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|  | **The origin of broad distribution of**  **breakdown times in polycrystalline thin film dielectrics** | |
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APPLIED PHYSICS LETTERS 101, 153511 (2012)

[The origin of broad distribution of breakdown times in polycrystalline thin film dielectrics](http://dx.doi.org/10.1063/1.4758684)

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The distribution of breakdown times of thin film dielectrics, stressed in a constant voltage mode, is generally interpreted in terms of percolation theory of dielectric breakdown. The percolation model suggests that relative distribution of failure times (normalized to the mean) should narrow down considerably for thicker dielectrics. Explicitly contradicting this prediction, we find a larger distribution of failure times even for relatively thick polycrystalline oxides. We use atomic force microscopy and conductive AFM measurements to confirm that breakdown in these films are primarily localized in the grain boundaries, decorated with large number of pre-existing defects. The classical percolation model—adapted to this specific situation of spatially localized trap generation—offers an intuitive explanation of the breadth of the failure time distribution in thick polycrystalline dielectric. The theory offers an opportunity to optimize the intrinsic trade-off between variability and reliability in polycrystalline films. V C 2012 American Institute of Physics.

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As the device dimension approaches the atomistic limit, it is well-known that the present and the future electronic devices will face increasing variability and reliability chal-lenges. The origin of such challenges lies not only on the non-scalability of the defect size and thermal voltage level (despite device dimension and voltage scaling) but also due to the fact that the devices are getting more heterogeneous from material and structural point of view.1In recent years, for example, homogeneous gate dielectric (SiO2) has been replaced by composite stacks of SiO2 and other high-k dielectrics such as HfO2,2planner device structure has been replaced by three-dimensional devices3and interconnects such as Through-Si-Via (TSV).4Thus, it is important to re-examine our understanding of the classical reliability phe-nomena in the modified scenario of new heterogeneous materials and devices.

One of the classical damage mechanisms of dielectrics in different electronic devices is time dependent dielectric breakdown (TDDB).5–9In this paper, we wish to examine the phenomenon of dielectric breakdown in the context of heterogeneous, polycrystalline oxide, which have applica-tions not only in gate dielectric (HfO2) of transistors but also in memory (e.g., ferroelectrics in FRAM10) and in embedded passive devices (e.g., on-chip very high-k capacitors11).

For homogeneous dielectrics (e.g., amorphous SiO2), the TDDB phenomenon is understood by random generation of defects under an applied bias, and subsequent formation of one or more percolation paths to create an electrical short through the dielectric (percolation model).5Since defect gen-eration is a stochastic process, the time to form a percolation path differs from one sample to the next, and therefore the time-to-breakdown is represented by a probability distribu-tion. The classical theory explains not only the shape of the

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breakdown distribution (Weibull) but also the linear scaling of the Weibull shape factor (b) with oxide thickness. Accord-ing to the percolation model, as the oxide thickness increases, it becomes less likely to accidentally align a set of defects to form a percolation path, and thus the relative dis-tribution of breakdown time becomes narrower (or, equiva-lently, the Weibull slope becomes higher) with increasing oxide thickness. The predictions of the classical percolation

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| theory | have | been | extensively | validated | by | many |
| experiments.7 | |

We were surprised, therefore, when we observed that the distribution of breakdown in polycrystalline oxides is significantly broader than that predicted by percolation theory. An extensive review of the literature suggests that this is a generic phenomenon and many groups have previ-ously observed such broad distribution of failure time in sim-ilar technologies and oxides.11–13Some progress has been made towards understanding this puzzle by Chentir et al.,12 who have rightly pointed out to the non-homogeneity of the polycrystalline oxide, and attempted to model the breakdown in the presence of conducting voids within the oxide. How-ever, the simulated results still showed significant discrep-ancy between the theory and the experiments. Therefore, despite its importance for technology scaling, the phenom-enon has never been interpreted consistently. In the follow-ing discussion, we explore the physical origin of this anomalously broad failure time distribution in polycrystal-line films, and confirm that the pre-existing defects, espe-cially the “hot spots” in the grain boundaries (GBs), play a crucial role in broadening the distribution of failure times. We also provide an analytical formulation by extending the classical percolation model to interpret the experiments.

