Invited paper

Energy concepts involved in MOS characterization

Olof Engström, Tomasz Gutt, and Henryk M. Przewłocki

Abstract— Starting from a quantum statistical reasoning, it is demonstrated that entropy properties of silicon/silicon dioxide interface electron traps may have a strong influence on measured distributions of interface states, depending on measurement method used. For methods, where the Fermi-level is used as a probe to define an energy position, the scale is based on free energy. On the other hand, methods based on thermal activation of electrons give the distribution on an enthalpy scale. It is shown that measured interface state distributions are influenced by the distribution of entropy, and that common features of measured energy distributions may be influenced by entropy variations. These results are used to interpret experimental data on the energy distribution of electron capture cross sections with an exponential increase followed by a more or less constant value as the energy distance of the traps from the conduction band edge increases. Such a relation is shown to be consistent with a situation where the emission and capture processes of electrons obey the Meyer-Neldel rule.

Keywords—C-V technique, capture cross sections, interface states, Meyer-Neldel rule, MOS, thermally stimulated current.

1. Introduction

Techniques commonly used for investigating electrical interface properties of metal-oxide-semiconductor (MOS) structures can be divided into two different classes depending on the energy concepts involved. In the capacitance versus voltage (C-V) [1], charge pumping [2] and conductance techniques [1], the position of the Fermi-level at the oxide-semiconductor interface is used to define the energy scale when determining the position of an interface electron state in relation to the semiconductor band edges. Another class includes methods where the energy position is measured by thermal activation, as for example in a thermally stimulated current experiment [3]. It has been shown that the energy quantities in these two cases are to be interpreted as different thermodynamic potentials; in the first case as free energies, in the second case as enthalpies [4–7]. A third energy scale is governed by quantum mechanical energy level calculations, where only the electronic potentials of the defect systems are taken into account. This corresponds to the electronic eigenvalues of the interface states and gives rise to a third energy scale. In principle, these three different energy quantities are identical only at zero absolute temperature, $T = 0$ K.

When measuring the interface electron state densities, D_{it}, of MOS structures by Fermi-probe technique, often two characteristic features are obtained when D_{it} is plotted as a function of the Fermi-level position at the interface: a U-shaped over-all distribution and two peaks at about 0.3 eV from the conduction and valence bands, respectively [1]. The latter are generally associated with the Pb-center [8]. It has been suggested that the peaks may be influenced by the merging of states on a free energy scale, due to a varying entropy along the interface state distribution [6]. Another common feature is the energy dependence of capture cross sections. In a number of works, a distribution has been found where the capture cross section increases exponentially with increasing energy distance to the conduction band and rolls over to a more or less constant value for energies deeper than about 0.3 eV [9–15].

In the present paper, we study interface electron states by taking into account the influence of total entropy changes when electrons are emitted to and captured from the conduction band. This treatment will be used as a background to explain experimental electron capture data for MOS interfaces. By adding new experimental data to that existing in the literature, we demonstrate that the energy dependence of capture cross sections can be considered as a result of interface states obeying the Meyer-Neldel rule (MNR) [16].

2. Carrier statistics of interface states

2.1. Occupation probabilities in the grand canonical ensemble

The probability, $P(1)$, to capture one electron, for an interface trap capable to hold one electron, can be expressed in the grand canonical ensemble as [17]

$$P(1) = \frac{Z_1 \exp\left(\frac{1\mu}{\mathrm{k}T}\right)}{\sum_{r=0}^{1} Z_r \exp\left(\frac{r\mu}{\mathrm{k}T}\right)}, \tag{1}$$

where μ is the Fermi-level at the interface, k is Boltzmann's constant and T is absolute temperature. Z_r is the partition function of the canonical ensemble and is given by

$$Z_r = \sum_{i=1}^{I} \exp\left(-\frac{E_{i,r}}{\mathrm{k}T}\right) \equiv \exp\left(-\frac{G_r}{\mathrm{k}T}\right). \tag{2}$$

Here $E_{i,r}$ is the eigenenergy of the trap in state i with a maximum of I available states and with r electrons captured. The partition function can also be expressed by introducing free energy G_r as defined in Eq. (2). Using Eq. (2)

in Eq. (1) and introducing the definition of a "free energy level of the trap", G_T, as the free energy difference for $r = 1$ and $r = 0$,

$$G_T \equiv G_1 - G_0\,, \tag{3}$$

one finds

$$P(1) = \frac{1}{1 + \exp\left(\frac{G_T - \mu}{kT}\right)}\,. \tag{4}$$

From this equation, it can be seen that the occupation probability, $P(1)$, of the trap is determined by the relation between the free energy position, G_T, and the Fermi-level, μ. Especially, when these two quantities coincide, the probability of trap occupation is $1/2$. From this we can make the following important statement: *When the Fermi-level is used as a probe to select interface state energy positions, the energy scale of the distribution obtained is based on a free energy*. It has been argued that for deep bulk energy levels this quantity can be identified as the Gibbs free energy [18].

