Large Enhancement of Sensitivity by Strong Electric Field

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Abstract

We observed a large enhancement of photographic sensitivity when a light pulse and a high voltage pulse were synchronously applied to a layer of a photographic emulsion. Unsensitized cubic AgBr grains which have an edge length of 0.8μm were prepared and exposed to a light and a high voltage pulse. Photographic sensitivity of the emulsion layer was measured in terms of quantum sensitivity (i.e., number of absorbed photons /grain, which rendered 50% of existing grains developable). The quantum sensitivity of the emulsion, which was measured as $\sim$ 400 absorbed photons/grain in the absence of the voltage pulse, reached to the sensitivity of 4 $\sim$ 5 absorbed photons/grain in the presence of high voltage pulse. The achieved sensitivity is higher than that of a typical level of sulfur-plus-gold-sensitized emulsion.

1. Introduction

About thirty five years ago, there was a tide to study pulsed electric field effects on photographic sensitivity. This trend was triggered by J.Rothstein et al [1], they reported an enhancement of photographic sensitivity by electric field pulse and suggested that the mechanism was an avalanche multiplication of generated photo-electrons [1].

The following studies [2-8] confirms the effect, but not only enhancement but also desensitization were observed, and the mechanism proposed in these studies is not an avalanche multiplication. They proposed a mechanism that the electric field affects the latent image formation process through polarization of electron, hole and mobile ions. Finally the avalanche multiplication mechanism was almost ignored because the electric field sensitization did not exceed the chemical one and new mechanism was not required. Then the tide was almost vanished.

However, several Russian group have been reporting successive study of these effects during '80th. They said that at much higher electric field above 1MV/cm, there is a strong sensitivity enhancement which can be explained by avalanche multiplication[9-13]. But partially because of the conclusion of the studies in '60th and '70th, these studies were not paid any attention for a long time, no further proof or study of their results has been done by another groups and no additional study was reported using these effects until today.

We started this study in order to check the possibility to control the sensitivity of nuclear emulsion, which have been used to study elementary particle physics, by any physical method [16]. In order to check the possibility of electrical sensitivity control as reported by Russian group, we have constructed devices to apply high electric field ($\geq$ $1MV/cm$) pulses in coincide with light pulses and did tests using chemically unsensitized AgBr grains. In order to measure the effect quantitatively and keep the possibility to compare the results with some models, we decided to utilize quantum sensitivity measurement. The quantum sensitivity is defined as the number of absorbed photons/grain which rendered 50% of existing grains developable.

Details of the experimental methods will be described in section 2, our results will be presented in section 3 and in section 4 some discussion will be made to investigate the mechanism of the enhancement observed in this paper.

2. Experimental Method

2.1. Samples

Unsensitized cubic AgBr grains with the edge length of 0.8 μm were used as the sample. The AgBr volume occupancy in dried emulsion was diluted to be 0.65% in order to separate individual grains and developed silver grains. Therefore, we can count them under optical microscope.

These counts are converted to the quantum sensitivity as described later.

An emulsion layer with 27 μm thickness was coated on the transparent electrode (ITO glass).

2.2. Setup

Fig.1. shows a schematic view of the experimental setup. It consists of a light source (Xenon flash lamp), a monochro meter, a timing generator, a high voltage pulse generator, a monitor(oscilloscope), a sample holder and an metal electrode. The time duration of the flash light is about $0.3\mu sec$.

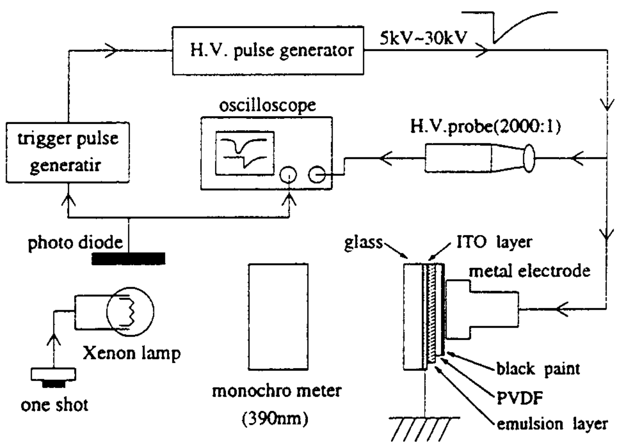

Figure 1: Setup

Because unsensitized AgBr grains are used for the test, the center wave length is tuned to be 390 nm using monochro meter of which slit width corresponds to 12 nm.

