The current waveform in reactive high power impulse magnetron sputtering

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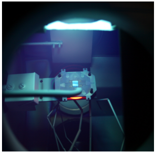
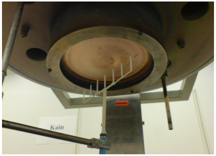
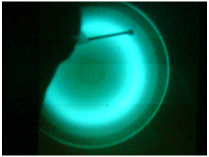
4th Magnetron, Ion processing & Arc Technologies European Conference,
14th International Symposium on Reactive Sputter Deposition,
Paris, France
December 11, 2015

Introduction



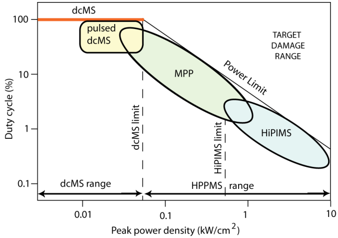




- Magnetron sputtering has been the workhorse of plasma based sputtering methods for over four decades
- For many applications a high degree of ionization of the sputtered vapor is desired
  - controlled ion bombardment of the growing film – controlled by a negative bias applied to the substrate
  - collimation – enhanced step coverage
- Common to all highly ionized magnetron sputtering techniques is a very high density plasma

Introduction

- In a conventional dc magnetron discharge the power density is limited by the thermal load on the target
- High power pulsed magnetron sputtering (HPPMS)
- In a HiPIMS discharge a high power pulse is supplied for a short period
  - low frequency
  - low duty cycle
  - low average power

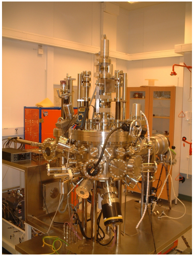


From Gudmundsson et al. (2012), JVSTA **30** 030801

- Power density limits
  $p_t = 0.05$ kW/cm^2 dcMS limit
  $p_t = 0.5$ kW/cm^2 HiPIMS limit

Introduction

- Reactive sputtering, where metal targets are sputtered in a reactive gas atmosphere to deposit compound materials is of utmost importance in various technologies
- In reactive sputtering processes a reactive gas O_2, N_2, or CH_4 etc. is mixed to the noble working gas for oxide, nitride, or carbide deposition
- The high electron density in the HiPIMS discharge is expected to enhance the dissociation of the molecular gas

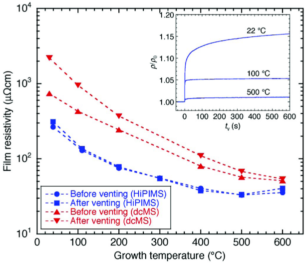


Gudmundsson (2016) Plasma Phys. Control. Fusion **58** 014002

Reactive HiPIMS - Applications

Application – Film Resistivity

- TiN as diffusion barriers for interconnects
- HiPIMS deposited films have significantly lower resistivity than dcMS deposited films on SiO_2 at all growth temperatures due to reduced grain boundary scattering
- Thus, ultrathin continuous TiN films with superior electrical characteristics and high resistance towards oxidation can be obtained with HiPIMS at reduced temperatures

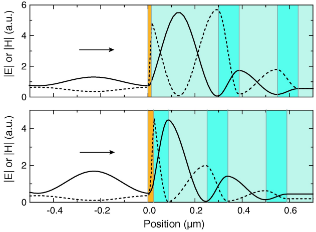


From Magnus et al. (2012) IEEE EDL **33** 1045

Application – Bragg mirror

- Multilayer structures containing a high-contrast (TiO_2/SiO_2) Bragg mirror fabricated on fused-silica substrates
  - reactive HiPIMS TiO_2 (88 nm)
  - reactive dcMS SiO_2 (163 nm)
  - capped with semitransparent gold
- Rutile TiO_2 (n = 2.59) and SiO_2 (n = 1.45) provide a large index contrast
- Smooth rutile TiO_2 films can be obtained by HiPIMS at relatively low growth temperatures, without post-annealing

