▸원 저◂

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Transport parameters in *a*-Se : As films for digital X-ray conversion material using the moving-photocarrier-grating technique

— moving-photocarrier-grating 기술을 이용한 디지털 X-선 변환물질 *a*-Se : As의 수송변수 —

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— Abstract —

The effects of As addition in amorphous selenium (*a*-Se) films for digital X-ray conversion material have been studied using the moving photocarrier grating (MPG) technique. This method utilizes the moving interference pattern generated by the superposition of the two frequency shifted laser beams for the illumination of the sample. This moving intensity grating induces a short circuit current, j_{sc} in *a*-Se : As film. The transport parameters of the sample are extracted from the grating-velocity dependent short circuit current induced in the sample along the modulation direction. The electron and hole mobility, and recombination lifetime of *a*-Se films with arsenic (As) additions have been obtained. We have found an increase in hole drift mobility and recombination lifetime, especially when 0.3% As is added into *a*-Se film, whereas electron mobility decreases with As addition due to the defect density. The transport properties for As doped *a*-Se films obtained by using MPG technique have been compared with X-ray sensitivity for *a*-Se : As device. The fabricated *a*-Se(0.3% As) device film exhibited the highest X-ray sensitivity out of 5 samples.

Key Words : *a*-Se : As, moving photocarrier grating, X-ray conversion material, carrier mobility, recombination lifetime, X-ray sensitivity

Ⅰ. Introduction

Amorphous selenium (*a*-Se) has a special attraction in a wide variety of electronic device application not only because of its commercial importance as a xerographic photoreceptor material but also because of its very interesting physical property[1-2]. There are a number of potential applications for selenium-based glassy alloys in imaging devices[3], optical storage[4], and fiber optics[5]. While traditionally *a*-Se was employed in xerography[6], more recently this material has been used as the X-ray photoconductor in flat-panel X-ray image detectors[7]. The amorphous selenium film that is currently being studied for use an X-ray photoconductor is

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not pure a-Se but rather a-Se alloyed with 0.2–0.5% As (normally 0.3% As) and doped with chlorine (Cl) in the 10–20 ppm range, also known as stabilized a-Se[8-9]. A small amount of As in a-Se film is added to enhance the thermal stability of the amorphous state. But a high As addition induces the undesirable hole traps in a-Se : As film. It is well known that doping a-Se film with small amounts of Cl in the ppm range is necessary to reduce the deep traps associated the arsenic atoms.

The mobility and the recombination lifetime of electrons and holes in semiconductors are important parameters that determine the performance of many devices, such as solar cells or thin film transistors[10-11]. The moving photo-carrier grating (MPG) technique allows us to determine the carrier mobility and recombination lifetime of electrons and holes in semiconductors[11-12]. This technique utilizes a moving intensity grating that is generated by the superposition of frequency shifted laser beams for the illumination of the sample. While a several MPG measurements have been carried out on the transport properties of amorphous silicon (a-Si) sample in the past, the transport phenomena for a-Se films using MPG method have not been accomplished yet.

We focus on the transport properties of a-$se_{1-x}As_x$($x = 0.001$, 0.003, 0.01, 0.05, and 0.1) films related to the underlying electron and hole drift mobility and recombination lifetime. The electron drift mobility and the hole drift mobility were obtained as a function of As addition in a-Se films to find out the a-Se based X-ray detector with the high performance. The dependence of recombination lifetime on the As addition in a-Se film is also reported in this study. The X-ray sensitivity for a-Se : As film is carried out to compare with the transport property of a-Se : As films obtained by MPG measurement.

