On the relation between disorder and homogeneity in an amorphous metal

Z. Ovadyahu

Racah Institute of Physics, The Hebrew University, Jerusalem 9190401, Israel

Abstract

Disorder and homogeneity are two concepts that refer to spatial variation of the system potential. In condensed-matter systems disorder is typically divided into two types: those with local parameters varying from site to site (diagonal disorder) and those characterized by random transfer-integral values (off-diagonal disorder). Amorphous systems in particular exhibit off-diagonal disorder due to random positions of their constituents. In real systems diagonal and off-diagonal disorder may be interconnected. The formal depiction of disorder as local deviations from a common value focuses attention on the short-range components of the potential-landscape. However, long range potential fluctuation are quite common in real systems. In this work we seek to find a correlation between disorder and homogeneity using amorphous indium-oxide ($\text{In}_x\text{O}$) films with different carrier-concentrations and with different degree of disorder. Thermal treatment is used as a means of fine tuning the system disorder. In this process the resistance of the sample decreases while its amorphous structure and chemical composition is preserved. The reduced resistivity affects the Ioffe-Regel parameter $k_F\ell$ that is taken as a relative measure of disorder in a given sample. The homogeneity of the system was monitored using inelastic light-scattering. This is based on collecting the Raman signal from micron-size spots across the sample. The statistics of these low-energy data are compared with the sample disorder independently estimated from transport measurements. The analysis establishes that heterogeneity and disorder are correlated.

INTRODUCTION

Disorder plays a major role in the properties of condensed-matter systems [1–9]. The experimental study of disorder-induced phenomena present a challenge in terms of being able to control, characterize, and quantify it. Effort in this vein was mainly invested in the field of electronic transport. In particular, this issue has been a major concern in studies of the metal-insulator transition (MIT) and the superconductor-insulator transition (SIT). The system conductivity is frequently used as an empirical measure of disorder in these studies. The conductivity of a solid is arguably its most sensitive property and it may be affected by different means, not all of them may be attributed to disorder. A change in carrier-concentration for example, would affect the conductivity while causing only a small change in the disorder.

An aspect of disorder frequently referred to but rarely actually measured is homogeneity. It should be realized that the two concepts are related; spatial disorder is tantamount to inhomogeneous. Strong disorder, such as required to Anderson-localize a system with a relatively high carrier-concentration, is naturally accompanied by significant spatial variations of the potential. We shall refer to this type of inhomogeneity as ‘inherent’. A system may however be inhomogeneous with little or no short-range potential-fluctuations being present. This form of ‘technological’ inhomogeneity [10, 11] may lead to non-trivial transport effects in otherwise ‘clean’ systems. In principle, the two types of inhomogeneities may be told apart; changing the disorder should show a concomitant change of inhomogeneity if the disorder is inherent.

As a relevant example, we present in this note experiments to study the relation between disorder and spatial inhomogeneity in amorphous indium-oxide ($\text{In}_x\text{O}$) films. Using a purely amorphous system for this project is natural; the only structurally imposed length-scale is the nearest-neighbor distance which turns out to be much smaller than any dimension relevant to the problem. This research focused on the version of the material that is relatively rich in carrier-concentration. This version of $\text{In}_x\text{O}$ has been extensively studied and its transport parameters are well known [12, 13]. In particular, near the critical point of the MIT the magnitude of the disorder may be quantitatively determined from the condition for the Anderson transition. The heterogeneity at this disorder is compared with another version of $\text{In}_x\text{O}$ that topologically is nearly identical while much less disordered. Subjecting both systems to thermal treatment that modifies their resistivities allows for another way to test the effect of disorder on system homogeneity. The sample disorder before and after heat treatment is characterized by the Ioffe-Regel parameter $k_F\ell$. Low-energy Raman-spectroscopy is employed to probe the system spatial homogeneity in each case for tracking the change associated with the modified disorder. The results support the expected notion that heterogeneity is an inherent property of the disordered system.

