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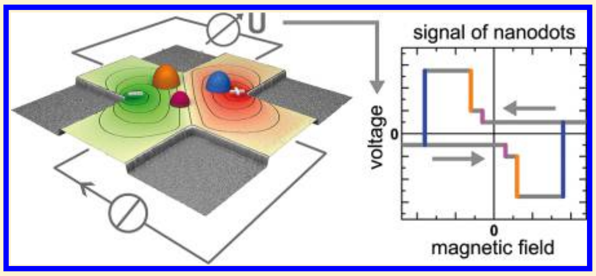
Probing the Magnetic Behavior of Single Nanodots

Alexander Neumann,[†,*] Carsten Thönnißen,[†] Axel Frauen,[†] Simon Heße,[†] Andreas Meyer,[‡] and Hans Peter Oepen[†]

[†]Institut für Angewandte Physik, Universität Hamburg, Jungiusstraße 11, 20355 Hamburg, Germany
[‡]Institut für Physikalische Chemie, Universität Hamburg, Grindelallee 117, 20146 Hamburg, Germany

S Supporting Information

ABSTRACT: In this paper, a method is presented that has the sensitivity to measure magnetization behavior of single nanostructures. It is demonstrated that the technique gives the ability to separate different signals of single nanodots from a small ensemble of structures. Our method is based on the anomalous Hall-Effect and allows for resolving signals from spherical nanoparticles with diameter down to 3.5 nm. The method gives access to magnetic properties of particles in a wide thermal and dynamical range. The potential of the technique is demonstrated utilizing particles that are created from Co films sandwiched by Pt layers.



KEYWORDS: *Magnetic nanostructure, magnetic nanoparticles, magnetic reversal, bit patterned media, anomalous Hall-Effect, nanosphere lithography*

Nowadays a big challenge in nanoscience is the quantification of interactions between nanoparticles and to pinpoint their influence on the behavior of an ensemble of namely identical particles. The main issue is to get rid of latent ambiguities that are concerned with variations of size and properties of the individual particle and the arrangement and mean separation of the particles when investigating large ensembles. The importance of interactions is generally downplayed in two respects. At first, as the dipole interactions scale with the separation d like d^{-3} the interaction is commonly assumed to terminate at a distance roughly on a scale of the particle dimensions. Second, the arrangement of the particles is assumed to be isotropic and fixed to a mean separation. The reduction of the interaction volume is acceptable as long as the system is in a stable state. As soon as the local energy minimum gets weak compared to competing interactions, like, for example, thermal energy, even the smallest interactions will have dramatic effects on the behavior of individual particles. Such very delicate situations are to be expected in thermally driven phase transition. As a large variety of scales coexists in an ensemble of particles an exact description of the critical thermal behavior is almost impossible. The best approach to get access to nanoscale systems in critical states (independent of the driving parameter) is to know very accurately the geometry and measure the behavior of the individual particles simultaneously.

The fundamental prerequisite for such investigations is to have probing technique at hands that has single particle sensitivity. This in its own is a real challenge. In the case of magnetic systems, only a few methods have been proven to have the sensitivity for detecting the magnetization behavior of single nanoparticles below 35 nm.[1,2] While these techniques image the magnetic structure of the particle predominantly in remanence, a new approach was developed that relies on microsized signal pick up to measure quantitatively magnetic values, that is, the micro- or nanoSQUID.[3,4] The latter technique has been successfully used to investigate the magnetization behavior of very small units. Up to now, however, the method has not been proven to give direct access to individual magnetization curves when more than one particle are simultaneously studied.

In this paper, we introduce a new approach utilizing the anomalous Hall-effect (AHE)[5−8] that has single particle sensitivity and demonstrate its potential to study the state of magnetization of the individual nanostructures in a collection of a few particles. As the approach is not limited in temperature and field strength it is well suited to investigate the phase transition between blocked and superparamagnetic state of single particles and particularly to study the influence of the magnetic environment on the transition. In the examples given below, variations of the switching field of single particles are found. Recent models,[9] which describe the distribution of the switching fields of single particles as a function of temperature, can be proven by similar experiments including a quantification of the influence of interactions. The latter item is important for the understanding of the switching field distribution in large ensembles, which is a big issue in new storage media based on bit pattern media.[10−12]

