaller than the lattice constant. Therefore, it was often suggested that Umklapp scattering alone is unlikely to be sufficiently strong to account for the observed small mean-free path (12, 27, 28). Furthermore, due to their structural complexity, complex oxides are susceptible to a variety of local disorder effects, which may further enhance relaxation processes and thus degrade thermal transport. These observations lead to a suggestion that the phonon thermal diffusivity is limited by a Planckian relaxation time \( \tau \sim h/k_B T \) (12, 19, 20). This proposal may first seem at odds with the fact that the temperature range where this behavior occurs is of order of or exceeding the acoustic phonons’ Debye temperature and thus should be considered as “classical.” However, a closer investigation of the phonon band structure of perovskites and similar complex materials shows that new phonon bands, many of them dispersive, continue to be activated much above 1,000 K, where much of the decrease in phonon mean-free path comes from anharmonic and phonon–phonon scattering. For example, \( \text{MgSiO}_3 \) (29) and \( \text{SrTiO}_3 \) (30) exhibit phonon bands all of the way to \( \sim 1.5 \) 500 K, while for \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) phonon bands exceed \( \sim 1,100 \) K (31). By contrast, material systems such as PbTe exhibit very low thermal diffusivity due to their low sound velocity, but are farther away from \( \tau_p \) (12).

Table 1 is a compilation of the linear behavior parameters of the inverse diffusivity of several examples of insulating perovskites compared to well-known highly crystalline “simple” insulators: BeO, silicon, and diamond. Table 1 is constructed from data in the range of 250 K \( \lesssim T \lesssim 700 \) K by assuming a phonon-only thermal diffusivity behavior of

\[
D_k = D_{\text{ph}} = \frac{1}{3} v_{\text{ph}}^2 \tau = s \frac{\hbar v_{\text{ph}}^2}{k_B T}.
\]

Here \( d \) is the relevant dimensionality in the standard expression for the diffusivity, but it is embedded in the numerical constant \( s \) once we impose a Planckian relaxation time \( \tau \sim h/k_B T \). Furthermore, in this expression, \( v_{\text{ph}} \) is taken as the compressional sound velocity with the rationale that much of the heat is transported by the longitudinal acoustic (LA) mode, since it involves excitations of atoms along the direction of heat propagation (similar to ref. 20). Also we add in Table 1 an estimate of the phonon mean-free path, calculated at room temperature from the expression \( D_k = D_{\phi} = \frac{1}{3} v_{\text{ph}}^2 \tau \).

The most important point to note from Table 1 is that complex oxides are very different from the simple, highly crystalline insulators even though the high-temperature diffusivity in both cases is inversely proportional to temperature. Their complex optical phonon spectrum includes numerous phonon bands that strongly interact with the heat-carrying acoustic phonons and result in a complex many-body quantum system that is likely obeying Wigner–Dyson statistics (15, 16). Thus, the observation that they also saturate the thermal relaxation bound at the Planckian rate suggests that they satisfy the conditions to exhibit chaos and quantum thermalization (17). This is an important observation, as we next superimpose the electronic contribution to the thermal transport.

Bad Metals Embedded in Complex Insulators. In the presence of electrons, phonon–electron scattering rate may be the dominating high-temperature cause of the observed short phonon mean-free path in complex correlated materials (27, 48). Indeed, in 2 recent studies of hole-doped (19) and electron-doped (21) cuprates, Zhang et al. (19, 21) have shown that the high-temperature thermal diffusivity of these complex oxides exhibits electronic as much as phononic character. These perovskite material systems are known to have many phonon bands that are excited at temperatures above the resistively determined MIR limit, while also possessing substantial electron–phonon interaction. However, the main discovery of these 2 papers has been

