



The microscopic physics of electronic paper revealed

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Abstract

For many years, it was thought that passive matrix addressing of electrophoretic image displays (EPIDs) was impossible due to the absence of a threshold. We proved that there are ways to bypass this [Proc. 22nd Int. Display Res. Conf. (2002) 251]. However, the physical basis was not fully understood. This article presents new insights in the physics of EPIDs. By applying the theory of dielectrophoresis (DEP), several discrepancies between theory and measurement, that occurred in explaining properties of EPIDs, are solved. We introduce a new terminology: DEPIDs. For the first time the theory of DEP is applied to model the movement of pigments in electronic paper.

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1. Introduction

Electrophoretic image displays (EPIDs) are one of the main competitors in the quest for electronic paper. Their functioning is based on the movement of colored pigments in a solvent under the influence of an electric field. Pigments are dissolved as a colloidal solution in a colored solvent; the solution is placed in a narrow space (approximately 125 μm thick) between two transparent electrodes. The pigments move when an electric field is applied, generated by a voltage difference between the electrodes. When the field is applied in one direction, the pigments move towards the observer and the pixel has the color of the pigment. When the field is applied in the opposite direction, the ITO-electrode closest to the observer is covered with colored solvent. If the light extinction coefficient of this solvent is high enough to make sure that the pigments on the opposite electrode cannot influence the light reflection, the pixel will have the color of the solvent. By giving pigments and solvent a contrasting color, one can construct a reflective display which looks like ink on paper.

These displays provide all the advantages desired from electronic paper. The pigments are bistable, once attracted to an electrode. Therefore an image written onto a display is retained for a long time without applying voltages on

the electrodes, hence without consuming power. In contrast to other frequently used display types (CRT, LCD) they show a high contrast in an outdoor environment. Constructed as reflective displays, with an ink-on-paper appearance, their image quality does not suffer from bright surrounding light, as is the case in an outdoor sunlight environment. They have a potential for combination with flexible substrates. In their intrinsic construction, nothing impedes flexibility: both the colloidal liquid, the medium in which it is sealed and the ITO electrode coating can be made flexible. This does not only improve the sturdiness, but also makes EPIDs one of the potential solutions where flexible displays are needed.

Due to the absence of a threshold in the switching properties, it was long thought that passive matrix addressing of EPIDs was impossible. Indeed, the pigments within each pixel move when a non-zero electric field in the right direction is applied. This would make EPIDs less attractive for commercial use, since the development of active matrix addressing in flexible large area applications is still in an early phase. Direct line addressing is too complex when high resolution displays need to be addressed. We proved however that passive matrix addressing is indeed possible [2]. This was shown on a 5×7 prototype display.

The switching speed remained a problem. The properties of the switching scheme delayed the switching speed of each pixel to much lower values than the optimal value. Also, since there was no insight in the internal physics of

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EPIDs, no sound explanation for the results could be given. Even though there was still no threshold in the switching properties, we did design a passive matrix addressing scheme based on other physical properties of the display. The origin of these properties was unknown. This paper reveals the microscopic properties and behavior of a switching pixel. For the first time a theoretical explanation for unexplained properties is given.

2. Problem definition

Applying the theory of electrophoresis to the results found from optical and electrical measurements on displays, leaves some discrepancies between theory and practice.

2.1. Removal time

There is no explanation for the measured delay time between application of the electric field and the optical response of the display [1,8]. In Fig. 1, it can be seen that the optical response lags behind the application of the external field. Hopper and Novotny [8] found a theoretical model for the optical response of a reflective electrophoretic display. They assume a normal distribution for the pigment velocity during switching. The pigments have an average velocity v_a and the standard deviation in the velocity is v_s . This gives a normal distribution for the position of the pigments along the axis of the electric field. If this the x -axis, then the number of pigments at position x at time t is given by:

$$n(x, t) = \frac{N}{\sqrt{2\pi}v_s t} \exp\left(-\frac{(x - v_a t)^2}{2(v_s t)^2}\right)$$

N is the total number of particles in the system.