In this study, we use a metal-oxide-metal capacitor with a polycrystalline ferroelectric as the oxide, see Fig. 1(a). The oxide material is Pb(Zr,Ti)O3 (PZT), deposited onto Iridium (Ir) bottom electrode by metal oxide chemical vapor

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| 153511-2 | Masuduzzaman et al. | Appl. Phys. Lett. 101, 153511 (2012)  FIG. 1. (a) Schematic of a polycrystal-line PZT oxide. (b) Experimental data show a wide distribution of breakdown time. (c) Cell-based model for TDDB. The GB regions have higher pre-existing | | | | | |
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| defects | (darker | cells) | and | thus | the |
| required number of defects for a break-down is smaller than that in grain regions. (d) The simulation results of Weibull distribution for the polycrystal-line oxide. | | | | | |

deposition (MOCVD) technique, and has a thickness of 70 nm.14The polycrystalline nature of the oxide is character-ized by scanning electron microscopy (SEM) of the cross-section, as well as the atomic force microscopy (AFM) of the topography of the oxide. The capacitors show the typical

In order to explain the above discrepancy, we first note that the classical theory is based on the defect generation on a homogeneous (amorphous) dielectric. However, the mate-rial under study is a heterogeneous (polycrystalline) dielec-tric containing grains and grain boundaries. The theories of

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| capacitance-voltage | (C-V), | transient | leakage | (I-t), | and | crystal growth anticipate larger density of defects related to |

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| hysteresis | characteristics, | appropriate | for | ferroelectric |
| materials.14 |

To determine the TDDB lifetime of the capacitors, we apply a constant voltage stress across the capacitor at room temperature of 20�C, and monitor the leakage current as a function of time.15When the leakage exceeds a certain threshold, the time is noted as the breakdown time (TBD). This use of leakage current as a monitor of dielectric lifetime is standard and widely used in the industry.7Fig. 1(b) shows the Weibull plot of TBD, for breakdown measurements of 35 samples of the capacitor. Here, the vertical axis represents the Weibit, defined as W ¼ lnð�lnð1 � FÞÞ, where F being the cumulative failure fraction associated with 35 samples. As shown in the figure, the lifetime distribution is spread over more than one order of magnitude.

According to the classical theory, the slope of the Weibull distribution depends as b ¼ aM,16where a is the time exponent of the defect generation, and M is the number of defect cells in the vertical stack of a percolation path through the oxide [Fig. 1(c)]. For example, if the defect size (ld) is �1 nm, and the oxide thickness is 70 nm, then M ¼ TOX=ld ¼ 70: The value of a for gate oxides of modern transistors is well-characterized, and typically ranges from 0.1 to 1.17Presuming similar kinetics of defect generation, one expects a minimum value of b to be �7. As shown in smaller than the expected value of Weibull slope in the poly-Fig. 1(b), however, the observed value (b ¼ 1.54) is much crystalline oxide. A similar mismatch between theoretical and experimental b of about a factor of 5 on similar oxide is also reported in Ref. 12.

unsatisfied dangling bonds at the boundary between disori-ented grains. This has been extensively verified by first prin-ciple simulation as well as experimental techniques.18–20 Also, there are reports of void formation within the grain boundaries, which could introduce additional defects.12As the pre-existing defects reduce the required numbers of defects to generate for a breakdown, one can ask if this heter-ogeneous defect density explain the anomalously large distri-bution of breakdown times.

First, we need to confirm the existence of higher defect density at the grain boundary sites for the particular PZT ox-ide. Since local leakage current across the oxide depends on the defect density, we used the conductive AFM (CAFM) technique to spatially resolve the leakage current over the oxide. Indeed, such technique has been used to show larger defect density in grain boundaries of polycrystalline gate dielectric (such as HfO2) for transistor applications.21The CAFM measurement requires specially prepared capacitor-like test structure whose top electrode has been removed so that the surface is accessible to AFM tip. We scan the surface of the bare oxide in contact mode of the AFM, and simulta-neously measure the local topographic information, as well as the leakage current under a given bias. Fig. 2 shows the results of such measurements for a typical fresh sample. The topological variation [Fig. 2(a)] is related to the surface roughness, as well as the interface (grain boundary) between two or more grain regions. The simultaneous leakage current measurement on the same area [Fig. 2(b)] clearly shows that the current is much higher (>100 nA) through regions which correlate to the grain boundaries of the surface [Fig. 2(a)].

