Change of Energy Distribution of He^+ Induced Electrons from MgO Thin Film with Ion Irradiation

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The change in the energy distribution of ion-induced secondary electrons (IISEs) emitted from a MgO surface upon 1 keV He^+ irradiation and the effects of electron irradiation on the IISE emission were investigated. The self-sustained electron emission induced by the field emission (FE) due to charging was observed during non-ion irradiation. The electron spectra obtained during both ion irradiation and non-ion irradiation were found to consist of two peaks. One stays at the same energy and the other shifts toward the lower-energy side. The former and latter peaks might be attributed to the Malter and Townsend-Avalanche effects. In addition, the recovery of the charging condition, i.e., the discharge, was confirmed under irradiation of 10 keV electrons, of which the energy is much higher than those used for the neutralizer in surface analysis equipment. The present results revealed that there is a contribution of FE to SE yield during the SE yield measurement for the evaluation of a MgO film as a protective layer in a plasma display panel (PDP) cell. The FE may introduce uncertainty into the evaluation of the MgO film by the IISE yield measurement.

1. Introduction

Secondary electron (SE) emission from an insulator thin-film surface under ion irradiation has received renewed attention over the last few years. One of the driving forces of studies on SE emission from an insulator thin-film surface is that the SE emission properties of MgO films, which are used as a protective layer of the dielectric layer in a plasma display panel (PDP) cell, are one of the most important key factors for furthering the development of PDPs. For the further development of PDPs, a reduction in power consumption is required and it can be achieved by lowering the firing voltage of the plasma. It has been reported that MgO film, having a high ion-induced secondary electron (IISE) yield γ, provides a low firing voltage [1] and the γ measurement of the MgO thin film has been intensively performed [2-4]. However, several studies have pointed out that there is no correlation between γ and the firing voltage [5]. This can be attributed to the fact that, in most studies dealing with γ measurement, the effects of charging are not taken into account.

As regards the SE emission from the insulator surface under charged particle irradiation, the measurements of SE yield during and after the irradiation of charged particles have been intensively studied [6-14]. The most distinguishable features observed in the SE emission from the insulator surface are the abnormally high SE yield during the irradiation of charged particles and the self-sustained electron emission (SSEE) after irradiation. One of the most well-known mechanisms describing such phenomena is the so-called Malter effect observed for thin solid insulator films [6,7]. Another is the Townsend-Avalanche effect observed for porous thin insulator films [8,9]. Both mechanisms are based on the field emission (FE) induced by the positive surface potential due to the positive charge accumulated in the insulator surface under the irradiation of charged particles. These studies have revealed that the SE emission is strongly affected by the surface potential that correlates with charging. In this regard, the systematic investigation of the SE emission properties of MgO films under ion irradiation should be performed by taking into account the influence of charging.

From the point of view of charging the insulator surface, the measurement of the SE spectrum under ion irradiation is one of the most effective approaches. Although several studies have dealt with the energy distribution of

SEs, only a few studies have been conducted in the measurement of the surface potential [15]. The authors have systematically studied the energy distribution of SEs under the irradiations of electrons and ions. The results revealed that the measurement of the SE spectrum is crucial to understand the SE emission from the insulator surface since it provides information about the surface potential as well as the energy distribution of SEs [16-18]. In the present study, therefore, we aimed at investigating the variation in the energy distribution of SEs during He^+ irradiation to study the mechanism of the SE emission properties of MgO films as a protective layer in a PDP cell.

2. Experiment

Measurements were performed using a scanning Auger microscope, JAMP-3 (JEOL). The apparatus was equipped with an electron gun for AES measurement, an ion gun and a cylindrical mirror analyzer. The primary energy of electrons for the AES measurement was 10 keV. The incident angles of electrons and ions were 45 and 50° from the sample surface normal, respectively. The beam diameters of electrons and ions at the normal incidence were ~10 μm and ~1 mm, respectively. The base pressure of the apparatus was ~2 × 10^{-7} Pa. The pressure of the apparatus during ion irradiation was maintained below ~1 × 10^{-6} Pa with the help of a differential pumping system equipped with an ion gun [19]. Details of the experimental setup are described elsewhere [16,20,21].

