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Development of a Radiowave Photoconductivity Apparatus with Improved Sensitivity and Stability for Studies of Photocarriers in Silver Halide Grains

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Although radiowave photoconductivity measurement was expected to be sensitive enough to detect the transient behavior not only of photoelectrons but also of positive holes in silver halide grains, it was not necessarily successful owing to the lack of its stability and reproducibility. In this investigation, we made a theoretical study and simulation of this measurement technique and developed a new apparatus for it, with significantly improved stability and reproducibility. The coefficient of variation of the photoconductivity of AgBr grains with positive holes as carriers by the new apparatus was 3.6%, while that by the old one was 33.8%. It was demonstrated that the new apparatus was sensitive and reproducible enough to measure small changes in photoconductivity of silver halide grains with positive holes as carriers. These changes were undetectable by the previous one.

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Introduction

In photographic materials, silver halide grains capture an image by absorbing incident photons and causing photoelectric effect to create photoelectrons and positive holes. In collaboration with interstitial silver ions, photoelectrons take part in the formation of latent image centers composed of Ag clusters on the grain surface to memorize the captured image. On the contrary, positive holes are usually undesirable for photographic sensitivity, since they recombine with photoelectrons and bleach already formed latent image centers. Attention is paid to the fact that positive holes enhance the formation of latent image centers in reduction sensitized emulsions according to a hypothesis proposed by Lowe[1] and experimentally proved by one of the present authors (TT).[2] Namely, a positive hole oxidizes a reduction sensitization center composed of a dimer of silver atoms,[2–5] and the oxidized center dissociates to produce an additional free electron, which contributes to the formation of a latent image center. It is therefore very important to study the transient behavior of positive holes as well as that of photoelectrons in silver halide emulsion grains from the viewpoint of photographic science and engineering.[2]

Various types of microwave photoconductivity apparatuses with time resolution of a few nanoseconds were developed and have been effectively used to study transient behavior of photoelectrons in silver halide grains in the time period ranging from a few nanoseconds to submicroseconds.[6–9] In this time range, photoelectrons are responsible for the photoconductivity, since photoelectrons have much larger mobility than positive holes.

A radiowave photoconductivity apparatus was developed by Kellogg and co-workers[10] as a tool with higher sensitivity for studying the behavior of photocarriers over a longer time range than is possible with a microwave photoconductivity apparatus. Actually, the time resolution of a radiowave photoconductivity apparatus is several microseconds. Thus, a radiowave photoconductivity measurement was expected to make it possible to observe both photoelectrons and positive holes in silver halide grains, depending on the kind of samples and experimental conditions. In the case of small AgBr grains[8,11] and grains with electron trapping dyes adsorbed on the surface,[11,12] photoelectrons disappeared within a microsecond and did not contribute to the radiowave photoconductivity in the time range of several microseconds. Under those conditions, a radiowave photoconductivity apparatus detected positive holes, whose mobilities were too small to be detected by a microwave photoconductivity apparatus. On the other hand, photoelectrons mainly contributed to the radiowave photoconductivity of AgCl grains,[13] since they still appeared in the conduction band of the grains in the time range of several microseconds.

After Kellogg and others,[10] Hirano[12] developed another type of a radiowave photoconductivity apparatus equipped with phase-selective detection in order to achieve high sensitivity. However, his apparatus had difficulty in maintaining high stability and reproducibility. In the present study, we theoretically analyzed and simulated a radiowave photoconductivity apparatus, and developed a new apparatus with significantly more stable and reproducible performance on the basis of the theoretical analysis.

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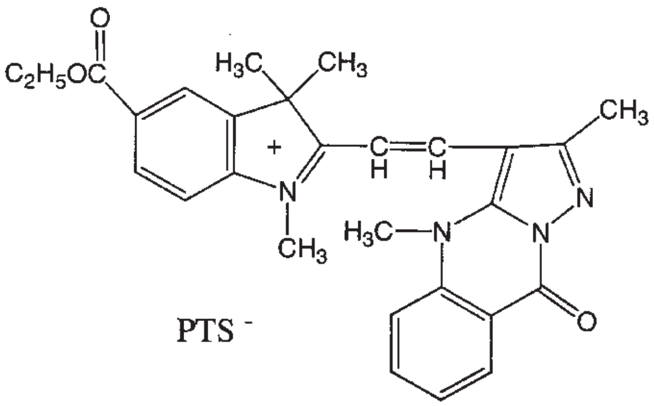


Figure 1. The molecular structure of Dye 1, which was used in this study to inject positive holes into silver halide grains on excitation.

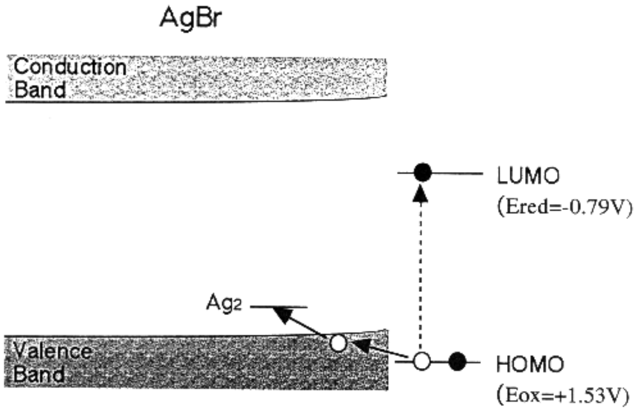


Figure 2. Electronic energy level diagram of Dye 1 and AgBr in terms of reduction potential (E_{re}) and oxidation potential (E_{ox}), where closed and open circles denote an electron and a positive hole, respectively.