EXPERIMENTAL

Samples preparation and measurements techniques

The $\text{In}_x\text{O}$ films used in this work were e-gun evaporated onto room-temperature substrates using 99.999% pure $\text{In}_2\text{O}_3$ sputtering-target. Deposition was carried out at the ambience pressure of $3\pm 0.5\times 10^{-5}$ to $4\pm 0.5\times 10^{-4}$ Torr oxygen-pressure maintained by leaking 99.9% pure $\text{O}_2$ through a needle valve into the vacuum chamber (base pressure $\simeq 10^{-6}$ Torr). Undoped silicon wafers with (100)
orientation were used as substrates for electrical and Raman measurements. Carbon-coated copper grids were used for transmission electron-diffraction. During film deposition, the grids were anchored to 1mm glass-slides by small indium pellets later removed for mounting the grids in the microscope. The deposited film on the rest of the slide was used for monitoring the sample resistance for comparison. X-ray interferometry was performed on samples deposited on a 2.8mm float-glass.

Rates of deposition in the range 0.3-2.5 Å/s were used to produce films with different compositions; The \( In_2O \) samples had carrier-concentration \( N \) that increases with the ratio of deposition-rate to the oxygen-partial-pressure. For the rates_pressures used here \( N \) was in the range \( 1x10^{19} \text{cm}^{-3} \) to \( 9x10^{20} \text{cm}^{-3} \) as measured by Hall effect at room-temperature using a Hall-bar control-sample prepared simultaneously for each sample deposition. Lateral sizes of samples used for transmission electron-diffraction measurements was typically \( 1x2.5 \text{mm}^2 \) (width x length respectively), and \( 1x1 \text{cm}^2 \) for the Raman scattering experiments. The evaporation source to substrate distance in the deposition chamber was 45±1mm. Therefore, thickness variations across the samples are unlikely to be related to geometrical consideration in the deposition process.

To afford reasonable resolution for electron-microscopy thickness of the films used for the electron-microscope work was \( d=20 \pm 1 \text{nm} \).

Two batches of \( In_2O \) with carrier-concentrations of \( N=(1.5 \pm 1)x10^{19} \text{cm}^{-3} \), and \( N=(8 \pm 1)x10^{20} \text{cm}^{-3} \) were deposited for the Raman scattering experiments. In the following these will be referred to as "low-\( N \)" and "high-\( N \)" versions of \( In_2O \) respectively. The high-\( N \) version may be also referred to in this work as the "indium-rich" phase of the compound. These version of \( In_2O \) are representative for the substance used in recent electron-glass experiments with typically \( N \approx 10^{19} \text{cm}^{-3} \) and the \( In_2O \) version with \( N \approx 10^{20} \text{cm}^{-3} \) commonly used in superconductor-insulator studies [14-22].

The Ioffe-Regel parameter \( k_F \ell = (9 \pi^4 / N)^{1/3} \rho_{RT} / R_Q \) where \( R_Q=h/e^2 \) is the resistance quantum and \( \rho_{RT} \) is the resistivity at room-temperature, is used here as a measure of the sample relative disorder. This parameter is monotonic with disorder, and it is well defined experimentally. It is descriptive of the static disorder even when its value is smaller than unity where neither \( k_F \) nor \( \ell \) are "good" parameters (which is the case in all our "as-made" samples). The important caveat that should be borne in mind in this regard is that when \( k_F \ell \ll 1 \), \( \ell \) must not be interpreted as a semiclassical mean-free-path. At room temperatures, where our measurements are performed, it is safe to say that the transport length-scale (whether it is the hopping-length or the mean-free-path) is much smaller than the size of the region probed by the laser beam.

Sample homogeneity on a micron-scale was characterized by the intensity variations of the Raman signal over the energy range 70-420cm\(^{-1} \). These were carried for \( In_2O \) samples with different \( N \) as well as for the as-made and the annealed samples where disorder was modified by thermal-treatment.

