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TITLE: **ON PROPERTIES OF BOUNDARIES AND ELECTRON CONDUCTIVITY IN MESOSCOPIC POLYCRYSTALLINE SILICON FILMS FOR MEMORY DEVICES**

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On Properties of Boundaries and Electron Conductivity in Mesoscopic Polycrystalline Silicon Films for Memory Devices

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Abstract

We present the results of MD modeling on the structural properties of grain boundaries (GB) in thin polycrystalline films. The transition from crystalline boundaries with low mismatch angle to amorphous boundaries is investigated. It is shown that the structures of the GBs satisfy a thermodynamical criterion suggested in [9]. The potential energy of silicon atoms is closely related with a geometrical quantity – tetragonality of their coordination with their nearest neighbors. A crossover of the length of localization is observed To analyze the crossover of the length of localization of the single-electron states and properties of conductance of the thin polycrystalline film at low temperature. we use a two-dimensional Anderson localization model, with the random one-site electron charging energy for a single grain (dot), random non-diagonal matrix elements, and random number of connections between the neighboring grains. The results on the crossover behavior of localization length of the single-electron states and characteristic properties of conductance are presented in the region of parameters where the transition from an insulator to a conductor regimes takes place.

Recently it was demonstrated that thin polycrystalline silicon films are promising materials for future room temperature single-electron devices [1]- [4]. Main reasons which make this material so attractive are the following: I). Usually, the film's thickness varies from 1 to 5 nm, and the average lateral grain size is 10 nm or less. In this case, the energy of an electron in a single grain, E_e, is bigger than the thermal energy even at room temperature, $E_e > T = 300^o K$. II). It is believed that in these films, the characteristic resistance of the potential barriers between the grains is big enough, $R_b > R_c = h/e^2 \sim 25k\Omega$. If both these conditions are satisfied, an electron is strongly localized in the grain. At the same time, one can regulate (up to some extent) the electron conductivity in these films by varying the gate voltage, and creating a current channel. Those electrons which are stored in the grains (storage dots) create a Coulomb repulsion for those electrons which are involved in the current channel. This allows one to implement memory operations in these films at room temperature using a Coulomb blockade effect. Different implementations of these ideas have been discussed, for example, in [1]- [4]. To satisfy both mentioned above conditions, the boundaries between the nanocrystalline grains in these films play important role. For example, one of the most important characteristics of the electron transport in polysilicon films is connected with the distribution of crystalline and amorphous grain boundaries (GB) [5]. At room temperature, the main factor which determines the structure of a GB is a mutual misorientation of the neighboring crystalline grains. At present there does not exist a consistent theoretical approach for the description of the GBs in thin polycrystalline films.