Experimental

The photographic emulsion used primarily in this study contained octahedral AgBr grains in an aqueous gelatin solution, and was prepared by a controlled double-jet method,[14] according to which aqueous solutions of 1N $AgNO_3$ and 1N KBr were poured into an aqueous gelatin solution at pH 2.0 at 75°C for 60 min. The pAg of the reaction solution was maintained at 8.3 during the precipitation of AgBr grains. The pAg and pH of the emulsion thus prepared were adjusted to be 8.9 and 6.7, respectively, at 35°C before coating. The diameter of the circle whose area was equal to the average projection area of the grains was determined as 0.18 µm by an electron microscope.

An emulsion containing tabular AgBrI grains with average equivalent circular diameter of 1.16 µm and average thickness of 0.17 µm was also used in some experiments. Their average iodide content was 6 mole %. This emulsion was also made in principle by a controlled double jet addition of solutions of $AgNO_3$ and KBr(KI). Reduction sensitization was carried out by digesting the above-stated AgBrI emulsions in the presence of a reduction sensitizer for 60 min at 60°C. The reduction sensitizer used was dimethyl amineborane (DMAB). Thin layers of the emulsions described above were prepared by coating and drying them on triacetate cellulose (TAC) film bases with 1.74 g $AgBr/m^2$ and 1.27 g $gelatin/m^2$, and subjected to various measurements.

In order to make sure that only positive holes are responsible for the radiowave photoconductivity signal of silver halide grains, an electron trapping and hole injecting dye was adsorbed on the grains. The molecular structure of such a dye, i.e., Dye 1, is shown in Fig. 1. The electronic energy levels of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of Dye 1 were represented in terms of its oxidation potential E_{ox} and reduction potential E_{red} according to the following relationships,

$$\varepsilon_{HOMO} = -E_{ox} + C \tag{1}$$

$$\varepsilon_{LUMO} = -E_{red} + C' \tag{2}$$

where ε_{HOMO} and ε_{LUMO} are the electronic energy levels of HOMO and LUMO, respectively, and C and C' are electrochemical constants.[15] The reduction and oxidation potentials of Dye 1 were obtained by means of phase-selective second-harmonic voltammetry.[16,17] The observed values of E_{ox} and E_{red} were +1.53 V and –0.79 versus standard calomel electrode (SCE), respectively. The energy level diagram of Dye 1 and AgBr are schematically shown in Fig. 2 on the basis of the observations that the top of the valence band and the bottom of the conduction band of AgBr grains corresponded to E_{ox} of +1.40 V versus SCE[18] and E_{red} of –1.24 V versus SCE,[19] respectively. It is therefore understood from the energy level diagram that when Dye 1 is excited a positive hole created at its HOMO level is efficiently transferred to the valence band of AgBr grains, since the HOMO level of the dye is lower than the top of the valence band of the grains. It is also obvious that an electron excited to the LUMO level of Dye 1 is not injected into the conduction band of AgBr grains, since the LUMO level of the dye is much lower than the bottom of the conduction band of the grains.

Figure 3 shows a block diagram of a radiowave photoconductivity apparatus which was developed by Hirano.[13] Its operating frequency was 100 ~ 120 MHz. A sample film was sandwiched between two electrodes comprised of an ITO-coated glass and a copper plate, and a pair of the two electrodes functioned as a condenser. The resonant circuit was tuned to make its complex impedance equal to 50Ω in the dark in order to balance the bridge. When a sample was excited by a Xe flash lamp, the photoconductivity generated in the sample brought about an unbalanced signal in the bridge, which was amplified and detected by a mixer. A mixer was adjusted to phase-selectively detect the change in the radiowave photoconductivity signal due to the photoconductivity of a sample. The transient signal thus detected was monitored and recorded on a digital storage scope. The half width of a Xe flash lamp used in this study was 2 µs, and responsible for the time resolution of our apparatus. A sharp-cut filter SC54 made by Fuji Photo Film Co. Ltd or O54 made by Toshiba Electric Co., Ltd., which transmits light with wavelength longer than 540nm, was used to selectively excite Dye 1 adsorbed on the surface of silver halide grains. Color filters UVD33S and V40 made by Toshiba Electric Co. were used to excite AgBr grains.