Electron diffraction patterns were taken with the Philips Tecnai F20 G2 operating at 200kV. The Raman spectra were taken with a Renishaw inVia Reflex Spectrometer using a laser beam with a wavelength of 514 nm and edge-filter at \( \approx 70 \text{cm}^{-1} \). These measurements employed beam-spots of 2μm and 5μm. In each case, fifty spectra were taken at different spots across a rectangle area of the sample for obtaining statistics of the Raman signal magnitude. The laser intensity was checked to be in the linear response and low enough to avoid structural changes during exposure [23]. During these preliminary tests, the same spot on the sample was measured several times. Traces were taken from this spot with different laser intensities. The spot could be identified in the microscope as it changed color after several exposures. However, the Raman traces taken from it registered the same spectrum over the range studied with intensity variations of typically ±0.5%. Therefore the variations of signal intensity reported below are mostly due to inhomogeneities rather than due to time dependence.

The deposited samples, after being characterized, were heat treated to modify their disorder at \( T=360 \pm 5 \text{K} \) for 24-48 hours. Fuller details related to heat treatment of \( In_2O \) films are described elsewhere [24].

**RESULTS AND DISCUSSION**

Local Raman-spectra for the high-\( N \) version \( In_2O \) film are shown in Fig. 1 for the as-made, indium-rich sample. As mentioned above, this version of \( In_2O \) has been often used for the disorder-driven SIT. This transition occurs when the Ioffe-Regel parameter \( k_F \ell \) of the system is 0.29-0.32 [23]. Figure 2 shows the respective set of data for the same sample after it has been heat-treated and its \( k_F \ell \) value has increased from 0.1 to 0.42. The data in Fig. 1 and Fig. 2 pertain therefore to the insulating and superconducting side of the SIT respectively.

Two features stand out in these data: First, the local intensity of the Raman signal fluctuates considerably from point to point (Fig. 1a and Fig. 1c) manifesting the spatial heterogeneity of the sample. Secondly, homogeneity of the sample visibly improves after heat-treatment (Fig. 2a and Fig. 2c). The sample heterogeneity parameter \( \gamma \) is defined here as a ratio: \( \gamma=\text{[standard-deviation]/[mean]} \) of a normal-distribution fitted to the intensity-histogram. The use of normal distribution to fit the histogram data is just to allow an impartial estimate of the sample heterogeneity. To determine the precise functional form of the distribution would require many more data plots than used here. Nonetheless, the ob-
tained data are consistent and systematic enough to test
the basic conjecture concerning the disorder-homogeneity
relation. Note that \( \gamma \) is smaller for the annealed sample,
and it is smaller for the larger spot-size as might be ex-
pected from the better ensemble averaging.

Before proceeding, the natural question is what is re-
flected by the magnitude of the Raman signal?

At the energy range studied \( \varepsilon = 70-420 \text{ cm}^{-1} \), the signal
is composed of two components associated with differ-
ent inelastic light-scattering mechanisms. For energies
\( \varepsilon \leq 250 \text{ cm}^{-1} \), the main contribution comes from the boson
peak of the amorphous material. This feature, re-
lated to the phonon system of In\(_2\)O\(_3\), has been studied pre-
viously as function of disorder and composition [23]. The
other component is presumably inelastic scattering from
the electronic states, either extended or weakly-localized
in the presence of quenched disorder.

To illustrate the relevance of this electronic mechanism
for this material, consider the Raman data shown in Fig.
3. Note the different spectra for the two polycrystalline
versions of indium-oxide. These samples share the same
crystal structure with a body-centered cubic symmetry
[33]. The grain-size of the In\(_2\)O\(_3\) may be somewhat larger
than in the deposited film In\(_2\)O\(_3\), but this cannot explain
the grossly different Raman spectra. A likely relevant
difference between the compounds is their chemical sto-
ichiometry; the vacuum-deposited film In\(_2\)O\(_{3-x}\) is 8-10%
oxygen deficient [33] that, in turn, renders the system metallic with carrier-concentration \( N \approx 5 \times 10^{19} \text{ cm}^{-3} \). The
In\(_2\)O\(_3\) sample on the other hand is actually an intrinsic
semiconductor with \( N \approx 10^{16} \text{ cm}^{-3} \) as measured by Hall
effect at room-temperature. It is then plausible to asso-
ciate the distinctive spectrum of the In\(_2\)O\(_{3-x}\) with an
electronic mechanism for inelastic light-scattering [34],
similar to the findings in underdoped cuprate [35].