| Sample             | \( v_{\text{ph}} \), \( 10^2 \text{cm/s} \) | \( \tau \) | \( \ell_{\text{ph}}(300 \text{ K}) \), Å |
|--------------------|---------------------------------|--------|-------------------------------|
| \( \text{SrTiO}_3 \) (20) | 7.87                             | 2.7    | 5.1                           |
| \( \text{LaAlO}_3 \) (24, 32) | 6.72                             | 2.9    | 3.86                          |
| \( \text{KTaO}_3 \) (24, 33) | 7.5                              | 3.1    | 5.56                          |
| \( \text{K NbO}_3 \) (24, 34) | 7.0                              | 1.6    | 2.69                          |
| \( \text{NdGaO}_3 \) (24, 35) | 6.5                              | 1.65   | 2.61                          |
| \( \text{YAlO}_3 \) (24, 36) | 8.25                             | 1.8    | 3.54                          |
| \( \text{MgSiO}_3 \) (37, 38) | 8.0                              | 1.05   | 2.1                           |
| Disordered \( \text{SrTiO}_3 \) (24) | 7.87                             | 1.9    | 3.57                          |
| \( \text{GGG} \) (39, 40) | 6.55                             | 2.5    | 3.98                          |
| \( \text{PbWO}_3 \) (41, 42) | 3.47                             | 3.0    | 2.65                          |
| \( \text{BeO} \) (43, 44) | 11.3                             | 11     | 46                           |
| \( \text{Si} \) (20) | 8.43                             | 23     | 202                           |
| Natural diamond (45–47) | 18.0                             | 50     | 55,000                        |

Using known compressional speed of sound for \( v_{\text{ph}} \) \( s \) is determined from \( D_{\text{ph}} = \frac{1}{3} v_{\text{ph}}^2 \tau \). The room-temperature phonon mean-free path is calculated from \( \ell_{\text{ph}} = \frac{D_{\text{ph}}}{\rho_{\text{ph}}} \). Note that for BeO, Si, and diamond, linear inverse diffusivity is observed only above \( \sim 750 \) K.
that over a very wide range of thermal-modulation frequency the data could be fitted with a single diffusivity constant of \( D_{\text{eff}} \) corresponding to the interacting electron–phonon system. Moreover, analyzing the temperature dependence of the thermal diffusivity, Zhang et al. (19, 21) noted that in the case of electron-doped cuprates (\( L_2-x\)-CeO\(_2\)CuO\(_4\), with \( L = \text{Nd}, \text{Sm}, \text{and Pr} \)), the measured inverse thermal diffusivities at high temperatures are linear in temperature, satisfying

\[
D_{\text{eff}}^{-1} = \left( \frac{\hbar v_s^2}{k_B T} \right)^{-1} + D_0^{-1},
\]

where \( s \) plays the same role as in the pure phonon case. The \( T \) linearity is directly attributed to a Planckian relaxation rate \( \tau = h/k_BT \), and the slope of the linear term gives rise to a velocity \( v_s \), which was interpreted as the velocity of an incoherent mixture ("soup") of electrons and phonons. The difference between Eq. 2 and Eq. 1 is the \( T \to 0 \) extrapolation which is close to 0 for the insulators, but finite for the systems that include electrons. The constant term \( D_0^{-1} = \lim_{T \to 0} D_{\text{eff}}^{-1} = (h/3m^*)^{-1} \) was interpreted as the residual electron diffusivity contribution at the MIR limit (21). The presence of electrons must imply that \( s \), or the velocity, or both need to be modified to include the electrons. There are 3 possible scenarios for this case: 1) We assume that the velocity remains the sound velocity. In that case we can show that in the range of materials we measured \( s \) must increases 4 to 6 times from its present value, implying an increasing mean-free path. This does not make sense since the presence of electrons tends to increase the phonon relaxation through electron–phonon interaction and hence is expected to reduce \( s \). Furthermore, from our YBCO study we know that the thermal diffusivity follows the resistivity and not the sound-velocity anisotropy and thus should have an implicit character of the electron contribution. 2) We can assume that the electrons indeed push the relaxation time to the Planckian limit with \( s = 1 \). While this is certainly possible, and would yield \( E_g \) much larger than \( v_s \), we choose to be more conservative and choose the following: 3) We assume that since the system is dominated by phonons, similar to the parent insulator, only the velocity is changing, taking \( s \approx 2.0 \) calculated as an average value of many different perovskites (e.g., the average of the systems in Table 1). Thus, determining \( s, v_E \) can be calculated using Eq. 2. Table 2 is a summary of results for a few materials reported in that study (21).

The most important feature in Table 2 is that \( v_E \gtrsim v_{\text{phon}} \) by roughly a factor of 2, which translates to a factor of 4 in the diffusivity slope (although, as discussed above, the difference can be larger if \( s \) is closer to unity). Furthermore, note that the extracted \( \ell_{\text{eff}} \) in Table 2 is larger than for the pure insulators summarized in Table 1. For example, NdGaO\(_3\) exhibits similar sound velocity, but much smaller extracted phonon mean free path than any of the LCCO samples. This can be understood as a result of the contribution of the electrons to the thermal transport, which is another reason to argue for their role in energy transport even at these high temperatures.