The time-resolved photoconductivity of octahedral AgBr grains with photoelectrons as carriers was also

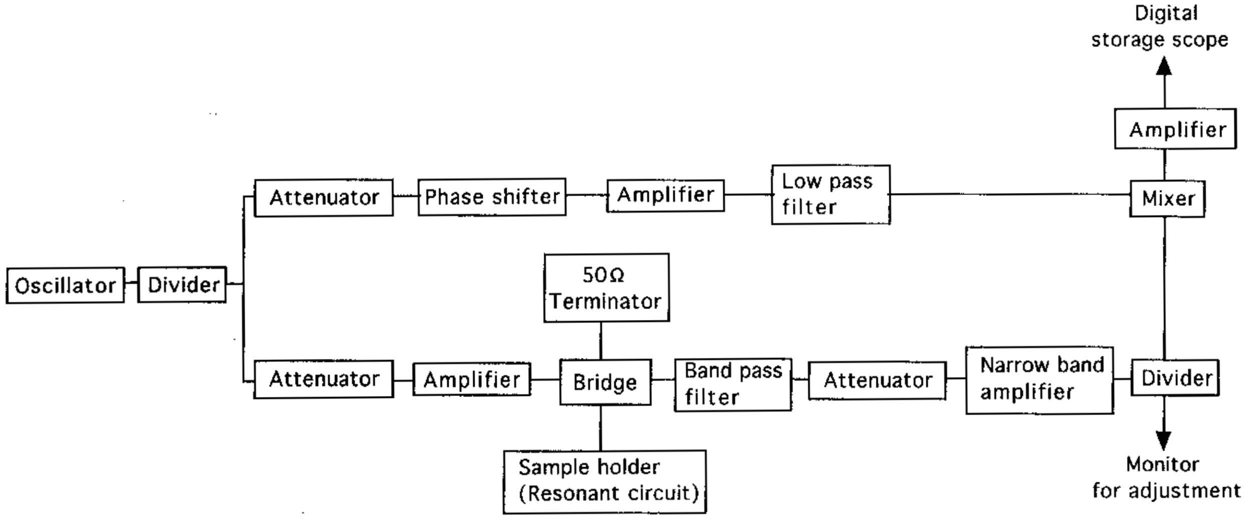
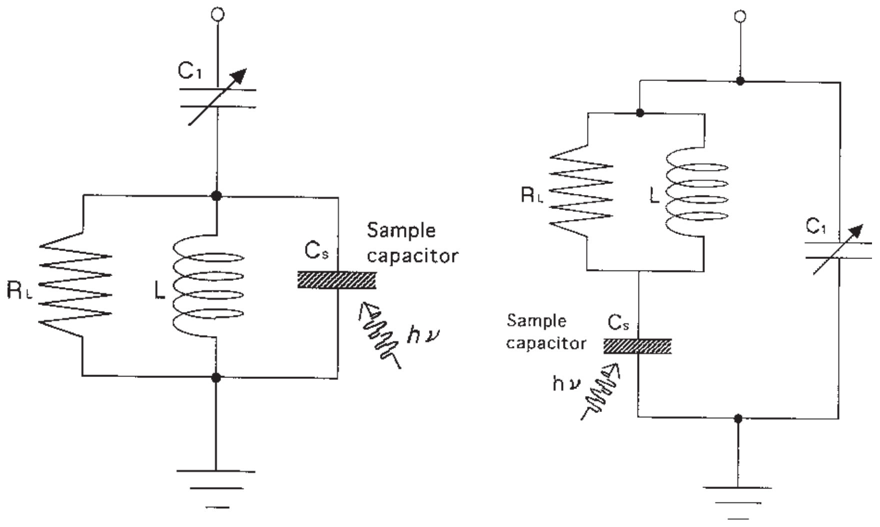


Figure 3. Block diagram of a radiowave photoconductivity apparatus.



(a) (b)

Figure 4. (a) A sample holder circuit which was used in the apparatus developed by Kellogg and co-workers.[10] and comprised of a coil with resistance R_L and impedance of *L,* a variable condenser with capacitance C_1 and a sample condenser C_s. (b) A sample holder circuit which was used in the apparatus developed by Hirano.[12]

measured by use of 9GHz and 35 GHz microwave photoconductivity apparatuses, details of which were reported elsewhere.[20,21]

Theory

Kellogg and co-workers[10] and Hirano[12] developed radiowave photoconductivity apparatuses with block diagrams similar to that shown in Fig. 3 and resonant circuits shown in Figs. 4(a) and (b). In the case of the measurement by Hirano, the apparatus was tuned to produce zero output voltage in the bridge with two arms by adjusting the complex impedance of the resonant circuit to 50Ω, and was used to measure the time-resolved photoconductivity of a sample as the imaginary part of its dielectric susceptibility by means of phase selective detection. This adjustment was made by changing the operating radiowave frequency (RF) of the apparatus and the capacitance of a variable condenser.

This adjustment does not, however, allow photoconductivity measurement with high stability and reproducibility. In this measurement, it is necessary to optimize the reference phase by monitoring photoconductivity under irradiation. Thus, the apparatus is adjusted to get the optimum phase for a typical sample under irradiation, and then the adjusted apparatus is used to measure the photoconductivity of other samples. It is noted however that for each measurement, not only the capacitance of a variable condenser, but also RF have to be adjusted to tune the apparatus with the circuit as shown in Fig. 4(b). The change in the RF would cause the deviation of the phase and signal intensity from their optimum condition and the deterioration of the accuracy of the measured values. Similar adjustment might be carried out for an apparatus developed by Kellogg and co-workers.

