Evidence for Interactions in Nonergodic Electronic Transport

A. Vaknin and Z. Ovadyahu
The Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel

M. Pollak
Department of Physics, University of California, Riverside, California 92521
(Received 27 February 1998)

It is shown that the nonergodic effects observed in field-effect experiments on insulating indium-oxide films (as well as in other Anderson insulators) depend in a systematic manner on the carrier concentration n. The results are interpreted as evidence for interactions. [S0031-9007(98)06660-5]

PACS numbers: 72.80.Sk, 72.20.Ht

The existence of a glassy phase in electronic systems was theoretically predicted by several authors [1]. Experimental evidence for nonergodic behavior in such systems, called Fermi glasses, was recently observed in field-effect experiments in Anderson-localized crystalline and amorphous-indium-oxide films [2], granular Au films [3], and granular Pb films [4] where an anomalous symmetric component appeared in the $G(V_g)$ plots around the gate voltage V_g at which the sample was cooled down to low temperatures. A qualitative explanation of this anomaly based on the sluggish relaxation inherent to electronic transport in the hopping regime was given in Ref. [2]. On the other hand, the quantitative aspects of these effects, and, in particular, the dependence of the relaxation time on material parameters, have not received due attention.

In this Letter we report on a systematic study of these anomalies in amorphous-indium-oxide (InO_x) films as a function of carrier concentration n. It is shown that a typical relaxation time τ in these experiments changes from 10^3 s to ≈ 1 s when n is reduced from 10^{21} to 4×10^{19} cm^{-3}. We discuss these results and argue that interactions must play a dominant role in the observed slow dynamics. In addition, the characteristic width, Γ, of the cusplike minimum in $G(V_g)$ is observed to depend on n in a systematic manner. When compared with results obtained for other systems, this dependence appears to follow a common trend which attests to the ubiquitous nature of these anomalies.

The present experiments employed a MOSFET-like structure, InO_x film serving as the semiconductor, and a 0.5 μm thick thermally grown SiO_2 as the dielectric separating it from a heavily doped Si wafer serving as a gate electrode. The amorphous-indium-oxide films used here were prepared by e-gun evaporation of 99.999% pure In_2O_3 by a method fully described elsewhere [5]. This method results in amorphous structures without any trace of free In inclusions that plagues other preparation techniques. Furthermore, by a judicious choice of evaporation rate and partial O_2 pressure, the carrier concentration n (estimated from Hall-effect measurements at room temperature) of the film can be varied [5] between 10^{19}–10^{22} cm^{-3}. Over this range the O/In atomic ratio, as measured by Rutherford backscattering, covers the range 1.38–1.12, respectively. InO_x samples with nine different n were prepared during this study, and at least two samples for each n were measured. The samples had lateral dimensions of 0.5×0.5 mm^2 and thickness of 200 ± 20 Å. The degree of disorder for a specific composition was deliberately fine-tuned by thermal annealing [6] in such a way that at $T = 4.11$ K all samples had similar sheet resistance [7] R (typically, $R \approx 1$–10 MΩ). (The reason for aiming at a constant G will become clear below.) All data reported below were taken at this convenient temperature.

The film's conductivity G ($G \equiv R^{-1}$) was measured using a two terminal ac technique employing a ITHACO 1211 current preamplifier and a PAR 124A lock-in amplifier tuned to the measuring frequency. Source-drain voltages were such that the conductance G is in the Ohmic regime.

A main tool in this work is the two-dip experiment (TDE) [8]. This involves the following procedure (cf. Fig. 1 as an example). The sample is cooled to the measuring temperature with a voltage V_g^0 held at the gate, and is allowed to equilibrate for several hours. Then, a $G(V_g)$ trace is taken by sweeping V_g across a voltage range straddling V_g^0. The resulting $G(V_g)$ exhibits a minimum centered at V_g^0 which reflects an inherent feature of a hopping system—its equilibrium conductance is at a local minimum [2]. At the end of this sweep, a new gate voltage, V_g^n (differing from V_g^0 by typically few volts), is applied and maintained at the gate between subsequent V_g sweeps that are taken consecutively at latter times (measured from the moment V_g^n was first applied). Each such sweep reveals *two* minima; one at V_g^0 which fades away with time, and the other at V_g^n whose magnitude increases with time. The TDE then amounts to studying the dynamics of the "forming" of a cusp at a newly imposed V_g^n and the "healing" of an "old" cusp at V_g^0. A characteristic time τ is defined as the time at which the amplitude of the cusp at V_g^n just equals the amplitude of the cusp at V_g^0. This τ is easily obtained by interpolation between $G(V_g)$ scans taken