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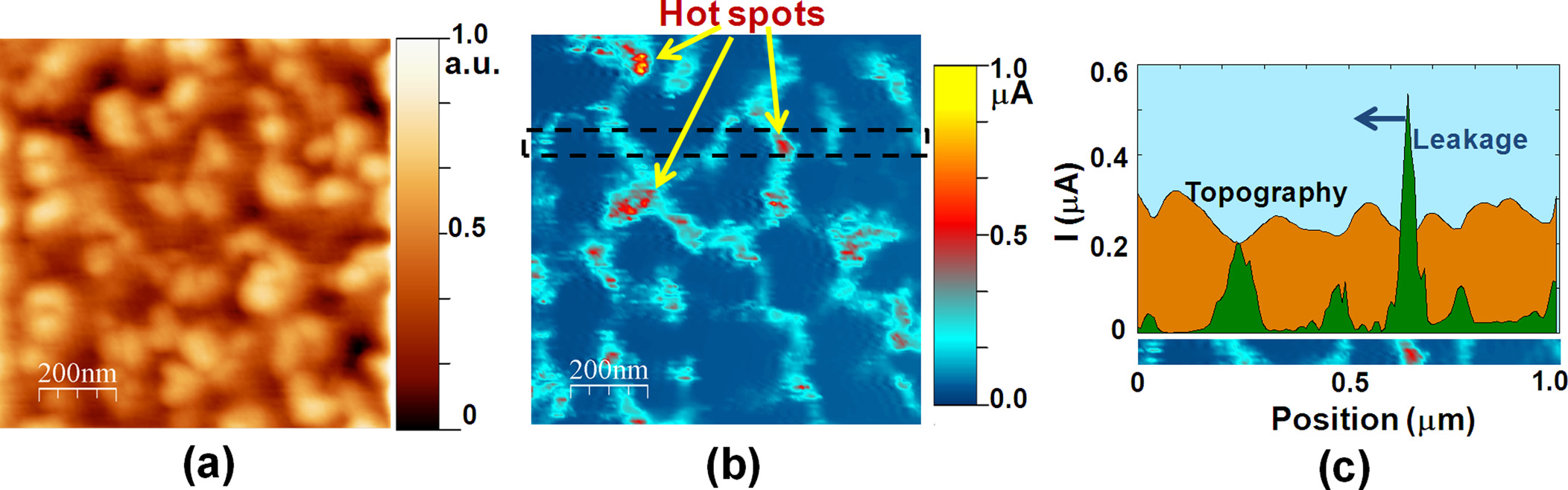


FIG. 2. Top view (x-z plane) of the PZT oxide using conductive AFM technique. (a) Topography of the oxide shows the grain (lighter) and grain boundary (darker) regions. (b) Leakage map on the same area shows that the grain boundaries have higher leakage current due to larger pre-existing traps. Moreover, some grain boundary locations have even higher leakage current (hot spots), indicating a distribution of defect density along the grain boundaries. (c) The leak-age current values, along a horizontal line located in the region as highlighted in (b), is shown along with the topographical background in the same region. The current peaks clearly correlate with the topographic valleys due to the grain boundaries.

On the other hand, the typical leakage current along the grain regions is at least an order of magnitude lower (�10 nAÞ than those from the grain boundary regions. The correlation between grain boundary and excess leakage current is shown [Fig. 2(c)] for a particular location as highlighted in Fig. 2(b). Such correlation has been observed in all the samples we measured. Thus, the experiment confirms that the grain boundaries have a higher pre-existing defect density that facilitates excess leakage current through the grain bounda-ries as compared to the bulk grain regions. We also note that the leakage along the grain boundaries is not uniform: Some locations (hot spots) have higher leakage than others, sug-gesting a distribution of defect density along the grain boun-daries. Such distribution is expected if we consider the fact that different grain boundaries have different degrees of angle mismatch among the adjacent grains, and the corre-sponding strain and interfacial energies should also be differ-ent for different grain boundaries.18   
 To understand how the higher defect density at grain boundaries influence the generation of new defects and con-sequent localized breakdown, we made the following meas-urements. We apply a constant DC stress voltage in the AFM tip, and repeatedly measure the leakage current through the oxide. Fig. 3(a) shows the topographic map, and Figs. 3(b)