I. MD simulation of grain boundaries at room temperature.

The initial configuration of the polysilicon film was generated using the Voronoi algorithm: first, the centers of the grains were defined; then each grain was obtained by filling the space, nearest to the center of the grain, with the diamond lattice at a chosen orientation. The atoms with very high potential energies were eliminated from the GBs. The film had the dimensions $16.7 \times 16.7 \times 3.1$ nm, and contained 44174 atoms in 12 grains (Fig. 1). The majority of the grains in our model were oriented randomly, so they had both tilt and twist misorientations. Four grains, namely the 3,4,7 and 8-th were generated so that the misorientation tilt angle between the 3-rd and 4-th grains was $\alpha_{3-4} = 11.0^o$; between 4-th and 7-th grains $\alpha_{4-7} = 11.0^o$; and between 7-th and 8-th grains, $\alpha_{7-8} = 15.0^o$. The GB_{3-4} and the GB_{7-8} are close to (1,1,1)-interface, and the GB_{4-7} is close to (0,0,1)-interface. The (1,1,1)-planes in these four grains are perpendicular to the plane of the figure. The average linear size of a grain in our model, $d_{gr} \simeq 4.8$ nm, is of the same order as for the grains in the experimental polycrystalline silicon films obtained recently by thermal annealing of amorphous Si deposited by decomposing silane on SiO_2 substrate [4]. Periodic boundary conditions were used in the X and Y directions, and free boundary conditions were used in the Z - direction. It is shown that the potential energy of the atoms closely correlates with the degree of their tetragonal coordination with the nearest neighbors. To estimate the quality of the tetragonal coordination of the j-th atom, we calculated the value, $\mathcal{T}_j = \sum_{l<k}(l_{jl} - l_{jk})^2)/15\bar{l}_j^2$, which is called the *tetragonality* [7]. Here l_{jk} is the length of k-th edge of the tetrahedron of general shape formed by the four nearest neighbors of the j-th atom; $\overline{l_j}$ is the average length of the edge. By definition, $\mathcal{T}_j$ is equal to zero for an ideal tetrahedron (as it is the case of ideal diamond structure), and increases if the shape of the tetrahedron is distorted. The value, $\mathcal{T}_j$, is correlated with the dispersion of the edge lengths of the tetrahedron. According to the criterion suggested in [7], the spatial figure formed by four points can be recognized as having the "good tetrahedral shape" if $\mathcal{T}_j$ is less than or equal to $\mathcal{T}^{(c)} = 0.018$. The value of tetragonality of an atom is very sensitive to the number of atoms in the first coordination shell, z. In the bulk amorphous phase of silicon, for which $z_a \simeq 4.04$, the average tetragonality is equal to $\mathcal{T}_{am} = 0.015$ which is slightly less than the critical value $\mathcal{T}^{(c)}$. In liquid silicon (in which the first coordination shell consists in average of $z_{lq} \simeq 4.46$ neighbors), the average value of tetragonality is, $< \mathcal{T} >_{lq} \simeq = 0.034$. In Table I, we present the data for the potential energy per atom, density and average tetragonality, for the longest GBs (for which it was possible to make measurements with reasonable accuracy). Densities of the tilt crystalline GBs between the 3, 4, 7 and 8-th grains are practically the same, or slightly smaller, than the density of the bulk crystal, ρ_{cr}. The densities of other GBs are systematically larger than ρ_{cr}. In covalent materials with friable structure, such as silicon and germanium, some GBs, indeed, contract. This was observed in X-ray diffraction experiments for (1,0,0) twist GBs in Ge [8] and in MD simulation of the (1,1,1)-twist boundaries in silicon [10]. It is remarkable that the contraction is observed for the twist boundaries. In such GBs, the covalent bonds are most probably stretched rather than squeezed, and the system demonstrates a tendency to restore their lengths at the cost of a reduction of the GB's volume. MD simulations show that in our model of polysilicon film, the majority of the grain boundaries between the grains with random mutual orientation are disordered. The grains with small mutual misorientations are connected by crystalline grain boundaries. The grain boundaries with "medium-angle" misorientation have a complex inhomogeneous structure. These boundaries consist of crystalline connections interspersed with disordered regions. In disordered regions, the misfit between the crystal structures of the neighboring grains is compensated by non-crystalline arrangements of atoms, such as 5- and 7- fold rings. In general, the structure of the grain boundaries satisfies the thermodynamical criterion suggested in

[9]. The potential energy of the silicon atoms closely correlates with the tetragonality, $\mathcal{T}$, of their coordination with their nearest neighbors. The majority of atoms in the polysilicon grain boundary are well coordinated tetrahedrally, even if they are arranged in a non-crystalline manner in the high-angle grain boundary. From the observed small values of the average tetragonality, it follows that the high-angle disordered grain boundaries can be characterized as amorphous.

The results obtained are important for better understanding of structural properties of thin polycrystalline silicon films, which have been used recently for memory devices. However, for obtaining more reliable data, larger polysilicon simulations should be considered.