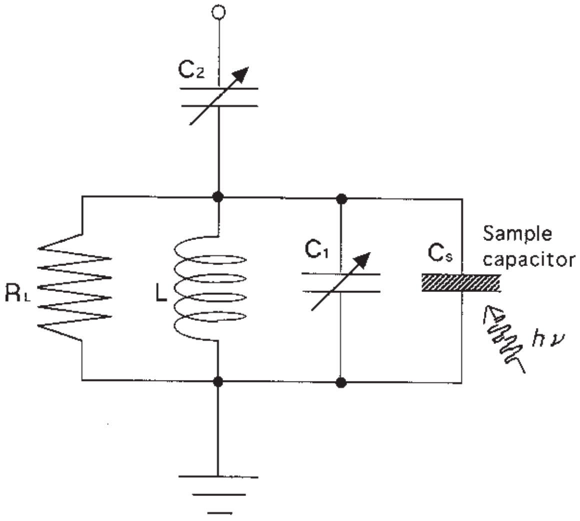


Figure 5. A sample holder circuit in our apparatus, which is basically the same as the one shown in Fig. 4(a) except that two variable condensers are used.

It was therefore expected that the systems developed by Kellogg and co-workers.[10] and by Hirano[12] would be improved by introducing a circuit, whereby the apparatus could be tuned without changing RF. This article is undertaken to propose a system with a circuit which includes two variable condensers in order to make it possible to produce the resonant condition which adjusts the complex impedance of the circuit to be 50Ω by changing the capacitances of two variable condensers C_1 and C_2 without changing RF. The proposed resonant circuit is shown in Fig. 5.

A certain treatment in the field of NMR[22] was useful as a reference to introduce the new circuit to a radiowave photoconductivity apparatus. We define the Q-value of a coil by the following equation.

$$Q \equiv \frac{R_L}{\omega L} \tag{3}$$

where ω is the angular radiofrequency, R_L and L are the resistance and inductance components of an equivalent circuit of a coil, respectively, under the condition that R_L and L are parallel in the circuit.[23] The complex impedance Z of the resonant circuit in Fig. 5 should be adjusted to 50Ω before each measurement. Therefore the resonant condition for it is represented as follows:

$$\operatorname{Re} Z = 50\Omega \tag{4}$$

$$\operatorname{Im} Z = 0 \tag{5}$$

In the context, Eqs. 4 and 5 are called the matching and tuning conditions, respectively.

The complex impedance Z of the resonant circuit in Fig. 5 is represented as follows:

$$Z = \frac{1}{j\omega C_2} + \frac{1}{j\omega(C_1 + C_S) + \frac{1}{j\omega L} + \frac{1}{R_L}}. \tag{6}$$

By transforming Eq. 6, the following equation is derived,

$$Z = \frac{\frac{\omega C_2}{R_L} - j\left[\left\{\omega(C_1 + C_S + C_2) - \frac{1}{\omega L}\right\}\left(\omega(C_1 + C_S) - \frac{1}{\omega L}\right) + \left(\frac{1}{R_L}\right)^2\right]}{\omega C_2\left\{\left(\omega(C_1 + C_S) - \frac{1}{\omega L}\right) + \left(\frac{1}{R_L}\right)^2\right\}} \tag{7}$$

where $j = \sqrt{1}$, C_1 and C_2 are the capacitances of variable condensers C_1 and C_2 in Fig. 5, respectively, and C_S represents the capacitance of a sample condenser. From Eq. 7, the tuning condition derived from Eq. 5 is represented as follows:

$$\omega(C_1 + C_S + C_2) - \frac{1}{\omega L} = 0 \tag{8}$$

$$\omega(C_1 + C_S) - \frac{1}{\omega L} = 0 \tag{9}$$

where it is assumed that $(1/R_L)^2 \approx 0$ for the derivation of Eqs. 8 and 9.

By use of Eq. 8, Z is represented as follows:

$$Z \approx \frac{1}{(\omega C_2)^2 R_L} \tag{10}$$

This equation is derived for the condition that

$$\frac{1}{\omega C_2} << R_L.$$

By using this equation, the matching condition due to Eq. 4 is represented as follows:

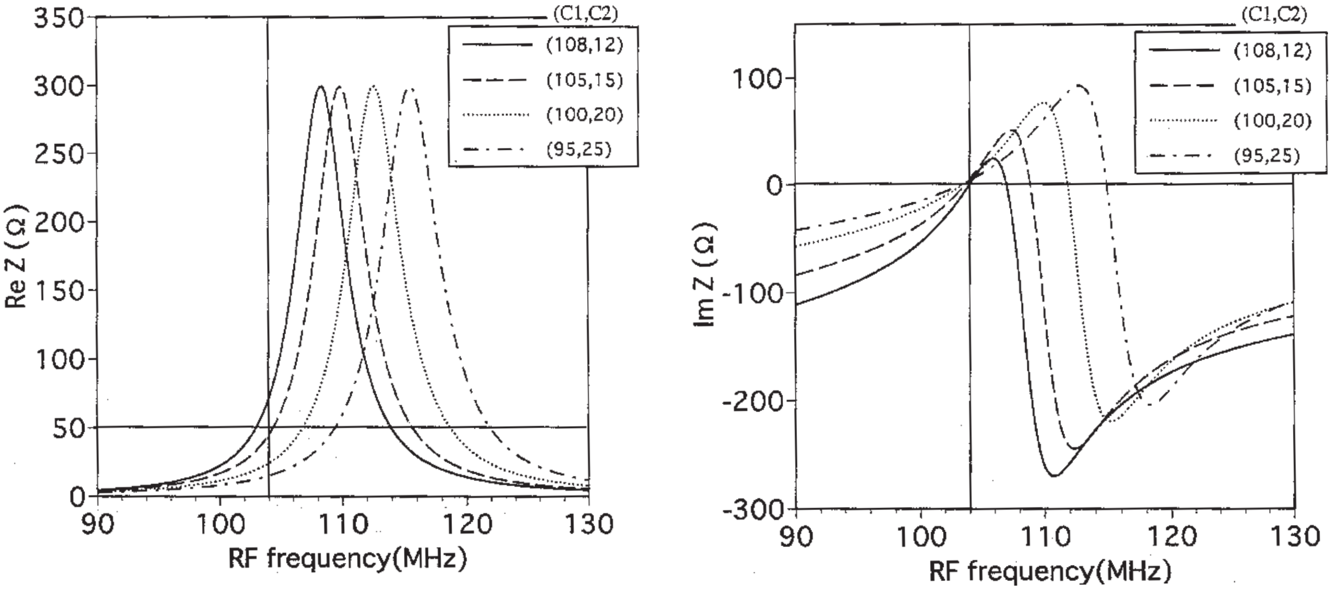
$$C_2 = \frac{1}{\sqrt{50\omega^2 R_L}} = \frac{1}{\sqrt{50\omega^3 QL}}. \tag{11}$$