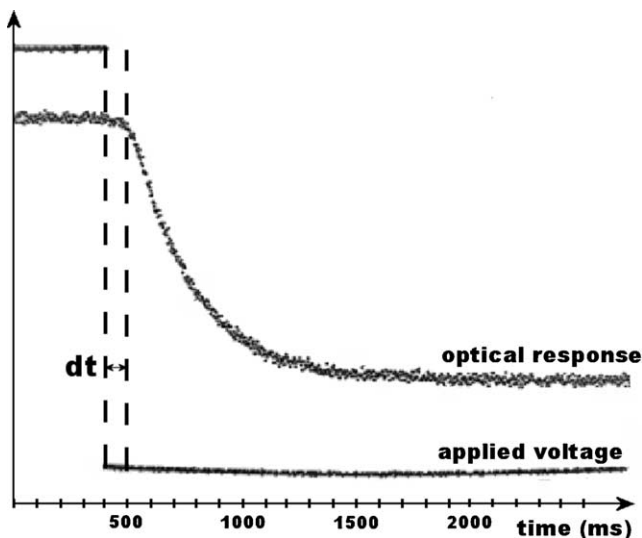


Fig. 1. Delay time between optical response and application of external electrical field.

The intensity of the reflected light varies exponentially with the distance to the observer. The formula in case all particles move at the same velocity in a densely packed group towards the observer is:

$$I(t) = NI_0 s e^{-\alpha a(d-vt)}$$

I_0 is the input intensity originating from the light source, α is the extinction coefficient of the medium, s is the scattering efficiency and a is a geometrical factor that varies with the incidence angle of the light and dielectric properties of the medium.

Hopper and Novotny had to include a removal time in their model to make it accurate. The particles are not removed instantaneously from the electrode after application of the electric field. They introduce a function $R(t)$ to model the normal distribution of the removal time:

$$R(t) = \frac{1}{\sqrt{2\pi}t_s} \exp\left(-\frac{(t - t_a)^2}{2t_s^2}\right)$$

particles have an average removal time t_a ; the standard deviation in the removal time is t_s . When we replace t in the formula $n(x, t)$ by $t - t_{\text{removal}}$ and combine this formula with these for $I(t)$ and $R(t)$, we find for the intensity, hence the optical response of a switching pixel:

$$I(t) = I_0 s \int_{-\infty}^{\infty} R(t_u) \left[\int_0^d n(x, t, t_u) e^{-\alpha a(d-x)} dx + \int_d^{\infty} n(x, t, t_u) dx \right] dt_u$$

this formula is for pigments moving towards the observer. The formula for movement away from the observer is similar.

This formula is more accurate than the one found by Hopper and Novotny and is in accordance to the results found in Ref. [1].

Hopper and Novotny cannot explain what the physical origin of the removal time $R(t)$ is. They suppose a redistribution of charge on the particle surface, leading to a change in the image force, has something to do with it. No certain explanation is given.

The theory of electrophoresis predicts no such delay time. Colloidal particles, such as the pigments in a pixel, that move under the laws of electrophoresis respond immediately when an electric field is applied. This is the first discrepancy between theory and practice.

2.2. Unrealistic physical properties

It is impossible to match measurements to theory without using non-realistic values for physical properties. Matching the measured speed and force to the laws of electrophoresis gives values for the zeta-potential above 1 V and values for the charge on each pixel that exceed normal values [6]. We apply the formulas used by Comiskey et al. in Ref. [3].

The Reynolds number is given by:

$$Re = \frac{\rho v r}{\eta}$$

ρ is the internal fluid density, r is the particle radius, η is the internal fluid viscosity and v is the particle velocity. In our system, we have $\rho = 0.804 \text{ g/cm}^3$; $v = 100 \text{ }\mu\text{m/s}$; $r = 0.4 \text{ }\mu\text{m}$; $\eta = 0.003875 \text{ kg/ms}$. Values for the fluid density and viscosity came from the data sheet of the product: Isopar P. The fluid velocity came from matching of the Hopper and Novotny model [8] to the results found in Ref. [1]. This gives a value for the Reynolds number of $1.04\text{E-}5$, it is therefore safe to presume a laminar flow.