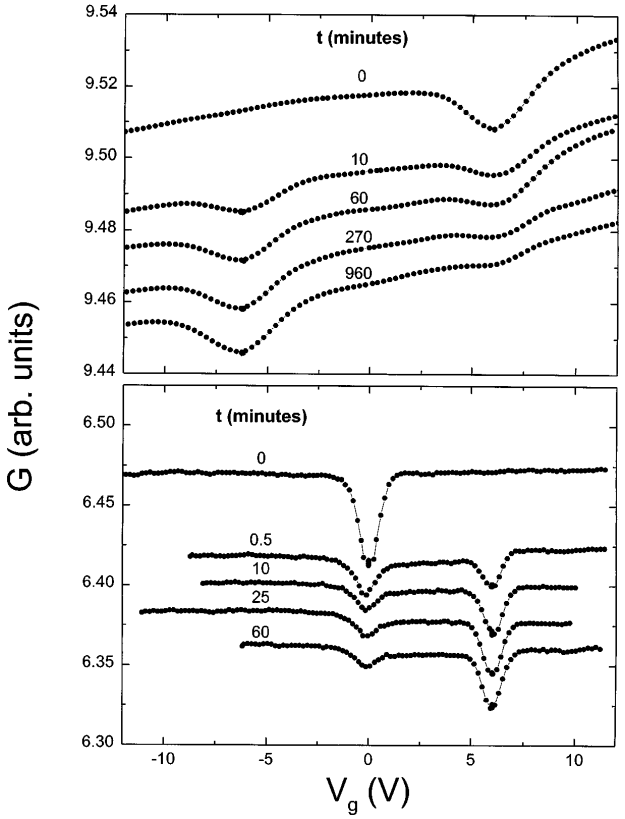

FIG. 1. Conductance versus gate voltage for a TDE of two samples with n of 4×10^{20} and 1×10^{20} cm^{-3} (upper and lower graphs, respectively). In each graph, the first trace was taken $\approx$12 hours after the initial cooldown with V_g^0 imposed (6 and 0 V for the upper and lower graphs, respectively). Then, V_g^n was applied and maintained between subsequent sweeps (-6 and 6 V, respectively). The other traces (shifted for clarity) are labeled by the time elapsed since V_g^n was first applied. Typical scan rate was 0.1–0.2 V/s.

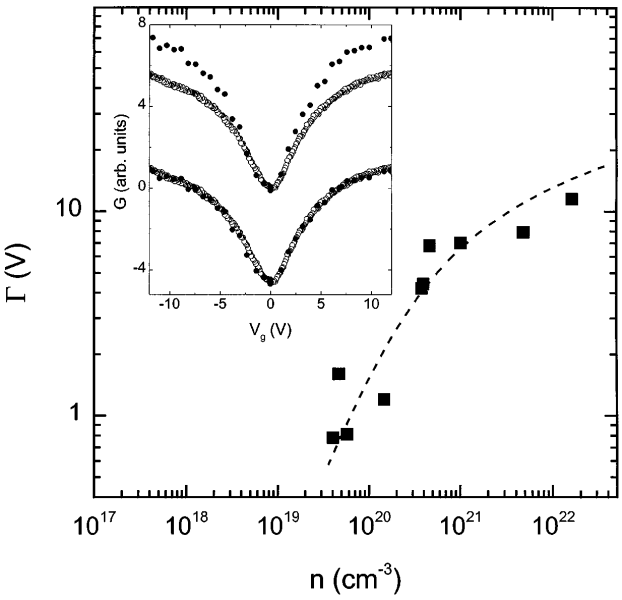

FIG. 2. The width of the cusp Γ as a function of n. Γ is taken as the width at half-height of the cusplike anomaly in $G(V_g)$ measured for each of the samples following a cooldown to 4.11 K with $V_g = 0$ imposed and maintained for a few hours. The dashed line is a guide for the eyes. Inset: The effect of sweep rate on the cusp shape for a typical sample. The top two curves depict $G(V_g)$ traces taken with sweep rates of 0.05 V/s (empty circles) and 0.6 V/s (full circles). Lower traces (shifted for clarity) are the same data after amplitude normalization by a numerical constant.

at different times, it is experimentally well defined, and it is fairly independent of the particular relaxation law.

