PHYSICAL REVIEW B **85**, 235114 (2012)

Determination of characteristic relaxation times and their significance in glassy Anderson insulators

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(Received 16 January 2012; published 8 June 2012)

We revisit the field effect procedure used to characterize the slow dynamics of glassy Anderson insulators. It is shown that in the slowest systems, the procedure fails and the "characteristic" time values extracted are not intrinsic but rather determined by the experimental procedure itself. In other cases (like lightly doped indium oxide) qualitative indications about the dynamics might be obtained; however, the times extracted cannot be seen as characteristic relaxation times of the system in any simple manner, and more complete experiments are necessary. Implications regarding the effect of carrier concentration on the emergence of glassiness are briefly outlined.

DOI: 10.1103/PhysRevB.85.235114 PACS number(s): 72.80.Ng, 61.20.Lc, 73.23.Hk

I. INTRODUCTION

Typical glassy phenomena such as long relaxation times, memory of history, and physical aging exist at low temperature in the conductance field effect anomaly observed in thin film disordered insulators such as microcrystalline and amorphous indium oxide[1] and granular metals.[2,3] Recent experiments on discontinuous Ni films, consisting of ferromagnetic metallic islands, have recently shown that the slow dynamics can be significantly altered by the magnetic field.[4]

These phenomena are believed to be the signatures of a Coulomb glass state of the charge carriers, which was first suggested more than two decades ago[5] and may result from the strong Coulomb interactions among the poorly screened localized carriers of these highly disordered insulators.

Although a large amount of theoretical work has been performed recently, there is no established precise scenario for the appearance of the glassy behavior. One of the most important open questions is why the typical experimental signature (slow logarithmic growth of a field effect anomaly after a quench) has not yet been observed in standard doped semiconductors, the archetype of Anderson insulators. One suggestion is that high-enough carrier concentrations are necessary, which cannot be realized in standard doped semiconductors without getting into the metallic state. This suggestion was based on a series of experiments on indium oxide samples with various oxygen deficiencies and thus various electron concentrations. It was shown that a characteristic relaxation time τ, defined in the way we describe and discuss later, increases drastically when the electron concentration exceeds a few $10^{19}\,\mathrm{cm}^{-3}$ (Ref. 6) and then saturates above a few $10^{20}\,\mathrm{cm}^{-3}$.

The experimental implementation of τ is now used to characterize and compare the dynamics of other glassy disordered insulators. However, in the present paper, we stress that the definition used for τ can be problematic. In particular, we show with new measurements that in "highly doped" samples, its value is determined by the experimental procedure so that the apparent saturation of the dynamics is an artifact. However, in "lightly doped" samples, different values for τ are observed, their meaning is not straightforward, and more complete experiments are necessary to form conclusions about the nature of the changes observed in the dynamics. Some implications of these findings are briefly discussed.

II. EXPERIMENTAL DEFINITION OF τ

One general property of the systems under interest is that after a quench from high to liquid He temperature, their electrical conductance is not constant but decreases logarithmically with time, with no reported saturation for up to several days or weeks of measurement. As is well known, this corresponds to a $1/\tau_i$ effective time distribution, which has no characteristic value.[7] It is also difficult to directly compare the dynamics of different samples, or of one given sample at different temperatures, because the physics of the prefactor determining the amplitude of the relaxations (the slope in a logarithmic plot) is not yet established. In an attempt to circumvent these difficulties, a "two dip" protocol was used to define a "characteristic" time τ of indium oxide samples.[6] It is based on the slow dynamics of the field effect anomaly observed in all glassy systems of interest here and is shown in Fig. 1.

For such experiments, the samples consist of metal–oxide–semiconductor field effect transistor (MOSFET)–like devices with a thin film of the disordered insulator constituting the (poorly) conducting channel. After a sample has been quench cooled at a given gate voltage $V_g = V_{g1}$ and has been allowed to relax for a given time t_{w1}, any sudden change of V_g away from V_{g1} re-excites the system, which was slowly approaching equilibrium and restores the higher off-equilibrium conductivity it had before relaxing. However, the system keeps a memory of its relaxation under V_{g1}. Thus, a fast V_g scan reveals a conductivity "dip," as seen in the upper curve of Fig. 1(b).

