PHYSICAL REVIEW B **82**, 195404 (2010)

Intrinsic electron glassiness in strongly localized Be films

Z. Ovadyahu,[1] Y. M. Xiong,[2] and P. W. Adams[2]
[1]*Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel*
[2]*Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803, USA*

(Received 24 August 2010; revised manuscript received 13 October 2010; published 3 November 2010; corrected 12 November 2010)

We present results of out-of-equilibrium transport measurements made on strongly localized Beryllium films and demonstrate that these films exhibit all the earmarks of intrinsic electron glasses. These include slow (logarithmic) relaxation, memory effects, and more importantly, the observation of a memory dip that has a characteristic width compatible with the carrier concentration of beryllium. The latter is an empirical signature of the electron glass. Comparing various nonequilibrium attributes of the beryllium films with other systems that exhibit intrinsic electron-glasses behavior reveals that high carrier concentration is their only common feature rather than the specifics of the disorder that rendered them insulating. It is suggested that this should be taken as an important hint for any theory that attempts to account for the surprisingly slow relaxation times observed in these systems.

DOI: 10.1103/PhysRevB.82.195404 PACS number(s): 72.20.Ee, 72.20.Ht, 72.70.+m

I. INTRODUCTION

The interplay between static disorder and Coulomb interactions may precipitate a glassy state in an Anderson insulator. This "electron-glass" scenario was discussed in several papers.[1–4] In theory, this property is generic to *all* degenerate Fermi systems with localized states interacting via a Coulomb potential. Experimental evidence for these glassy effects, however, has been somewhat scarce, presumably due to specific material requirements. It turns out that only systems with relatively high carrier concentration n exhibit relaxation times that can be conveniently monitored by transport measurements. Conductance relaxations that persist for many seconds, and memory effects characteristic of *intrinsic*[5] electron glass, seem to be peculiar to systems with $n > 10^{19}$ cm^{-3}.[5] A prominent group of materials that exhibit electron-glass behavior with long relaxation times are granular metals; Al,[6] Bi,[7] Pb,[7] Ni,[8] and Au,[9] all having high carrier concentration $n \geqslant 10^{22}$ cm^{-3}.

Hitherto, the only nongranular systems that exhibited intrinsic electron-glass behavior were crystalline and amorphous indium-oxide films (In_2O_{3-x} and In_xO respectively),[5] which are ionic compounds. In this work we report on the low-temperature transport properties of strongly localized Be films and demonstrate that they exhibit intrinsic glassy effects. This is the first nongranular monatomic system to show these effects. These include logarithmic relaxation of the out-of-equilibrium conductance and, more importantly, a memory dip that has all the earmarks of intrinsic electron glass. Although beryllium, like a typical metal, has a Fermi energy E_F of few electron volts, it has an unusually low density of states at E_F.[10] A signature of the Be peculiar density of states, namely, at the Fermi energy, the density of states decreases with energy, is actually observed in our field-effect measurements as will be demonstrated below. The low density of states of beryllium is presumably the main reason why strong localization is achievable in this material just by making the sample thin enough (yet still physically continuous).

II. EXPERIMENTAL

Samples used in the experiments reported here were 18 ± 5 Å films deposited as described elsewhere[11] on 140 μm glass slides. These were silver painted on their backside so as to form a gate for the field effect measurements. The samples were typically 400-μm-wide and 600-μm-long strips and had sheet resistance $R_\square$ in the range 23–120 kΩ at 295 K and 100 kΩ–160 MΩ at ≈4 K. The strongly localized nature of the films at this temperature range was tested by measuring their conductance versus temperature dependence $G(T)$ in the range 4–50 K. Below ≈10 K, this dependence is consistent with $G(T) \propto \exp[-(\frac{T_0}{T})^{1/2}]$ with T_0 in the range 100–900 K. Figure 1 illustrates this behavior for two of the samples that were used in this study.