Ⅱ. Experiment

The starting materials for sampleswere prepared by mixing 99.999% a-Se (Nippon Rare Metal Co, Japan) and 99.999% As (Nippon Rare Metal Co, Japan) in a weight ratio of 0.1, 0.3, 1, 5, and 10%. As-doped a-Se films were deposited on the Corning glass by the thermal evaporation technique under 10^{-6} Torr. Prior to film deposition, the Corning glass was washed by ultrasonic cleaner and rinsed with deionized water and, finally blown dry with N_2 gas. The transmission for 30 μm a-$Se_{1-x}As_x$(between $x = 0$ and 0.1) films was measured to determine the absorption of a-Se : As films using the visible spectrophotometer. The bandgap energy E_g of a-$Se_{1-x}As_x$ was estimated from these absorption spectra. The thickness of a-Se : As films for MPG measurement was about 100 μm. The parallel gold electrodes with 1 mm spatial separation were coated by thermal evaporation to measure the weak current flowing in a-Se : As film. For dark- and photo-current measurements, a-Se (30 μm thickness) based X-ray detectors for 3 As additions ($x = 0.001$, 0.003, and 0.01) were also fabricated to obtain the X-ray sensitivity as a function of applied electric field. Polyparaxylylene layer (10 μm) as a dielectric layer was coated on the a-$se_{1-x}As_x$ films of a-Se based X-ray detectors using Paraxylylene Deposition System (PDS 2060, SCS Co, USA). Au top electrode for I–V measurement was evaporated on the paraxylylene layer by thermal evaporation.

The experimental setup used for the MPG measurement is shown in Fig. 1. Two coherent laser

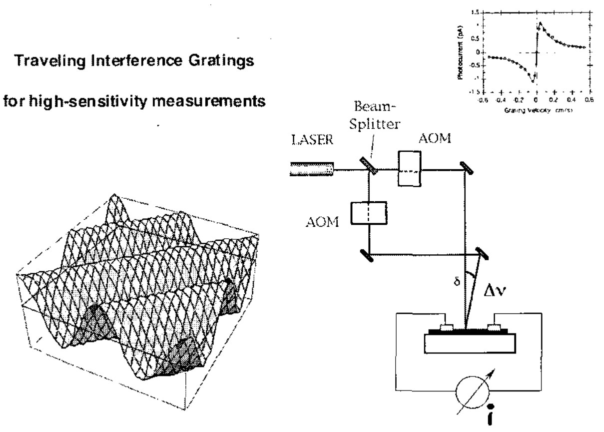

Fig. 1. Experimental set-up for the moving photo-carrier grating measurement

beams of wavelength λ that form an interference pattern on the sample surface. The beams impinge under an angle δ resulting in an intensity grating with spatial period $\Lambda = \lambda/[2\sin(\delta/2)$. A well-defined frequency difference Δf is introduced between the two beams so that the intensity grating moves with a velocity $\upsilon_{gr} = \Lambda\Delta f$. This movement is achieved through acousto-optic modulators in each laser beam. The approach to calculate the current density induced by the moving intensity grating is described in Ref[11].

The light intensity at the surface of the sample (the x coordinate) has a spatial and temporal dependence[11,13]:

$$I(x,\ t) = (I_1 + I_2) + 2\sqrt{I_1 I_2}\cos(kx - \omega_{gr}t),$$

where I_1 and I_2 are the intensity of the two beams, and k is the spatial frequency ($k = 2\pi/\Lambda$), ω_{gr} is the angular grating velocity ($\omega_{gr} = 2\pi\upsilon_{gr}/\Lambda$). MPG measurements were performed by using the line $\lambda = 532$ nm.

This technique utilizes a spatially and temporally modulated light intensity for the generation of photocarriers, which leads to a modulation of the carrier density. The different mobilities of electrons and holes introduce a phase shift between the charge distributions. The resulting internal electric field produces a grating-velocity dependent short circuit current j_{sc}. The very existence of j_{sc} points to different mobilities of electrons and holes and the analysis for the sign and the shape of $j_{sc}(\nu_{gr})$ allow the determination of the values of the carrier mobility and their recombination lifetime. The short circuit current in a-Se sample is typically in the range of 10^{-12} - 10^{-13}A, which can be measured using an electrometer. The MPG technique was applied to a-$Se_{1-x}As_x$(between $x = 0$ and 0.1) films. The laser angle δ for MPG measurement samples was 21.1°, which gives the grating period of $\Lambda = 1.45\,\mu$m.