Agnarsson et al. (2013) TSF **545** 445

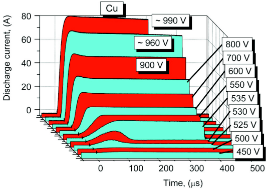


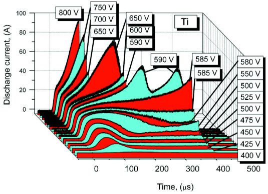
From Leosson et al. (2012) Opt. Lett. **37** 4026

Reactive HiPIMS - Voltage - Current - Time characteristics

HiPIMS - Voltage - Current - time

- In **non-reactive** discharge the current waveform shows an initial pressure dependent peak that is followed by a second phase that is power and material dependent
- The initial phase is dominated by gas ions, whereas the later phase has a strong contribution from self-sputtering
- For some materials, the discharge switches into a mode of **sustained self-sputtering**



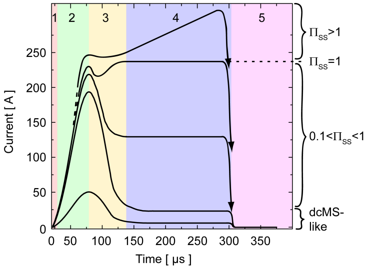


From Anders et al. (2007),

JAP **102** 113303 and JAP **103** 039901

HiPIMS - Voltage - Current - time

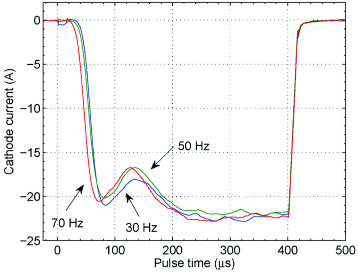
- A schematic illustration of the discharge current assuming square shaped voltage pulses
- The current is generally characterized by an initial peak followed by a more or less stable current plateau (bottom current curves)
- In other cases it shows an initial peak followed by a second increase of the discharge current (top current curves)
- The non-reactive case is well understood



From Gudmundsson et al. (2012), JVSTA **30** 030801

HiPIMS - Voltage - Current - time

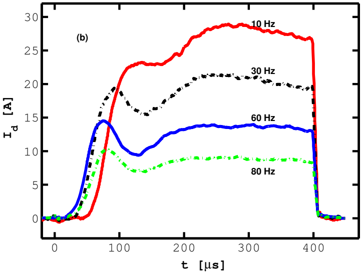
- Ar discharge with Ti target
- The initial peak in current results large flux of atoms from the target
- Collisions of the sputtered atoms with the working gas result in heating and expansion of the working gas – **rarefaction**
- A significant fraction of the sputtered atoms experience electron impact ionization (the ionization mean free path $\sim$ 1 cm) and are attracted back to the target to participate in the sputtering process – **self-sputtering**



From Magnus et al. (2011) JAP **110** 083306

HiPIMS - Voltage - Current - time

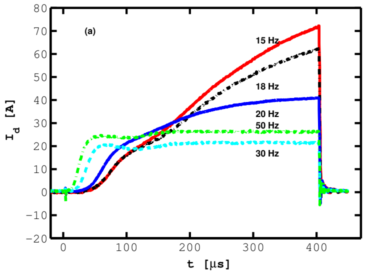
- During reactive sputtering, a reactive gas is added to the inert working gas
- The current waveform in the reactive Ar/N_2 HiPIMS discharge is highly dependent on the pulse repetition frequency, unlike for pure Ar
- N_2 addition changes the plasma composition and the target condition can also change due to the formation of a compound on its surface



After Magnus et al. (2011) JAP **110** 083306

HiPIMS - Voltage - Current - time

- Similarly for the Ar/O_2 discharge, the current waveform is highly dependent on the repetition frequency and applied voltage which is linked to oxide formation on the target
- The current is found to increase significantly as the frequency is lowered

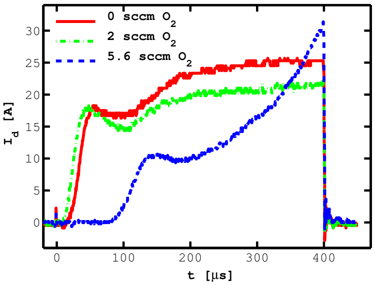


After Magnus et al. (2012), JVSTA **30** 050601

HiPIMS - Voltage - Current - time

- As the oxygen flow is increased a transition to oxide mode is observed – The delay in the onset of the current increases, the initial current peak is lowered and a transition to a self-sputtering runaway occurs
- It has been confirmed that in the oxide mode, the discharge is dominated by O^+-ions, due to oxygen atoms sputtered off the target surface