High voltage pulse (5-30kV: in this time 15kV was used) is created by a spark gap discharger. The pulse has an exponentially decaying tail following a fast rising edge. The rise time of the fast edge is around 80nsec and the decaying time is 500 nsec.

Our estimation of the electric field strength in AgBr grain is about 1.8MV/cm by the similar calculation as described in [16]. However exact value of dielectric constant of the PVDF is not known especially in the case of impulse voltage. Therefore it is safe to say the estimated field strength is a rough value.

2.3. Sample holder

Fig.2(a) shows details of the sample holder. We use a glycerin pail with glass window, in which the electrode and the sample are immersed. Glycerin is one of the liquid that have a high dielectric constant(ε=43). Using these kind liquids, we can avoid air break down and lightning caused by electric discharges. The sample(emulsion layer on ITO glass) was sandwiched by a glass (3mm thickness) and an metal electrode (W-Cu,1cm ϕ) which is pushed to the sample by a spring. Between the sample and the metal electrode, a plastic film (PVDF, Poly Vinylidene Fluoride which has high dielectric constant of 6.4) was installed as an insulator. The film back was coated by a black paint in order to cut-the reflection light from the metal electrode.

Two kinds of optical filters (20%,50%) and shading are put in front of the glass window in order to make various exposure regions with different light intensity as shown in Fig.2(b).

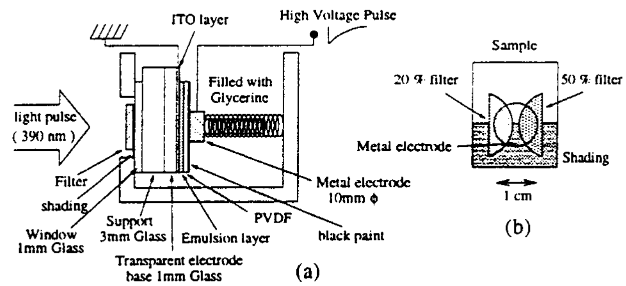

Figure 2: (a)Sample holder, (b)Exposure condition

2.4. Exposure condition

A sample has eight exposure regions, i.e. (light+electric field; no filter, 20%, 50%filter), (only electric field),(only light; no filter,20%, 50%filter),(no light and no electric field), in order to compare the single effect of light or electric field and their combination effect under the same conditions(the same light intensity, the same electric field strength and the same development condition). A schematic figure of the configuration of these regions in a sample is shown in Fig.2(b).

2.5. Measurement of the number of absorbed photons per grain

The number of absorbed photons per grain (:N_{ab}) is calculated by using following parameters measured individually, i.e. light intensity(number of incident photons per unit area through the materials in front of the sample (:F), the absorption coefficient of the emulsion layer (:A), the reflectivity of the PVDF (:R) and the number of grains in unit area of the emulsion layer (:n). Then $N_{ab} = F \times (1+R) \times A/n$. F was measured by a photodiode of which quantum efficiency is calibrated by a standard one. A and R were measured by using optical integral spheres (R is smaller than 0.01). The n was counted under optical microscope as described above. The total error of the calculated absorbed photon number per grain is estimated to be within 15%.

2.6. Exposure

At first, the Xenon lamp is manually triggered. The emitted light is detected by a photodiode which is installed at the side of the Xenon lamp. The signal from the photodiode is fed by a timing generator which create a trigger signal to fire the high voltage pulse. Then the trigger signal triggers the high voltage pulse generator to apply HV pulse to the sample. The delay time between the light and the HV pulse was tuned to be 400nsec. The light intensity, the applied voltage, and the delay time were measured by the oscilloscope.

2.7. Development

Because amidol developer is frequently used in the development of nuclear emulsion, we used it in this study. The containments are Na_2SO_3 $7.2g/l$, $NaHSO_3$ $1.1.g/l$, KBr $3.2g/l$ and $Amidol$ $3.2g/l$. The developing time is 20min at 20^oC. The characteristic curve obtained is almost the same as MAA-1 5min 20^oC development.