II. Crossover behavior of the localization length of single-electron states, and the properties of conductivity.

To model the properties of conductive electrons in thin polycrystalline silicon film at low temperature, we use an approach based on the two-dimensional Anderson localization model. Each nano-silicon grain (with the coordinates (m,n) in (X,Y)-directions) is considered as an individual dot. The characteristic energy of quantization in the Z-direction, E^z, for the lowest level, is of the order, $E^z \sim 1-300$ meV [1,2]. The charging energy of an electron $E_{m,n}^{(charge)}$ in the individual dot is a random variable, and depends on the capacitance $C_{m,n}$ of the dot, $E_{m,n}^{(charge)} = e^2/2C_{m,n}$. The average capacitance of the dot is of the order $< C_{m,n} >\sim$ 1aF. Then, $< E_{m,n}^{(charge)} >\sim 50-70$meV. We assume that the total one-site electron energy is: $E_{m,n} =< E > +\delta E_{m,n}$, where $\delta E_{m,n}$ is a random variable (as the size of the dot fluctuates). At low temperature, the mechanism of electron conductivity is provided by quantum tunneling between the dots, which we describe by the matrix elements, $V_{m,n}^{(m',n')}$. Below we consider only the simplest case of the neighboring transitions: $V_{m,n}^{(m\pm1,n\pm1)} =< V > +\delta V_{m,n}$, where $< V >$ is the average value of the non-diagonal matrix elements, and $\delta V_{m,n}$ describes the fluctuations connected with the random characteristics of the potential barriers between the neighboring dots. Let us now estimate the number of conductive electrons which occupy an individual dot. The maximum value of a 2D electron density in the film can be chosen as: $n_e \sim 10^{11} - 10^{12} cm^{-2}$. Consider a sub-system of conductive electrons in a poly-silicon film as a two-dimensional one, with the size of the active region 100nm×100nm [1,2]. Then, the number of conductive electrons, $\mathcal{N}$, in this region is of the order: $\mathcal{N} = 10-100$. If the average size of the grain (dot) in (X,Y) plane is of the order 10nm, then one has, on average, 100 dots in the active region. So, the average number of conductive electrons in a dot is of the order, 0.1-1. In this paper, we consider the conductive electrons as non-interactive. To describe the electron sub-system at low temperature, we use a 2D Anderson localization model with the Hamiltonian, $\mathcal{H} = \sum_l E_l|l >< l| + \sum_{l,k} V_{l,k}|l >< k|, \quad (E_l = E_1^{(z)} + E_l^{(charge)})$. Fig. 2 shows the dependences of the average length of localization, $< L >$, and the dispersion of the length of localization, $D(L) =< (L- < L >)^2 >$, of the single-electron states, $\Psi^{(i)}(m,n)$, as a function of the average value of the matrix element $< V >$. (The size of the lattice is: (M,N)=(18,12). Zero boundary boundary conditions were chosen.) In Fig. 2, $< E >= 50$ meV; $E_{m,n} \in [50-12.5; 50+12.5]$ meV; $\delta V_{m,n} \in [0.9; 1.1] < V >$ meV. For each state, $i = 1, ..., MN$, the length of localization was introduced as: $L^{(i)} = (L_m^{(i)} + L_n^{(i)})/2$. For $L_m^{(i)}$ we used: $L_m^{(i)} = \sqrt{\sum_{m,n=1}^{M,N} (m - \bar{m}^{(i)})^2 |\Psi(m,n)|^2}$, where $\bar{m}^{(i)} = \sum_{m,n=1}^{M,N} m|\Psi(m,n)|^2$. (Similar expressions were used to calculate $L_n^{(i)}$.) As one can see from Fig. 2, the fluctuations of the average localization length, $D(L)$, exhibit a characteristic maximum at $< V >\approx< V_c >= 3.5$ meV. For $< V ><< V_c >$, the system is more close to the insulator. For $< V >>< V_c >$, the system exhibits metallic properties. We are recently investigating this crossover behavior in connection with the metal-insulator transition (MIT). Finally, in Fig. 3, we present the characteristic results of numerical simulations of the dimensionless conductance, $\sigma = \sum |T_{\alpha,\beta}|^2$, in this system, where $T_{\alpha,\beta}$ is the transmission amplitude from channel α to channel β. The method we used is based on a Green's functions approach [12]. The lattice size is (11,11), with 11 channels in the left and the right electrodes. In Fig. 3, the horizontal axis is the dimensionless value of the diagonal disorder, δE. The dimensionless non-diagonal matrix element was chosen, $V = 1$ in the active region and in the electrodes. The results shown in Fig. 3 correspond to the dimensionless Fermi energy: $E_F = 0$. In Fig. 3 the data indicated by $\diamond$ (a) corresponds to the randomization only of two boundaries: $(n = 1,\ m = 1, ..., M)$, and $(n = N,\ m = 1, ..., M)$. The data indicated by + (b) corresponds to the randomization of the whole active region. One can see that increasing the diagonal disorder leads to a crossover behavior of the conductance in the case when only the boundaries are randomized. In the case presented in Fig. 3, the asymptotic behavior of the data indicated by $\diamond$ for large δE corresponds to $\sigma = 9$; in this case only 9 channels give a contribution to the conductance.

