Evidence for the defect-pool model from induced recombination level shifts in undoped *a*-Si:H

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The measured dependence of the mobility-lifetime products for electrons and holes, on the relative position of the dark Fermi level E_F in *undoped* hydrogenated amorphous silicon, *a*-Si:H, is reported here. The corresponding measurements, which also included the E_F dependencies of the light-intensity exponents γ and S, became feasible due to the use of a combination of metal-oxide-semiconductor and photocarrier grating configurations. We found that the experimental results of the $S(E_F)$ dependence are very helpful in determining which of the models proposed for the recombination-center distribution in the mobility gap of *a*-Si:H is the most appropriate one.

Since the measured values of the mobility-lifetime $\mu\tau$ products in an insulator may be a result of numerous recombination scenarios,[1] one needs many independent types of data in order to confirm that a particular model scenario is pertinent to the system under investigation. In hydrogenated amorphous silicon, *a*-Si:H, the two types of experimental data commonly used for this evaluation were the light-intensity dependence[2,3] and the temperature dependence[4,5] of the photoconductivity. However, the results obtained by these methods were not good enough even for the determination of the gross features of the energy distribution of the dominant recombination centers. In fact, in the early works these dependencies were interpreted in terms of very different recombination-center distributions.[2-5] A useful concept introduced into some of the above studies, as another handle for the evaluation of the "correct" model, was the effect of the position of the equilibrium ("dark") Fermi level E_F on the photoconductivity and its light-intensity dependence.[2,3] The variation of E_F enables us to scan the forbidden gap, and thus to change the *a priori* equilibrium occupation of the recombination centers in the gap.[2,3] Indeed, it appears now that this approach is quite useful and one can get some of the model predictions to fit quantitatively the experimental data.[2,3,6,7] On the other hand, the agreements obtained depend on the parameters chosen in the models, and as such they do not yield convincing evidence for a specific scenario. In particular, we note that very different deep-level models have been claimed to account quantitatively for the experimental data.[2-7]

Since the recombination process is a two carrier process, and since the photoconductivity[8-11] is determined essentially by the mobility-lifetime product of the majority carriers (electrons in undoped *a*-Si:H), it is to be expected that the determination of the minority carrier mobility-lifetime product can provide additional, complimentary, information regarding the recombination process in *a*-Si:H. The development of the photocarrier grating (PCG) technique[8-10] has indeed enabled[10,11] the measurement of the effect of the E_F shift on the $\mu\tau$ products of the two carriers. Again, two different discrete level models, which were used previously[6,12] for the interpretation of the photoconductivity data, have been suggested[13,14] for the interpretation of the experimental results. However, the relative shift of E_F, in the studies reported thus far[10,11] for the $\mu\tau$ products of the two carriers was induced by *doping*. This approach has a serious drawback when one is trying to learn about the electronic structure of undoped *a*-Si:H, for which the available models have been suggested,[13,14] since it is well known[15,16] that such doping (in concentrations necessary to shift E_F) does not only significantly increase the midgap defect density, but it changes also the energy profile of the state distribution. Moreover, it is the undoped material which is better characterized in the experimental studies (the "device quality" material) and for which there is a much better theoretical understanding.[15] To overcome this major drawback one can use a metal-oxide-semiconductor (MOS) structure, such that the applied gate voltage enables a relative shift of the localized defect levels with respect to E_F without altering the (undoped *a*-Si:H) material. Indeed this approach has been used already[7,17] for the study of the dependence of the mobility-lifetime product of the electrons on E_F, in undoped *a*-Si:H. As we show below, however, this dependence does not provide enough information for the purpose of narrowing down the choice of "correct" recombination level models.