2.2. Thermal emission rates

At thermal equilibrium, the emission of electrons from an interface energy level, G_T, to the conduction band is balanced by a capture process governed by the average thermal velocity of electrons in the condution band, v_{th}, and the capture cross section, σ_n, of electrons according to the following equality:

$$e_n P(1) = v_{th}\sigma_n n\big(1 - P(1)\big)\,, \tag{5}$$

where e_n is the thermal emission rate for electrons from the trap level to the conduction band and n is the concentration

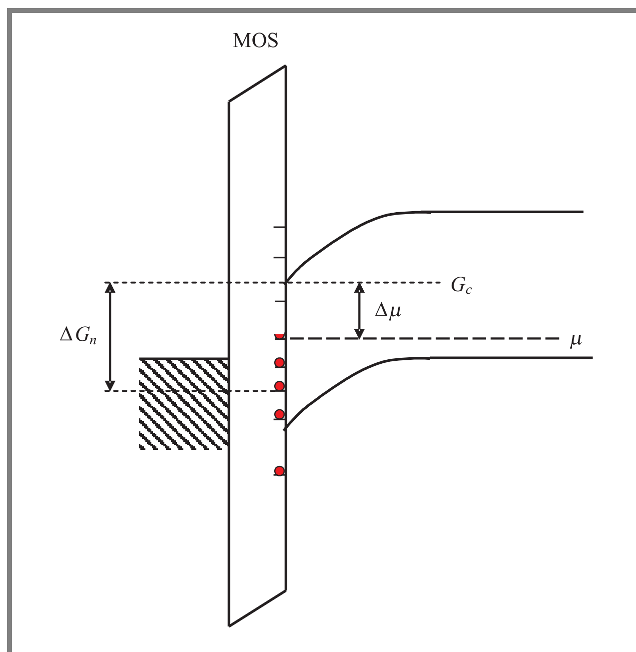

Fig. 1. Band model of the MOS structure on a free energy scale.

of electrons in the conduction band. The latter quantity follows the standard expression

$$n = N_c \exp\left(-\frac{G_c - \mu}{kT}\right), \tag{6}$$

where N_c is the effective density of states in the conduction band and G_c is the free energy of carriers in the conduction band (equal to the "conduction band edge position" at $T = 0$ K), respectively. Introducing the definition $\Delta G_n \equiv G_c - G_T$, as depicted in Fig. 1, we find from Eqs. (5) and (6):

$$e_n = v_{th}\sigma_n N_c \exp\left(-\frac{\Delta G_n}{kT}\right). \tag{7}$$

Taking into consideration that the electron emission from the interface trap system may be connected to a change

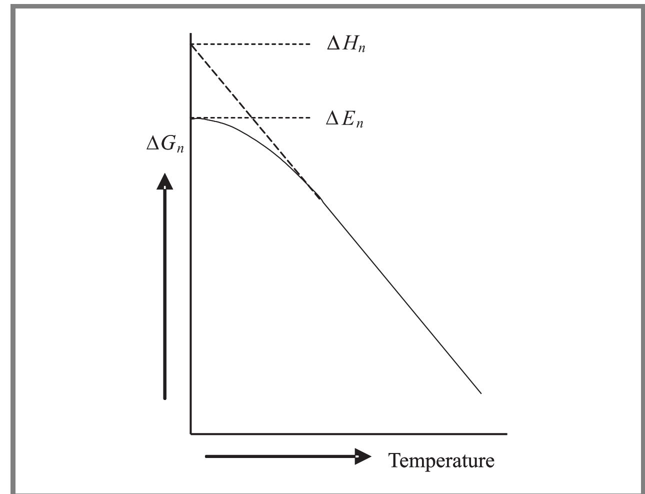

Fig. 2. Temperature dependence of the free energy position, ΔG_n of an interface trap often occurring in semiconductors [19]. For not too low temperatures, $\Delta G_n(T)$ approaches a linear function. As the entropy is $\Delta S_n = -\partial(\Delta G_n)/\partial T$, this quantity is temperature independent for the higher temperatures. At $T = 0$, the free energy and the eigen energy coincide. In an activation measurement, where the slope of an Arrhenius plot is ΔH_n, this quantity is the extrapolated value of ΔG_n in the temperature interval where the measurement is made. For simplicity, in the present treatment, we assume that ΔG_n is linear down to $T = 0$, which makes $\Delta G_n = \Delta H_n = \Delta E_n$ at this temperature.