The sample was an MgO film of ~600 nm thickness (~1.7 × 10^{10} Ω・cm) deposited on the Si substrate by electron beam deposition. The sample was cut into the size of ~10 × 10 mm^2. No pretreatment was performed before the sample was introduced into the apparatus. The sample surface was cleaned by sputter cleaning with 1 keV Ar^+ ions until no AES peaks of contaminants were visible.

The IISE spectra were measured under the irradiation of 1 keV He^+. The beam current was ~70 nA. The experimental procedure for the measurement of the IISE spectra is schematically shown in Fig. 1. First, He^+ was irradiated for 5 min. Then, ion irradiation was stopped for 5 min. Again ion irradiation was performed for 5 min and stopped for 5 min. The 5-min irradiation and 5-min non-irradiation were repeated 7 times. The acquisition time required for measuring one SE spectrum was ~30 s. The SE spectra were measured continuously from the first to seventh repetition during both ion irradiation and non-ion irradiation.

The surface potential of the sample was measured from the onset of the SE spectrum [16-18], which is based on the SE method conventionally used for the work function measurement of metals [22,23]. The bias voltage applied to the sample, which is required for the SE method, was -23 V. The measurement of the surface potential of the insulator sample under the irradiation of charged particles is described in detail elsewhere [16-18].

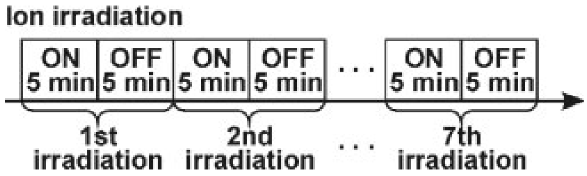

Fig. 1. Schematic diagram of experimental procedure of measurement of IISE spectra.

3. Results and Discussions

3-1. Electron Emission under 1 keV He^+ Ion Irradiation

Figures 2(a) and (b) show the electron spectra obtained during 1 keV He^+ irradiation and non-ion irradiation, respectively. The IISE spectra shown in Fig. 2(a) revealed that the profile changes upon ion irradiation. These changes are due to charging of the sample surface induced by ion irradiation. One of the most marked features is that the IISE spectra consist of two peaks. One peak stays at ~23 eV. Another peak shifts toward the lower-energy side upon ion irradiation and disappears during the fourth ion irradiation. The behaviors of the two peaks are different, indicating that the two peaks are due to electrons emitted by different mechanisms.

A shift in the onset energy and a decrease in the intensity of the IISE spectra can be observed in Fig. 2(a), and the onset energy and maximum intensity of the IISE spectra are plotted in Figs. 3(a) and (b), respectively. In Fig. 3(a), the onset energies of the spectra for the fifth to seventh ion irradiations are determined from the peak at ~23 eV because the peak shifting toward the lower-energy side disappears during the fourth ion irradiation. At the beginning of ion irradiation, the shift in the onset energy of the IISE spectra toward the lower-energy side is confirmed. This is attributed to the positive change in surface potential due to charging induced by ion irradiation and SE emission, resulting in a decrease in the intensity of SE spectra upon ion irradiation as confirmed in Fig. 3(b). Both the decrease in intensity and the shift in the onset energy of the SE spectra are rapid for the initial 0-10 min of ion irradiation and, then, gradually approach asymptotic values. No significant variation of the spectra was observed during the seventh ion irradiation. These findings indicate that charging is significant at the initial stage of ion irradiation and gradually approaches the steady state of charging [16].