Evidently, the electronic contribution to the Raman
signal is substantial. It overwhelms the Raman-active
material vibration-modes and those that appear are
shifted in energy from the positions of their stoichiomet-
ic compound.

Note that the prominent boson peak portrayed by the
amorphous sample is absent (or is very weak) in either
crystalline specimen. This is a demonstration of the dif-
ference sensitivity of phonons and electrons to different
types of disorder; the grain-size in both, In\(_2\)O\(_{3-x}\) films and
In\(_2\)O\(_3\) sputtering-target pieces is at least 0.5 \( \mu \text{m} \), much
larger than the mean-free-path for electrons in samples
where \( k_F \ell < 1 \). The large wavelength of phonons makes
them less sensitive to the short-range fluctuations of
the chemical composition which strongly affects charge
carriers. Hence the effective Ioffe-Regel parameter for
phonons \( (k_F \ell)_{\text{ph}} \) may be larger than unity, thus unfavor-

FIG. 1: Raman spectra for the as-made In\(_2\)O sample with
\( N \approx 8 \times 10^{20} \text{ cm}^{-3} \). The sheet-resistance of the film
\( R_{\square} = 18.3 \text{ k}\( \Omega \) at room-temperature which yields
\( k_F \ell = 0.1 \). (a) and (c) show the Raman shift versus energy for fifty different local
sampling-points using 2 \( \mu \text{m} \) and 5 \( \mu \text{m} \) spot-size respectively.
The black line is the average of the 50 plots. (b) and (d)
exhibit the histogram of the Raman intensity-plots averaged
over the interval 350-400 \( \text{cm}^{-1} \) for each of the local readings
related to the 2 \( \mu \text{m} \) and 5 \( \mu \text{m} \) spot-size respectively. The his-
tograms in (b) and (d) are fitted to normal distributions de-
picted as dashed lines. These fits yield heterogenous param-
eters (see text for definition) \( \gamma = 9.7\% \) for the 2 \( \mu \text{m} \) spot-size
and \( \gamma = 5.1\% \) for the 5 \( \mu \text{m} \) spot-size.

FIG. 2: Raman spectra for the In\(_2\)O sample with
\( N \approx 8 \times 10^{20} \text{ cm}^{-3} \) after being heat-treated at \( \approx 380 \text{K} \) for \( \approx 48 \text{ hours} \). The sheet-resistance of the film changed to:
\( R_{\square} = 3.95 \text{ k}\( \Omega \) which gives \( k_F \ell = 0.42 \). (a) and (c) show the Raman
shift versus energy for fifty different local sampling-points
using 2 \( \mu \text{m} \) and 5 \( \mu \text{m} \) spot-size respectively. The black line is the average
of the 50 plots. (b) and (d) show the histogram of the Raman intensity-plots averaged
over the interval 350-400 \( \text{cm}^{-1} \) for each of the local readings related to the 2 \( \mu \text{m} \) and
5 \( \mu \text{m} \) spot-size respectively. The histograms in (b) and (d)
are fitted to normal distributions depicted as dashed lines.
These fits yield heterogenous parameters (see text for defi-
nition) \( \gamma = 4.9\% \) (down from \( \gamma = 9.7\% \) in the as-made sample)
for the 2 \( \mu \text{m} \) spot-size and \( \gamma = 2.5\% \) (down from \( \gamma = 5.1\% \) in the
as-made sample) for the 5 \( \mu \text{m} \) spot-size.
The concentration as demonstrated in high-temperature and low-frequency radiation depends mainly on the charge-distribution of the sample and is still appreciable. This aspect has consequences for the size areas with carrier-concentrations that differ by 20-30% as shown in Fig. 1b and Fig. 2b. Moreover, it is evident from the distribution in Fig. 2b that the probability to find micron-size areas with carrier-concentrations that differ by 20-30% is appreciable. This aspect has consequences for the inherent aspect of disorder in these In$_x$O films. Table 1 summarizes the values of $\gamma$ for the respective samples and states.