If the gate voltage is set at another value V_{g2}, subsequent fast V_g scans (separated by "long" periods with $V_g = V_{g2}$) show a fading dip at V_{g1} and a slowly emerging one at V_{g2}, indicating the system's relaxation to a new equilibrium state under $V_g = V_{g2}$ [lower curve of Fig. 1(b)]. Using this two-dip protocol, a characteristic time τ was defined as the time to be spent at V_{g2} until the two dips are of equal amplitude. It was determined by regularly performing fast V_g scans to detect when the two cusp amplitudes have crossed. The first step was performed during a time t_{w1} of 12 h, supposedly long enough for the system to have "equilibrated" before V_g is changed to V_{g2}.

A systematic study showed that monitoring the electron concentration by changing the oxygen deficiency of indium

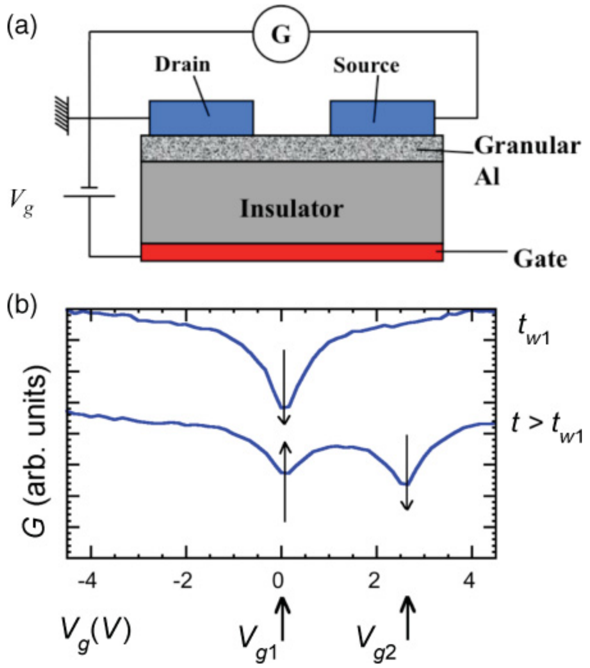

FIG. 1. (Color online) (a) MOSFET device used to measure the field effect dip in granular aluminum. (b) Two-dip protocol (see text). The upper curve shows the field effect dip formed at V_{g1} during the first step of the protocol (sample cooled and maintained under V_{g1} for t_{w1}). The lower curve shows the fading V_{g1} dip and growing V_{g2} dip during the second step of the protocol (sample maintained under V_{g2}).

oxide films has a dramatic effect on the value of τ: while it is on the order of 10^3 s for samples with $n_e > 10^{20}$ cm^{-3}, it seems to abruptly decrease when $n_e < 10^{20}$ cm^{-3} (see Fig. 3 of Ref. 8). This is believed to be an important clue to why the gate voltage dip and its glassy behavior are not observed in standard doped semiconductors: these have carrier concentrations that are too low in their insulating state to have macroscopic relaxation times.

Below we discuss difficulties associated with the definition and interpretation of τ.

A. Highly doped indium oxide and granular Al

We need to distinguish the cases of lightly and highly doped indium oxide samples. We first discuss the highly doped ones, as well as granular aluminum. We associate these two materials in the discussion because they share the same glassy phenomenology. First, recall that the width of the field effect dip in indium oxide strongly increases with the electron concentration,[6] and that the one observed in granular aluminum (when expressed in the gate field) is the same as that of the most highly doped indium oxide samples.[2] Moreover, when the protocol described previously is applied to granular aluminum-based MOSFETs, a value for τ is obtained similar to that for highly doped indium oxide.

If the systems under study were fully equilibrated during the first step of the protocol (i.e., if t_{w1} was longer than the longest relaxation time of the systems), then the time τ obtained from the two-dip protocol would indeed characterize the systems' dynamics.[9] But such is not the case. Both highly doped indium oxide and granular aluminum, following a quench, display logarithmic relaxations for durations far exceeding the t_{w1} used in the two-dip protocol. Thus, at t_{w1}, the samples are generally not equilibrated and are still logarithmically relaxing. We may think this is unimportant for practical purposes as long as the residual relaxation is very slow (logarithmic), but this turns out to be incorrect.[10] Suppose that in the first step, the first dip amplitude grows like $A\ln(t/\tau_{\text{micro}})$; then, its amplitude at time t_{w1} is $A\ln(t_{w1}/\tau_{\text{micro}})$. Here, τ_{micro} is the minimum relaxation time of the system. In the second step of the protocol, the second dip at V_{g2} also grows like $A\ln(t'/\tau_{\text{micro}})$, while the first one at V_{g1} decreases like $A(\ln[t_{w1}/\tau_{\text{micro}}] - \ln[t'/\tau_{\text{micro}}])$, where t' is the time spent in the second step. Equating the two amplitudes gives