To end the discussion on thermal diffusivity, we show in Fig. 1 a log-log plot of the diffusivity vs. temperature above 100 K for several insulating samples compared to Nd\(_{1.85}\)Ce\(_{0.15}\)CuO\(_4\) (NCCO). First we note that silicon as a highly crystalline material approaches \( D_g \sim T^{-1} \) at high temperatures, but this can be attributed to standard Umklapp scattering (53). On the other hand, YAlO\(_3\) and NdGaO\(_3\) are complex insulating materials with noncubic crystal structure similar to NCCO, with diffusivity that follows Eq. 1, and with a typical proportionality constant \( s \) (Table 1). The NCCO crystal which is also a complex perovskite with a very similar sound velocity shows also linear dependence of the inverse diffusivity, but with a finite intercept. Subtracting that intercept, its linear dependence on the same log-log plot is evident. Also evident is the higher slope, by about a factor of 2 to 3 over the insulators. Keeping the prefactor constant (see above), this is a manifestation of the higher velocity extracted from the slope. To emphasize the range of phonon bands in these materials, we note, e.g., that for Nd\(_{2}\)CuO\(_4\) (54) (the parent compound of NCCO) or YAlO\(_3\) (55), room temperature lies low within the rather wide (extending much above 1,000 K) complex band structure of the phonons, some of which are strongly dispersive.

**Charge Diffusivity and Resistivity.** In the presence of itinerant electrons, such as in the aforementioned strongly correlated systems, we can discuss separately the issue of charge transport and its relation to the MIR limit. For the strongly correlated hole- or electron-doped cuprates studied by Zhang et al. (19, 21), it was argued that both charge and lattice degrees of freedom are strongly scattered, and hence the scattering rate is pushed to the Planckian bound (56). Applying a temperature gradient to the sample, that system of electrons and phonons reacts together, and heat is flowing as an overdamped diffusing soup of phonons and electrons. While the very high density of phonons dominates the thermal transport, the electrons with higher velocity but the
same scattering rate are able to increase the effective velocity of that fluid.

Considering next charge transport, an electric field applied to the system will cause charges to move, but will influence the phonon system only indirectly through electron–phonon interaction. This in turn may result only in some renormalization of the electron’s effective mass, but will not change the velocity that determines the charge diffusivity, that is, the Fermi velocity, by much. Therefore, establishing that \( \nu_{ph} \lesssim v_B \ll \nu_{e} \), we conclude that in general \( D_C \gtrsim D_E \).

To further explore the above idea, we compare resistivity and thermal diffusivity data, particularly on materials that show \( D_C \sim \propto T \). Using the Einstein relation for the resistivity, the charge diffusivity is given by \( D_C = [\rho^E (dn/dp)]^{-1} \). If the relaxation time is bound at \( \tau \sim h/k_B T \), the resistivity will show \( \rho \propto T \), and if the density of states is known, the velocity associated with the charge diffusivity can be calculated. It is evident from the resistivity data compiled by Bruijn et al. (57) that the extracted velocity associated with \( D_C \) is approximately the Fermi velocity. One concrete example is \( B_2S_3 \) which is a hole-doped cuprate that is less susceptible to oxygen loss and thus was measured to temperatures much above room temperature. In particular, for near-optimal doping a linear dependence of the resistance and inverse thermal diffusivity have been observed. While the thermal diffusivity yields a velocity \( v_B \approx 7.4 \times 10^5 \text{ cm/s} \), analysis of the linear resistivity on very similar crystals (58) yields a velocity very close to the known Fermi velocity for this system, \( v_F \approx 2.8 \times 10^5 \text{ cm/s} \) (59).

A more striking difference between electron and charge diffusions is observed in many of the electron-doped cuprates where above the MIR limit the inverse thermal diffusivity is linear in temperature (21), while the resistivity behaves as \( \rho \propto T^{1+y} \), with \( y \gtrsim 0 \) (60–62). Imposing a Planckian relaxation bound suggests that in these materials the electronic compressibility varies as well above the MIR limit as has been recently proposed in refs. 63 and 64.