It is remarkable that significant fluctuations of carrier-concentration are observable over a scale of microns in the high-N sample; $\gamma$ for the 2$\mu$m area is $\approx$10% in the as-made sample and is still $\approx$5% after being annealed (Fig. 2b). Moreover, it is evident from the distribution in Fig. 2b that the probability to find micron-size areas with carrier-concentrations that differ by 20-30% is appreciable. This aspect has consequences for the low temperature transport in such systems in particular, when the observed property is sensitive to the carrier-concentration (for example; superconductivity). Indeed, high-N versions of In$_x$O exhibit anomalous transport effects near their SIT that are ascribed to superconducting islands embedded in an insulating matrix reflecting areas where $N$ is larger than the average value of the sample. Similar emergence of disorder induced "granularity" was found in other metal-oxides like high-T$_C$ compounds.

It is illuminating to compare the before-and-after change in $\gamma$ for this $N$=8x10$^{20}$cm$^{-3}$ version of the material (Fig. 1 and Fig. 2) with the respective behavior in the low-N version sample shown in Fig. 4 and Fig. 5 respectively.

Just by eyeing the raw data (compare Fig. 2 and Fig. 4) it is clear that even the as-made low-N version of In$_x$O is more homogenous than the heat-treated specimen of the indium-rich material. It is gratifying to see that the respective values of $\gamma$ are consistent with this observation. This is in line with the notion that heterogeneity is an inherent aspect of disorder in these In$_x$O films.
Heat-treated ∆(k_Fℓ) parameter for two batches of In_xO samples labeled by their respective N. Dashed lines are guides to the eye. Note the much steeper dependence of E_g versus a change in k_Fℓ for the sample with the larger N.

![Raman spectra for the In_xO sample](image)

FIG. 5: Raman spectra for the In_xO sample with N≈10^{19} cm^{-3} after being heat-treated at ~380K for ~48 hours. The sheet-resistance of the film changed to R_{∥} = 23kΩ which gives k_Fℓ≈0.29. (a) and (c) show the Raman shift versus energy for fifty different local sampling-points using 2µm and 5µm spot-size respectively. The black line is the average of the 50 plots. (b) and (d) show the histogram of the Raman intensity-plots averaged over the interval 350-400 cm^{-1} for each of the local readings related to the 2µm and 5µm spot-size respectively. The histograms in (b) and (d) are fitted to normal distributions depicted as dashed lines. These fits yield heterogenous parameters (see text for definition) γ = 2.8% (compared with γ = 2.7% in the as-made sample) for the 2µm spot-size and γ = 1.6% (down from γ = 2.1% in the as-made sample) for the 5µm spot-size.

In particular, the steeper ∂γ/Δ(k_Fℓ) in the indium-rich version, we need to expound on the connection between disorder and k_Fℓ.

| Spot-size | As-made γ (%) | Heat-treated γ (%) | ∆(k_Fℓ) |
|-----------|---------------|--------------------|---------|
| High-N    | 9.7±0.5       | 5.1±0.5            | 4.9±0.5 | 2.5±0.5 | 0.32 |
| Low-N     | 2.7±0.4       | 2.1±0.4            | 2.8±0.4 | 1.6±0.4 | 0.29 |

Table 1: The inhomogeneous parameter γ (defined in the text), and the change of the sample Ioffe-Regel parameter ∆(k_Fℓ) by heat treatment for the two studied samples. The carrier concentration for the high-N and low-N samples are N = (8±1)×10^{20} cm^{-3} and N = (1.5±1)×10^{19} cm^{-3} respectively.