$$t' = \tau = \sqrt{t_{w1}\tau_{\text{micro}}}.$$

This simple derivation assumes that the measurement (V_g scans) is instantaneous and has no influence on the dips. However, this is not the case; because a typical measurement time is the duration t_{scan} of a V_g scan, we might expect (see the Appendix for justification)

$$t' = \tau \approx \sqrt{t_{w1}t_{\text{scan}}}. \tag{1}$$

Thus, the measured τ should be determined by the experimental protocol, not solely by the system's dynamics.

We performed new measurements on a granular aluminum film with R (4K) = 20 GΩ to check this. In Fig. 2, we show the t_{w1} dependence of τ. We clearly see the square-root dependence of τ with t_{w1} up to the longest time measured (2.59 10^5 s) which corresponds to three days.

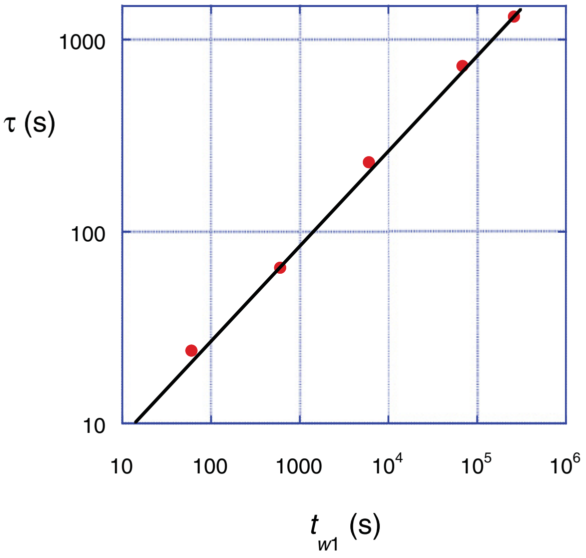

FIG. 2. (Color online) t_{w1} dependence of τ in a granular aluminum thin film. V_g scans are performed each minute. Here, a V_g scan consists of three conductance measurements performed at $V_g = -5$ V (baseline conductance), $V_{g1} = 0$ V, and $V_{g2} = +5$ V. The straight line has the slope 1/2, indicating square-root dependence.

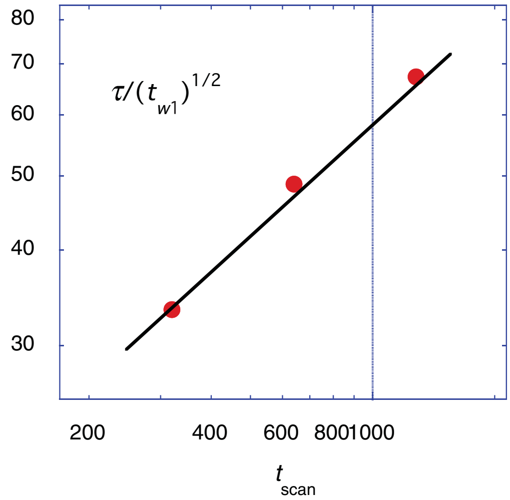

FIG. 3. (Color online) t_{scan} dependence of τ in the same granular aluminum thin film as in Fig. 2. For these measurements, the V_g scans consisted of 31 measurements equally spaced between $V_g = -5$ V and $V_g = +5$ V. The different scan durations correspond to different single measurement durations (stays at the measured V_g scans). Because for the three values of t_{scan} the t_{w1} times were slightly different, we plot $t/t_{w1}{}^{1/2}$ versus t_{scan}. The straight line has the slope $1/2$ indicating square-root dependence.

In Fig. 3, we show the t_{scan} dependence of τ for the same sample. We again see clear dependence close to a square root.

This establishes that in granular aluminum the value of the two-dip characteristic time is determined by the experimental protocol and not by the systems' dynamics.

We wonder whether such an artifact is also present with highly doped indium oxide. This should be tested directly by future experiments, but we tend to think the answer is yes. Indeed, the value estimated using Eq. (1) (with $t_{\text{scan}} =$ 100–200 s and $t_{w1} = 12$ h)[6] gives $\tau \approx 2$–$3\ 10^3$ s, close to the experimental values (1–2 10^3 s).