In this paper we report results obtained by the combined application of the MOS and PCG configurations, which enabled *simultaneous* study of the mobility-lifetime product of the electrons, $(\mu\tau)_e$, and the holes, $(\mu\tau)_h$, and their light-intensity exponents γ and S (see below), as a function of E_F in *undoped* *a*-Si:H. The results show that the anticorrelations and correlations between these two sets of quantities do indeed yield additional information. The conclusion derived from these behaviors, although semiqualitative in nature, is, for the reasons given above, more convincing than the "quantitative" agreement of the measured $\mu\tau$ products with the suggested model calculations.[13,14]

The basic MOS structure used in this study was similar

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to the ones described in previous studies of the corresponding photoconductivity.[7,17] In this structure, as illustrated in Fig. 1(a), an n^+ crystalline silicon wafer was thermally oxidized to form a 0.2-μm-thick SiO_2 layer. On this layer a 1-μm (or a 0.5-μm) -thick film of device quality undoped a-Si:H has been deposited using rf glow discharge decomposition of silane.[18] The gate electrode, to which the gate voltage was applied, consisted of In which was soldered onto the etched Si wafer, and the source and drain contacts, which were electron-beam evaporated on top of the a-Si:H film, were made of NiCr. The separation between the contacts was 0.4 mm, to ensure that the applied He-Ne laser beam, which has a diameter of 2.5 mm, will yield a uniform illumination between them. The measurement in this configuration was carried out with a source-drain voltage of 0.5 V. The experimental setup of the PCG was described,[8-10] and the relation between $(\mu\tau)_h$ and the measured ambipolar diffusion length L (see below) was discussed,[9,10] in previous works. The important point in the present context is that the carrier grating was obtained by having two coherent He-Ne laser beams interfere between the coplanar source and drain contacts as indicated in Fig. 1(b).

In the MOS structure the heavily doped Si crystal serves as a "metallic gate" and the effect of the forward gate bias ($V_G > 0$) is to bend the bands and localized levels "down" as shown in Fig. 1(c). While this band bending has been discussed in detail previously,[7,17] no discussion was presented thus far regarding the effect of the above, band and localized level, bending on the occupation of the recombination centers. This effect is therefore considered here in some detail. For simplicity let us assume one type of recombination center in the pseudogap, located at E_r, which (due to atomic forces) "bends in parallel" to the band edges, E_c and E_v. We have chosen E_r to lie below E_F, since this is the common conclusion from previous works,[3,12,14] and also since this is the only way to get a consistent picture between our data and previous data (see below). Considering a previous analysis of the space-charge region[7] and the agreement of the theoretically predicted[1,3] variation of γ from 1 to 0.5, with the experimentally observed[4,16,17] variation of this exponent with V_G (as we also find below), we can scale the band bending. Our forward bias variation of V_G between $V_G = 0$ and 6 V corresponds to a relative band bending at the a-Si:H/SiO_2 interface of about 0.3 eV, i.e., to an increase of $\Delta = E_F - E_r$ by about 0.3 eV at this interface. Under this forward bias E_r is lowered with respect to E_F, which is equivalent (apart from the objection discussed above) to raising E_F by increasing the phosphorus doping, or to lowering E_F by decreasing the boron doping (see below). Under reverse bias, in the -6 V $\leq V_G \leq 0$ range, photoelectron depletion takes place at the surface, and thus one actually measures the photoconductivity of the bulk where Δ is hardly effected by variations in V_G.

Since a one-level model is the *simplest* recombination center model possible, let us first consider the expectations for the behavior of the corresponding system as a function of the separation $\Delta = E_F - E_r$, assuming that the concentrations of photogenerated free electrons and free holes are much smaller than the concentration of recombination centers, N_r. For simplicity and brevity we outline these expectations following an idea discussed many years ago by Rose.[19] A more detailed discussion of the system, which is not necessary for the present work, will be given elsewhere.

The conductivity of a semiconductor is given by

$$\sigma = q(\mu_e n + \mu_h p) , \tag{1}$$

where q is the magnitude of the electronic charge, μ_e (μ_h) is the band mobility of the electron (hole), and $n(p)$ is the free-electron (hole) concentration. In a photoconductor in which the charge carrier concentrations in the dark are negligible, one obtains that in the steady-state conditions established under uniform illumination

$$n = G\tau_e \tag{2}$$

and

$$p = G\tau_h , \tag{3}$$

where G is the (illumination excited) electron-hole pair generation rate and τ_e (τ_h) is the free-electron (hole) lifetime (or recombination time). Under the domination of one carrier in the transport (say, the electrons) the photoconductivity [see Eqs. (1) and (2)] is given by