Aiempanakit et al. (2013), JAP **113** 133302



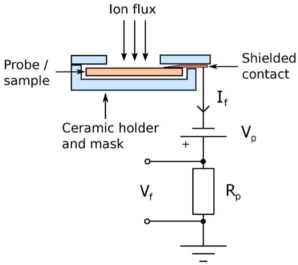
The current waveforms for an Ar/O_2 discharge with a Ti target where the oxygen flow rate is varied – 600 V, 50 Hz and 0.6 Pa

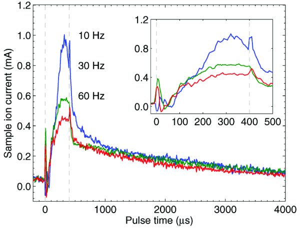
From Gudmundsson et al. (2013), ISSP 2013, p. 192

Gudmundsson (2016) Plasma Phys. Contr. Fus. **58** 014002

HiPIMS - Voltage - Current - time

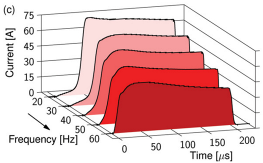
- The observed changes in the discharge current are reflected in the flux of ions impinging on the substrate

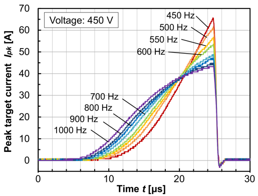




From Magnus et al. (2011), JAP **110** 083306

HiPIMS - Voltage - Current - time





- Similar behaviour has been reported for various target and reactive gas combinations
  - The current increases with decreased repetition frequency
  - The current waveform maintains its shape for Ar/O_2 discharge with Nb target

  From Hála et al. (2012), JPD **45** 055204

  - The current waveform becomes distinctly triangular for Ar/N_2 discharge with Hf target

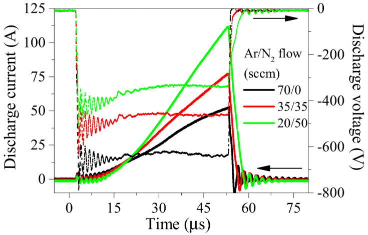
  From Shimizu et al. (2015), `arXiv:1509.07002`

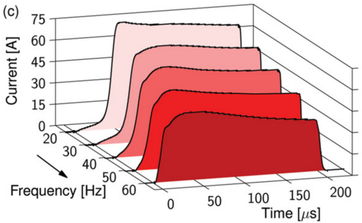
HiPIMS - Voltage - Current - time

- The current increases with increased partial pressure of the reactive gas
    - The current waveform becomes distinctly triangular for Ar/N_2 discharge with Al target

      From Moreira et al. (2015), JVSTA **33** 021518
    - The current waveform maintains its shape for Ar/O_2 discharge with Nb target

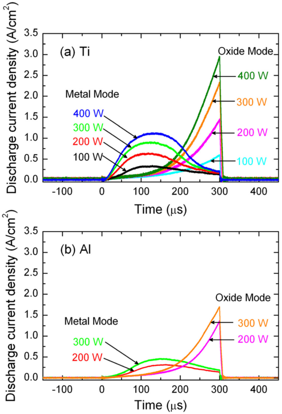
      From Hála et al. (2012), JPD **45** 055204





HiPIMS - Voltage - Current - time

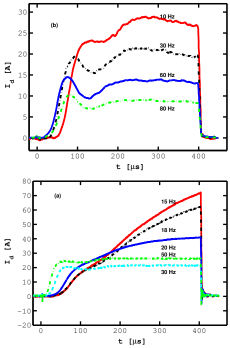
- The shape of the current waveform depends on the mode of operation
  - The current waveform becomes distinctly triangular for Ar/O_2 dischage with both Al and Ti target