**Discussion**

In a recent publication Werman et al. (23) introduced the idea that strongly coupled incoherent bad metals are also strongly chaotic. In their model an electron system interacts with a parametrically much larger system of optical phonons via local electron–phonon interaction. The root-mean-squared phonon frequency and velocity averaged over all phonon bands were set to be \( \omega_0 \) and \( \nu_0 \), respectively, while similarly, the root-mean-squared Fermi velocity averaged over all electron bands was set to be \( v_F \). Their calculations, which were performed for the cases of dispersive and dispersionless phonons, concentrated on the temperature regime where \( \omega_0 \ll k_B T \ll \epsilon_F \), in which electronic quasiparticles are no longer well defined. With the above assumptions, their primary result was that the thermal and chaos diffusivity constants \( D_B \) and \( D_C \) are always comparable \( D_B \approx D_C \), while this is not necessarily the case for charge diffusion. More recently similar results were also obtained by Guo et al. (65), who introduced to the aforementioned Sachdev–Ye–Kitaev (SYK) model (11), in addition to the electronic coupling of the SYK islands, also a local coupling of each island to a large number of low-energy “well-defined” phonons.

**Quantum Chaos and Thermalization.** A closed, many-body interacting quantum system without well-defined quasiparticle excitations is argued to exhibit quantum chaos (6). In this scenario local perturbations in the system’s initial conditions are “scrambled” exponentially into nonlocal degrees of freedom with a Lyapunov exponent rate \( \Gamma = 1/\tau_{\chi} \) and spread out to affect the whole system at the butterfly velocity \( v_B \) (1, 5–7, 10). Consider a perturbed subsystem of a larger many-body interacting quantum system. It is then possible to assign a thermal distribution to the subsystem, while the rest of the system serves as a heat bath that allows the subsystem to thermalize. Scrambling and the butterfly velocity then describe the initial amplification of local perturbations and the spread of chaos through quantum entanglement and the loss of memory of the initial state of the whole system. Macroscopic diffusion that originated from the microscopic scrambling processes emerges, with a diffusion constant

\[
D_{\chi} \approx v_B^2 \tau_{\chi}
\]

that controls global thermalization. Fig. 2 shows a cartoon of this sequence, which was further argued to describe charge and energy diffusion (5, 6, 8).

**Quantum Chaos and Solid-State Systems.** While it is reasonable to expect quantum chaos in strongly interacting electron systems, its observation, as well as that of a scrambling time, is generally not expected to be measurable in solid-state systems. However, it was recently proposed that either the charge diffusivity \( D_C \) (5, 66) or energy diffusivity \( D_E \) (7), or both (25), is related to \( D_{\chi} \). These connections followed a proposal by Hartnoll (4) that for incoherent nonquasiparticle transport, both diffusivities are subject to a fundamental quantum mechanical bound of \( D \gtrsim \hbar v_B^2 / k_B T \), where the Fermi velocity was introduced as the natural velocity for the problem of electronic transport in a metal, and the relaxation time was set at the shortest timescale allowed by the uncertainty principle \( \tau \sim h/k_B T \) (67). An immediate consequence of this approach is that for a constant electronic compressibility, the resistivity is linear in temperature, which has been ubiquitously observed in many bad metals (57).

While the above proposal seems to work for charge diffusivity \( (D_C) \), it was further suggested that the above bound applies to energy diffusivity \( D_E \) and that \( D_E \sim D_C \sim D_{\chi} \), with the proviso that electron–phonon interaction can be neglected. However, in calculations of charge and energy diffusivities in specific models of metals without quasiparticles, a critical Fermi surface (7), and the SYK model (11), it was found that “quantum chaos, as characterized by the butterfly velocity and the Lyapunov rate, universally determines the thermal diffusivity, but not the charge diffusivity” (11). This seems natural in the context of the studied models since quantum chaos originates from local energy fluctuations.

Obviously, an interacting many-electron system in an itinerant solid is not in general a closed quantum system, since at least phonons need to be incorporated into the problem. Moreover, any experiment on a solid-state system in which the temperature is well defined implies a connection to a heat bath which in general is dominated by the phonons. Thermalizing the system is a
delicate balance between internal electron–phonon interaction and connection to the environment through, e.g., the electrical leads. Thus, even if we invoke a saturation of the relaxation time at \( h/k_B T \), the notion of a unique velocity that governs scrambling of local perturbations into nonlocal degrees of freedom, that is, the “spread of chaos,” may not be applicable to solid-state systems. However, we can still use the concept of quantum chaos in situations that fulfill certain conditions. In particular, a state that at least mimics a quantum chaotic system may be realized in a regime where the full electron–phonon system includes electron–phonon interactions and also exhibits strong momentum degradation.