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	GB_{3-4}	GB_{4-7}	GB_{7-8}	GB_{3-6}	GB_{6-7}	GB_{4-1}	GB_{1-5}	GB_{5-2}	GB_{9-10}	GB_{10-11}	GB_{9-12}	$a-Si$	$cr-Si$
U (eV/at), ± 0.04	-4.24	-4.18	-4.20	-4.19	-4.16	-4.17	-4.18	-4.09	-4.21	-4.15	-4.14	-4.15^*	-4.335
ρ (g/cm^3), ± 0.02	2.29	2.31	2.34	2.39	2.39	2.41	2.38	2.36	2.37	2.38	2.38	2.29^*	2.324
$< T >$, ± 0.0003	0.005	0.014	0.011	0.010	0.013	0.013	0.011	0.017	0.012	0.015	0.017	0.015^*	0

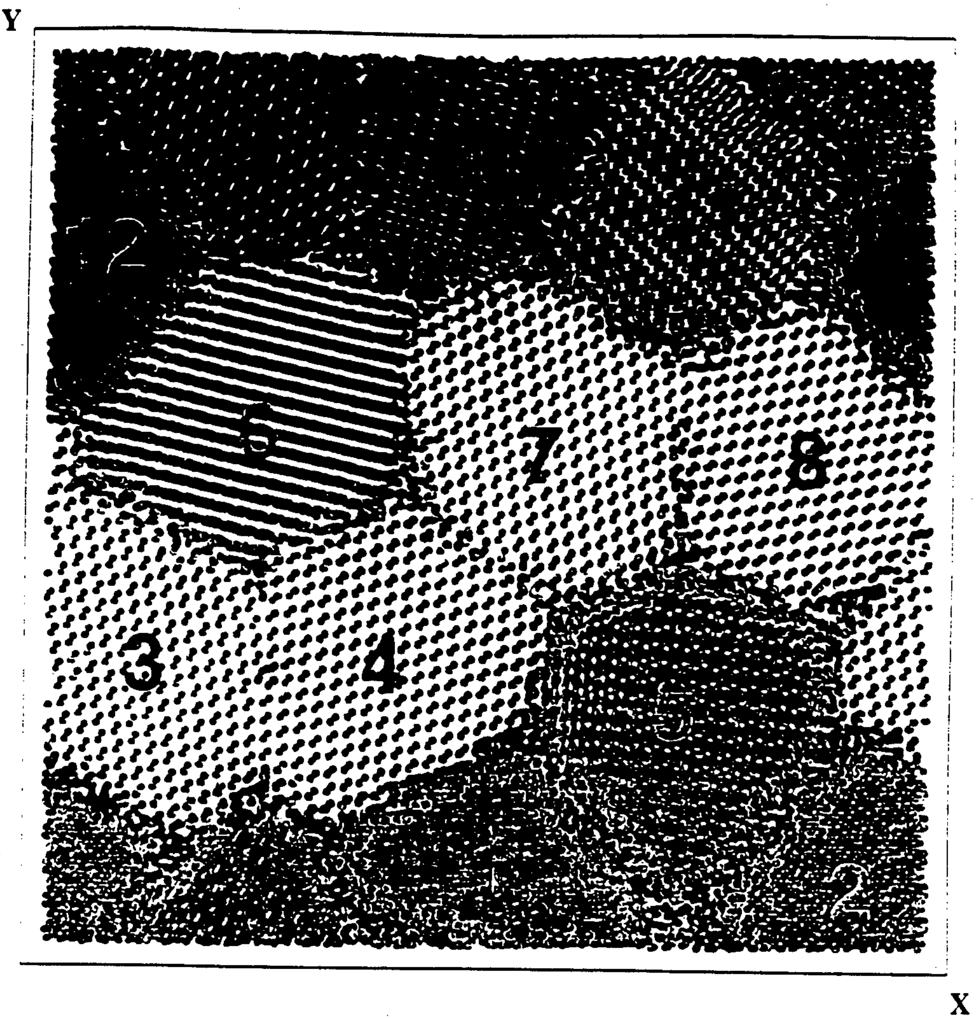
Table I: Potential energy, density and average tetragonality of grain boundaries in the polysilicon model. *Data for a-Si obtained from the model [6]