The formula for particle velocity in a laminar flow (in regime) is:

$$\mu = \frac{v}{E} = \frac{\epsilon \zeta}{6\pi\eta} = \frac{q}{12\pi r\eta}$$

E is the electric field, ζ is the zeta-potential, ϵ is the dielectric constant of the internal fluid and q is the charge per particle. Using the results from Ref. [1], we find values for the zeta-potential above 2.5 V: this is not a realistic value. Normal values for zeta-potential are well below 1 V. For the charge per particle, we find equally unrealistic high values.

This second discrepancy between theory and practice leads us to believe that the theory of electrophoresis is not sufficient to explain the properties of a switching pixel.

2.3. Response to AC fields

There seems to be an optical response, hence a non-zero net force on the pigments, when a pure AC field is applied. This was measured in Ref. [1] and is shown in Fig. 2. This only occurs at sufficiently high frequencies. Applying the theory of electrophoresis, one would expect no net movement of the pigments, since every cycle of the field exercises an exactly opposite force on the pigments. Even at higher frequencies—we measured approximately to 20 Hz—the theory of electrophoresis predicts no net force when a square wave with zero DC value is applied. The force exerted on a particle by an electric field is instantaneous and

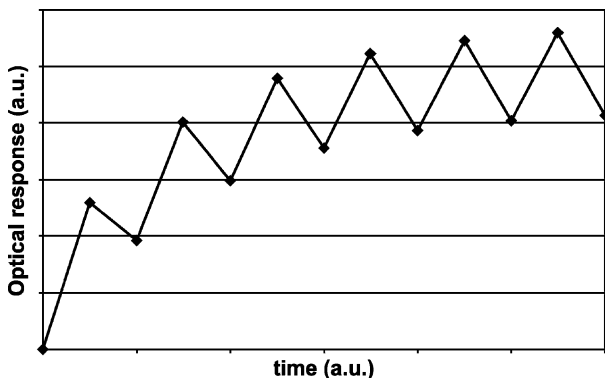


Fig. 2. Optical response when a pure AC voltage is applied.

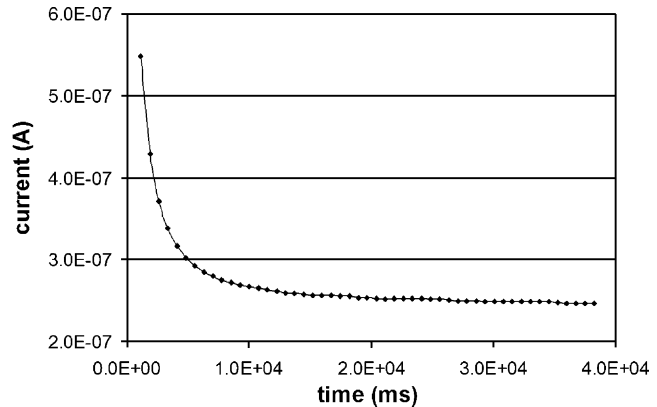


Fig. 3. Electric current in a pixel during switching.

proportional to the applied field. When the direction of the field inverts (as is the case every half period of a square wave), electrophoresis predicts that the force on the particle also changes direction, but that the magnitude stays the same. If you apply this opposite force for the same time as the first half period, the particle returns to its original position. Therefore, according to the theory of electrophoresis, it is impossible to cause a net movement of a particle by applying a block wave with a zero DC value.

This third discrepancy between theory and practice confirms that there must be other forces at work than only these of electrophoresis.

2.4. RC-model

We measured current and voltage between the electrodes of a 3×3 display during switching. The results are shown in Fig. 3. It is possible to match a macroscopic RC-network to these measurements to model the electrical response. The network of Fig. 4 gives good results. Matching of the parameters of the model based on the time-constant, steady-state current and start-current gives a deviation of less than 1% with the measurements. Matching this to the physics of electrophoresis, one cannot find a source for the capacitive part of the macroscopic model. There is no microscopic physical part of electrophoresis that has a delay

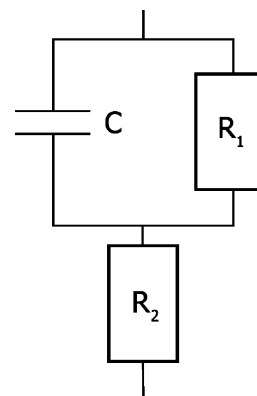


Fig. 4. Equivalent RC-network to fit the current measurements.

time, as a capacitor does. As mentioned in the previous paragraph, the response of a particle to a field is instantaneous. The model predicts accordance to loading a capacitor: an exponential behavior where a certain property reaches an asymptotic end value with a typical time constant.