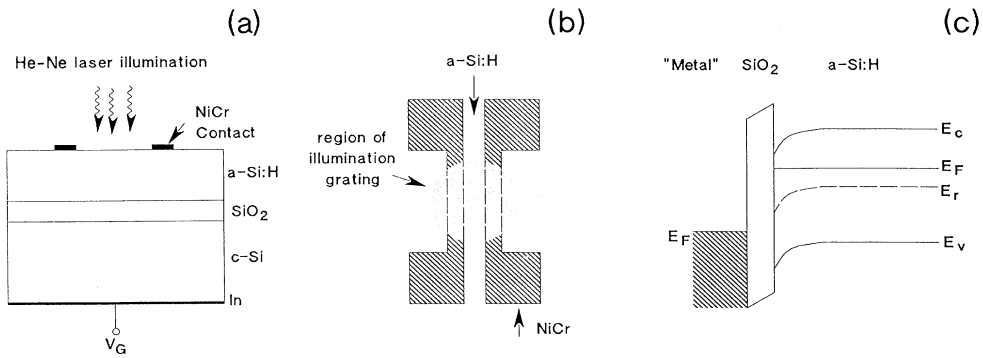

FIG. 1. The MOS structure (a) and the PCG configuration (b) used in the present study. Also shown (c) is a schematic illustration of the a-Si:H bent bands and localized level, under the application of forward bias ($V_G > 0$).

$$\sigma = q\mu_e\tau_e G \equiv q(\mu\tau)_e G \ , \tag{4}$$

where $(\mu\tau)_e$ is the electron mobility-lifetime product $(\mu_e\tau_e)$. The experimental determination of the latter product, i.e., that of the majority carrier, is essentially straightforward from the measured $\sigma/(qG)$ ratio. In contrast, the determination of the minority-carrier mobility-lifetime product $[\mu_h\tau_h \equiv (\mu\tau)_h]$ requires more elaborate procedures.[9] In our case this procedure is the PCG measurement in which the ambipolar diffusion length L is determined. The value of L gives essentially the $(\mu\tau)_h$ product via the relation[9,10]

$$L^2 = (2kT/q)(\mu\tau)_h \ , \tag{5}$$

where kT is the thermal energy. Since the band mobilities are independent of the carrier concentration [and therefore of G, see Eqs. (2) and (3)] the dependencies of σ [or $(\mu\tau)_e$] and L [or $(\mu\tau)_h$] on G and E_F are essentially the corresponding dependencies of τ_e and τ_h.

In the case of a single (deeply lying) recombination level, E_r, such as the one illustrated in Fig. 1(c), the electron lifetime is well known to be given by[1]

$$\tau_e = (p_r v_e s_e)^{-1} \ , \tag{6}$$

where p_r is the steady-state concentration of electron unoccupied (or hole occupied) centers, v_e is the thermal velocity of the electrons, and s_e is the effective cross section for the electron capture by an electron-empty center. Similarly, the lifetime of the hole is given by

$$\tau_h = (n_r v_h s_h)^{-1} \ . \tag{7}$$

Since $v_e s_e$ and $v_h s_h$ are constants the variations of τ_e and τ_h with changing G and E_F are the variations due to the dependencies of p_r and n_r on these parameters.

Let us follow now the variations of p_r and n_r due to a shift of E_F with respect to E_r (or vice versa). If these two levels coincide (i.e., $\Delta = 0$) we have that in the dark (i.e., in equilibrium) $p_r \approx n_r \approx N_r/2$. Recalling our assumption of low enough illumination intensities (a low enough value of G) such that $n,p \ll N_r$, the changes in p_r or n_r, due to the capture of photogenerated electrons and holes, are minute. Correspondingly, as is apparent from Eqs. (6) and (7), τ_e (and thus σ) and τ_h (and thus L) are constants independent of G. If we now define the light-intensity exponents γ and S according to

$$\sigma = q(\mu\tau)_e G \propto G^{\gamma} \tag{8}$$

and

$$L \propto (\mu\tau)_h^{1/2} \propto G^{-S} \ , \tag{9}$$

we get that in the $\Delta = 0$ case, $\gamma = 1$ and $S = 0$.