and 3(c) show the leakage current at the beginning and at the end of the stress. Note the particular grain boundary region (circled) in Fig. 3(a). The region already has high leakage current (hot spot) before the stress [Fig. 3(b)]. As the stress continues, defects are presumed to generate everywhere in the sample. However, since the hot spots require fewer defects to complete the percolation path, one of the hot spots is most likely to experience TDDB breakdown ahead of the other regions in the oxide [Fig. 3(c)]. This experiment, there-fore, demonstrates that the grain boundaries not only have higher pre-existing defect density and spatially isolated hot spots but also appear to break-down earlier than the grain regions, despite the fact that the total area of the former is only a fraction of that of the later.

Given the two observation regarding the grain bounda-ries, we will see how this translates to the distribution of the TDDB breakdown. The classical theory of dielectric break-down in a homogeneous material can be explained by an array (N � M) of potential defect sites within the dielectric, where N and M are proportional to the area and thickness of the dielectric, respectively [Fig. 1(c)]. Random defect gener-ation increases the probability of defect as- qðtÞ ¼ ata, for a given location. The probability of at least one breakdown at any given time [FðtÞ] is expressed as5

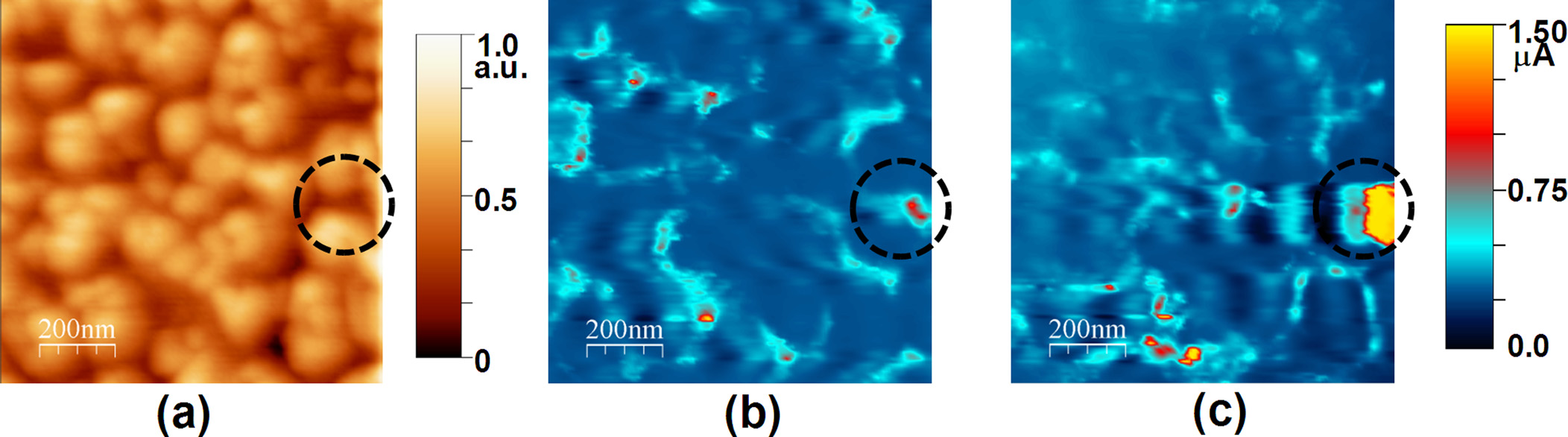


FIG. 3. The topography (a), and the leakage current before (b) and after (c) voltage stress clearly show (circled) that the breakdown happens on region coin-cides with a grain boundary. The stress has been applied using the conductive AFM tip by repeated scanning with a stress bias.