According to Hopper and Novotny [8], the steady-state current is controlled by ionization of charge control agent in the solution. This is indeed probably the reason for the non-zero value of the steady-state current, but it gives no explanation for the capacitive behavior. Not only would an exponential behavior of the ionization current lead to an exponential augmentation of the current (in stead of an exponential decay as in Fig. 3), also the values for the time constants are different. Molecular ionization under the influence of an electric field happens within a timescale of some nano-seconds, more than a million times faster than the time scales measured in the current-curves.

Clearly another phenomenon than the ionization is responsible for the steady-state current that is the source of the capacitive behavior. Electrophoresis does not have such a behavior and therefore does not provide an explanation.

2.5. Passive matrix addressing

Applying solely electrophoresis gives no explanation for the proven passive matrix addressing scheme [2]. For many years, it was thought that passive matrix addressing was impossible since no threshold was measured and since electrophoresis did not predict one. We proved that passive matrix addressing is indeed possible by finding a difference in the optical response between application of a voltage step and an AC-signal with the same DC-value as the voltage step [2]. We found that a pigment responds faster to an AC-signal with a non-zero DC-value than to a DC-step. By using the right combination of block waves on the rows and columns, this principle was used to develop a passive matrix driving scheme. The principle was proved on a 5×7 prototype.

3. Solutions and results

All the discrepancies between measurement results and electrophoretic theory can be solved by applying the theory of *dielectrophoresis (DEP)*.

3.1. What is dielectrophoresis?

Dielectrophoresis is the motion of polarizable particles in a locally non-uniform field and arises from the interaction of the electric field and the induced dipole [5]. The induced dipole from a particle symmetrically surrounded by counter ions under the influence of an external field, is modeled in Fig. 5 [7].

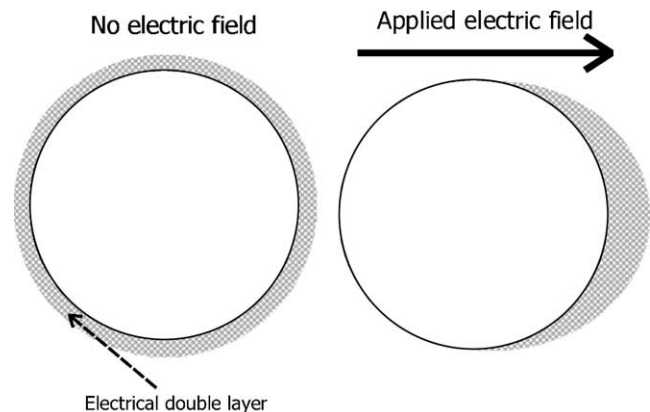


Fig. 5. Model of the polarization of a pigment and its counter ions in an electrical field.

The force on such a polarized particle is given by:

$$F = 2\pi\epsilon_m R^3 \text{Re}(f_{CM}) \nabla |E^2|$$

ϵ_m is the permittivity of the medium in which the pigments are dissolved, R is the radius of the pigments, $\text{Re}(f_{CM})$ is the real part of the Clausius–Mossotti factor and $\nabla |E^2|$ is the gradient of the square of the electric field. Inside a pixel, the gradient in the local field comes from the presence of other particles in the proximity. Wang et al. derived a formula for the dielectrophoretic force acting on two neighboring particles resulting from dipole–dipole interactions induced by an applied electric field [12]. In our configuration, the applied electric field by the electrodes is uniform. It is the presence of other particles and the induced dipole moment in these particles that makes the field locally non-uniform. It is this induced non-uniformity in the electric field that provides the gradient in E^2 needed to exert a dielectrophoretic force.