From Aiempanakit et al. (2013), JAP **113** 133302

HiPIMS - Voltage - Current - time

- HiPIMS differs significantly from dcMS, due to the fact that self-sputtering quickly becomes dominant and the working gas ions (mostly Ar^+ and N_2^+ or O_2^+) are depleted from the area in front of the target, due to rarefaction
- The secondary electron emission yield is governed by the composition of the target (Ti or TiN or TiO_2) and the type of ions that are bombarding it

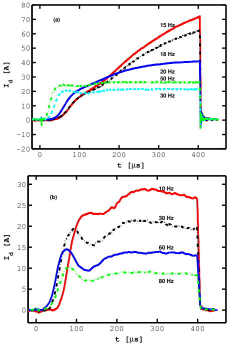


After Magnus et al. (2011), JAP **110** 083306

and Magnus et al. (2012), JVSTA **30** 050601

HiPIMS - Voltage - Current - time

- γ_{see} is practically zero for singly charged metal ions impacting a target of the same metal
- γ_{see} will be higher for self sputtering from a TiN or TiO_2 target, where N^+-ions or O^+-ions are also present, than for self-sputtering from a Ti target, where multiply charged Ti ions are needed to create secondary electrons

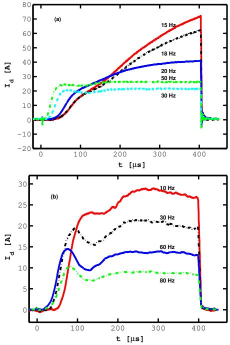


After Magnus et al. (2011), JAP **110** 083306

and Magnus et al. (2012), JVSTA **30** 050601

HiPIMS - Voltage - Current - time

- At high frequencies, nitride or oxide is not able to form between pulses, and self-sputtering by Ti^{+}-ions (singly and multiply charged) from a Ti target is the dominant process
- At low frequency, the long off-time results in a nitride or oxide layer being formed on the target surface and self-sputtering by Ti^{+}- and N^{+}-ions or O^{+}-ions from TiN or TiO_2 takes place



From Magnus et al. (2011), JAP **110** 083306

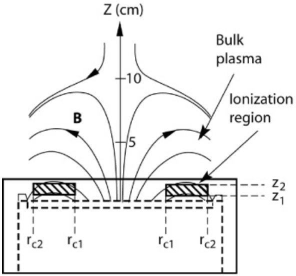
and Magnus et al. (2012), JVSTA **30** 050601

Gudmundsson (2016) PPCF **58** 014002

Ionization region model studies of reactive HiPIMS

Ionization region model studies of reactive HiPIMS

- The ionization region model (IRM) was developed to improve the understanding of the plasma behaviour during a HiPIMS pulse and the afterglow
- The main feature of the model is that an ionization region (IR) is defined next to the race track
- The IR is defined as an annular cylinder with outer radii r_{c2}, inner radii r_{c1} and length $L = z_2 - z_1$, extends from z_1 to z_2 axially away from the target

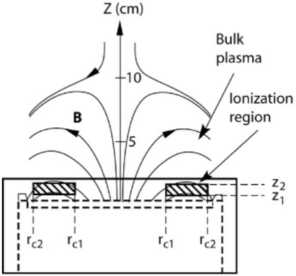


The definition of the volume covered by the IRM

From Raadu et al. (2011), PSST **20** 065007

Ionization region model studies of reactive HiPIMS

- Geometrical effects are included indirectly as loss and gain rates across the boundaries of this annular cylinder to the target and the bulk plasma
- The temporal development is defined by a set of ordinary differential equations giving the first time derivatives of the electron energy and the particle densities for all the particles
- The electron density is found assuming quasi-neutrality of the plasma



The definition of the volume covered by the IRM

From Raadu et al. (2011), PSST **20** 065007

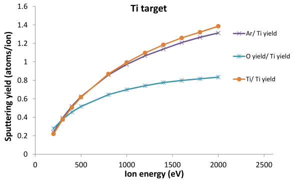
Ionization region model studies of reactive HiPIMS