In the context of transport, one usually weighs disorder on an energy scale while k_Fℓ is dimensionless. A quantitative estimate of disorder is therefore limited to special values of k_Fℓ such as its value at the Anderson MIT where the disorder-energy may be assessed with respect to E_F [42]. This means that, without specific information on auxiliary properties of a given system, k_Fℓ is merely a relative measure of disorder.

Moreover, k_Fℓ may be relied on to be indicative of quenched-disorder when \( \rho_{RT} \) is strictly determined by elastic scattering, a condition that may be obeyed for k_Fℓ values at the vicinity of the MIT. The critical k_Fℓ in In_xO occurs at k_Fℓ=0.31±0.2 essentially independent of the carrier-concentration [25] presumably because the disorder in these samples is always larger than the interaction [43, 44]. On the other hand, versions of this material with different N have different degrees of disorder for the same value of k_Fℓ. In particular, the disorder that brings the system near the Anderson transition, is greater the larger N is. Recall that the critical disorder is associated with potential-fluctuations that are large enough to overcome the Fermi-energy E_F [42] that in turn increases with the carrier-concentration of the system.

For the same reason, for higher N, a larger degree of disorder is required to achieve a given change in k_Fℓ. This may be illustrated by observing the change in the material optical properties accompanying a reduction of resistivity following heat-treatment. Figure 6 shows how the optical-gap E_g changes after several heat-treatments [45] that progressively increase k_Fℓ by reducing \( \rho_{RT} \). E_g signifies the energy scale for transitions between the valence and the conduction bands of In_xO. The figure compares E_g vs. k_Fℓ dependence for two versions of In_xO with different carrier-concentrations. A change of E_g is a consequence of the system densification [24]. Rearrangement of oxygen vacancies take place concomitantly. Both processes reduce the system disorder. Figure 6 shows that a larger degree of structural change, reflected by \( \Delta E_{g} \), is required to induce a similar change in k_Fℓ for the high-N version of In_xO. This is an example of a relation between the energy change required to affect ∆(k_Fℓ) in a specific system.

The two versions of the material differ by their chemical composition, explicitly, by their O/In ratio. For the
higher sensitivity of the conductance of the low-N carrier-concentration than the low-N version (the “Hooge law” [47]). This means that their Fermi-energies differ by a factor of 9 which accounts for the difference in the slopes for ∆E_g/∆(k_Fℓ) of the samples. The higher sensitivity of the conductance of the low-N system to changes in the ions-potential was offered [48] as the physical reason for the observation that the magnitude of the 1/f-noise often scales with the inverse of the carrier-concentration (the ”Hooge law” [47]).

The fact that, for a similar k_Fℓ, the high-N sample is more disordered than the low-N sample is not evident from the diffraction patterns of the samples. From the point of view of topology, these two versions are difficult to tell apart; the diffraction patterns comparing between low-N and high-N versions of In_2O thin films shown in Fig. 7 reveal amorphous structures with nearly identical nearest-neighbor distance for both versions (judging by the strong diffraction-ring diameter).

The additional source of disorder that accounts for the difference between the two versions in Fig. 7 is their deviation from the chemical stoichiometry of the compound. This property is carried by the relatively few valence electrons while diffraction patterns are dominated by scattering from the plentiful inner-shell electrons. Charge-carriers that are involved in transport, on the other hand, are equally sensitive to both types of disorder.

To understand the origin of the disorder associated with the lack of stoichiometry, recall that the value of N is determined by the O/In ratio. Oxygen deficiency in indium-oxide specimens is of the order of 10-20% while the observed N is typically smaller by 2-3 orders of magnitude. Therefore some indium atoms must have a valency of +1 rather than the more common +3 to account for the observed N and preserve chemical neutrality [52]. The relative number of these ions determines the carrier-concentration while the way they are arranged in space affects the disorder perceived by charge-carriers.