According to Wang et al., the force on a pigment at position (x, z) under the influence of the induced dipole of a particle on position $(0, 0)$ by an electric field E in the z -direction is:

$$\bar{F} = 2\pi\epsilon_m R^3 E^2 (0.5 \text{Re}(f_{CM}) |f_{CM}|^2 \nabla(P^2 + Q^2) - |f_{CM}|^2 \nabla P)$$

$$\text{where } P(x, z) = R^3 \frac{x^2 - 2z^2}{(x^2 + z^2)^{2.5}} \text{ and } Q(x, z) = 3R^3 \frac{xz}{(x^2 + z^2)^{2.5}}$$

are spatial functions.

R is the pigment radius, ϵ_m is the permittivity of the medium in which the pigments are dissolved and f_{CM} is the Clausius–Mossotti factor. E is the applied electric field, the force is proportional to the square of the electric field.

Disruption of an external uniform field to form a locally non-uniform field is modeled in Fig. 6.

3.2. Realistic physical properties

Comparison between dielectrophoretic theory and measurements gives realistic values for all physical properties [9,12]. When applying the measurement values

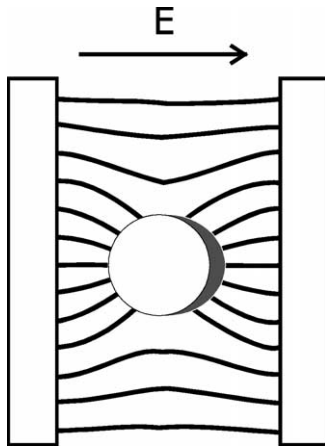


Fig. 6. Inhomogeneities in a uniform field due to the presence of a pigment.

from Ref. [1] to the formulas derived by Wang et al. [12], one finds a satisfying accordance. Fig. 7 shows the velocity of a particle at position (x, y) in the non-uniform field induced by a particle in $(0, 0)$. The plotted values are \log_{10} of the velocity values in micron per second. The velocity drops fast with the distance between the particles; this is due to the spatial functions $P(x, z)$ and $Q(x, z)$ in the formulas from Wang et al. Yet at distances of many times the particle radius the velocities still have satisfactory values to correspond with the measurements.

Contrary to electrophoresis, DEP does provide a match between theory and practice when it comes to realistic values for physical properties. Filling in the physical properties of our display material into the formulas for dielectrophoretic force yields the right values for force and hence velocity of the pigments.

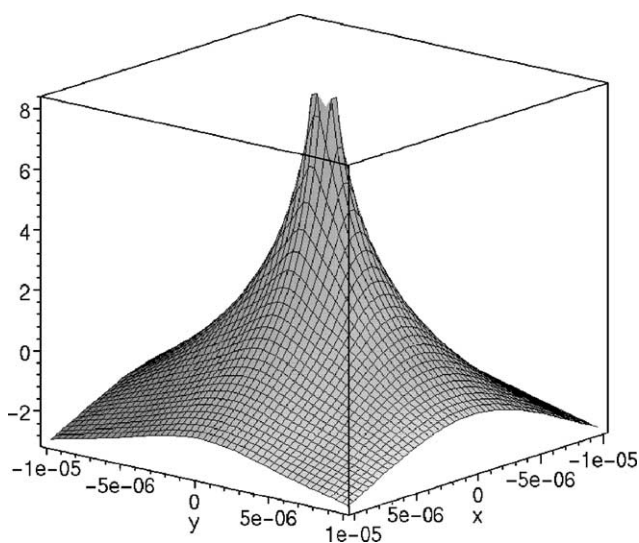


Fig. 7. Velocity of pigment in position (x, y) due to non-uniformity in the electric field induced by a pigment in $(0, 0)$. The plotted values are \log_{10} of the velocity in micron per second.

3.3. Delay time explained

DEP gives an explanation for the occurrence of a delay time in the switching properties. Schwarz's formula gives a value for the time constant needed to induce a dipole from a particle at rest [9,10].

$$\tau = \frac{R^2 e}{2\mu k T}$$

R is the pigment-radius, e is the elementary charge, k is the Boltzman-constant, T is the temperature and μ is the surface mobility of the counter ions. As derived by Hu and Chen in Ref. [9], the polarization and hence the dipole moment of the particle in an electric field evolves exponentially to an asymptotic value. The time constant for this evolution is given by Schwarz's formula. Since we have no accurate data on the surface mobility of the counter ions around the pigments, we can only estimate a value for this mobility using the measured values for the time constant. This gives satisfactory results for the surface mobility of the same magnitude as surface mobilities in other systems.