Figure Captions

Fig. 1. The (X, Y)-projection of the polycrystalline film.

Fig. 2. Demonstration of the crossover behavior of the average localization length of the single-electron states. Dependence of the average localization length $< L >$ (a), and of the dispersion $D(L)$ (b) on the average value of the matrix element, $< V >$. $(M, N) = (18, 12)$; $< E >= 50$ meV; $\delta E = 25$ meV; $V =\in [0.9; 1.1] < V >$ meV.

Fig. 3. Dependence of the conductance, $\sum T_{\alpha,\beta}$, on the disorder parameter δE. The lattice size $(M, N) = (11, 11)$. 11 channels in the left and right electrodes were used. Dimensionless parameters: $V = 1$; $E_F = 0$; (a) Two of the boundaries are randomized: $(n = 1, m = 1, ..., M)$, and $(n = N, m = 1, ..., M)$; (b) The whole active region is randomized.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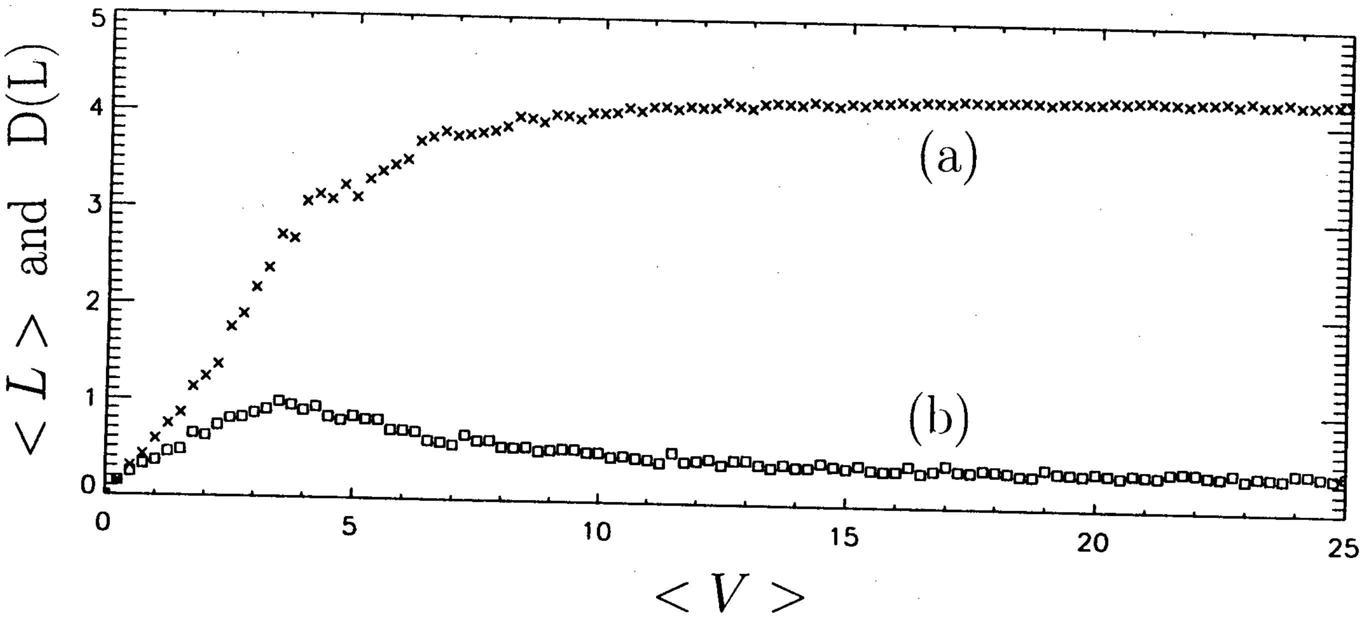