Thus, it is highly probable that the constant value found for τ in the highly doped indium oxide samples (with $n_e > 10^{20}$ cm^{-3}) is also solely determined by the experimental protocol. It does not indicate that the dynamics (which may be characterized by, e.g., the longest relaxation times of the systems) is constant. It merely indicates that the maximum relaxation times are larger than the experimental t_{w1} and that the relaxations are all close to logarithmic with nearly the same prefactor A [as assumed in our derivation of Eq. (1)].

B. Lightly doped indium oxide

We discuss the more interesting case of indium oxide with $n_e < 10^{20}$ cm^{-3}, for which the two-dip τ is significantly different (smaller) than Eq. (1). This may mean either of the following:

(1) The maximum relaxation times are smaller than the experimental times.

(2) The dips relaxations are not symmetrical (unequal prefactors) or are not simply logarithmic.

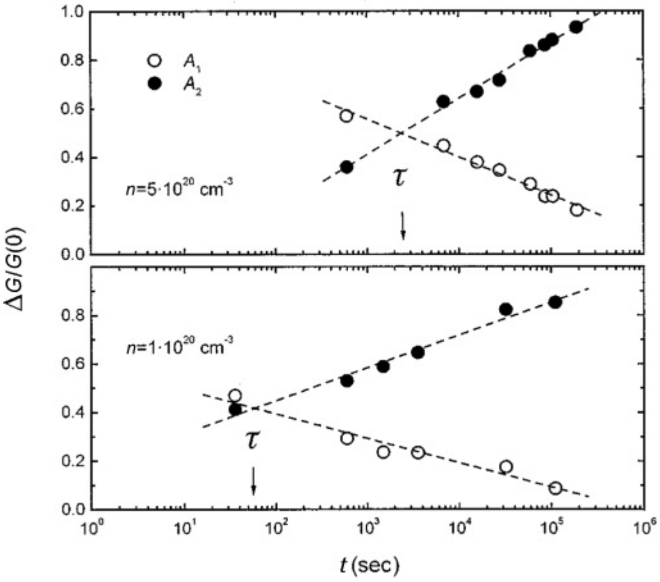

FIG. 4. Time evolutions of the V_{g1} and V_{g2} dips during the second step of the two-dip protocol for a highly (top) and a lightly (bottom) indium oxide sample (Reprinted from Ref. 11).

The published data give clues about what is happening. In Fig. 2 of Ref. 11, reproduced here in Fig. 4, the two-dip evolutions are shown for two indium oxide samples, a highly doped one (with $n_e = 5\ 10^{20}$ cm^{-3} and $\tau = 2$–$3\ 10^3$ s) and a lightly doped one (with $n_e = 10^{20}$ cm^{-3} and $\tau = 50$–60 s). We immediately see that hypothesis 1 is not correct, because the two-dip amplitudes in the lightly doped sample, much like in the highly doped one, are seen to relax with no clear sign of saturation up to times of 10^5 s, which is larger than the t_{w1} used. Surprisingly, hypothesis 2 also seems to be contradicted by the data. As emphasized in the text, within the experimental accuracy, the relaxations are logarithmic in time for both samples, with nearly symmetrical evolutions for each sample.

Moreover, the slopes are smaller for the lightly doped sample than for the highly doped one. If we accepted that we can compare the slow dynamics of samples with similar resistances at the same temperature by comparing the slopes of the logarithmic relaxations of $\Delta G/G$, we would conclude that the lightly doped sample (with the shortest τ) is the slower one.

The resolution of this paradox may come from the short time response after the V_g jump from V_{g1} to V_{g2}, which seems to be different in the lightly and in the highly doped systems. Judging from the starting points of the curves, in the lightly doped sample, the first dip seems to be quite rapidly erased after the gate voltage has been switched from V_{g1} to V_{g2} (it is reduced to a bit more than 40% of its initial value after 30 s), while the effect is smaller or slower in the highly doped one (60% of its initial value after 600 s). Similarly, the growth curve of the second dip starts from a higher value in the lightly doped sample (40% of amplitude at 30–40 s) than in the highly doped one (only 35% at 600 s).

Thus, it may be that the difference observed between highly and lightly doped samples comes from a "fast" response following V_g jumps, at times too short to be investigated by the experimental procedure. Lightly doped systems would then have a relaxation time distribution with a larger weight at short times and a smaller one in the long time (logarithmic) region compared to highly doped ones.