This finite time needed to induce the dipoles explains the delay time measured in the switching properties. When there is no electric field applied, the counter ions are symmetrically spread out around every pigment. When an electric field is applied, there is no immediate movement of the particles, since it takes a finite time to form the dipoles that induce the non-uniformity in the electric field. The polarization of the pigments takes place with a time-constant τ given by Schwarz's formula. The dielectrophoretic force reaches its steady-state value with the same time-constant. This is why there is no direct response when an electric field is applied and one has to introduce a removal time to model the optical response, just like Hopper and Novotny did in Ref. [8]. They assumed a redistribution of charge on the particle surface was at the basis of this removal time. They were right, since a particle-polarization is indeed a redistribution of the surface charge. Yet it is not the mirror charge of this redistributed charge that is at the basis of the removal time, but the finite time needed to redistribute this charge and form a dipole. Only when this dipole is formed, it can move under the influence of a non-uniform field induced by the other dipoles.

The theory of DEP again proves to be useful; this time to explain the phenomenon of the delay time.

3.4. Net movement in a pure AC field

Applying DEP to EPIDs also explains why at high frequencies there is a net pigment-movement, even though a pure AC signal is applied. This is modeled in Fig. 2. If the period of the applied field is lower than the delay time needed to induce the dipole, it seems as if the dipole

‘cannot follow’ the field. Schwarz’s formula gives a value for the time constant needed to induce a dipole around a pigment in an electric field. This time constant gives an approach for the minimum time a field has to be applied in a certain direction to fully induce a dipole in that direction. If the maximum dipole switching speed is exceeded by the external field, the dipole will not switch completely when the field switches, resulting in a net dipole-moment and therefore a net force in one direction. If you apply an electric field (let us say at $t = 0$) with half a period ($T/2$) shorter than the time-constant from Schwarz’s formula to a pigment at rest, the following will happen: at $t = T/2$, when the field direction switches, the dipole will not have reached its asymptotic value yet. The field direction switches and the formation of a dipole in the other direction commences. This time not starting from a symmetrical rest phase (as was the case at $t = 0$) but beginning from a partially formed dipole in the other direction. Therefore, at $t = T$, when the field switches back to the original direction, the dipole will not only be formed only partially; the dipole moment will also be less than on $t = T/2$. This is because the starting configuration on $t = 0$ is a symmetrical condition and on $t = T/2$ is an induced condition in the opposite direction. After application of several periods of such a block wave the dipole moment will have a non-zero net value. Therefore, there will be a net movement, even though the average value of the applied field is zero.

Fig. 8 shows how the contrast of a switching pixel varies with switching frequency. The contrast is the ratio of the optical response of an ‘on’ and an ‘off’ pixel. As the frequency increases, it is harder for pigments to follow the externally applied field so the contrast decreases. This difference in optical response between fast and slow driving signals, is one of the properties used in the driving scheme of Ref. [2].

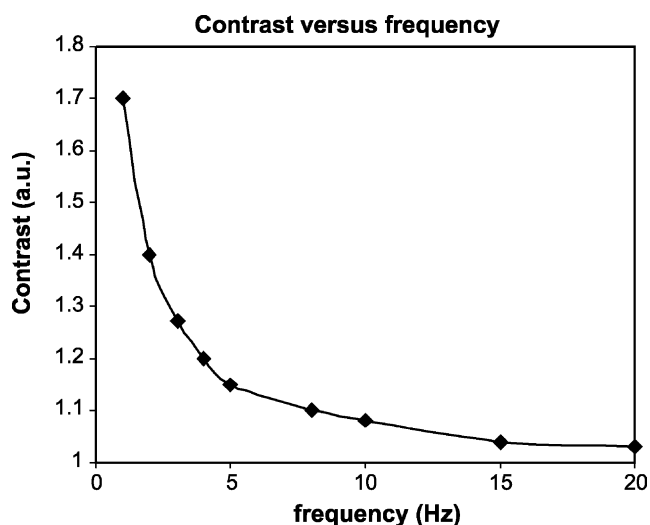


Fig. 8. Reachable contrast during display switching as a function of switching frequency.