In the following the main features of the method are explained. The basic constituent is the fabrication of the nanodots. In brief, the nanomagnets are carved out of a

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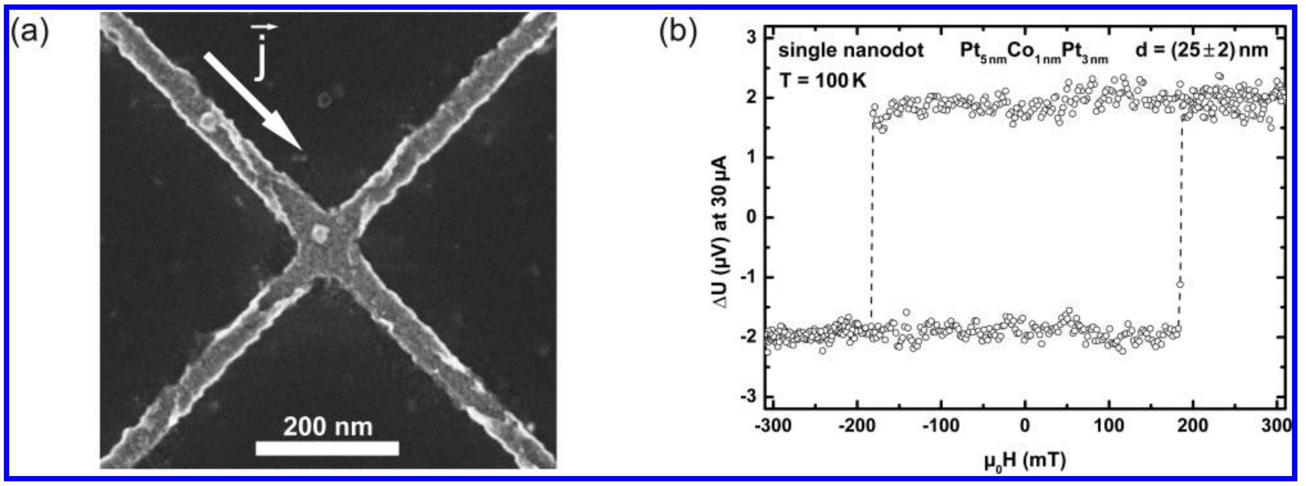

Figure 1. SEM micrograph and AHE signal versus field of a Hall cross containing a single dot. (a) The dimensions of the Hall cross that is created in the Pt seed layer are 3 nm/45 nm (thickness/width). The Co layer of the nanodot has a thickness of 1 nm and is sandwiched by Pt. The diameter of the dot is about 25 nm. (b) The AHE signal gives the magnetic response of the single dot on a field applied perpendicular to the plane of the cross. The measurement was performed at 100 K. The magnetization curve is taken performing a single field sweep. The dashed lines are meant as a guide to the eyes to demonstrate the instantaneous switching between the two stable states of magnetization orientation (up and down).