Unfortunately, this simple conclusion cannot be taken for granted from the existing data, because what happens in the short time regime is not measured. It is desirable to find ways to investigate this regime.

Moreover, we might wonder whether the experimental procedure chosen influences the results for the lightly doped systems. The two-dip protocol is clear if t_{scan} is much smaller than the waiting time after the first scan and between successive scans. But this condition is not met with the lightly doped system, so in that case the effective waiting times for the two dips are not well defined. Second, we might wonder whether phenomena due to the V_g jump (from V_{g1} to V_{g2}) influence the results. Indeed, it was already reported that V_g jumps can rapidly destroy the first dip memory in indium oxide (see Figs. 13 and 14 of Ref. 12), and there are indications that the effect is more prominent for lightly doped samples than for highly doped ones.[13] Although at that time no connection was made with the two-dip characteristic time, this phenomenon may influence the τ values obtained for the lightly doped indium oxide samples.

III. IMPLICATIONS

The present analysis has several potentially important implications for understanding glassiness in Anderson insulators. First, the dependence of the dynamics on the carrier concentration is probably not the one suggested by the curve τ (n_e) accepted until now—at least, the saturation above a "critical" concentration $\sim 10^{20}$ cm^{-3} is not real. The sole τ values bring either misleading or incomplete information about the systems' dynamics. To discuss two-dip experiments, we must compare the full relaxation curves, including the first step relaxation before t_{w1}.

Second, the nonobservation of any glassy field effect dip in "standard" semiconductors (e.g., doped silicon) was until now explained relying on the τ data, suggesting a very steep decrease of τ when n_e becomes smaller than 10^{20} cm^{-3}. For this reason, doped semiconductors (of a carrier concentration less than 10^{19} cm^{-3} in their insulating regime) were believed to have immeasurably fast relaxation times. This explanation has to be reconsidered or refined in light of our findings. If V_g changes indeed have strong detrimental effects on the memory, then in principle no gate voltage dip can be measured in these systems using V_g scans. It might then be more interesting to search for slow relaxations after the quench at constant gate voltage.

Finally, experiments aimed at comparing the dynamics of different systems using the two-dip protocol should be considered with much care. We must remember that by construction the two-dip protocol does not provide an absolute measurement of a given system's dynamics; instead, it only provides a comparison of its dynamics in the first and second steps, i.e., before and after the gate voltage jump.[14] Thus, it may be used to study the effect of an external parameter like temperature or magnetic field on the dynamics of one given system, provided that we change the external parameter value at the same time as the V_g jump (e.g., see the case of T changes in Ref. 2) and that possible effects of the V_g jump on the dynamics can be dealt with. An even better way is probably to study whether the growth of a given dip, submitted to different values of the external parameter during succeeding time intervals, is cumulative (like in Fig. 10 of Ref. 15). This procedure seems to be the most reliable envisaged so far.

IV. CONCLUSION

We have revisited the main experimental procedure used so far to characterize the dynamics of the glassy disordered insulators. We have shown that the characteristic times τ extracted from the two-dip protocol can be misleading. For systems behaving like highly doped indium oxide (including granular aluminum), values obtained for τ are useless because they are determined by the experimental procedure [Eq. (1)] and not by the systems' properties. For the lightly doped indium oxide case, the small values of τ seem to be related to short time effects following the gate voltage jump in the two-dip protocol. The characteristic time τ alone does not constitute a good indicator of the systems' dynamics. Indeed, it is observed that both large and small τ-valued indium oxide samples have slow logarithmic relaxations extending to very large times (at least 10^5 s).

In general, to understand departures of τ from the value given by Eq. (1), it is important to compare the full relaxation curves of all steps of the experiments protocols.

In the case of lightly doped indium oxide samples, future experiments seem necessary to better understand the short time responses. Relaxation measurements after a V_g jump without performing complete V_g scans (which take time and prevent the short times from being explored), as well as relaxation measurements after a quench (short time relaxation without any V_g jump), should bring many interesting results.

The common explanation for the nonobservation of any gate voltage dip in standard doped semiconductors might also be revisited in the light of the present remarks.

ACKNOWLEDGMENT

We acknowledge financial support from the Université Joseph Fourier (SMINGUE 2010 program), as well as from the Region Rhône-Alpes (CIBLE 2010 program). Discussions about this topic with Ariel Amir, Stefano Borini, and Zvi Ovadyahu are also acknowledged.