The dark current flowing in fabricated a-Se : As based X-ray detector was measured at dark state (without X-ray exposure) while applying an electric field from 2 to 10 V/μm. The experimental setup for measuring dark current as shown in Fig. 2(a) was composed of a current amplifier (Keithley Model 428) for measuring small dark current, and a power supply (EG&G 558H) for applying high electric field. The measurement of photo-current as shown in Fig. 2(b) was similar to that of dark current, with the addition of X-ray exposure. X-ray exposure conditions were 70 kVp, 100 mA and 0.003s, respectively. Al layer with 2.5 μm thickness was also used as an X-ray absorption layer to control the X-ray dose exposing on the a-Se : As film. The exposure dose on the surface of the X-ray film was monitored with an ion chamber 2060 (Radical Cooperation, USA) during measurement. Experimental setup was protected by a custom made Al case. After applying high electric field of DC 1-5 kV to the fabricated X-ray detecting device, the output terminal of the

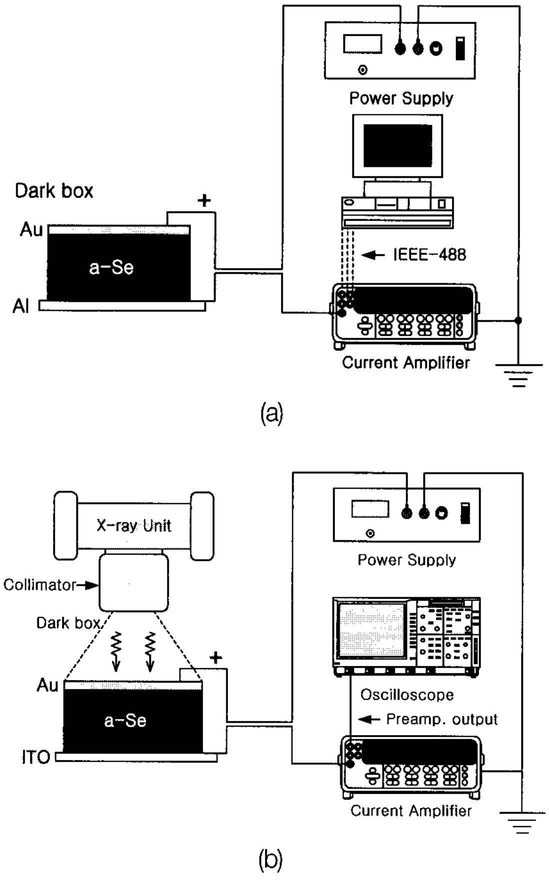

Fig. 2. Block diagram for (a) dark current measurement (b) photo-current measurement

current amplifier was connected with input of an oscilloscope to collect the electrical signal.

The X-ray sensitivity for a–$Se_{1-x}As_x$ can be obtained from the photocurrent divided by dark current. The X-ray sensitivities for a–$Se_{1-x}As_x(x = 0.001,\ 0.003,\ 0.01,\ 0.05$, and $0.1)$ films were measured as a function of applied electric field to investigate the X-ray response with respect to the As additions.

Ⅲ. Results and Discussions

Arsenic (As) atoms have only five electrons in their outermost shells ($4s^2 4p^3$), while selenium (Se) atom have six ($4s^2 4p^4$). Arsenic presence in Se leaves vacancies called holes in the electron structure of Se atom. Therefore, As addition in a–Se film increases the hole mobility because the presence of As in a–Se film provide accepter energy level, just above the highest filled band. The bandgap energy E_g of a–$Se_{1-x}As_x(0.001 \le x \le 0.1)$ was estimated from absorption spectra obtained by the visible spectrophotometer. The energy band of a–$Se_{1-x}As_x$ films decreased from 2.22 eV to 1.83 eV when 0.001–0.1% As were added to a–Se films.

The short circuit currents measured for a–$Se_{1-x}As_x$ $(0.001 \le x \le 0.1)$ films as a function of υ_{gr} are plotted in Fig. 3. The short circuit currents is zero for $\nu_{gr} = 0$ and decreases linearly for small values of ν_{gr}. After reaching a minimum, j_{sc} increases steadily up to the highest grating velocity used in this study. MPG curves for a–Se : As film exhibit the different behavior when compared with those for a–Si : H[5]. The inverted MPG curves of a–Se : As film compared with the MPG curves of a–Si : H are due to the positive photocarrier charges, holes. The dominant mobility carriers are holes for a–Se films, whereas those are electrons for a–Si : H films[13].