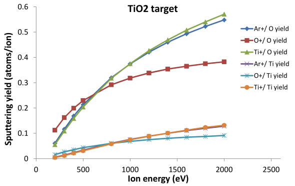
- The species assumed in the IRM are
  - electrons
  - argon atoms $Ar(3s^2 3p^6)$, warm argon atoms in the ground state Ar^W, hot argon atoms in the ground state Ar^H, Ar^m ($1s_5$ and $1s_3$) (11.6 eV), argon ions Ar^+ (15.76 eV), doubly ionized argon ions Ar^{2+} (27.63 eV)
  - titanium atoms $Ti(a\,^3F)$, titanium ions Ti^+ (6.83 eV), doubly ionized titanium ions Ti^{2+} (13.58 eV)
  - oxygen molecule in the ground state $O_2(X^3\Sigma_g^-)$, the metastable oxygen molecules $O_2(a^1\Delta_g)$ (0.98 eV) and $O_2(b^1\Sigma_g)$ (1.627 eV), the oxygen atom in the ground state $O(^3P)$, the metastable oxygen atom $O(^1D)$ (1.96 eV), the positive ions O_2^+ (12.61 eV) and O^+ (13.62 eV), and the negative ion O^-

Toneli et al. (2015) J. Phys. D. **48** 325202

Ionization region model studies of reactive HiPIMS

- The sputter yield for the various bombarding ions was calculated by TRIDYN for
  - Metal mode – Ti target
  - Poisoned mode – TiO_2 target
- The yields correspond to the extreme cases of either clean Ti surface and a surface completely oxidized (TiO_2 surface)

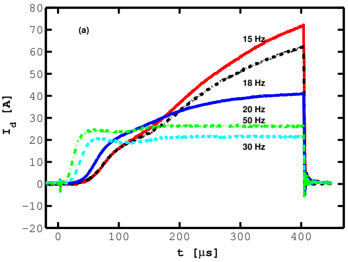




The sputter yield data is from Tomas Kubart, Uppsala University

Ionization region model studies of reactive HiPIMS

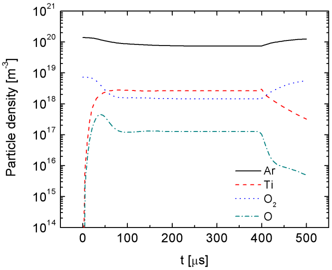
- The model is applied to explore Ar/O_2 discharge with Ti target in both metal mode and oxide (poisoned) mode
- The IRM is a semi-empirical model in the sense that it uses a measured discharge current waveform as a main input parameter
- For this study we use the measured curve for Ar/O_2 with Ti target at 50 Hz for metal mode and at 15 Hz for poisoned mode



After Magnus et al. (2012), JVSTA **30** 050601

Ionization region model studies of reactive HiPIMS

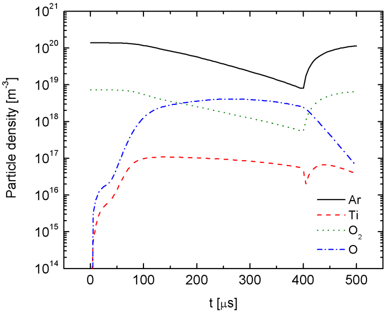
- The gas rarefaction is observed for the argon atoms but is more significant for the O_2 molecule
- The density of Ti atoms is higher than the O_2 density
- The atomic oxygen density of is over one order of magnitude lower than the molecular oxygen density – the dissociation fraction is low



The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O_2 discharge with Ti target in **metal mode**.

Ionization region model studies of reactive HiPIMS

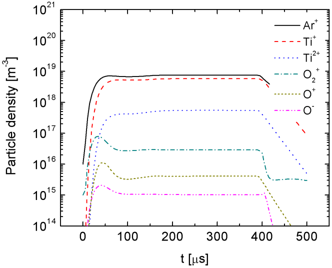
- Gas rarefaction is observed for both argon atoms and O_2 molecules
- The density of Ti atoms is lower than both the O_2 density and atomic oxygen density
- The atomic oxygen density is higher than the O_2 density in towards the end of the pulse



The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O_2 discharge with Ti target in **poisoned mode**.