Turning to the other extreme, i.e., that E_F lies at least a few kT above E_r, we have that under equilibrium almost all the states at E_r are occupied by electrons. Hence the concentration of the captured electrons n_{r0} is very close to N_r while, due to the exponential decrease of p_{r0} with Δ/kT, the concentration of captured holes p_{r0} is very small in comparison with N_r. Under illumination, even though $n,p \ll N_r$, we have that for large enough Δ/kT, the value of n may approach or surpass the value of p_{r0}. If we assume that the recombination centers are neutral when occupied by electrons, and positively charged when empty of electrons (or occupied by holes), the requirement of charge neutrality means that $n = p + p_r$. However, since $n_r \gg p_r$ we get from Eqs. (6) and (7) that $\tau_h \ll \tau_e$ and thus p is negligible with respect to n. Hence the neutrality condition is essentially

$$n \approx p_r \ . \tag{10}$$

Substitution of Eq. (10) in Eq. (6) yields that $\tau_e \propto 1/n$, and thus from Eq. (2) one finds that

$$n \propto G^{1/2} \ , \tag{11}$$

i.e. [according to definition (8)], that $\gamma = \frac{1}{2}$. On the other hand, since $n_r \approx N_r$ for the entire G range under consideration, τ_h does not vary with G [see Eq. (7)] and correspondingly L is a constant, yielding, according to definition (9), that $S = 0$.

From the knowledge of the extreme cases outlined above we can follow the behaviors of τ_e, τ_h, γ, and S with the increase of Δ from $\Delta = 0$ to Δ of a few kT. The corresponding increase by a factor of about 2 of n_r during this Δ increase means [see Eq. (6)] that τ_h will decrease by a factor of 2. On the other hand, since $n_r \gg n,p$, the value of τ_h will be independent of G, i.e., $S = 0$, *throughout* the entire Δ range. The exponential decrease of p_r with Δ/kT will yield at first a fast increase of τ_e [see Eq. (6)], then, the charge neutrality condition will dominate the increase of p_r thus yielding a slowing down in the increase of τ_e. Correspondingly [see Eqs. (8) and (11)] γ will decrease from $\gamma = 1$ to $\gamma = \frac{1}{2}$.

For completeness let us consider the same variation of the above-discussed forward bias but in the case where E_r lies a few kT above E_F (i.e., the case where $\Delta < 0$ and where the holes are the majority carriers) [see Eqs. (6) and (7)]. The application of the same forward bias will now cause a variation of $|\Delta|$ from a few kT to $\Delta = 0$, and will have the effect of bringing E_r and E_F together. Correspondingly, the behavior of the above two $\mu\tau$ products will reverse. There will be a decrease of p_r from almost N_r to about $N_r/2$ and thus a factor of 2 increase in $(\mu\tau)_e$. Simultaneously there will be a strong decrease of $(\mu\tau)_h$. This behavior, as can be gathered from the $\Delta > 0$ case discussed above, will yield that $(\mu\tau)_e$ will be independent of G while $(\mu\tau)_h$ will be dependent on G. Returning to Eqs. (8) and (9) let us now reassociate the γ and S exponents with the $\mu\tau$ products of the corresponding carriers, regardless of being a majority or a minority carrier, i.e., for consistency we define them by

$$(\mu\tau)_e \propto G^{\gamma - 1} \tag{12}$$

and

$$(\mu\tau)_h \propto G^{-2S} \ . \tag{13}$$

Hence, in the case under consideration, γ will be a constant, $\gamma = 1$, and $2S$ will vary from $\frac{1}{2}$ to 0, as $|\Delta|$ is made to decrease by the application of the forward bias.

Summarizing the two one-level cases discussed above

we derive the most important, and new, consequence of our model, i.e., that there cannot be a simultaneous variation of γ and S with E_F in a system which contains only one recombination level. Following Eqs. (12) and (13) it is either that $\gamma=1$ or that $S=0$ throughout the entire range of Δ variation.

Let us examine now whether the above predictions are fulfilled by our experimental results. In Fig. 2(a) we show the measured increase of $(\mu\tau)_e$ with forward bias, while in Fig. 2(b) we show the measured decrease of L [i.e., of $(\mu\tau)_h$] with forward bias (on a 1-μm-thick film). We see, in accordance with previous studies, in which phosphorus and/or boron dopings were used,[10,11] that under forward bias, $(\mu\tau)_e$ increases by orders of magnitude, while L^2 [$\propto(\mu\tau)_h$] has a much more moderate decrease. These results are qualitatively in agreement with the predictions of the one-level model which were given above. We note, however, that the amplitudes of the changes are much smaller than those reported in doped materials[10,11,14] for the same range of Δ, and we thus conclude that in doped materials the quantity, the nature, and the energy profile of the recombination center distribution may be different from those in undoped a-Si:H.