The carrier mobilities μ_n and μ_p are obtained by fitting the measured short circuit current to the theoretical expression derived by U. Haken et al. [11]. The electron and hole drift mobility for a–$Se_{1-x}As_x$ films are plotted as a function of As addition in Fig. 4. The hole drift mobility exhibits the apparent increase at the As addition of $x = 0.003$ between $x = 0.001$ and $x = 0.01$, whereas electron drift mobility decreases with As addition. The hole mobility decreases due to defect density of deep hole traps when $x = 0.003$ exceeds, after hole mobility increases in low As addition$(0.001 \le x \le 0.003)$.

The dependence of the recombination lifetime on the As addition in a–Se films is shown in Fig. 5. This behavior is understood, if one considers that holes dominantly recombine, after they were excited from localized into extended states. We have found the apparent increase of τ_R at the $x = 0.003$ between $x = 0.001$ and $x = 0.1$, and τ_R increases with respect to As addition when As addition exceeds $x = 0.01$. We assign this change to the contribution of two factors : a small additions of As in a–Se films up to $x =$

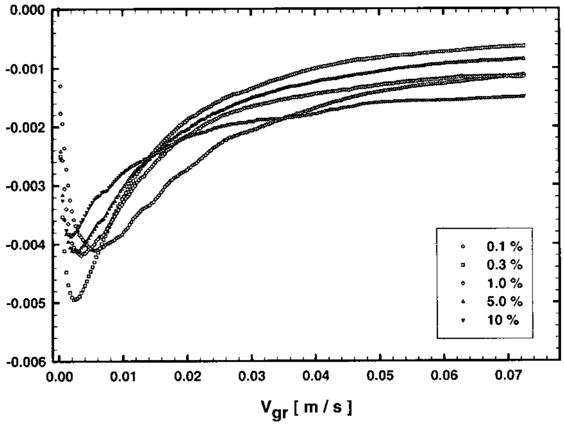

Fig. 3. Current density as a function of for

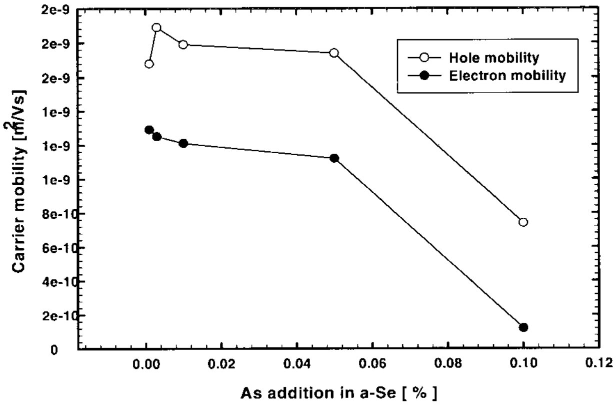

Fig. 4. The electron and hole drift mobility as a function of As addition

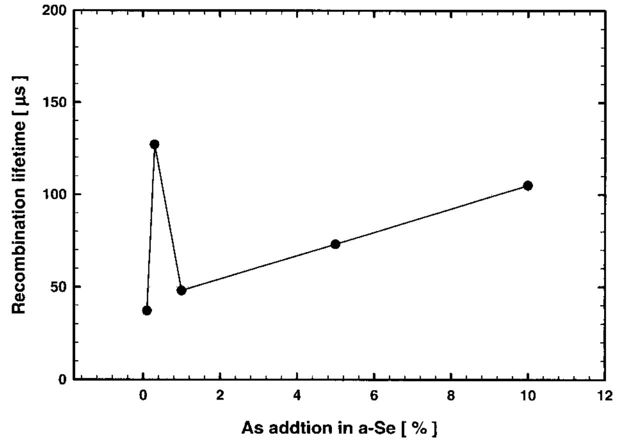

Fig. 5. The recombination lifetimes for a-Se : As films as a function of As addition