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| 1 � FðtÞ ¼ | �1 � qðtÞM�N | : | (1) |

For the polycrystalline oxide, since the grain and grain boundary regions have different pre-existing trap, and trap generation characteristics, the probabilities of defect genera-tion can be written as qi, in general, with the parameters ai and ai, where i ¼ 1; 2 for grain and grain boundary regions, respectively [Fig. 1(c)]. Also, since there are already some pre-existing defects in grain boundary regions, the required number of defects (M2) to create a vertical percolation path should be smaller than that in grain regions (M1). Moreover, the area of the two regions, and hence N1 and N2 are differ-ent. Therefore, Eq. (1) is modified as

the device area.21,23,24A reduction in grain-size, however, increases the net area of the grain boundary, with higher number of hot spots and reduced lifetime. Therefore, the theory developed in this paper offers an opportunity to opti-mize this intrinsic tradeoff between variability and reliability of the devices based on polycrystalline films by optimizing the size of the grains as well as the operating voltage that define dielectric lifetime.

In summary, in this paper, we have explored the origin of anomalously wide TDDB distribution in polycrystalline PZT oxide that we observed, and have been reported by many groups in the literature. We used AFM technique to separate the respective leakage contributions of grain and grain boundary regions. From the observed higher leakage

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| 1 � FðtÞ ¼ | Y ið1 � qi | Mi ÞNi: | (2) | current through the grain boundaries, we confirm that the |
| grain boundary regions of polycrystalline oxides have a large |
| number of pre-existing defects including some high-defect |
| areas or hot spots. We also find that with voltage stress, the |

To determine the relative area of the grain and grain boundary regions (N1=N2), we use the topographic profile as found from the AFM measurements. As for the time expo-nents of defect generation (ai), we assume that both of these are equal to 0.1—a value close to that for the high-j oxides.17Fig. 1(d) shows the Weibull distribution of lifetime of the polycrystalline oxide based on the analytical formula-tion as described above, as well as Monte-Carlo simula-tion.22The solid line on the right [Fig. 1(d)] represents the distribution for a hypothetical homogeneous film, where the parameters for grain and grain boundary regions are identi-cal. Note that when the grain boundaries have the same prop-erties as of a grain (e.g., do not have pre-existing defects, or M2 ¼ M1 ¼ M), the slope of the distribution is steep (bhomo ¼ aM ¼ aTOX=ld ¼ 7). However, once we account for the experimentally observed fact that the grain boundary regions are populated with pre-existing defects, i.e., M2 < M1, then it follows that bhetero < bhomo [Fig. 1(d), left]. This explains why Weibull slope of polycrystalline films with significant localized defects is reduced below the value expected from homogenous percolation theory. The experi-mental value of b (�1.5) suggests characteristically high defect density (M2 � M1=5, i.e., �80% defective) along the hot spots of the grain boundary. As we have mentioned that the hot spots are the worst location along the grain boundary, where defect density is maximum, characterized by high initial current and eventual breakdown with stress. Finally, a high defect density at the hot spots of the grain boundaries may modulate the local electric field, and the defect generation rate can be even higher at these locations. In such a situation, breakdown will be confined even more exclu-sively to the grain boundary locations. However, further work is required to experimentally verify the difference in trap gen-eration rates in the bulk and at the grain boundaries.

The results discussed above imply a general considera-tion of reliability and variability for polycrystalline films, such as HfO2 in transistor gate dielectrics,21poly-Si thin film transistors,23and poly-Si solar cell.24It is well known that when the device area becomes comparable to the average grain size, a distribution of grain size introduces variability from one device to the next.21This variability can be sup-pressed by reducing the average grain size with respect to

hot spots within the grain boundary regions break down ear-lier than the bulk grain regions. We then generalize the clas-sical percolation model to account for pre-existing defects and find that the analytical results offer an intuitive explana-tion of the wider distribution of breakdown times observed typically in polycrystalline films. We conclude that the vari-ability related to grain size and the reliability associated with grain boundary hot spots are intrinsically correlated in such films, and therefore can and should be simultaneously opti-mized for high performance devices.

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