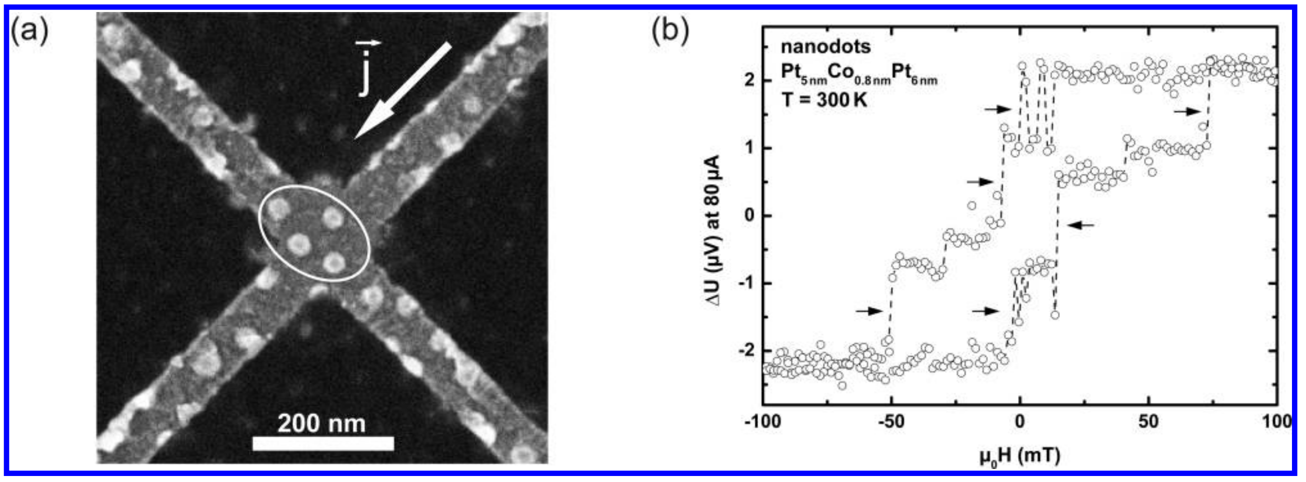

Figure 2. SEM micrograph and AHE signal versus field of a cross containing several dots. (a) The four dots within the crossing region that give signals in the AHE measurements are circled in the SEM micrograph. The Pt wire dimensions are roughly 75 nm in width and 3 nm in thickness. The dots are made from a Co (0.8 nm) film sandwiched by Pt layers. The four dots have almost the same diameter (32 nm). (b) Magnetic response in perpendicular field obtained via AHE at room temperature. The AHE measurement reveals voltage changes corresponding to the magnetic reversal of the four nanodots. Three out of the four dots create rather similar signal heights (marked by arrows.) while one signal is considerably smaller. The latter is caused by the position of the dot that is located at the periphery of the cross.[16]

Pt/Co/Pt multilayer via ion-milling. Low energy ions (Ar^+) are used to clone an ensemble of SiO_2 nanodots into the ferromagnetic film.[13,14] In the milling step the Pt cap and Co layer are removed in the range where the SiO_2 dots do not protect the multilayer. The milling is stopped within the Pt seed layer. The array of SiO_2 nanodots is created by spin coating the surface of the multilayer by diblock copolymer micelles that are filled with SiO_2 and removing the organic shell in oxygen plasma.[15] The magnetic properties of the film can be tuned on purpose via the growth conditions of the magnetic film or multilayer (polycrystalline, textured, magnetic anisotropy, easy axis of magnetization). Next, a Hall cross is fabricated by means of electron beam lithography within the seed layer that still covers the insulating substrate. Depending on dimensions of the Hall cross and separation of the particles (micelle diameter) it is possible to define the average number of particles that is stuck on the cross.

In Figure 1a a scanning electron microscopy (SEM) image of a Hall cross that consists of 45 nm wide platinum wires is displayed. Nanodots are scattered over the surface, some on top of the electrical leads (ferromagnetic) and some on the nonconductive background. The dot on the cross is made from a cobalt film (thickness of 1 nm) sandwiched by platinum layers exhibiting a perpendicular magnetic anisotropy. An electrical current is put through the wire and the transverse voltage is measured. The voltage is generated by the AHE that is proportional to the magnetization component perpendicular to the plane that is defined by the Hall cross, that is, the ferromagnetic nanodot in the center of the cross with diameter of 25 nm. A single scan of the magnetic field that is oriented perpendicular to the plane of the cross gives the hysteresis shown in Figure 1b. Depending on the orientation of the magnetization (up/down) two signal levels appear that indicate the two stable states in remanence of a uniaxial system. Instantaneous reversals appear in the single loop as multidomain states do not exist in such small magnets. The signal-to-noise ratio is ∼15 (amplitude ratio) yet not optimized. The signal is determined by the ratio of cross section of the leads with respect to the dot size and the conductivity of the involved materials.[6] A large signal is observed as the resistivity of the Co is smaller than the resistivity of the Pt. The electrons move partially through the cobalt in the dots where they experience the scattering that generates the AHE voltage. The single dot measurement allows for an estimation of the smallest detectable dot size. The nanodot used in the presented measurement contains roughly 45 000 cobalt atoms (with $V = 0.011$ nm^3 as the volume of a cobalt atom). Taking a signal-to-noise ratio of one, corresponding to the half of the total signal, the switching of a nanodot with about 3000 cobalt atoms could still be resolved. This corresponds to a spherical particle with a radius of about 3.3 nm or taking our cylindrical geometry a nanodot with a diameter of about 14 and 1 nm height can be studied.