The scattering centers for the conduction electrons are actually oxygen vacancies (and possibly, di-vacancies). Being the lighter element in the compound, oxygen atoms are also the dominant species to diffuse through the sample during heat-treatment. Measurement of carrier-concentration showed it to be essentially constant during treatment and therefore oxygen movement is essentially confined to within the boundaries of the sample. The constancy of the global carrier-concentration was confirmed by three independent means; Hall effect [12], optical absorption [12], and by monitoring the position of the boson-peak [23]. Therefore, migration of oxygen is plausibly the underlying mechanism for the changes in the spatial rearrangement of oxygen vacancies that take place during heat-treatment.

To be consistent with our observations, the oxygen diffusion-coefficient D_O should be no smaller than 10^{-12} cm^2/s; this will make it feasible for oxygen to diffuse over a region L≈5μm during a time τ≈two-days, the duration of heat-treatment in the present study. This value is comparable with conventionally measured D_O for oxygen diffusion in several solids [48] and in a polymer glass [49] (extrapolating results of these studies to the typical temperature used in the heat treatment, T≈360K). Actually, the presence of vacancies probably makes oxygen diffusion in these In_2O thin films faster than in the bulk solids we compare it with.

The dynamics of processes that occur on a microscopic scales depends on multi-stage bond-formation and local structure rearrangement [24, 50] that probably involves many-particle coordinated transitions.
In addition to the sample conductance, a feature that is affected by these short-range processes is the film surface roughness. This aspect may be probed by X-ray interferometry (XRR) using typically a sub-nanometer wavelength radiation. An example of XRR spectra taken before and after a brief heat-treatment of a high-N In$_x$O film is shown in Fig. 8.

The enhanced visibility of the XRR interference pattern following heat-treatment is the result of a more homogeneous distribution of defects. These processes take place on a microscopic scale, directly affecting the conductivity via changes in the basic transport parameters such as mean-free-path, or the hopping-length in the diffusive or hopping regime respectively.

The effect on conductivity due to homogenization of the mesoscopic regions probed by the Raman scattering is a more intricate issue. The length scale relevant for conductivity in In$_x$O films are much smaller than a few microns whether the system is on the metallic or insulating side of the transition even at cryogenic temperatures.

As mentioned above, the macroscopic carrier-concentration of the sample is essentially unchanged during heat-treatment. Local changes in $N$, on the other hand, do occur as evidenced by the Raman data (Fig. 1 and Fig. 2). These changes may increase the conductance of a given region while decreasing the conductance of another region (to be consistent with the global condition on $N$). The change in the macroscopic conductance due to these homogenization events will therefore be small except deep in the insulating side where the sample conductance would increase due to the logarithmic averaging inherent to the hopping regime.

In summary, we examined in this work the relation between disorder and spatial uniformity of the potential perceived by the charge carriers in an amorphous metal. Films of In$_x$O with different carrier-concentration but similar $k_F\ell$ were used to represent systems that differ significantly by their absolute degree of disorder. Both systems exhibit the same ‘frozen-liquid’ structure with essentially indistinguishable diffraction patterns but they differ in their ‘defects’ density determined by their chemical composition. It is shown that the film with the stronger disorder exhibits a higher degree of heterogeneity extending over mesoscopic scales. Moreover, reducing the disorder in the film by heat-treating it, resulted in a decrease in heterogeneity over both microscopic, and mesoscopic scales. In this case the change of disorder that resulted from the treatment is characterized by the Ioffe-Regel parameter acting as a relative measure of disorder. Disorder in In$_x$O is presumably dominated by the local concentration of oxygen vacancies that play the dual role as dopants and scatterers. Charge carriers that originate from off-stoichiometry conditions is a common scenario in many oxygen-deficient metal-oxides including high-Tc compounds. It would be interesting to see how general is the relation between disorder and heterogeneity in these compounds and in other disordered materials.