Ionization region model studies of reactive HiPIMS

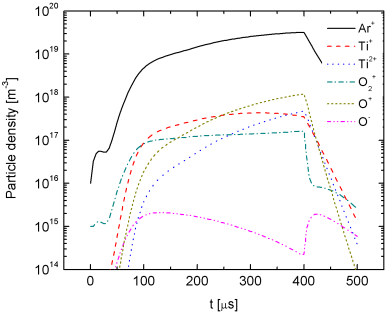
- Ar^+ and Ti^+-ions dominate the discharge
- Ti^{2+}-ions follow by roughly an order of magnitude lower density
- The O_2^+ and O^+-ion density is much lower



The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O_2 discharge with Ti target in **metal mode**.

Ionization region model studies of reactive HiPIMS

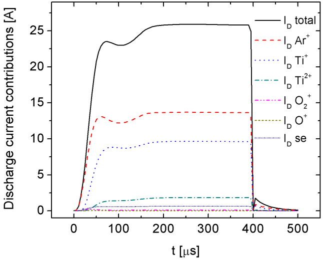
- Ar^+-ions dominate the discharge
- Ti^+, O^+, have very similar density, but the temporal variation is different, and the O_2^+ density is slightly lower
- The Ti^{2+}-ion density increases fast with time and overcomes the Ti^+ density towards the end of the pulse



The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O_2 discharge with Ti target in **poisoned mode**.

Ionization region model studies of reactive HiPIMS

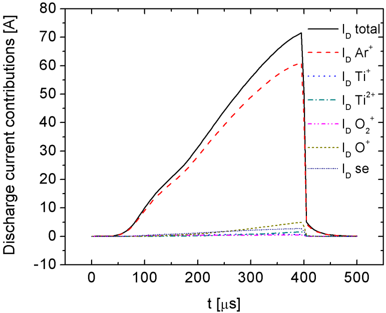
- Ar^+ and Ti^+-ions contribute most significantly to the discharge current



The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O_2 discharge with Ti target in **metal mode**.

Ionization region model studies of reactive HiPIMS

- Ar^+ contribute most significantly to the discharge current – almost solely
- The contribution of secondary electron emission is very small



The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O_2 discharge with Ti target in **poisoned mode**.

Summary

Summary

- The current-voltage-time waveforms in a reactive discharge exhibit similar general characteristics as the non-reactive case in some cases
  - the current rises to a peak, then decays because of rarefaction before rising to a self-sputtering dominated phase
  - in other cases the current develops a triangular shape as repetition frequency is lowered or the partial pressure of the reactive gas is increased
- At low repetition frequency, the long off-time results in a nitride or oxide layer being formed on the target surface and an increase in the discharge current

Summary

- An ionization region model was used to explore the plasma composition during the high power pulse
- Comparison was made between the metal mode and the poisoned mode
  - In metal mode Ar^+ and Ti^+-ions dominate the discharge and are of the same order of magnitude
  - In poisoned mode Ar^+-ions dominate the discharge and two orders of magnitude lower, Ti^+, O^+, have very similar density, with the O_2^+ density slightly lower
  - In the metal mode Ar^+ and Ti^+-ions contribute most significantly to the discharge current while in poisoned mode Ar^+ dominate
- Ar^+-ions are responsible for the increase in the discharge current in poisoned mode

The slides can be downloaded at

`http://langmuir.raunvis.hi.is/~tumi/hipims.html`

- The experimental work was made in collaboration with
  - Dr. Fridrik Magnus, Uppsala University, Uppsala, Sweden
  - Tryggvi K. Tryggvason, University of Iceland
- The modeling work is in collaboration with
  - Dr. Daniel Lundin, Université Paris-Sud, Orsay, France
  - Prof. Nils Brenning, KTH Royal Institite of Technology, Stockholm, Sweden
  - Prof. Tiberu Minea, Université Paris-Sud, Orsay, France
- We got help with the sputtering yields from
  - Dr. Tomas Kubart, Uppsala University, Uppsala, Sweden
- and the project is funded by
  - Icelandic Research Fund Grant No. 130029-053
  - Swedish Government Agency for Innovation Systems (VINNOVA) contract no. 2014-04876,

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Aiempanakit, M., A. Aijaz, D. Lundin, U. Helmersson, and T. Kubart (2013). Understanding the discharge current behavior in reactive high power impulse magnetron sputtering of oxides. *Journal of Applied Physics 113*(13), 133302.

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