On varying the dimensions of the Hall cross and/or the micelle diameter it is possible to tune the number of nanodots within the Hall cross. The total magnetic response of a system containing four dots with diameters of about 32 nm is shown in Figure 2b. In the transport measurement, the current is put through the wire as indicated in the SEM micrograph (Figure 2a). The voltage curve (determined by the perpendicular component of magnetization), obtained from a single field scan, is shown in the plot. For every sweep of the field (up/down), three abrupt switching events (marked by arrows) can be clearly distinguished while one dot causes a very small signal (switching field ∼40 mT). The latter is due to the position of the dot at the periphery of the cross where a reduced current density causes a smaller AHE voltage.[16] In addition, the magnetization reversal at small fields shows a back and forth switching, which indicates a temporal instability of one of the dots. The instability is most likely caused by the fact that the energy barrier due to the magnetic anisotropy is comparable to the thermal energy. In the language of superparamagnetism it means that the dot is close to its blocking temperature.[17,18] Because of the stochastical nature of the process the occurrence of switching is different in the two sweeping branches. The almost identical signal heights indicate that the magnetic volumes of the three dots contain nearly the same amount of magnetic material. The latter fact is insofar surprising as the magnetic behavior of one dot is strongly deviating from the behavior of the others.

The fact that three dots show similar signals indicates that an independent procedure is needed to allocate the particles to the individual switching events. For that purpose, simulations have been performed to learn about the signal strength of a magnetic dot at different positions on the Hall cross. A commercial finite element code[19] has been modified to describe the ferromagnetic dots. While the Hall voltage is implemented in the code, the AHE is not. Our modification of the code uses the existing conductivity tensors for the Hall-Effect, while the higher AHE constant for cobalt is used.[20] The latter transport properties are only applied within the dots (see Supporting Information for further details). The size of the dots and the cross are taken from the SEM micrographs. The difference in transverse voltage for up and down orientation of the magnetization in the dots gives the signal that is used to determine the sensitivity as a function of lateral position. The obtained results are almost identical to published analytical calculations and numerical treatments that were experimentally cross-checked.[21−23] The spatial variation of the sensitivity helps to identify the dots in the AHE signal as long as they are situated in regions with considerably different sensitivity. As the variation within the central part is only small,[21−23] the identification via signal heights is in general not possible. Therefore a new method to unambiguously assign the dots to their respective signal has been developed. On the basis of the modified code, we simulate the current and voltage distribution in geometries when the current is applied between two adjacent leads (Figure 3).

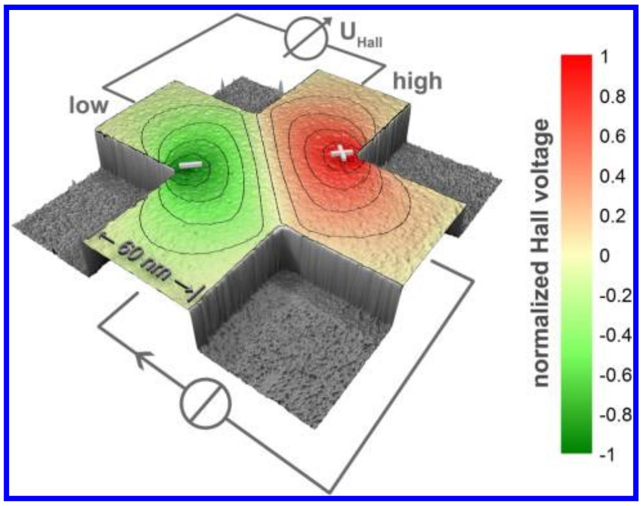

Figure 3. Simulation of the local AHE response. The current is applied between adjacent leads and the AHE voltage is measured between the opposite wires. The color coding represents the relative signal heights as well as the sign of the voltage (green/red) in the given geometry for variation of dot position The signal plotted represents the voltage change on reversing the orientation of magnetization from pointing down- to upward. The signal vanishes along the diagonal separating the two wires used for current injection and/or voltage measurement (white).