The different values of $R_\square$ in the studied series of samples were obtained by a judicious oxidation of the Be films in an

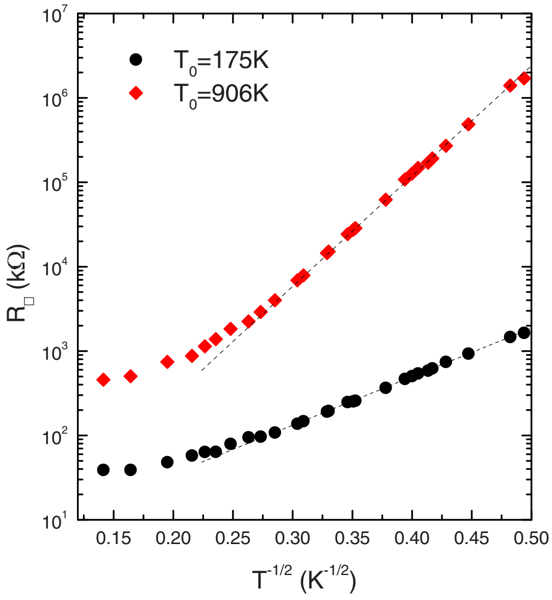

FIG. 1. (Color online) Resistance versus temperature plots for two of the beryllium films used in this study.

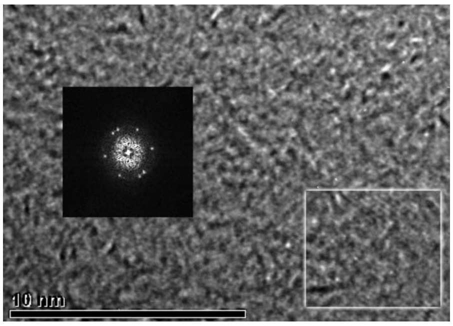

FIG. 2. Transmission electron microscope micrograph of ≈ 20 Å Be film deposited on carbon-coated Cu grid. The black square is a Fourier transform of the framed area on the right. The latter shows 2.33 Å fringes associated with the d spacing of the $\langle 100 \rangle$ planes of the hexagonal BeO (reflected as satellites in the Fourier transform plate).

oxygen-enriched chamber. The change in the samples resistance was constantly monitored during the oxidation process. At the range of sample thickness $d \approx 18$ Å, the room-temperature sheet resistance of the samples is ≈ 20 kΩ. Such samples are usually deep into the hopping regime at liquid helium temperatures and a small change in their thickness d translates into a large change of $R_\square$ at 4 K. The sheet resistance of samples much thicker than 18 Å may not reach the quantum resistance at 4 K (recall that $R_\square > \hbar/e^2$ is a prerequisite for electron-glass behavior[5]). For example, in a previous study[12] a beryllium film with a nominal thickness d of ≈ 20 Å, only slightly thicker than the films studied here, exhibited $G(T) \propto \exp[-(\frac{T_0}{T})^{1/2}]$ with $T_0 = 1.6$ K. This should be compared with $T_0 \gtrsim 100$ K in the samples used in this work. The much smaller value for T_0 and the associated larger localization length in this case means that strongly localized behavior is attained only at temperatures that are well below the range covered here. Such samples are not included in our present study where one of our goals is to compare the results with previously studied electron glasses, which were measured at or near 4 K.

The conductivity of the samples was measured using a two-terminal ac technique employing a 1211-ITHACO current preamplifier and a PAR-124 A lock-in amplifier. Measurements reported below were performed with the samples immersed in liquid helium at $T = 4.1$ K maintained by a 100 l storage-dewar, which allowed long-term measurements of samples as well as a convenient way to maintain a stable temperature bath. Unless otherwise indicated, the ac voltage bias was small enough to ensure linear response conditions (judged by Ohm's law being obeyed within the experimental error). Fuller details of measurements techniques are given elsewhere.[13]