3.5. RC-model

It is possible to match a macroscopic RC-model to the microscopic dielectrophoretic theory. Polarization of the pigment to form a dipole has the same properties as loading a capacitor [4,11]. By using a simple RC-network, it is possible to match the current-measurements shown in Fig. 3. Ramos et al. [11] compare the polarization of the pigments to the charging of a capacitor. Indeed, it is also physically evident that the evolution of a polarized pigment to its asymptotic end state in an exponential way is very similar to exponentially charging a capacitor with a non-zero resistance. In both cases, charges move exponentially to an asymptotic distribution. The macroscopic RC-model fitting the current measurements is shown in Fig. 4.

Here the pigment is modeled as a capacitor, the solvent surrounding the pigment is modeled by adding resistors in series and parallel. In fact, to model a complete pixel, one should connect a very large amount of these RC-networks in series and in parallel. This does not change the fundamental switching form, therefore this simple model is sufficient. The current through this network has to match the measured current from the display for the model to be correct. From Fig. 4, we see that on application of a voltage step V over this network, the current through the network evolves as:

$$I(t) = \frac{V}{R_2} e^{-t/\tau} + I_{ss}(1 - e^{-t/\tau})$$

I_{ss} is the steady state current given by

$$I_{ss} = \frac{V}{R_1 + R_2}$$

τ is a time constant given by

$$\tau = \frac{R_1 R_2}{R_1 + R_2} C$$

matching the $I(t)$ from this model to the measured values gives an estimate for the values of R_1 , R_2 and C .

3.6. Passive matrix addressing

The most important result is that the dielectrophoretic theory makes it possible to explain why the passive matrix addressing scheme works. Consider again the macroscopic RC-model from the previous paragraph. If we calculate the voltage over the capacitor when applying a voltage over the complete network, this voltage is representative for the force on a pigment and therefore the velocity of this pigment. The voltage over the capacitor when applying a voltage step V over the complete network is given by:

$$v(t) = V \frac{R_1}{R_1 + R_2} (1 - e^{-t/\tau})$$

τ is the same time constant as in the previous paragraph.

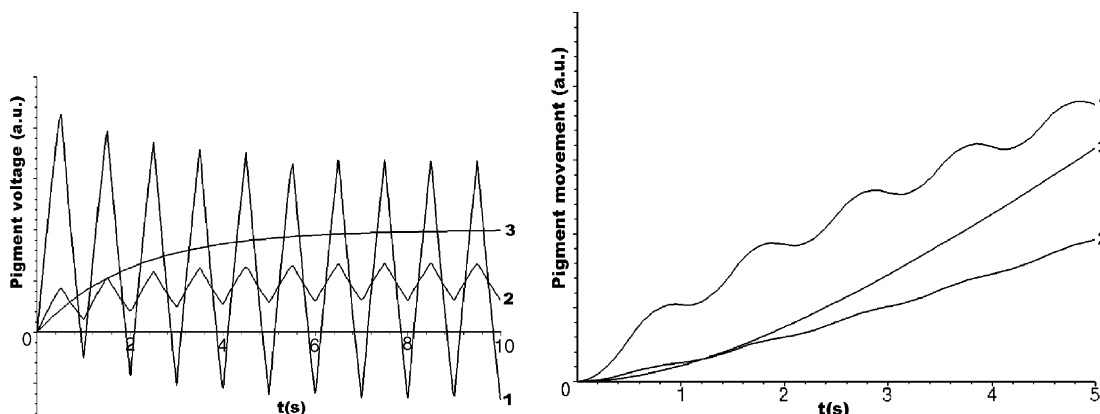


Fig. 9. Force on pigments and displacement of pigments according to the RC-model. 1: on application of a block wave voltage $+20\text{ V}/-18\text{ V}$, 2: on application of a block wave voltage $+4\text{ V}/-2\text{ V}$, 3: on application of a voltage step of $+2\text{ V}$.

This voltage in the macroscopic model can be matched to the field felt by every pigment, and therefore the force on every pigment. If we suppose the frictional force on a moving pigment to be proportional to the speed of this pigment, then the force applied on a pigment is proportional to the velocity. Therefore, the above RC-network not only allows to model force but also velocity. Integration of this velocity in time gives the movement of a pigment along the axis of the applied field.