The characteristic result of the simulation is displayed in Figure 3. The main outcome is that the sign of the signal depends on the position of the nanodot with respect to the diagonal of the cross. The diagonal that lies between the leads where the voltage is measured is a line of zero response. For particles above and below this line, the voltage response for a given magnetization orientation is reversed, that is, voltages of opposite polarity are generated. An equivalent symmetry appears for all four possible permutations of orientation of current and measuring leads. The different geometries can be applied to identify the particles. An example is presented in Figure 4

In Figure 4b, a single loop (blue) and an averaged voltage curve, consisting of 107 loops, are plotted for a cross containing three particles. Again the switching of two particles is evident from the plot. Traces of the third particle are apparently missing. A closer look reveals that the continuous change around zero is the trace of the switching activity of a particle. The particle behaves superparamagnetically like in the former case, however, with a switching frequency that is considerably higher than before. The individual switching processes cannot be resolved within the dwell time. The continuous change of signal is due to a shift of the probability of occupying the down or up state. To proceed as proposed the measurements utilizing adjacent leads have been made. The results displaying single loops as well as averaged magnetization curves are shown in Figure 4b,c. At first, it is evident that the switching of the dot

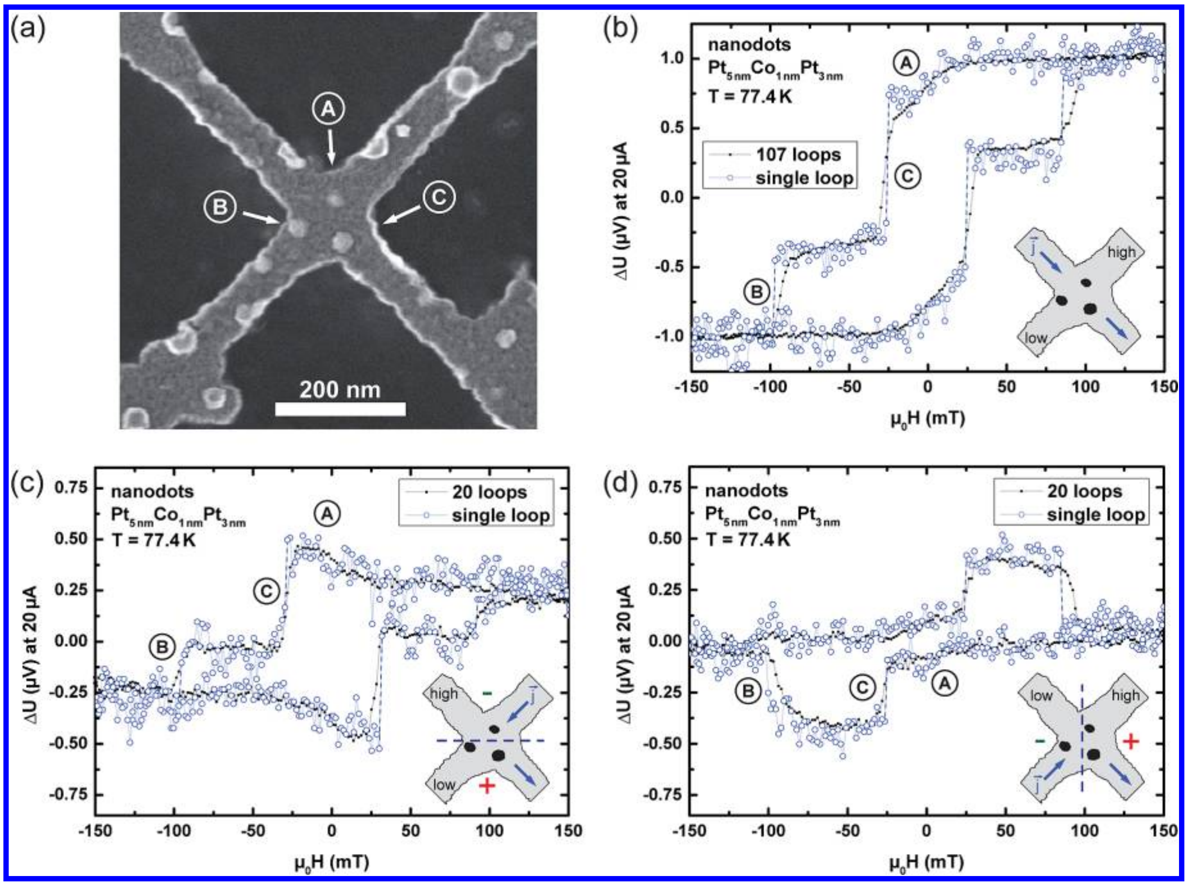

Figure 4. SEM micrograph and voltage versus field for different measuring geometries. (a) The SEM micrograph shows a Hall cross that contains three Co dots. The diameters of the dots are 18 (A), 22 (B), and 24 nm (C), respectively. In (b) the response of the three dots is shown in the conventional AHE geometry on applying a vertical field at 77.4 K. Two quite large, abrupt jumps (B,C) are notable. Additionally, a continuous change of AHE voltage (A) around zero is found. The magnetic response of all three dots is also found in the nonconventional geometry (panels c,d). The jumps appear at the same field strength as in panel b. The jump at about 100 mT has the same sign in (b) and (c) while in (d) the sign is reversed. From the symmetry features shown in Figure 3 this immediately allows the identification of the dot that is responsible for the signal, that is, the dot on the left-hand side of the cross.

with the largest coercivity changes sign in the two geometries while the dot with the second largest coercivity does not change sign. From the simulation, it is evident that the dot with the smaller coercivity is the one labeled "C". From the signals in the respective geometries the two others can also be easily assigned, that is, the small particle as "A" and the larger particle "B". As the latter two particles are very different in size the identification is also unambiguously possible from their magnetic behavior. The superparamagnetic particle should be the small one in complete agreement with