A variety of techniques were employed to characterize the films microstructure. Figure 2 shows a transmission electron microscope (TEM) micrograph of a Be film prepared in the same way and with similar thickness as the samples used for the transport studies. The physical continuity of the film is evident in the figure. On careful examination, the micrograph shows occasional fringes that indicate the presence of small crystals. These were identified as BeO by direct imaging and further, by their diffraction pattern (interestingly, the diffraction pattern of Be, being light on electrons, was presumably too weak to register a clear pattern over the background set by the amorphous carbon support film). The BeO crystals were clearly observable in TEM dark field imaging off their $\langle 100 \rangle$ diffraction line. This enabled an estimate of their crystallographic size and partial volume in the samples. Randomly distributed BeO crystallites of sizes up to 50–70 Å were observed in these dark-field images. We estimate that less than $\approx 10\%$ of the film area is occupied by fully oxidized Be crystallites and therefore transport presumably occur through nonoxidized Be phase. Yet, the insulating BeO crystallites, somewhat restrict the volume available for conductivity (much like punching holes in the film would). A result of this geometrically constrained structure is that the transport properties of the films show some mesoscopic effects that one usually encounters in smaller systems measured at similar temperatures and comparable degree of disorder.[14,19]

The physical continuity of the Be phase in the film was ascertained by performing local electron-energy loss spectroscopy (EELS) on the parts of the film that were not occupied by BeO crystals. The EELS spectra taken from theses areas was consistent with that of metallic Be. A slight shift of energy, $\leqslant +3\%$ of the peaks position in the spectra was detected, possibly due to strain. The presence of free Be in the samples was also confirmed by x-ray photoemission spectroscopy, which was carried out on the actual samples that were used for the transport measurements.

III. RESULTS AND DISCUSSION

We turn now to the nonequilibrium transport properties of the films. The first signature of glassy behavior in these films is encountered upon quench cooling the sample to 4 K; after an initial fast drop (reflecting the change in temperature), the conductance G keeps on decreasing slowly (logarithmically) long after the sample has reached the bath temperature. A typical quench-cooling protocol is shown in Fig. 3(a). The figure also illustrates the slow conductance excitation process upon "stressing"[15] the film with a non-Ohmic source-drain voltage Fig. 3(b). The ensuing relaxation of G after the source-drain voltage was set back to its Ohmic value is shown in Fig. 3(c). These excitation-relaxation curves are clearly similar to those previously observed in glassy In_2O_{3-x} samples.[15]

A controlled way to take the system out of equilibrium is a change in the potential difference between the sample and a near-by gate. This technique has been widely used in the study of several electron glasses.[5–7,13] Among other things, it may be used to estimate a typical relaxation time τ under a given set of conditions.[16] An example of such protocol is illustrated in Fig. 4. In this protocol, one uses the conductance relaxation law $\Delta G(t/t_0) = \Delta G(1\ \text{s}) - a \log(t/t_0)$, where t_0 is the experimental resolution time, and the equilibrium value of G at $V_g = 105$ V to extract the value of τ defined by $G(\tau) \equiv \frac{1}{2} V(1\ \text{s})$.

A characteristic feature that is believed to be common to all intrinsic electron glasses is a memory dip; this is a cusp-