By use of Eq. 9, Z is represented as follows:

$$Z \approx R_L. \tag{12}$$

In this case, it is not possible to satisfy the matching and tuning conditions simultaneously. Therefore, it is concluded that both matching and tuning conditions can be achieved only when C_1 and C_2 are adjusted to satisfy Eqs. 8 and 11.

Figures 6(a) and 6(b) show the results of the simulation of Eq. 6. These figures show that the resonant condition, as given by Eqs. 4 and 5, is satisfied at the frequency of about 104 MHz with C_1 and C_2 being approximately 105 pF and 15 pF, respectively. Since the sum of C_1 and C_2 is constant at the value of 120 pF, the frequency, which satisfies the tuning condition, is always equal to about 104 MHz. Namely, both the matching and tuning conditions can be achieved with keeping the reference phase at the optimum condition since the adjustment of C_1 and C_2, with their sum kept to be 120 pF, do not disturb the optimum phase and is effective to



(a) (b)

Figure 6. Results of a simulation showing frequency dependence of the real part (a) and imaginary part (b) of the complex impedance of the resonant circuit shown in Fig. 5 on the basis of Eq. 6. In this simulation, L and R_L were fixed at the values of 20 nH and 300 Ω, respectively, the combination of (C_1 + C_2) and C_2 were taken as (108 pF, 12 pF), (105,15), (100,20), and (95,25), respectively, with (C_1 + C_2) fixed at 120 pF.

achieve both the matching and tuning conditions at the same time.

The irradiation of a sample with a flash lamp results in the change in the dielectric susceptibility of the sample $\Delta\chi$, which is given as follows:

$$\Delta\chi = \Delta\chi' - j\Delta\chi'' \tag{13}$$

where $\Delta\chi'$ and $\Delta\chi''$ represent its real and imaginary parts. The change in the capacitance of the irradiated sample condenser is thus expressed as follows:

$$\Delta C_S = \frac{\Delta\chi}{\varepsilon_r} C_S = \frac{(\Delta\chi' - j\Delta\chi'')}{\varepsilon_r} C_S \tag{14}$$

where C_S is the capacitance of the sample condenser under the dark and ε_r is the relative dielectric constant of the sample. The change in the complex impedance of the total circuit in Fig. 5, ΔZ, is given as follows:

$$\Delta Z = \frac{\partial Z}{\partial C_S} \Delta C_S \tag{15}$$

$$\approx \frac{C_S}{C_1 + C_S + C_2} \cdot \frac{50Q}{\varepsilon_r} \cdot (\Delta\chi'' + j\Delta\chi') \tag{16}$$

It is shown in this equation that the photoconductive component of AgBr grains in a sample $\Delta\chi''$ appears as the real part of ΔZ, while its dielectric component $\Delta\chi'$ appears as the imaginary part of ΔZ. The photoconductive component can be selectively measured by adjusting the reference phase by a phase shifter as shown in Fig. 2.

For a highly sensitive apparatus, the Q-value of the circuit should be as high as possible. Therefore we tried to make the resistance of the circuit as low as possible. Consequently, we coated electric components with gold, kept their surfaces clean, and used copper plates to connect each component. A coil was made from Sn-coated wire with diameter of 2 mm. We used two air-gapped variable condensers with the capacitance of 30 pF. Rectangular ITO-coated glass with size of 21.5 mm × 9.5 mm was used as a transparent electrode, through which a sample was exposed to a Xe flash lamp.

By taking into account the fact that the sensitivity of an apparatus is proportional to the fill factor of a sample, it is considered that the main reason for high sensitivity of a radiowave photoconductivity apparatus was its large fill factor for a sample. In a microwave photoconductivity apparatus, a sample with volume of v is put in a cavity with volume of V, giving its fill factor of f, which is represented as follows:

$$f = c \cdot \left(\frac{v}{V}\right) \tag{17}$$

where c means the volume ratio of an emulsion layer to a total film sample. In the case of the 35 GHz microwave photoconductivity apparatus in our laboratories, v/V is evaluated to be as small as 0.016. In the case of a radiowave photoconductivity apparatus, v/V is almost equal to unity, since a sample was sandwiched between two electrodes. It is therefore concluded that the fill factor of a radiowave photoconductivity apparatus is much larger than that of a microwave photoconductivity one, and that the big difference in fill factor should allow the sensitivity of a radiowave photoconductivity apparatus to be much higher than that of a microwave photoconductivity apparatus.