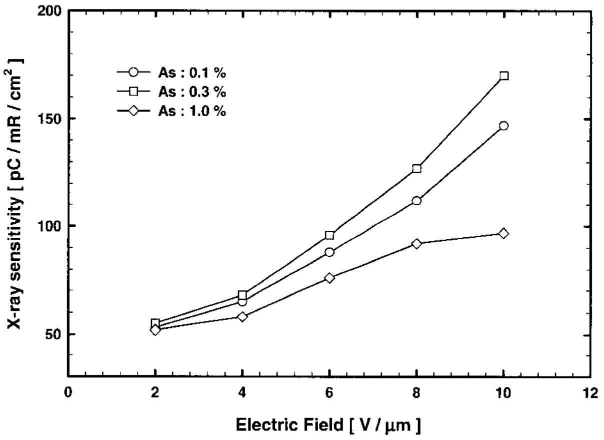

Fig. 6. X-ray sensitivity for a-Se : As films as a function of electric field

0.003 enhance the electric conductivity of *a*-$Se_{1-x}As_x$ films, while further As addition induces the undesirable hole traps in *a*-Se samples[14].

The X-ray sensitivity of the fabricated *a*-Se : As detectors for 3 As addition is shown as a function of electric field in Fig. 6. The X-ray sensitivity increases as the electric field increase from 2 to 10 V/μm. Experimental data indicates that the X-ray sensitivity for 0.3% As added *a*-Se detector exhibits higher value than those for others. The X-ray sensitivity for 0.3% As added *a*-Se detector was 170 pC/mR/cm^2. These results mean that the combined structure between *a*-Se and As effectively influences the recombination of the electron-hole pair created by X-ray exposure. The *a*-Se : 0.3% As film contributes a stable *a*-Se structure for collecting a created charges.

Ⅳ. Conclusion

The electron drift mobility and the hole drift mobility were obtained as a function of As addition in *a*-Se films to find out the *a*-Se based X-ray detector with the high performance. The hole drift mobility exhibits the apparent increase at the As addition of $x = 0.003$ between $x = 0.001$ and $x = 0.1$, whereas electron drift mobility decreases with As addition. A small additions of As in *a*-Se films up to $x = 0.003$ enhance the electric conductivity of *a*-$Se_{1-x}As_x$ films, while further As addition induces the undesirable hole traps in *a*-Se samples. Measurement indicates that the X-ray sensitivity for 0.3% As added *a*-Se detector exhibits higher value than those for others. The X-ray sensitivity for 0.3% As added *a*-Se detector was 170 pC/mR/cm^2. These results mean that the combined structure between *a*-Se and As effectively influences the recombination of the electron-hole pair created by X-ray exposure. The *a*-Se : 0.3% As film contributes a stable *a*-Se structure for collecting a created charges under X-ray exposure.

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• 국문초록

moving-photocarrier-grating 기술을 이용한 디지털 X-선 변환물질 *a*-Se : As의 수송변수

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moving photocarrier grating 기술을 이용하여 디지털 X-선 변환물질 *a*-Se : As 필름에서 As 첨가효과에 관하여 연구하였다. 이 방법은 시료를 조사하기 위하여 주파수를 변화시킨 2개 레이저 빔의 중첩으로 얻어진 움직이는 간섭패턴을 이용한다. 시료의 수송변수는 시료에서 변조 방향으로 유도되는 grating-속도에 의존하는 전류밀도로부터 얻어진다. As 첨가에 따른 *a*-Se 필름의 전자와 정공 이동도 그리고 재결합 수명을 구하였다. 전자의 이동도는 결합 상태 때문에 As 첨가에 따라 감소하는 반면, 특히 *a*-Se 필름에 0.3% As 첨가할 때 정공 이동도와 재결합 수명이 증가하였다. MPG 기술로 얻은 As가 첨가된 *a*-Se 필름의 수송성질을 *a*-Se : As로 제작한 X-선 디텍터의 X-선 감도와 비교하였다. 실험결과 0.3% As가 첨가된 *a*-Se으로 제작한 X-선 디텍터가 가장 우수한 X-선 감도를 나타내었다.

중심 단어 : *a*-Se : As(As가 첨가된 *a*-Se), moving photocarrier grating(MPG), X-선 변환물질, carrier mobility, 재결합 수명, X-선 감도