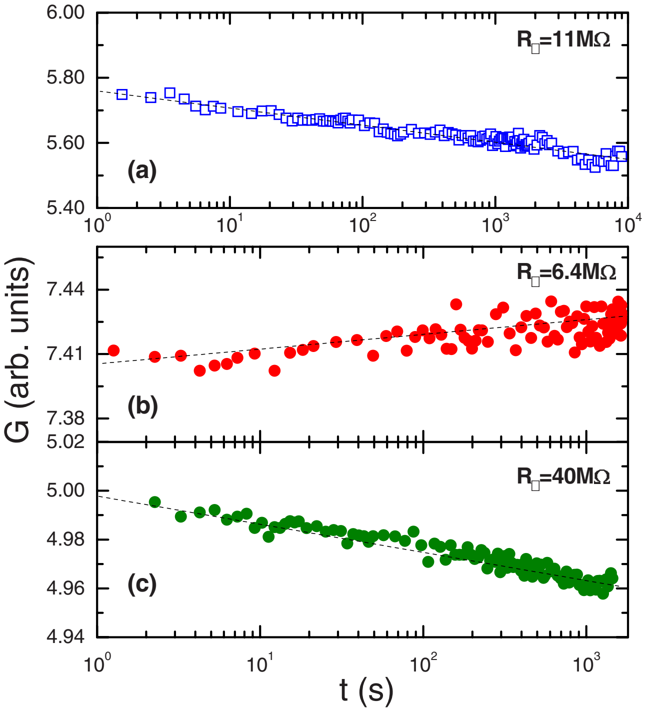

FIG. 3. (Color online) Nonequilibrium transport behavior of typical Be films under different protocols; (a) After a quench cool from $T \approx 100$ K to $T = 4.11$ K. (b) Starting from near-equilibrium, $G(T)$ was recorded from the instance the source-drain voltage was increased to 10 V_{ac} from the (Ohmic) 20 mV_{ac} bias. (c) Same sample as (b) after the 20 mV_{ac} has been restored. Each graph is labeled with the average $R_{\square}$ of a sample under the measurements conditions.

like minimum in $G(V_g)$ centered at the gate voltage V_g, where the system was allowed to equilibrate.[5–7,17] A conspicuous memory dip was consistently observed in all our Be films. Figure 5 shows this feature for two Be samples in the studied series. For both samples, $G(V_g)$ scans were taken after a $\simeq 24$ h equilibration under $V_g = 0$ V. Note first that the memory dips have the same shape and width independent of $R_{\square}$ and independent of whether the $G(V_g)$ scans were taken by sweeping V_g *through* the equilibrium V_g or *symmetrically* around it[13] (cf., the lower graph of Fig. 5).

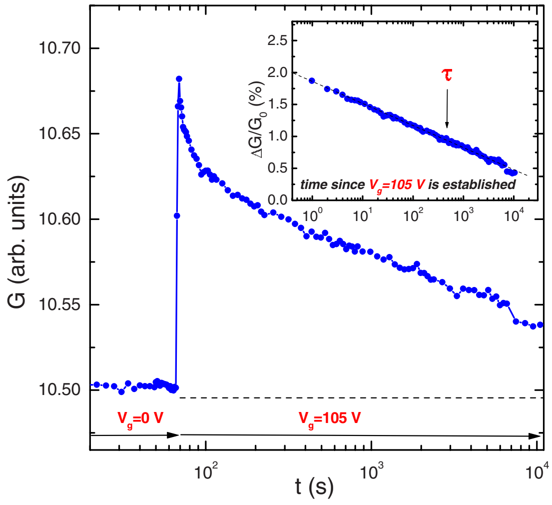

FIG. 4. (Color online) A gate-excitation protocol for a specific Be sample ($R_{\square} = 40$ MΩ). The inset depicts the characteristic log(t) dependence of the EG from which the typical relaxation time τ is estimated using the baseline conductance G_0 (105 V) (marked here by the dashed line) measured independently.