**Comparison to Experiments.** We return now to the comparison of our experimental observations in perovskites and other bad metals and complex insulators to the model of Werman et al. (23). In that model, strongly renormalized electrons are coupled to weakly interacting optical phonons, which are well-defined quasiparticles. This yields an average phonon frequency \( \langle \omega_{ph} \rangle \) \( \sim \omega_0 \ll k_B T \), and thus \( \tau_c \sim h/k_B T / \hbar \omega_0 \), which is far from the Planckian bound, thus should be considered only as “weakly” chaotic. Scrambling rate is therefore determined by the loss of phonon phase coherence, and the butterfly velocity of the electron–phonon system with dispersive phonons depends only on the phonons’ velocity. There are 2 main differences between the theoretical model and the conditions of the experimental data. First, concerning the phonon system, since the temperature range of interest lies in the middle of the complex phonon band structure, which also exhibits strong phonon–phonon interactions, we cannot assume that all phonons are well-defined quasiparticles, nor can we implement the theoretical assumption of \( k_B T \gg \omega_0 \). In fact, we argue that the complexity of the phonon system in the studied material systems implies phonon states that most likely exhibit chaotic statistics (15, 16). The magnitude of the measured diffusivities suggests that much of the heat is transported by dispersive phonon modes, most probably involving excursions of atoms along the direction of heat propagation, and the relaxation reaches \( \tau_c \sim h/k_B T \). Since in experiments \( k_B T \) determines both the excited phonon modes and relaxation rate, it is tempting to push the theoretical model to that limit, requiring that the average phonon frequency is determined by temperature, that is, \( \hbar \omega_0 \rightarrow k_B T \), which immediately implies \( \tau_c \sim h/k_B T \). This assignment also suggests that with no charge carriers, the velocity associated with the diffusivity must be similar to the phonon velocity, which we summarized in Table 1, yielding \( v_{ph} \sim v_{ph} \).

Adding the effect of the electrons for the further comparison with ref. 23, the fact that a single diffusion constant is observed suggests an increase in butterfly velocity \( v_{ph} \sim \sqrt{E \sum \omega_{ph}^2} \), but probably smaller than \( v_{ph} \). This again is understood as a consequence of the strongly interacting electron–phonon system, which diffuses as a soup of not well-defined quasiparticles (19), with a diffusion constant that is characterized by a relaxation time saturated at the Planckian time of \( h/k_B T \) and a velocity \( v_{ph} \gg v_{ph} \) due to the contribution of the much faster electrons. Indeed, Guo et al. (65) report that a more self-consistent inclusion of the electron–phonon and phonon–phonon interactions is expected to yield an “electron–phonon” soup, in which the strong dependence on the electron–phonon coupling disappears.

**Final Remarks.** There are 2 final remarks to make. First, it would be desirable to be able to separate the values of \( v_{ph} \) and \( s \) in Eq. 2. This may require measurements at shorter timescales to locate the actual time at which electrons thermalize with phonons. Second, we remark on the relationship between \( D_c \) and \( D_\alpha \). For energy diffusion, both electrons and phonons move along the temperature gradient, and for an overwhelming phonon system the electrons are a small contribution to the combined soup, providing enhanced velocity. On the other hand, an electric field will act only on the charges, which will move close to the Fermi velocity as conjectured in ref. 4. The phonons will then work to impede that motion through the interaction with the charges. Therefore, and in particular based on our experimental observations, we do not see a reason why \( D_c \sim D_\alpha \sim D_\phi \).

**Materials and Methods**

Data used in this paper were taken from published literature as indicated in the relevant references. Properties used to establish various claims have been calculated as described in the text.

**ACKNOWLEDGMENTS.** We thank Steve Kivelson, Subir Sachdev, and particularly Sean Hartnoll for many comments and insightful discussions. This work was supported by the Gordon and Betty Moore Foundation through Emergent Phenomena in Quantum Systems Initiative Grant GBMF45292 and by the US Department of Energy Office of Basic Energy Science, Division of Materials Science and Engineering at Stanford University under Contract DE-AC02-76SF00515.

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