In Fig. 3 we show the dependencies of the light-intensity exponents on the applied bias. The results for γ which are shown in Fig. 3(a) are in excellent agreement with those of previous works[7,17] on undoped a-Si:H, where γ was found to increase only slightly with $|\Delta|$ over the same reverse bias range, but to decrease quite sharply from $\gamma\approx1$ to $\gamma\approx0.4$ in the forward bias range of $0\le V_G\le6$ V. As mentioned above, such a decrease of γ in a-Si:H is expected, both experimentally[7,17] and theoretically,[3,6,13] to take place for Δ variation between $\Delta=0$ and $\Delta\approx0.3$ eV. All this enables us to set the correct energy scale, i.e., that a 0.3–0.4-eV variation in Δ corresponds to a 0–6-eV variation in V_G.

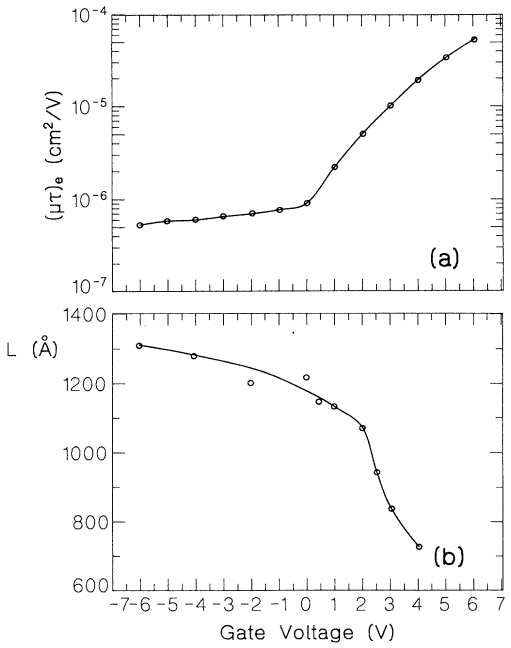

FIG. 2. The V_G dependencies of $(\mu\tau)_e$ and L. The first quantity was determined from the photoconductivity measurement while the second quantity was determined from the PCG measurement. The photon flux was $F=4\times10^{16}$ photons/cm^2 s.

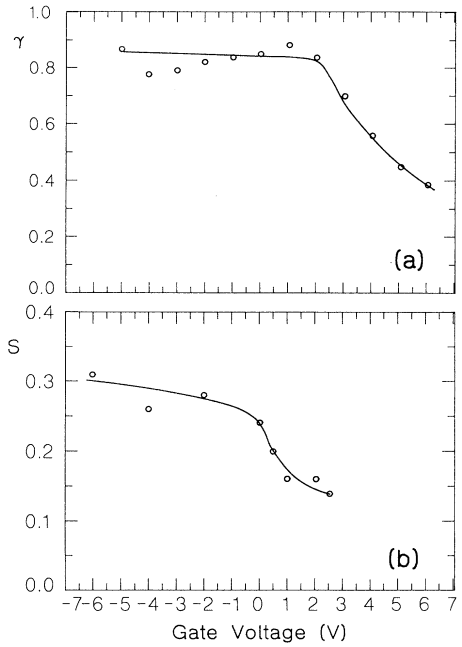

FIG. 3. The measured V_G dependencies of the exponents γ and S. The values of these exponents were obtained over the illumination range $3\times10^{15}\le F\le4\times10^{16}$ photons/cm^2 s.