In Fig. 1 we show such TDE plots for two samples, one typical of the high n materials and one of the low n materials. Comparing these plots makes it evident that the cusp width Γ is smaller for the material with the smaller n. The dependence of Γ on n turns out to be systematic as illustrated in Fig. 2. Over this range of n, the typical relaxation time τ, defined above, also varies systematically and extremely rapidly. The dependence of τ is shown in Fig. 3 as a function of Γ. There are two reasons for plotting τ versus Γ (rather than versus n). The carrier concentration n is based on Hall-effect measurements performed at room temperatures. It gives a reasonably good estimate of the carrier density on a relative basis. But when interactions are present (and, as is argued below, control the behavior at the temperatures of the experiments), it is more sensible to find a correlation between τ and Γ as both are measured at the *same* low temperature. Second, we believe that $\tau(\Gamma)$ expresses the essential physics of the problem in the sense that Γ is a measure of the interaction-strength which, in turn, governs the relaxation time that in our experiments is quantified by τ. Before elaborating on this issue we want to explain why the use of Γ as a parameter is meaningful from the empirical point of view. For a given sample composition and within the experimental accuracy, the value of Γ is constant, independent of disorder, and the sweep rate. (Note that all these experiments are performed at *a fixed* temperature of 4.11 K.) Changing the disorder

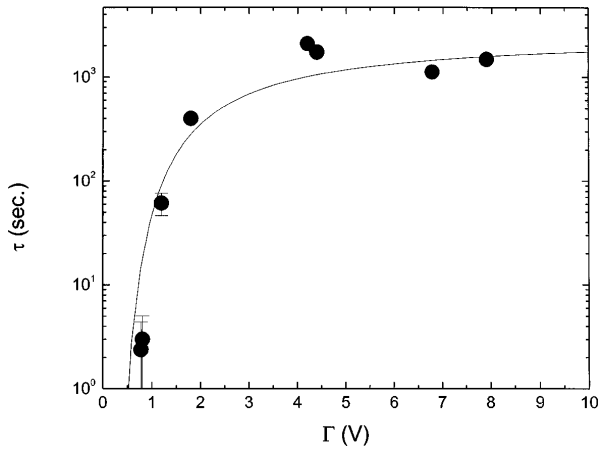

FIG. 3. The relaxation time τ extracted from a TDE of the respective sample (see text) as a function of the cusp width. The solid line is a guide for the eyes.

has virtually no effect on Γ (for example, changing R between 5 and 700 MΩ yielded the same Γ within our experimental accuracy). The only noticeable effect of the disorder is to change the *amplitude* of the cusp [2]. Similarly, for a given n, and disorder, the sweep rate (in the accessible range) used to obtain $G(V_g)$ has no measurable effect on the value of Γ or even on the shape of the cusp. As is shown in the inset of Fig. 2 the difference in amplitude due to a different sweep rate can be adjusted by changing the vertical scale which does not change the extracted Γ. We emphasize that it is *not* possible to scale these data by changing the voltage scale.

The first observation we make is that these results give strong evidence for the role interactions play in bringing about sluggish relaxation in the systems we investigated. In the absence of interaction, the time of relaxation τ is expected to be very similar for our samples which were chosen on the basis of similar values of G [9]. The argument for this is based on scaling. A similar G implies a similar distribution of microscopic hopping "conductances" g_i proportional to $e^{-2R_i/\xi}$ where R_i are hopping distances and ξ is the localization radius. The transition rates in a noninteracting system should depend on the distribution of g_i in a similar way, so the observed dramatic change in the relaxation times is not expected if interactions are unimportant.

Next, we address the physical meaning of Γ. As explained elsewhere [8] the cusp that appears in the $G(V_g)$ plots of such experiments has a compelling analogy with the dip formed on the surface of an "ordinary glass" due to the bearing action of a sharp tip. In the latter case, the width of the cusp is determined by surface tension (thus reflecting the interaction between atoms). Motivated by this picture, one is inclined to look for an energy-dependent quantity (reflecting some effective interaction between electrons) that plays an analogous role in the problem at hand. An obvious candidate to be considered is the density of states (DOS) [10].