Another corollary implicit to the arguments raised above is that local defects, the oxygen vacancies in the current system, contribute to $\rho_{RT}$ more than the lack of long-range-order associated with amorphicity. This means that, for the same $k_F\ell$, the larger is the In/O ratio, the larger is the contribution of the diagonal component to the system disorder. This makes In$_x$O a useful platform to study experimentally the relative role of the two types of disorder.

The assistance by Dr. Anna Radko with the Raman spectra work is gratefully acknowledged. This research has been partially supported by the 1030/16 grant administered by the Israel Academy for Sciences and Humanities.

---

[1] Bernard Derrida, Can disorder induce several phase transitions? Physics Reports, 103, 29 (1984).
[2] M. Imui, S. A. Trugman, and Eliahu Abrahams. Unusual properties of midband states in systems with off-diagonal disorder, Phys. Rev. B. 49, 3190 (1994).
[3] M. A. Ramos and S. Vieira, F. J. Bermejo and J. Dawidowski, H. E. Fischer, H. Schober, M. A. González, C. K. Loong and D. L. Price, Quantitative Assessment of the Effects of Orientational and Positional Disorder on Glassy Dynamics, Phys. Rev. Lett., 78, 82 (1997).
[4] T. Bellini, M. Buscaglia, C. Chiccoli, F. Mantegazzana, P. Pasini, and C. Zannoni, Nematics with Quenched Disorder: What Is Left when Long Range Order Is Disrupted? Phys. Rev. Lett., 85, 1008 (2000).
itions in highly crystalline two-dimensional superconductors, Nature communications, 9, 1 (2018).

[41] N. A. Lewellyn, I. M. Percher, J. Nelson, J. Garcia-Barriocanal, I. Volotsenko, A. Frydman, T. Vojta, and A. M. Goldman, Infinite-randomness fixed point of the quantum superconductor-metal transitions in amorphous thin films, Phys. Rev. B 99, 054515 (2019).

[42] Z. Ovadyahu, Slow dynamics of the electron-glasses; the role of disorder, Phys. Rev. B. 95, 134203 (2017).

[43] E. Yamaguchi, H. Aoki and H. Kamimura, Intra- and interstate interactions in Anderson localised states, J. Phys. C: Solid State Phys., 12, 4801 (1979); H. Kamimura, Theoretical model on the interplay of disorder and electron correlations, Progress of Theoretical Physics Supplement, 72, 206 (1982).

[44] Z. Ovadyahu, Interaction-induced spatial correlations in a disordered glass, Phys. Rev. B 105, 235101 (2022).

[45] The samples in figure 6 are part of a fuller study described in [42].

[46] O. Cohen and Z. Ovadyahu, Resistance Noise near the Anderson Transition, Phys. Rev. B 50, 10442 (1994).

[47] F. N. Hooge, 1/f noise is no surface effect, Phys. Lett. 29A, 139 (1969).

[48] R. Kirchheim, Metals as sinks and barriers for interstitial diffusion with examples for oxygen diffusion in copper, niobium, and tantalum, Acta Metallurgica, 27, 869 (1979).

[49] Lars Poulsen and Peter R. Ogilby, Oxygen Diffusion in Glassy Polymer Films: Effects of Other Gases and Changes in Pressure, J. Phys. Chem. 104, 2573 (2000).

[50] Yu. M. Galperin and V. L. Gurevich, Theory of low-temperature thermal expansion of glasses, 32, 6873 (1985).

[51] Hui Lin Zhao, Boris Z. Spivak, Martin P. Gelfand, and Shechao Feng, Negative magnetoresistance in variable-range-hopping conduction, Phys. Rev. B 44, 10 760 (1991).

[52] O. Entin-Wohlman, Y. Imry, and U. Sivan, Orbital magnetococonductance in the variable-range-hopping regime, Phys. Rev. B 40, 8342 (1989).