Results and Discussion

Figure 7 compares the radiowave photoconductivity signals of octahedral AgBr grains as measured by use of

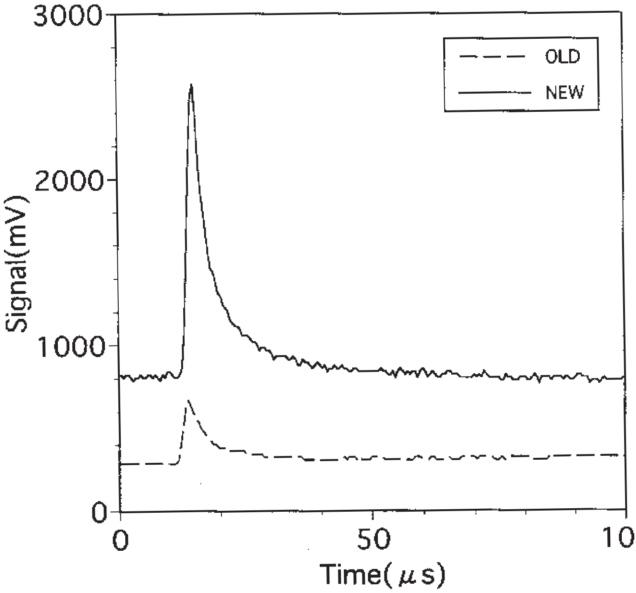
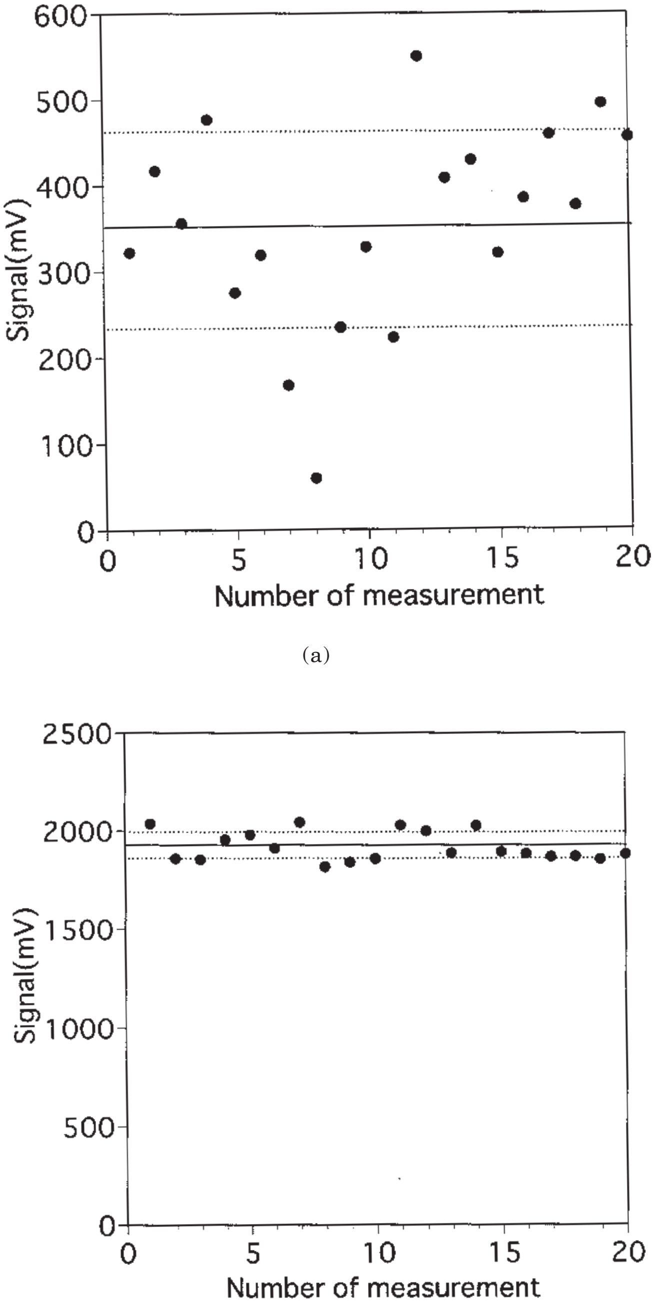


Figure 7. Radiowave photoconductivity signals of octahedral AgBr grains as measured by use of an old apparatus (solid curve) and a new one (broken curve). Dye 1 was adsorbed on the grain surface and excited to inject positive holes into the grains.