A one-particle DOS can be defined as the density of states at an energy E available for insertion of a new particle into a system in its ground state. In the presence of interactions the old particles in the system relax, in response to the new particle, to a new ground state by, say, an energy ε. The Fermi level E_F in a macroscopic system does not change due to the insertion of a single particle, so the new particle could be inserted only at ε above E_F. The energy ε varies with the site of insertion, in accordance with its environment in the ground state and with the nature of the interaction. For long range interactions the probability for $\varepsilon = 0$ is zero (i.e., there is a gap in the DOS), for short range interactions the probability for $\varepsilon = 0$ is finite, so there is only a dip [11].

It is clear that the one-particle DOS would be accurately reflected in the dip in $G(V_g)$ if the sweep rate would be more rapid than the relaxation. If not, inserted particles which already relaxed (i.e., particles partially "dressed" into quasiparticles) will have changed their contribution to ΔG, so $G(V_g)$ can no longer be expected to track exactly the one-particle DOS. Nevertheless, some signature of the one-particle DOS will remain for a reasonably fast scanning rate. The behavior of the observed dip in $G(V_g)$ is consistent with the notion that Γ is controlled by interactions. In the absence of interactions the dip in $G(V_g)$ is entirely due to the relaxation processes [2]. The shape and width of the dip are therefore expected to depend appreciably on the scanning rate. When interactions are important, the dip in $G(V_g)$ can be attributed in part to an interaction-induced dip (or gap), in the one-particle DOS, around E_F. Such a dip is expected to depend much less on the scanning rate, which is in keeping with our observations.

To the degree that our Γ is related to the one-particle DOS, it should correspond to the width Γ^* of the interaction-induced gap (or dip). To obtain an estimate for Γ^* from the experimentally measured parameter Γ we normalized the voltage scale taking into account the sample-to-gate capacitance C (which is directly measured for each sample), $N(0)$, the material DOS at E_F, and its screening-length λ [12]. $N(0)$ and λ were estimated from the material carrier concentration n using free-electron formulas. The values of Γ^* obtained in this way [13] are shown in Fig. 4 as a function of the intercarrier distance $\langle r\rangle \approx n^{-1/3}$. Interestingly, the data points for crystalline In_2O_{3-x}, granular-Au[3], and granular-Pb[4] samples seem to fall on a common curve with the data of the amorphous-indium-oxide samples used in this study. These data are compared in this figure with $\Gamma^*_{CG} = e^3[N(0)/\kappa^3]^{1/2}$, the width of the Coulomb gap [14] (where κ is an effective dielectric constant). Note that Γ^*_{CG} is roughly 1 order of magnitude larger than the experimental Γ^*. Observation of a much narrower Coulomb gap than predicted was also

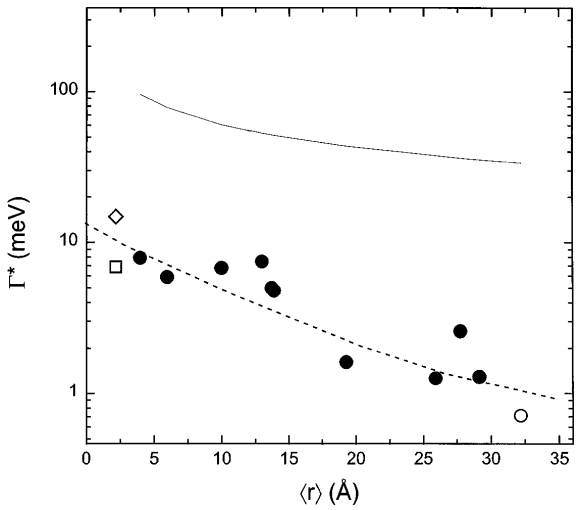

FIG. 4. The normalized values of the cusp width as a function of the intercarrier distance (see text). Full circles: InO_x films used in this study. Empty circle: Crystalline In_2O_{3-x}. Diamond: Granular Au (Ref. [3]). Square: Granular Pb (Ref. [4]). The solid line is a fit to $\Gamma^* = e^3[N(0)/\kappa^3]^{1/2}$ (Ref. [14]). The dashed line is a guide for the eyes.

reported in tunneling experiments [15] and in measurements [16] and simulations [17] of conductivity. The first may arise from screening by the tunneling counterelectrode [18], the other two from dynamical many-body effects, as explained in those papers. Similar dynamical effects are expected to play an important role in our experiments due to the finite sweep rate. Thus our experiment may not measure the full Coulomb gap, or otherwise the Coulomb interaction is not the dominant interaction. In particular, it may turn out that the dip we observe is due to polaron formation driven by the electron-phonon interaction or that it is a spin gap due to spin-spin interaction.