2.8. Measurement of the quantum sensitivity

Pre-developed and developed samples are investigated under microscope in order to count the grains and developed silver grains themselves. Using pre-developed samples, the AgBr grain density (n_O) per unit volume are estimated with the accuracy of $\sim$ 7%. For developed sample in each "light+electric field" region, developed grain density (n_D) per unit volume are measured with the accuracy of $\sim$ 7%. In "only electric field" region, we observed a slight increase of n_{DE} even though there is no light. We call them as "Electric fog". The value of n_{DE}/n_O is $\sim$ 0.03. The real fraction of developed grains are calculated as $(n_D - n_{DE})/n_O$.

3. Results

In Fig.3, the measured real fraction of developed grains in the "light+electric field" region are plotted as a function of absorbed photons. The quantum sensitivity(the number of absorbed photons/grain which rendered 50% of existing grains developable) was estimated to be 4 $\sim$ 5 from this figure.

In this figure, the original characteristic curve (without electric field only light) was also shown. This curve was measured by using an optical filter, a normal photographic flash lamp, and an optical wedge. The central wave length was tuned to be 420nm, using the filter of which half band width is 40nm. The duration time of the flash lamp was $250\mu sec$. A film sample(10μm emulsion layer on 90μm TAC base, 0.65% AgBr) was used to require this curve. The quantum sensitivity was estimated to be $\sim$ 400.

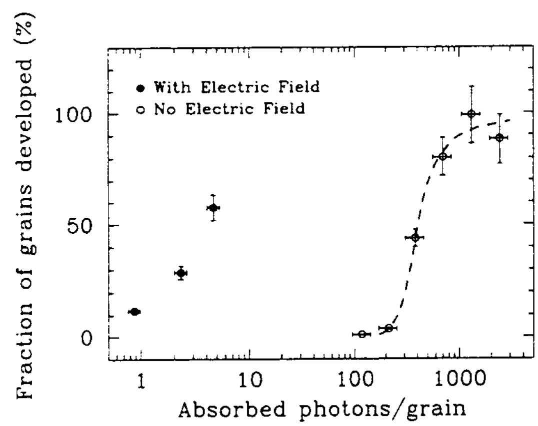

Figure 3: Quantum sensitivity curve of with and without electric field

Therefore the achieved sensitivity enhancement by the electric field is almost order 2.

4. Discussion

4.1. Achieved sensitivity

In Fig.4, typical quantum sensitivity of sulfur-plus-gold sensitization [14], sulfur-plus-gold and hydrogen hypersensitization [14] were compared with our results as a function of the grain size.

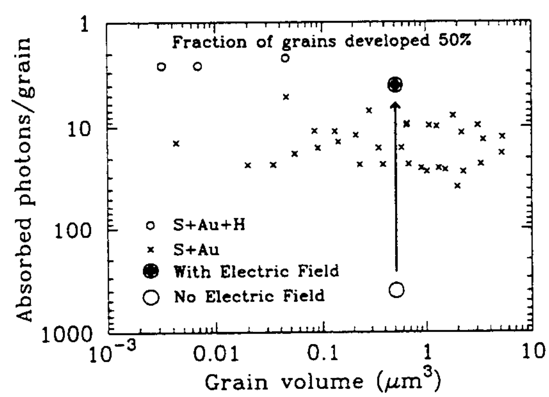

Figure 4: Quantum sensitivity VS. Grain size

We can say that our achieved sensitivity(4 $\sim$ 5) is higher than the typical level of Sulfur+Gold sensitization($\sim$ 10), but about 2 times smaller than sulfur-plus-gold and hydrogen hypersensitization. However in the case of lower exposure level around 1 absorbed photons/grain, our data was comparable to sulfur-plus-gold and hydrogen hypersensitization. (MAA-1 10min 20^oC) [15].

4.2. Consideration about the mechanism

In general, we can pick up two mechanisms for electric field sensitization. One is a cancellation of inefficiencies of the latent image formation and the other is an avalanche multiplication of created photo-electrons or holes. As described in the introduction, the avalanche mechanism was originally suggested by Rothstein, but it was abandoned in those days. However the Russian group insists that at higher electric field above 1MV/cm, there is a strong sensitivity enhancement which can be explained by avalanche multiplication[9-13]. They observed two kinds of enhancement, i.e. a small enhancement observed under 1MV/cm and a strong enhancement observed over 1MV/cm. They assigned the former as a cancellation of inefficiencies and the latter as an avalanche multiplication. The reasons why they assigned the latter as avalanche multiplication are as follows.
1. The dependence of the latter enhancement on the field strength obeys an exponential law as expected from an avalanche model [9].
2. The latter enhancement has a grain size dependence. The enhancement in larger grain appears at lower voltage than small grains, as expected from an avalanche model [11].