in entropy, ΔS_n, due to electronic degeneracy and ionic vibration [4, 7, 17, 18], we use the thermodynamic relation:

$$\Delta H_n = \Delta G_n + \Delta S_n T\,, \tag{8}$$

where ΔH_n is an enthalpy. Thus, an alternative expression for Eq. (7) is given by

$$e_n = v_{th}\sigma_n N_c \exp\left(\frac{\Delta S_n}{k}\right)\exp\left(-\frac{\Delta H_n}{kT}\right). \tag{9}$$

In an Arrhenius plot of the emission rate for different temperatures, the slope of the activation curve gives the enthalpy value if the temperature dependencies of the pre-exponential factors in Eq. (9) are known.

For traps in semiconductors, the entropy, ΔS_n, is often temperature dependent [19]. As the thermodynamic relationship between entropy and free energy is $\Delta S_n = -\partial \Delta G_n / \partial T$, this means that the temperature dependence of ΔG_n is nonlinear. In practice it exhibits a shape shown schematically in Fig. 2. In the present treatment, we assume for simplicity, that ΔS_n is temperature independent, meaning that ΔG_n is a linear function of T. For such a case it is readily shown from Eq. (2) that the mean value of the occupied eigenenergy levels, $E_{i,r}$, fulfills a relation to the free energy and the entropy as given by Eq. (8). For a one-electron trap with only one energy level, this means that the energy eigenvalue distance from the conduction band, ΔE_n of this level fulfills the relation:

$$\Delta E_n = \Delta G_n + \Delta S_n T \,. \tag{10}$$

Hence, the enthalpy and the energy eigenvalue are identical for this specific case.

2.3. Influence of entropies on the energy distribution of interface states

When measuring D_{it} by Fermi-probe technique, the energy resolution is determined by a weight function [4], $w = \partial P(1)/\partial(\Delta G_n)$, which is obtained from Eq. (4) as

$$w(\Delta G_n) = \frac{1}{\mathrm{k}T} \frac{\exp\left(\frac{\Delta\mu - \Delta G_n}{\mathrm{k}T}\right)}{\left\{1 + \exp\left(\frac{\Delta\mu - \Delta G_n}{\mathrm{k}T}\right)\right\}^2}, \tag{11}$$

where $\Delta\mu = G_c - \mu$.

The interface state density distribution on a free energy scale is then given by

$$D_{it}(\Delta G_n) = \int_0^{\Delta E_g} D_{it}(\Delta E_n)\left[w\left(\Delta G_n(\Delta E_n)\right)\right] d(\Delta E_n), \tag{12}$$

where the function $\Delta G_n(\Delta E_n)$ is given by Eq. (10) and ΔE_g is the semiconductor energy band gap.

In order to study the influence of a varying entropy, we assume that D_{it} calculated on an energy eigenvalue scale is constant. We will see that changes in the entropy distribution make clear changes in D_{it} when based on a free energy scale. As an example, taking the entropy distribution on an eigenenergy scale as shown in Fig. 3a, the corresponding interface state distribution is demonstrated in Fig. 3b on a free energy scale. Here we notice how $D_{it}(\Delta G_n)$ is merged towards the conduction band by the $\Delta S_n(\Delta E_n)$ function chosen in Fig. 3a. This shape of $\Delta S_n(\Delta E_n)$ gives the standard features often observed for D_{it} as measured by C-V technique: the increased interface state concentration close to the band edge and a peak at about 0.3 eV marking the Pb-center. The position of the 0.3 eV peak depends on the value of the constant part of the entropy curve in Fig. 3a while the step at about $\Delta E_n = 0.3$ eV determines the position of the flank at lower energies.