Applying the voltages used in the passive matrix addressing scheme to the macroscopic RC-model, confirms the results found in the previous paragraph. Fig. 9a shows the forces on a pigment on application of an AC square wave voltage $+20\text{ V}/-18\text{ V}$ (curve 1), AC square wave voltage of $+4\text{ V}/-2\text{ V}$ (curve 2) and application of a voltage step of $+2\text{ V}$. Fig. 9b gives the movement on application of these fields (integration of Fig. 9a). The period of the square wave was chosen smaller than the time constant τ found in the RC-model. This confirms the results found in Ref. [2]: even though both square waves have the same DC value, they give a different response. The magnitude of the AC-part also has an influence. Also frequency has an influence, since the effects on which the addressing scheme is based only occur at sufficiently high frequencies: higher than the one defined by $1/\tau$.

3.7. Optical response relaxation

When the applied voltage is removed after switching of a pixel, the optical response drops a little bit. Even if the voltage is applied long enough to make sure all pigments are placed at the electrode surface, one sees a relaxation in the optical response, once the voltage drops. The dielectrophoretic theory confirms this. The dipoles induced on application of the external field are in phase and therefore attract each other. Along the axis of the dipole an attractive force is felt, perpendicular to this axis a repulsive force is felt. Therefore, the dipoles arrange themselves

close to each other in chains along the axis of the applied field. If this field is removed, the pigment polarization also disappears and therefore also the attractive force. The pigments rearrange themselves in their symmetrical situation where every pigment is surrounded by an electrical double layer. Hence, the distance between the pigments increases a little bit and therefore the packing density at the electrode surface decreases. According to the theory of Hopper and Novotny [8], this leads to a decrease in the optical response.

3.8. Results

Fig. 10 shows a photograph of a 5×7 prototype display during switching. The display was addressed using the passive matrix addressing scheme as explained in Ref. [2]. Pixel size was approximately $1 \times 1\text{ cm}^2$. The 7 rows and 5 columns were addressed using a high voltage driver since voltages up to 28 V were required. The spacer placed between the 2 ITO-covered glass plates had a thickness of $127\text{ }\mu\text{m}$.

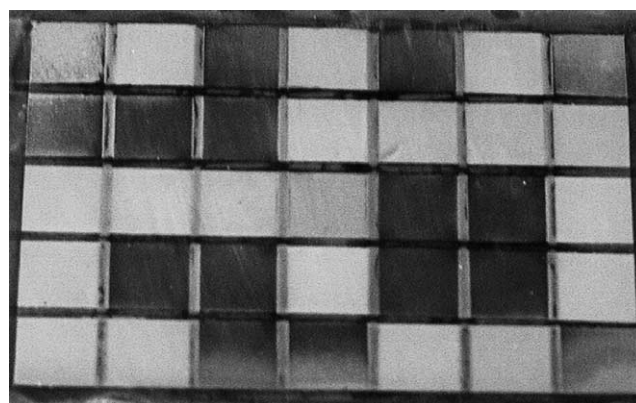


Fig. 10. Photograph of a 5×7 display during passive matrix switching.

One can see a degradation of image quality near the edges of the display. This is caused by problems with the display sealing.

4. Discussion

Of course, a better insight in the microscopic physics does not solve all problems. The switching speed, which is an important parameter for successful passive matrix addressing remains a drawback. Moving images on an electronic paper will not be seen in the near future. One can attempt to solve it as an electrical problem, by creating new addressing methods which use new wave forms or higher voltages. Alternatively, we can try handling it as a physical problem by using smaller spacer to reduce the pigment travel distance. Specially shaped areas in which the pigments are placed, can help to improve properties. Dealing with the problem as purely a chemical one by creating fast and stable pigments and solutions might help.

A stable, high resolution image on EPIDs is a first and important milestone that was reached; is it the last one?

5. Conclusion

This paper presents new insights in the internal physics of EPIDs. By applying the theory of DEP, several discrepancies between theory and practice can be solved. This is the first time the theory and properties of DEP are used in the field of electronic paper. One may wonder if we should not start talking about DEPIDs: dielectrophoretic image displays.

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