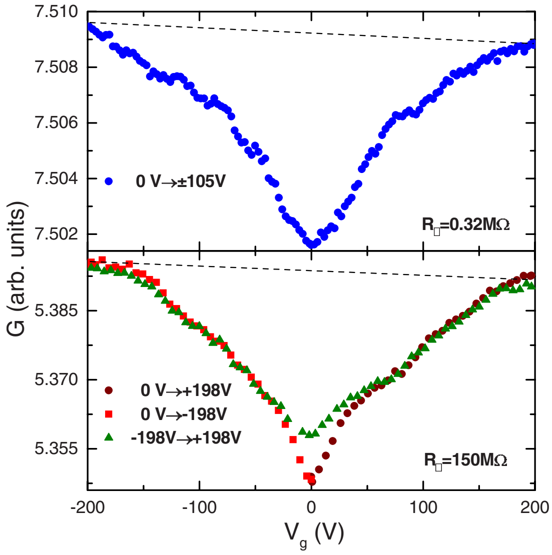

FIG. 5. (Color online) The conductance versus gate voltage for two Be films (labeled by their $R_{\square}$ illustrating the memory-dip structure. After the sample was allowed to equilibrate under $V_g = 0$ for ≈ 24 h, scans were taken either from $V_g = 0$ in each voltage direction [symmetrical $G(V_g)$ scans] or, from -198 to $+198$ V. Note that the structure is skewed due to the contribution of the antisymmetric (equilibrium) field effect (see text).

In previously studied electron glasses the width of the memory dip was found to systematically depend on the carrier concentration n of the system.[17] On the basis of the $G(V_g)$ data, we have estimated the typical width in our Be films in the same manner as was done in Ref. 17. This involves several stages; first the change in charge ΔQ associated with the cusp width is estimated from ΔV_g by taking heed of the sample-gate capacitance. The relevant energy is then calculated using the beryllium $(\partial n / \partial \mu)_{E_F}$ and the screening length. This procedure gave the energy width Γ^* (as defined in Ref. 17) as ≈ 8 meV. Using the empirical relation between the width and n (Fig. 4 of Ref. 17) such Γ^* corresponds to n of order $10^{21} - 10^{22}$ cm^{-3}. This is consistent with our Hall effect measurements that gave $n \cong 7 - 8 \times 10^{21}$ cm^{-3} as well as with the concentration predicted by band-structure calculations ($n = 0.016$ state/atom[18] tantamount to $n \simeq 1.7 \times 10^{21}$ cm^{-3}). This correlation between the width of the memory dip and the carrier concentration of the material is an important empirical test for the intrinsic nature of the electron glass.[5]

The $G(V_g)$ traces (Fig. 5) reveal some mesoscopic fluctuations (reproducible with V_g scans) superimposed on the memory dip (note e.g., the modulation of $G(V_g)$ around $V_g \approx -120$ V, and $+90$ for the 0.32 MΩ and the 150 MΩ samples, respectively). As alluded to above, these presumably result from the small effective volume for conductance due to the presence of the insulating BeO regions.

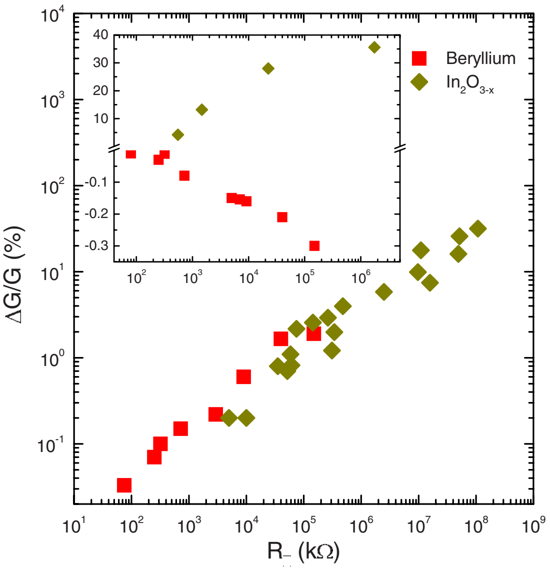

FIG. 6. (Color online) The relative magnitude of the memory dip versus $R_\square$ for the studied Be samples. The data of Ref. 15 are shown for comparison. Insert: The slope (relative change in G per 400 V of V_g) of the antisymmetric part of $G(V_g)$ compared with typical results for In_2O_{3-x} samples.