So far all the data discussed above are consistent *qualitatively* with the one-level model where E_F lies above E_r. Let us turn then to the results of the $S(E_F)$ dependence (reported here for undoped a-Si:H). As we see in Fig. 3(b), S decreases appreciably with increasing V_G (i.e., with increasing Δ). This result is in agreement with the decrease of S with *decreasing* boron content which we have reported previously,[10] since a decrease in boron doping amounts (in the still n-type material) to the relative raising of E_F, and thus to the increase of Δ in the present experiment. The similarity of the $\gamma(E_F)$ and $S(E_F)$ results, obtained for the doped bulk material and in the present MOS configuration, also indicates that in this configuration one measures the same minority-carrier diffusion length. Correspondingly this is an *a posteriori* proof that in our experiment ambipolar conditions[20] prevail in the near-surface region of the sample where the Fermi level shifts. The fulfillment of these conditions is necessary for the interpretation of the PCG measurement in terms of the minority-carrier diffusion length.[20] We should point out, however, that in a-Si:H under the MOS configuration the so-called accumulation conditions are not associated with a high free-carrier concentration (as in crystalline silicon) but rather with an accumulation of trapped carrier concentration.[7]

While the present and the above-mentioned experimental observations are consistent, the simultaneous varia-

tion of γ and S with E_F, and in particular the high nonzero value of S, is inconsistent with the main consequence of the above-given one-level model. Hence the *one-level model cannot account* for the $S(E_F)$ data and we should turn, when discussing *a*-Si:H, to the next model in complexity, i.e., to the well-known "standard" model.[3,6] In the "standard" model it is assumed that there are D^+ and D^0 dangling-bond centers which have the same energy level. This level lies below E_F. The other level is associated with the D^- dangling-bond center, and it lies above E_F. It is expected, however, that the correlated occupation statistics[21] in this case do not provide a situation which is significantly different from the one-level model, since the majority-carrier neutrality condition ($n \approx p_r$, see above), which controls the occupation of the D^+/D^0 level, also controls the correlated occupation of the D^- level. Indeed this simple conjecture has been confirmed by a detailed numerical study of the "standard" model.[13] In particular, our suggested $S=0$ value was found to hold throughout the entire Δ range under consideration[13] (i.e., as long as the electrons are majority carriers). Hence the $S\neq 0$ results shown in Fig. 3(b) indicate that *neither the one-level model nor even the "standard" model can account for the experimental observations.*

The preliminary requirement in order to account for the data is then to be able to explain the $S>0$ result and the simultaneous variation of γ and S with Δ. Taking one more step in increasing the model complexity beyond the "standard" model we are led to the conclusion that there are at least two occupation-independent levels involved in the recombination process. We assume then one recombination level which lies below E_F, and *another* (at least one) recombination level which lies above E_F. The first level accounts for the "electron charge neutrality" (i.e., the $p_r \approx n$ condition,[19] see above) while the other level accounts for the charge neutrality which is principally determined by the hole concentration (i.e., an $n_r \approx p$ condition). In such a two-level model the observed increase of $(\mu\tau)_e$ and the decrease of γ is consistent with the presence of a recombination level which lies below E_F, i.e., with an increase in Δ, while the observed decrease of $(\mu\tau)_h$ and S with increasing Δ is consistent with the presence of a second recombination center level which lies above E_F (see above). Now, the existence of at least two occupation-independent levels is not in accordance with the "standard" model[13] (see above), but it is in accordance with the "defect-pool" model.[12,14,15] The latter model assumes at least two independent groups of levels (or bands, if disorder broadening is considered), such that one lies above E_F and one lies below E_F. Our $S(E_F)$ data are compatible then with the "defect-pool" model and can be viewed as a strong support for it. We should mention that quantitative calculations for this model have been given thus far for the $\mu\tau$ products[14] but not for their light-intensity dependencies.

Following the fact that the present measurements are associated with a nonuniform carrier distribution which increases towards the *a*-Si:H/oxide interface, the question arises of how well do the present results represent the bulk rather than the (about 1000-Å-wide) region adjacent to this interface.[7] The most convincing evidence that our data reflect the recombination-center distribution in the bulk is that the S values in routine PCG measurements[10,22] of bulk material yield similar $S\neq 0$ values, and that the effects of dopings,[3,10] yield quantitatively (for the same E_F shift) the same variations in γ and S as those obtained in the present MOS configuration [and in previous MOS studies[7,17] of $\gamma(E_F)$].

In conclusion, the E_F dependencies of the phototransport properties have been measured on undoped *a*-Si:H. The measured dependence of the minority-carrier light-intensity exponent indicates that the uncorrelated occupation of two levels, which is also assumed in the "defect-pool" model, is a fundamental property of the recombination level structure in *a*-Si:H.

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