However, these features are not considered to be definite evidences to assign the enhancement as avalanche multiplication. It may be possible to device a cancellation model to explain these features. We considered what is the definite evidence of new mechanisms (avalanche). Our opinion is as follows. If avalanche multiplication occurred, the achieved quantum sensitivity could be higher than the ultimate chemical sensitization i.e. sulfur-plus-gold and hydrogen hypersensitization (quantum sensitivity ~ 3). We attempted to measure the achieved quantum sensitivity above 1MV/cm field strength. The quantum sensitivity was measured to be $4 \sim 5$ which is lower than the ultimate chemical sensitization. However in the case of low level exposure(i.e.~ 1.0), the obtained fraction of developed grains is around 12% which is comparable to sulfur-plus-gold and hydrogen hypersensitization, though we used unsensitized(not gold sensitized) AgBr grains. We intend to do extensive study by using chemically sensitized grains in order to check the additional effect of electrical sensitization on the chemical one.

5. Conclusion

The quantum sensitivity of our emulsion, which was measured as ~ 400 absorbed photons/grain in the absence of the voltage pulse, reached to $4 \sim 5$ absorbed photons/grain in the presence of high voltage pulse. The existence of large sensitivity enhancement above 1MV/cm which has been reported by Russian groups is confirmed by this result. But we could not confirm the mechanism as avalanche multiplication. However the obtained level of sensitivity at low level exposure (~ 1 absorbed photon /grain) is almost same as that of sulfur-plus-gold and hydrogen hypersensitization. Which may be difficult to explain by only cancellation of several inefficiencys by the applied electric field.

References

[1] J.Rothstein, Edgerton, Germeshausen and Grier, Photogr. Sci. Eng., 4,5(1960)

[2] M.A.Babalov,B.A.Dolgoshein,B.I.Luchkov, and F.R.Sosnin, Prib.Tekh.Eksperim.,5,178 (1962)

[3] V.I.Kalashnikova,D.M.Samoilovich, Yu.F.Pevchev, and K.G.Finogenov, Zh.nauch.prikl.Fotogr.Kinem., 9,464 (1964)

[4] J.F.Hamilton and L.E.Brady, Photogr.Sci.Eng.,10,1 (1966)

[5] M.Z.Peskova and P.V.Meiklyar, Zh.nauch.prikl. Fotogr.Kinem.,12,352 (1967)

[6] Yu.F.Pevchev,V.I.Kalashnikova, and L.P.Konovalova, Zh.nauch prikl.Fotogr.i Kinem., 15,250 (1970)

[7] A.E.Saunders, J.Photogr.Sci.,21,160 (1973)

[8] D.M.Samoylovitch and J.V.Ardashev, Photogr.Sci.Eng.,17,351 (1973)

[9] E.V.Kozhanov, V.M.Ulanov, Zh.Nauchn.Prikl. Fotogr.Kinematogr.,24, 292 (1979)

[10] V.M.Ulanov, Sov.Phys.Tech.Phys.,27,352 (1982)

[11] A.L.Kartuzhanskii,V.M.Ulanov, and A.F.Yurchenko, Phys.Stat.Sol.,(a)75,669 (1983)

[12] E.M.Gushchin,A.N.Lebedev,A.Yu.Lopyrew and S.V.Somov, Nucl.Instrum.Methods.,B16,515 (1986)

[13] E.M.Gushchin,A.N.Lebedev,V.N.Solovov,S.V.Somov, M.K.Timofeev and G.I.Tipografshchik, Nucl.Tracks Radiat.Mears., 21,391 (1993)

[14] R.K.Hailstone,N.B.Liebert,M.Levy,R.T.Mccleary, S.R.Girolmo,D.L.Jeanmaire and C.R.Boda, J.Imaging Sci., 32,113 (1988)

[15] T.A.Babcock and T.H.James, J.Photogr.Sci., 24,19 (1976)

[16] K.Teraoka, M,Nakamura, K,Hosino, K.Niwa, N.Saeki, S.Watanabe, T.Tani, J.Soc.Photogr.Sci.Technol.Jpn.,60,37 (1997) (in japanese)