The relative magnitude of the memory dip $\Delta G/G$ grows monotonously with $R_\square$. Interestingly, $\Delta G/G(R_\square)$ for Be is almost identical to that measured under the same conditions in other electron glasses. In Fig. 6 we compare the results of the current study with some old data[19] taken on In_2O_{3-x} films exhibiting quite similar behavior. A similar agreement is observed between that data of Fig. 6 and the results obtained on granular aluminum films (cf., Fig. 17 of Ref. 20).

On the other hand, the antisymmetric part (cf., Fig. 6) of the $G(V_g)$ for the beryllium samples has the *opposite* slope to that observed in In_2O_{3-x} and In_xO.[5,13] The sign of this slope is controlled by the energy derivative of the thermodynamic density of states $\partial n/\partial\mu$ at the Fermi level; $\partial G(V_g)/\partial V_g \propto \frac{\partial}{\partial E}(\partial n/\partial\mu)_{E_F}$. The antisymmetric part of $\partial G(V_g)/\partial V_g$ we observe in all our Be films (represented by the dashed curves in Fig. 5) is consistent with the negative slope of $\partial n/\partial\mu|_{E_F}$ found in theoretical calculations for the Be band structure.[21] These calculations assumed an ideal Be crystal which might not be relevant for the disordered structure. It is not uncommon however that band-structure features calculated for the perfect crystal persist in the disordered material [as actually observed for some optical properties of Be (Ref. 22)]. The magnitude of the slope depends also on the film resistance as is shown for both In_2O_{3-x} and Be in the insert to Fig. 6. Being a low density system, $\frac{\partial}{\partial E}(\partial n/\partial\mu)_{E_F}$ in In_2O_{3-x} is much larger than in metals, which in turn makes $\partial G(V_g)/\partial V_g$ larger.

It is interesting to note that the currently known electronic systems that exhibit intrinsic glassiness (with associated long relaxation times) are quite diverse in most other aspects. For example, in terms of microstructure, there are in this group representatives of all types of disordered structures; polycrystalline (In_2O_{3-x} and Be), granular or discontinuous (Al, Pb, Au, and Ni), and amorphous (In_xO and Bi). Most of these systems contain oxide, whether as an intrinsic part of the material (In_2O_{3-x}, In_xO), or to stabilize a granular structure (e.g., Al). However, the discontinuous Ni films, being prepared and measured under high-vacuum conditions,[8] is oxygen-free and show the same effects as the other intrinsic electron glasses.[8] In some of these systems there may be local order due to superconductivity (Pb, Bi, the high n version of In_xO, Be, and Al) or magnetism (Ni) at the temperatures of the experiments but not in others. Finally, all these systems obey some form of activated conductivity, $G(T) \propto \exp[-(\frac{T_0}{T})^\alpha]$ with $0.3 < \alpha < 1$, however, no single value of α is singled out in the group. In other words, the conductivity versus temperature law $G(T)$ exhibited by these systems is not due to a specific hopping mechanism. It does not set this group apart from other hopping systems that do not exhibit long relaxation times. Indeed, a $G(T)$ law that resembles is observed in many disordered semiconductors while their relaxation times are very short[23] [presumably due to their low n (Ref. 5)].

In fact, the *only* common feature of the materials that show long relaxation times appears to be their relatively high carrier concentration (in addition of course to being strongly localized thus exhibiting hopping conductivity). The common, out-of-equilibrium features that all these electron glasses exhibit are suggestive of a generic mechanism. The current work supports the conjecture that sluggish relaxation and the associated memory effects of the electron glass is intimately connected with high carrier concentration.[5] This should be the pivotal point of any theoretical model that purports to account for these phenomena.

ACKNOWLEDGMENTS

Z.O. and P.W.A. gratefully acknowledge discussions with the participants of the Electron-Glass program (organized by the Kavli Institute of Theoretical Physics) during which this manuscript was prepared. This research was supported by a grant administered by the U.S. Israel Binational Science Foundation and by the Israeli Foundation for Sciences and Humanities. P.W.A. acknowledges the support of the U.S. Department of Energy under Grant No. DE-FG02-07ER46420.