(a)

(b)

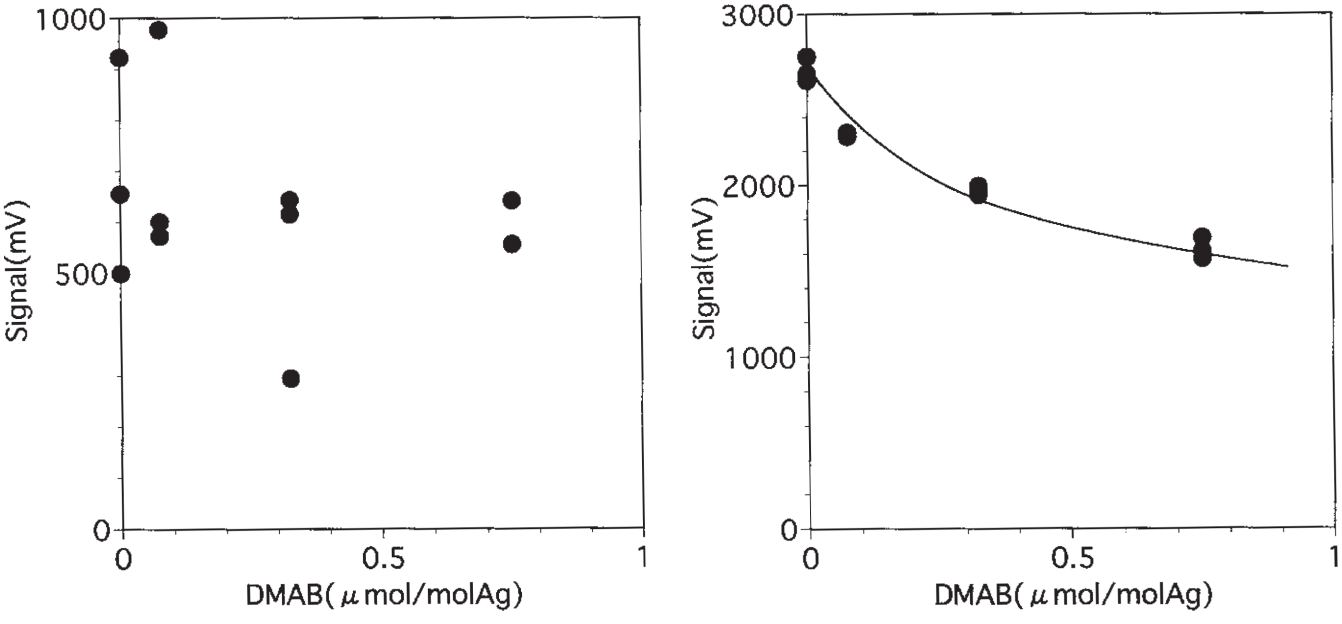
Figure 8. Reproducibility of intensities of radiowave photoconductivity signals of octahedral AgBr grains as measured by (a) an old apparatus and (b) a new one. Dye 1 was adsorbed on the grain surface, and excited by a Xe flash lamp through a filter SC-54 to inject positive holes into the grains.

the old apparatus developed by Hirano[12] and the new apparatus developed in this study. Dye 1 was adsorbed to the grains and excited to inject positive holes into the grains. The peak height of the signal measured by the new apparatus was higher by factor of about 5 than that by the old one, although the noise level of the former was a little bit higher than that of the latter. On the basis of the energy level diagram in Fig. 2 the observed signals were ascribed, not to photoelectrons, but to positive holes in the grains; fig. 7 thus clearly indicates significant improvement in the sensitivity of a radiowave photoconductivity apparatus for detecting positive holes in the grains.

Figures 8(a) and 8(b) compare the reproducibility of the peak height of the radiowave photoconductivity signals of octahedral AgBr grains as measured by use of the old apparatus and the new one. Dye 1 was adsorbed to the grains and excited by a Xe flash lamp through a filter of SC54 to inject positive holes into the grains. The average peak heights were 352.4 mV and 1917.4 mV, and their coefficients of variation were 33.8% and 3.6%, comparing the old apparatus and the new one, respectively. Namely, it was found that the reproducibility of signal intensities by the new apparatus was improved by approximately one order of magnitude, while the sensitivity of the new apparatus was five times larger than that of the old apparatus.

Figures 9(a) and 9(b) show an example of the result which was brought about by the improvement in the reproducibility of signal intensities of a radiowave photoconductivity apparatus in this study. The samples were tabular AgBrI grains, which were reduction sensitized in the presence of DMAB, with the amount indicated in the abscissa of these figures, and Dye 1 adsorbed on the grain surface. They were subjected to measurement of radiowave photoconductivity signal intensity by use of the old apparatus and the new one. Dye 1 was excited to inject positive holes into the grains, and injected positive holes were captured by reduction sensitization centers. As seen in Fig. 9(a), the reproducibility of signal intensity by the old apparatus was too poor to detect the behavior of positive holes in the presence of reduction sensitization centers, while the new apparatus was able to detect it.

As a result of the present investigation, we have the complete line-up of techniques for the study of



(a) (b)

Figure 9. Dependence of the intensity of radiowave photoconductivity signal of reduction sensitized tabular AgBrI grains upon amount of DMAB as measured by (a) an old apparatus and (b) a new one. Dye 1 was adsorbed on the grain surface and excited to inject positive holes into the grains.

photocarriers in silver halide grains in actual photographic emulsions. In order to confirm the usefulness of the line-up, we tried to apply the following four measurements to the study of photocarriers in reduction sensitized octahedral AgBr grains.

a. 9 GHz microwave photoconductivity apparatus at –150°C, at which ionic processes in silver halide grains are inactive.
b. 35 GHz microwave photoconductivity apparatus at room temperature
c. radiowave photoconductivity apparatus at room temperature
d. radiowave photoconductivity apparatus at room temperature for an emulsion layer with a strong electron-trapping dye on the grain surface.