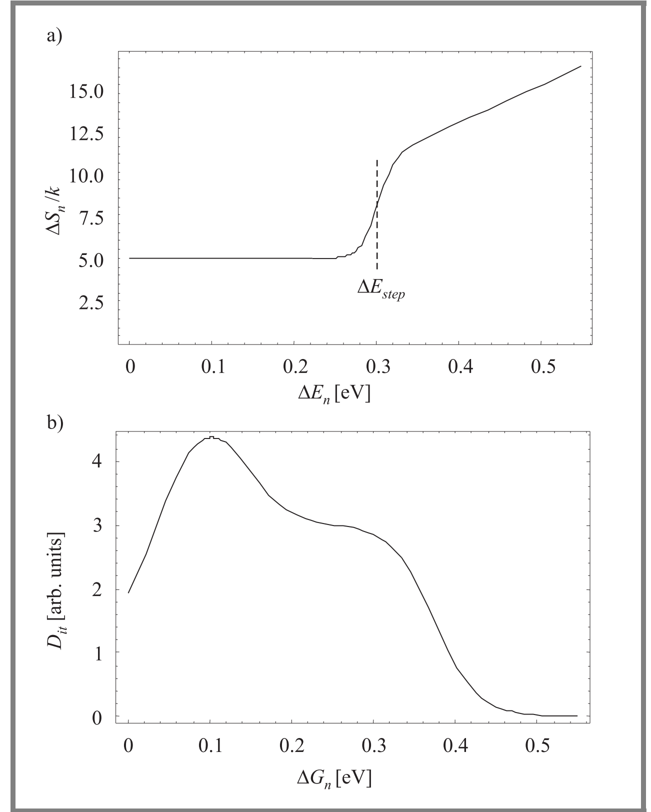

Fig. 3. Entropy in units of k as a function of eigenenergy (a) used for calculating the interface state density, D_{it} as a function of free energy (b) for a D_{it} distribution which is constant on an eigen energy scale.

2.4. Interface traps following the Meyer-Neldel rule

Considering a set $\{j\}$, constituting a "family" of electron traps with properties given by $(e_j, \sigma_j, \Delta S_j, \Delta H_j)$, the thermal emission rate for each member, j, can be expressed as

$$\begin{aligned} e_j &= v_{th}\sigma_j N_c \exp\left(\frac{\Delta S_j}{\mathrm{k}}\right)\exp\left(-\frac{\Delta H_j}{\mathrm{k}T}\right) \\ &= v_{th}\sigma_j N_c \exp\left(-\frac{\Delta G_j}{\mathrm{k}T}\right), \quad j = 1, 2, 3, \ldots \end{aligned} \tag{13}$$

Assuming that there exists an "iso-kinetic" temperature T_c such that the emission rate, e_c, is equal for all $j = 1, 2, 3, \ldots$, we have for $e_c \equiv e_j(T_c)$:

$$\begin{aligned} e_c &= v_{th}\sigma_j N_c \exp\left(\frac{\Delta S_j}{\mathrm{k}}\right)\exp\left(-\frac{\Delta H_j}{\mathrm{k}T_c}\right) \\ &= v_{th}\sigma_j N_c \exp\left(-\frac{\Delta G_j}{\mathrm{k}T_c}\right). \end{aligned} \tag{14}$$

For an Arrhenius plot, this means that all activation curves would intercept at the iso-kinetic temperature, T_c, with a common emission rate, e_c. Such a feature indicates that the family, $\{j\}$, obeys the Meyer-Neldel rule [16]. From Eq. (14), we find

$$\ln\sigma_j = \ln\frac{e_c}{v_{th}N_c} + \frac{\Delta G_j}{kT_c}. \qquad (15)$$

Hence, for a set of interface traps following the Meyer-Neldel rule, one would expect the logarithm of the capture cross sections to be a linear function of the free energy, ΔG_j. Such experimental data are commonly found for SiO_2/Si interfaces [9–15] and will be further discussed below.

From Eq. (15) it is also seen that increasing the iso-kinetic temperature, T_c, to high values makes the capture cross section less dependent on the free energy, ΔG_j, which may explain the constant range for capture cross sections often found for the deeper part of the D_{it} distribution of MOS interfaces, at energies larger than about 0.3 eV [9–15]. A second possibility would be that the enthalpy for releasing an electron from the trap is the product between T_c and ΔS_n, i.e., corresponds to the total heat for releasing the electron at this temperature:

$$\Delta H_j = \Delta S_j T_c. \qquad (16)$$

Such a relation has been discussed for deep bulk levels by Van Vechten and Thurmond [18]. As a motivation for MNR to be valid for interface states, it was also suggested by Johnston *et al*. [20] and was further used in [21] to explain the existence of MNR among a large number of bulk traps in GaAs. It means that the entropy is an increasing function of enthalpy or, in the theoretical treatment above, with the eigenenergy difference, ΔE_n. For a case where Eq. (16) is valid, we find from Eq. (13) that $\Delta G_j = 0$ which means that all traps of the family have the same capture cross section $\sigma_j \equiv \sigma_c$ for all $j = 1, 2, 3, \ldots$ However, for the present case we find from the slopes of the curves in Figs. 4 and 5 that $T_c \approx 300$ K, which is the temperature where the measurements were made. As ΔG_j is an independent parameter controlled by the applied gate voltage in this experiment, the former interpretation for the constant capture cross sections at higher energies is the most probable.

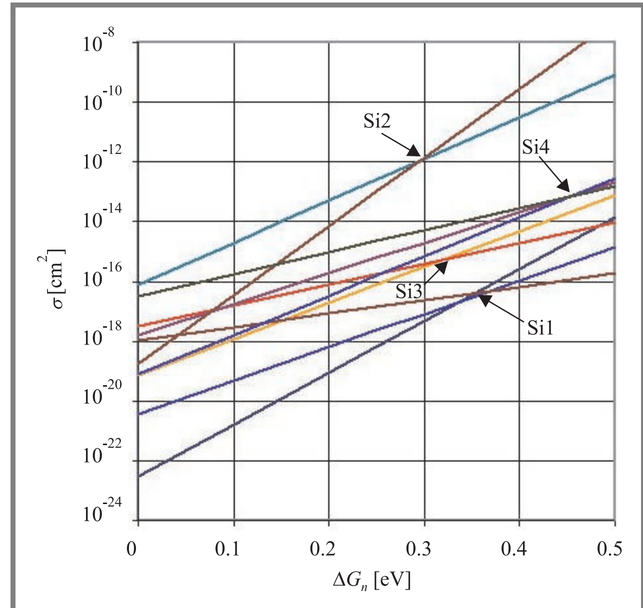

Fig. 5. Capture cross section versus free energy taken from the literature, showing the interception of curves. Each curve represents one interface state "family". Data intercepting at points marked in the diagram represent a "dynasty". The slopes of the curves give the iso-kinetic temperatures and the intercepts with the ordinate axis give the e_c values (Eq. (14)).

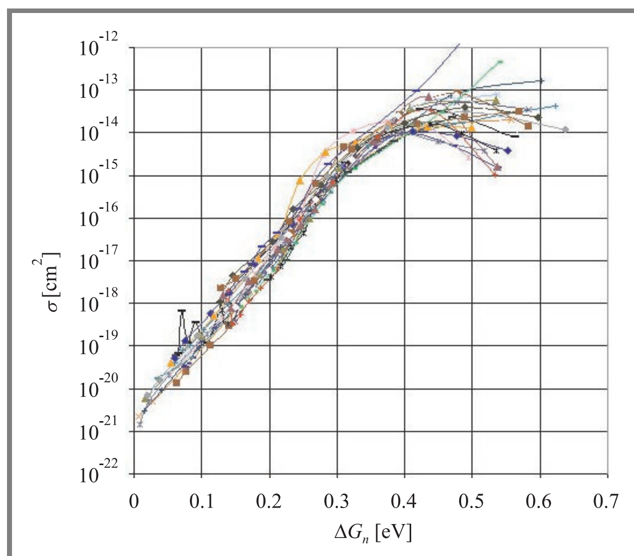

Fig. 4. Electron capture cross sections of Si/SiO_2 interface states as a function of free energy position in relation to the conduction band.

3. Experimental

In order to confirm the results earlier published in the literature [9–15], we repeated measurements on thermally oxidized MOS structures by using the conductance technique [1]. The samples were prepared by oxidizing 4-inch, n-type, phosphorus doped silicon wafers at 1000°C in an N_2/O_2 flow with 31/45 min ramp-up/ramp-down rates to obtain SiO_2 thicknesses of $t_{ox} = 60$ and 160 nm. The wafers were annealed afterwards in nitrogen for $t(N_2) = 0$, 10, 120 and 1440 min at the temperature $T = 1050$°C with 40/55 min ramp-up/ramp-down rates. Then, 416 nm of aluminium was deposited, followed by the standard post-metallization annealing at 450°C for 20 min in N_2/H_2.