the results from the symmetry considerations. In this case, the symmetry does not give conclusive results a comparison of experimental and theoretical signal heights have to be made. In the case discussed here, we proceed as follows. In the conventional Hall geometry (Figure 4b), the signals are normalized to the signal of dot "C". The following ratios are obtained from the experiment: $(49 \pm 4)\%$ (A) and $(77 \pm 6)\%$ (B), while the simulation yields 43% (A) and 71% (B). The agreement is reasonably good in particular when considering that the sensitivity scales with the volume, that is, the area of constant thickness, of the magnetic material.[22] The above deviation can be easily assigned to a small error in the "magnetic" diameter of the dot "C". To get rid of this uncertainty when comparing the other geometries (Figure 4c,d) the signals are normalized to the respective values in the conventional measurement (Figure 4b). For the geometry shown in Figure 4c the experimental values with respect to the straight measurement are$(-38 \pm 7)\%$ (A), $(29 \pm 3)\%$ (B), and $(53 \pm 3)\%$ (C) while the simulations give -39% (A), 27% (B), and 52% (C). Similar findings are obtained for the second geometry, that is, simulation/experiment $21\%/(26 \pm 7)\%$ (A), $-58\%/(-50 \pm 4)\%$ (B), and $30\%/(31 \pm 4)\%$ (C). Vice versa, the close agreement between simulation and experiment can be utilized to cross check whether the magnetic volume of the particles are identical to the size obtained from SEM images.

In summary, it is demonstrated that the method presented here has the sensitivity and potential to study single nanodot behavior. Utilizing the unconventional Hall geometry the magnetic switching can be assigned to individual particles in an ensemble of a few particles. The method gives access to experiments covering a wide thermal and dynamical range. A more promising aspect is the capability to investigate interactions and mutual impact due to certain arrangements and/or varying properties of particles. First, traces of such effects have already been found. As demonstrated in the above examples, it turns out that apparently identical particles are behaving different. Even when the dots seem to have same volumes, the signal height or the reversal characteristics can vary when revealing some impact of the fabrication process. The direct measure of the latter nanodot properties proves the superiority of this approach in research on nanoscale devices over studies on ensembles. The underlying concept can also be applied to more complex geometries, for example, crosses with an increased number of leads. The more leads that are used the better the localization of the source of a particular signal can be achieved, which allows the assignment of the signals in ensembles with even larger number of particles.

ASSOCIATED CONTENT

Supporting Information

Additional Information on the anomalous Hall-Effect and the simulation procedure. This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*E-mail: aneumann@physnet.uni-hamburg.de.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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