Finally, the correlation between τ and Γ (Fig. 3) is very plausible once it is recognized that Γ is indeed a measure of the interaction strength. It would be of much interest to see whether such a dependence is observed in other systems where n can be varied over an extended range (e.g., semiconductors). The exponentially fast decrease of τ when n becomes smaller than $\approx 10^{20}$ cm^{-3} is noteworthy as it may hint to the reason for the difficulty of observing these anomalies in lightly doped semiconductors. For insulating semiconductors n is typically much smaller than the carrier concentrations used here, and this may result in τ being extremely short. Indeed, relaxation times of the order of only $\approx$10 msec were reported in insulating ($n < 10^{18}$ cm^{-3}) GaAs samples [19]. These issues clearly deserve further study.

In summary, we have presented data that strongly suggest the relevance of interactions in giving rise to the surprisingly long-relaxation phenomena observed in disordered electronic systems. The nature of the dominant interaction is yet unknown, and further work is clearly necessary to identify its specific origin.

We gratefully acknowledge discussions with E. Pazy, O. Agam, and B. Spivak. This research was supported by a grant administered by the U.S.-Israel Binational Science Foundation and by the Israeli Academy of Science.

[1] J. H. Davies, P. A. Lee, and T. M. Rice, Phys. Rev. Lett. **49**, 758 (1982); M. Pollak, Philos. Mag. B **50**, 265 (1984); M. Pollak and M. Ortuño, Sol. Energy Mater. **8**, 81 (1982); M. Grünewald, B. Pohlman, L. Schweitzer, and D. Würtz, J. Phys. C **15**, L1153 (1982).
[2] M. Ben-Chorin, Z. Ovadyahu, and M. Pollak, Phys. Rev. B **48**, 15 025 (1993).
[3] C. J. Adkins, J. D. Benjamin, J. M. D. Thomas, J. W. Gardner, and A. J. McGeown, J. Phys. C **17**, 4633 (1984).
[4] G. Martinez-Arizala, D. E. Grupp, C. Christiansen, A. Mack, N. Markovic, Y. Seguchi, and A. M. Goldman, Phys. Rev. Lett. **78**, 1130 (1997).
[5] Z. Ovadyahu, J. Phys. C **19**, 5187 (1986).
[6] Z. Ovadyahu, Phys. Rev. B **47**, 6161 (1993).
[7] Note that over the range of n covered the Fermi energy changes considerably. Therefore the samples must also differ in disorder energy to exhibit a similar G.
[8] Z. Ovadyahu and M. Pollak, Phys. Rev. Lett. **79**, 459 (1997).
[9] Our argument is not vulnerable to some variation in the value of G within the series of samples because a change of G in a given sample (i.e., fixed n) has only a small effect on τ (see also Ref. [8]).
[10] See also Sven Rogge, Douglas Natelson, and D. D. Osheroff, Phys. Rev. Lett. **76**, 268 (1996).
[11] We comment that the DOS in the vicinity of the electrode from which particles are inserted may differ from the bulk DOS due to the screening of the interaction by the electrode.
[12] N. F. Mott and E. A. Davis, *Electronic Processes In Non-Crystalline Materials* (Clarendon Press, Oxford, 1979), p. 245.
[13] An alternative normalization, based on the thermal filling of the gap over a width $\sim kT$ [J. H. Davies, P. A. Lee, and T. M. Rice, Phys. Rev. B **29**, 4260 (1984); Q. M. Li and P. Phillips, Phys. Rev. B **49**, 10 269 (1994); M. Sarvestani *et al.*, Phys. Rev. B **52**, R3820 (1995)], yields comparable values for Γ^* but this could be reliably applied only for the samples with $\tau \geq 500$ s.
[14] A. L. Efros and B. I. Shklovskii, J. Phys. C **8**, L49 (1975).
[15] J. G. Massey and M. Lee, Phys. Rev. Lett. **75**, 4266 (1995).
[16] A. G. Zabrodskii and A. G. Andreev, JETP Lett. **58**, 759 (1993).
[17] A. Perez-Garrido *et al.*, Phys. Rev. B **55**, R8630 (1997).
[18] See, e.g., E. Cuevas *et al.*, Philos. Mag. B **70**, 1231 (1994).
[19] Don Monroe *et al.*, Phys. Rev. Lett. **59**, 1148 (1987).