---

[1] J. H. Davies, P. A. Lee, and T. M. Rice, Phys. Rev. Lett. **49**, 758 (1982).

[2] M. Grünewald, B. Pohlman, L. Schweitzer, and D. Würtz, J. Phys. C **15**, L1153 (1982).

[3] M. Pollak and M. Ortuño, Sol. Energy Mater. **8**, 81 (1982); M. Pollak, Philos. Mag. B **50**, 265 (1984).

[4] A. Amir, Y. Oreg, and Y. Imry, Phys. Rev. B **77**, 165207 (2008); Phys. Rev. Lett. **103**, 126403 (2009).

[5] Z. Ovadyahu, Phys. Rev. B **78**, 195120 (2008).

[6] T. Grenet, Eur. Phys. J. B **32**, 275 (2003); T. Grenet, J. Delahaye, M. Sabra, and F. Gay, Eur. Phys. J. B **56**, 183 (2007).

[7] G. Martinez-Arizala, D. E. Grupp, C. Christiansen, A. M. Mack, N. Markovic, Y. Seguchi, and A. M. Goldman, Phys. Rev. Lett. **78**, 1130 (1997); G. Martinez-Arizala, C. Christiansen, D. E. Grupp, N. Markovic, A. M. Mack, and A. M. Goldman, Phys. Rev. B **57**, R670 (1998).

[8] A. Frydman (private communication).

[9] C. J. Adkins, J. D. Benjamin, J. M. D. Thomas, J. W. Gardner, and A. J. McGeown, J. Phys. C **17**, 4633 (1984). Note however that no time dependence of the effects has been reported in this paper.

[10] G. Ahlers, Phys. Rev. **145**, 419 (1966).

[11] P. W. Adams, P. Herron, and E. I. Meletis, Phys. Rev. B **58**, R2952 (1998).

[12] V. Yu. Butko, J. F. DiTusa, and P. W. Adams, Phys. Rev. Lett. **84**, 1543 (2000); V. Yu. Butko and P. W. Adams, Nature (London) **409**, 161 (2001).

[13] A. Vaknin, Z. Ovadyahu, and M. Pollak, Phys. Rev. B **65**, 134208 (2002).

[14] V. Orlyanchik and Z. Ovadyahu, Phys. Rev. B **75**, 174205 (2007).

[15] V. Orlyanchik, A. Vaknin, and Z. Ovadyahu, Phys. Status Solidi B **230**, 67 (2002); V. Orlyanchik and Z. Ovadyahu, Phys. Rev. Lett. **92**, 066801 (2004).

[16] Z. Ovadyahu, Phys. Rev. B **73**, 214208 (2006).

[17] A. Vaknin, Z. Ovadyahu, and M. Pollak, Phys. Rev. Lett. **81**, 669 (1998).

[18] J. H. Tripp, P. M. Everett, W. L. Gordon, and R. W. Stark, Phys. Rev. **180**, 669 (1969).

[19] M. Pollak and Z. Ovadyahu, J. Phys. I **7**, 1595 (1997).

[20] J. Delahaye, T. Grenet, and F. Gay, Eur. Phys. J. B **65**, 5 (2008).

[21] E. Wimmer, J. Phys. F: Met. Phys. **14**, 681 (1984); G. Pari Vijay Kumar, A. Mookerjee, and A. K. Bhattacharyya, J. Phys.: Condens. Matter **11**, 4291 (1999); R. Boča, P. Hajko, and L. Benko, Czech. J. Phys. **44**, 897 (1994).

[22] O. Hunderi and H. P. Myers, J. Phys. F: Met. Phys. **4**, 1088 (1974).

[23] D. Monroe, A. C. Gossard, J. H. English, B. Golding, W. H. Haemmerle, and M. A. Kastner, Phys. Rev. Lett. **59**, 1148 (1987); V. K. Thorsmølle and N. P. Armitage, *ibid.* **105**, 086601 (2010).