APPENDIX

Here, we justify Eq. (1). Suppose a sample is quench cooled at $t = 0$, relaxed under V_{g1} for t_{w1} and then relaxed for a time t_{w2} under V_{g2}. Then, a V_g scan is performed. For simplicity, we suppose that the value of t_{w2} was wisely chosen so that the two dips are found to be of equal amplitude (i.e., $t_{w2} = \tau$). We choose (like in our experiments) to perform the V_g scan in the direction of increasing gate voltage. In general, as long as V_g

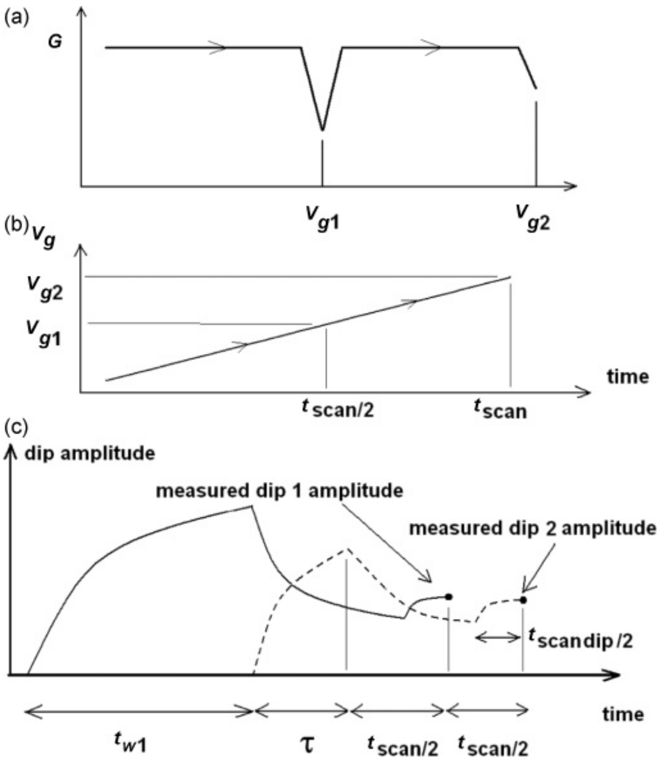

FIG. 5. (a) and (b) V_g scan during the second step of the two-dip protocol. (c) Time evolution of the dips' amplitudes in the two-dip protocol, with only one V_g scan started at time τ after t_{w1}. Time intervals are not to scale.

is nearly equal to a given value V_{gi}, the ith dip amplitude (centered on V_{gi}) increases with time while the jth dip amplitude (if any) decreases. Using the "superposition principle" discussed in Ref. 3, the time evolution of the dips' amplitudes is a sum of positive and negative logarithms. The evolutions of the two-dip amplitudes may be shown as in Fig. 5.

Their expressions are

$$\begin{aligned}\Delta G(V_{g1}) &= A[\ln(t) - \ln(t - t_{w1}) + \ln(t - t_{w1} - \tau \\ &\quad - (t_{\text{scan}} - t_{\text{scandip}})/2)]\\ \Delta G(V_{g2}) &= A[\ln(t - t_{w1}) - \ln(t - t_{w1} - \tau) \\ &\quad + \ln(t - t_{w1} - \tau - t_{\text{scan}} + t_{\text{scandip}}/2)]\end{aligned}$$

where t_{scandip} is the time it takes to scan a dip width. The τ_{micro} denominators have been omitted in the logarithms because they cancel in the calculation. Equating the two dips amplitudes each at the time it is measured gives a relation whose solution is

$$\tau = {}^1\!/_2\sqrt{(t_{\text{scan}}/2)^2 + 4t_{\text{scan}}t_{w1}} - t_{\text{scan}}/4$$

which is close to Eq. (1) as long as $t_{\text{scan}} \ll t_{w1}$.

Unlike in the simple case just considered, in real measurements, the value of τ is not known *a priori* and several V_g scans are performed before the dips' amplitudes cross, the crossing point being obtained by interpolation. Moreover, the relaxations are not exactly logarithmic (aging effect[3]). The real situations can be simulated numerically. The value of τ then also depends on the waiting time between each V_g scan (if t_{scan} is not negligible compared to it), but we find that Eq. (1) still gives a good estimate. Thus, τ is expected not to be a constant but to strongly depend on the measurement protocol, as observed experimentally.

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