The AgBr grains were excited for all the above measurements.

The measurements (a) and (b) are expected to detect the behavior of photoelectrons in the range of several nanoseconds to several sub microseconds in the absence and presence of ionic processes, respectively, since photoelectrons have much larger mobility than positive holes and have their lifetime in the above-stated time range. The measurements (c) and (d) are expected to detect the behavior of positive holes in the regime of several tens of microseconds in the absence and presence of an electron-trapping dye, respectively. In support of this expectation was we observe that photoelectrons could not be detected by microwave photoconductivity apparatuses in the above-stated time range during the lifetime of positive holes in the grains,[8,20,21] and that the sensitivity of the radiowave photoconductivity apparatus is sufficient to detect behavior of positive holes in the same time range, even though the mobility of positive holes is much lower than that that of photoelectrons.[9] It has been already proposed[2–5,8] that reduction sensitization provides silver halide grains with hole trapping centers, i.e., *R* centers, and electron trapping centers, i.e., *P* centers, depending upon the experimental condition. Therefore this combination of samples and experiments should be very effective for confirming both the above proposal for the mechanism of reduction sensitization and the usefulness of the line-up of the techniques for the study of photocarriers in silver halide grains.

Figure 10 shows the results of the four photoconductivity measurements compared with sensitometric results. As described in a series of articles,[4,5,8] photographic sensitivity increases with increasing the amount of DMAB through two steps, the first and second steps being ascribed to the sensitizing effects caused by *R* centers and *P* centers, respectively. It is seen in this figure that, with increasing amounts of DMAB, the radiowave photoconductivity at room temperature began to decrease at the same amount of DMAB as corresponded to the beginning of the sensitizing effect by *R* centers. The reproducibility of the old apparatus was too poor to detect the change in the radiowave photoconductivity of these grains due to the formation of *R* centers on the grain surface. This result clearly confirms the formation of *R* centers which act as traps for positive holes, contributing to the sensitivity increase in the first step in Fig. 10, and also supports the usefulness of a new radiowave photoconductivity apparatus developed in this study for detecting the behavior of positive holes in AgBr grains. It is also seen in Fig. 10 that, with increasing amount of DMAB, the microwave photoconductivity in the presence and absence of ionic process began to decrease for the same amount of DMAB as corresponds to the beginning of the sensitizing effect by *P* centers. This result clearly confirms the formation of *P* centers which act as traps for photoelectrons, further contributing to the sensitivity increase in the second step in Fig. 10, and also supports the validity of the microwave photoconductivity measurement even in the presence of an

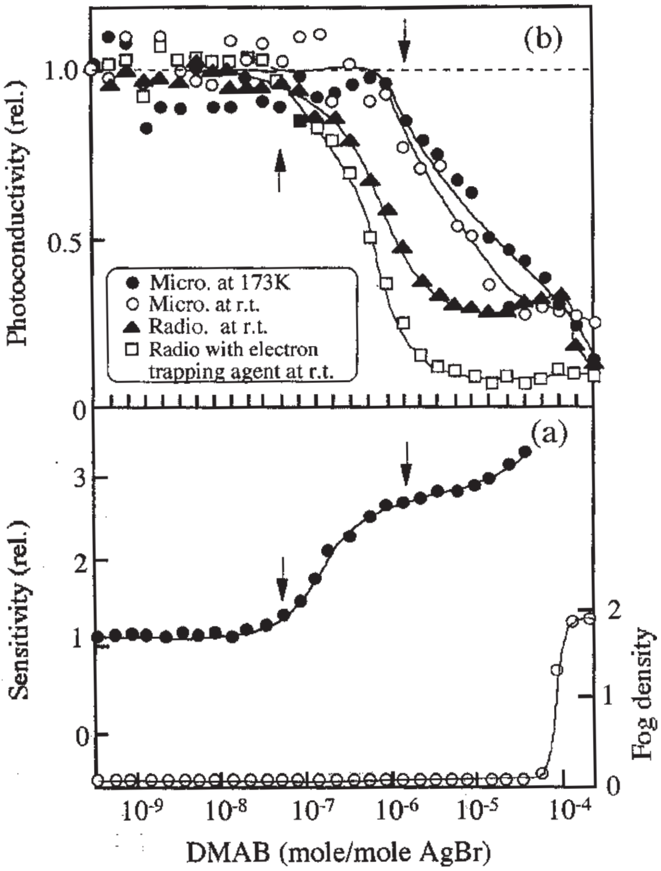


Figure 10. (a) Sensitivity and fog density and (b) photoconductivity of reduction-sensitized octahedral AgBr grains as a function of amount of DMAB. The photoconductivity measurements were made by using four methods [9 GHz microwave photoconductivity apparatus at –150°C, 35 GHz microwave photoconductivity apparatus at room temperature, radiowave photoconductivity apparatus at room temperature with and without addition of an electron-trapping dye (Pinakryptol Yellow; 1.1 mmole/mole AgBr) to emulsion layers].

ionic process, for detecting the behavior of photoelectrons in the grains.

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