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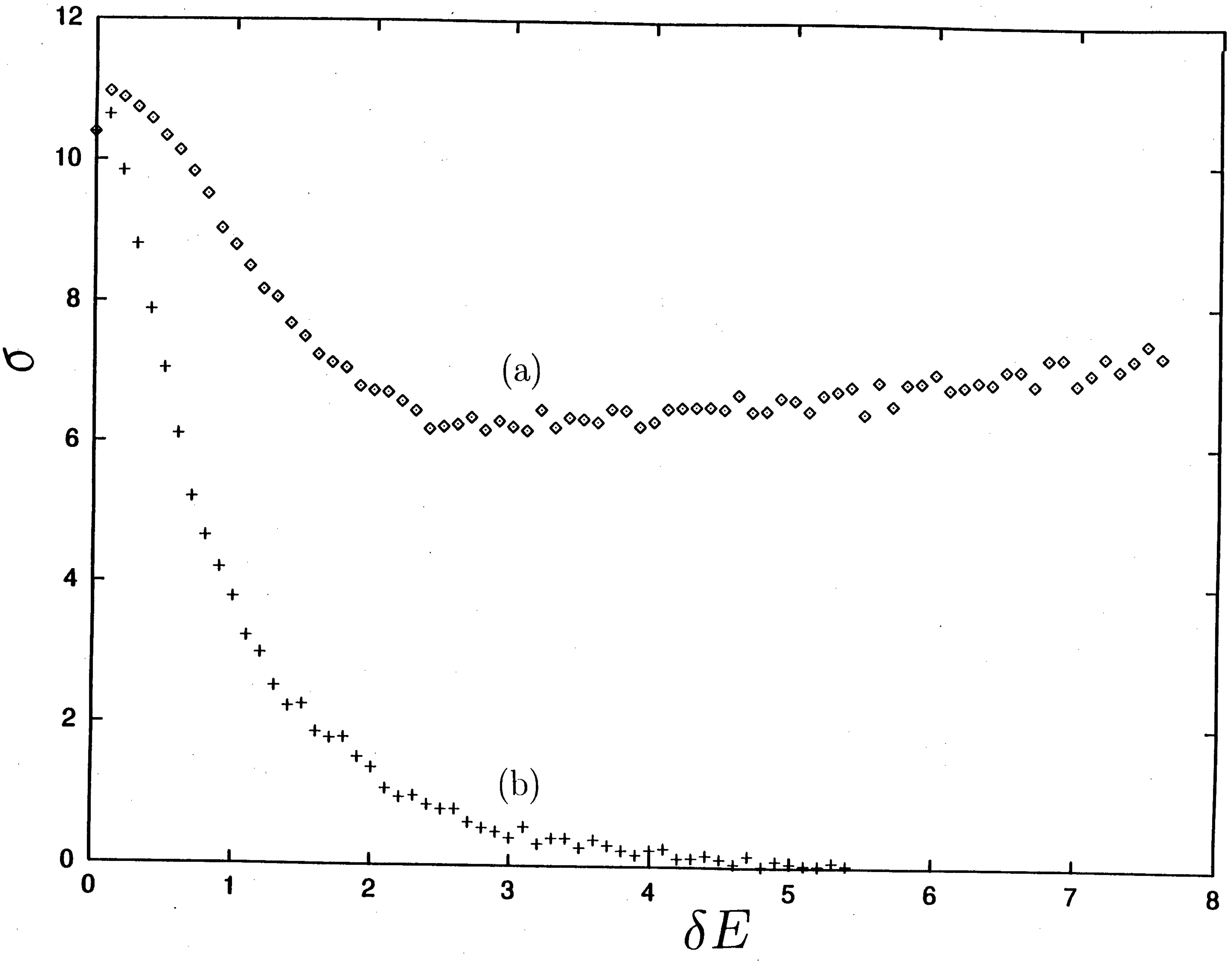

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