Figure 4 shows the capture cross section for electrons as a function of free energy distance from the conduction band for a large number of samples taken from this series of wafers. For the lower energy part, the capture cross sections increase exponentially over more than six orders of

magnitude. At $\Delta G_n = 0.4$ eV, the increasing feature rolls off and becomes either close to constant or decreases with increasing energy. Often in the literature, a more constant behavior is observed in this energy range [9–15].

Figure 5 depicts corresponding data for the low energy part, redrawn from literature [9–15]. An interesting feature of these results is that the different families, representing different MOS systems, in a couple of cases seem to cluster into common crossing points at about 0.3–0.4 eV. From the slopes of the curves, the iso-kinetic temperature T_c can be calculated while the intercepts at the ordinate axis gives the emission rate e_c, in accordance with Eq. (14). Each family has its own values of these quantities but, interesting enough, in some cases they seem to cluster into "dynasties", i.e., at one common ΔG value some families have one common σ value. Moreover, the different families originate from the results of different authors! The data indicate that for each dynasty there exists at least one common free energy value, ΔG_j with a common capture cross section, governed by the crossing points in Fig. 5. This means that plotting the e_c values versus the inverse T_c values in an Arrhenius plot would give one activation curve for each dynasty. As seen in Fig. 6, this is indeed the case. Because all ΔG_j values at the crossing points in Fig. 5 were taken at about the same room temperature, the slope of the activation plots in Fig. 6 is expected to be equal to the free energies as deduced from Eq. (13).

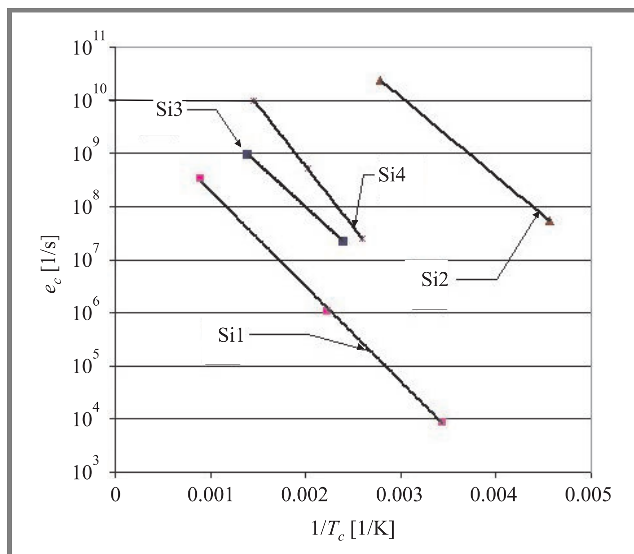

Fig. 6. Dynasties represented in an Arrhenius plot with data taken from Fig. 5.

4. Discussion

The theoretical $D_{it}(\Delta G_n)$ curves in Figs. 3a and 3b were based on an assumption that the value of $D_{it}(\Delta E_n)$ is constant. There is no experimental or theoretical support for such an approach. The idea is to demonstrate the strong influence of entropy variation among the interface states on measured $D_{it}(\Delta G_n)$ distributions. Such consideration is seldom taken into account when comparing theoretically calculated $D_{it}(\Delta E_n)$ data with measured $D_{it}(\Delta G_n)$ results. Theoretical calculations of, for example, the U-shaped interface state distribution by Sakurai and Sugano [22] or of the Pb-center by Edwards [23] were made for eigenenergy scales. Even if these results demonstrate the occurrence of specific distribution characteristics, it is the aim of the present work to point out the influence of electron state degeneracies and ionic vibrational changes [7], together manifesting the total entropy of interface states.

The entropy distribution as shown in Fig. 3a, with a constant value for lower energies and a linear behavior for energies above about 0.3 eV, gives rise to a $D_{it}(\Delta G_n)$ distribution which qualitatively agrees with the features found from Fermi-probe measurements like the C-V technique. Furthermore, it gives a motivation for the shape of the free energy distribution of capture cross sections normally found at SiO_2/Si interfaces by assuming that this kind of states belong to "families" obeying the Meyer-Neldel rule. A fascinating observation is that these families can be portioned into clusters of "dynasties", including centers with equal capture cross sections at equal free energy positions. The condition for making up a dynasty is that one capture cross section is identical for the members of each family. An interesting observation in Figs. 5 and 6 is that this occurs at ΔG_n values of about 0.3–0.4 eV, where the Pb-center is normally found. This would mean that centers giving rise to what is normally labeled as the "Pb-center" may have widely different capture cross sections as given by the interception points between the curves in Fig. 5.

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