The electron spectra obtained during non-ion irradiation shown in Fig. 2(b) confirmed the electron emission during non-ion irradiation. This is the so-called SSEE induced by the FE due to the accumulation of the positive charge in the surface region [1,6-9]. The profiles of the

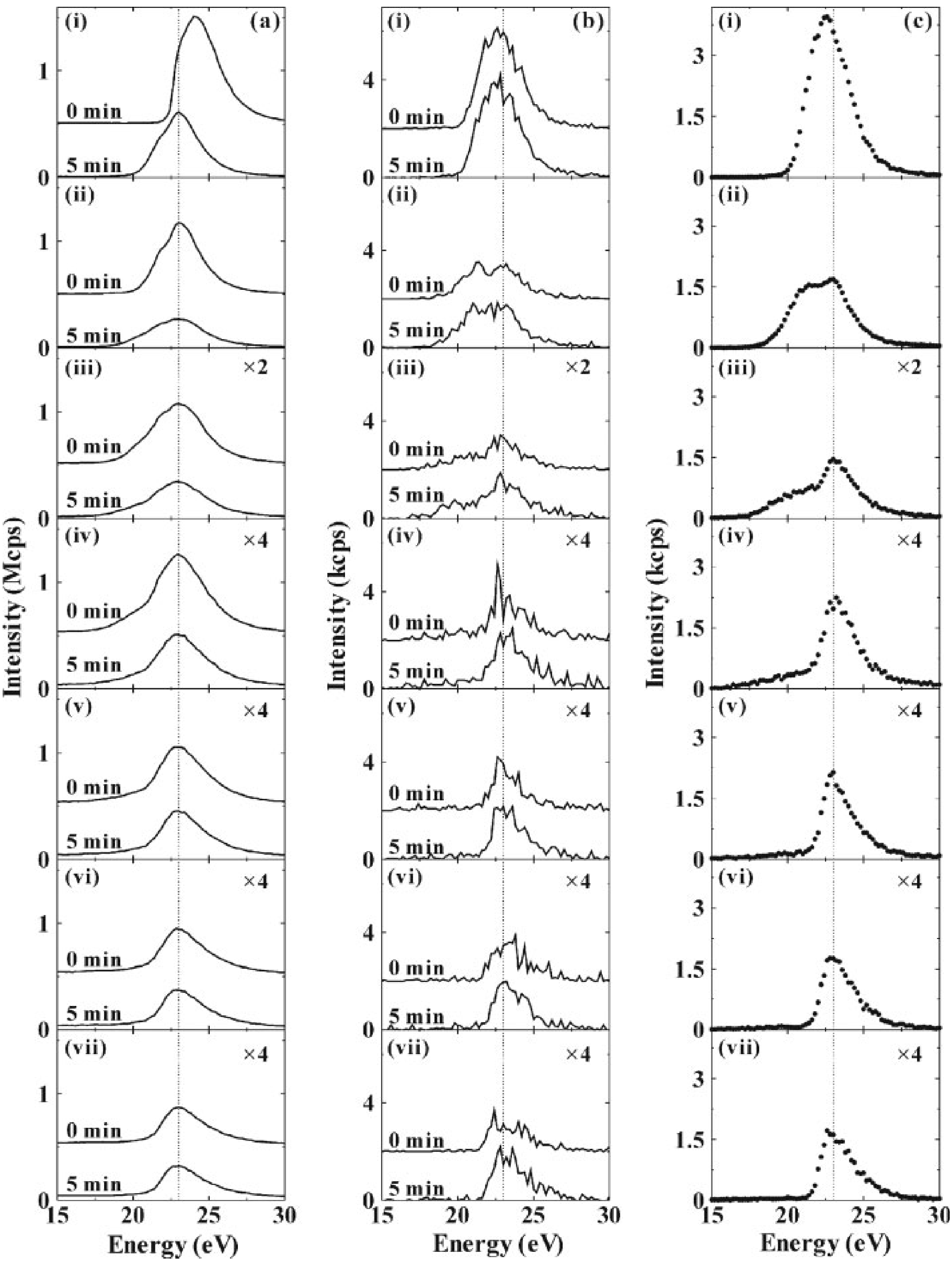

Fig. 2. (a) IISE spectra obtained under 1 keV He+ irradiation. (b) Electron spectra obtained during non-ion irradiation. (i) to (vii) correspond to the spectra obtained during the first to seventh ion irradiations. In each figure, the spectra obtained at 0 and 5 min during each ion irradiation are shown. (c) The spectra obtained by summing up all the spectra measured during non-ion irradiation. The intensity is multiplied by the factor as labeled in each figure.

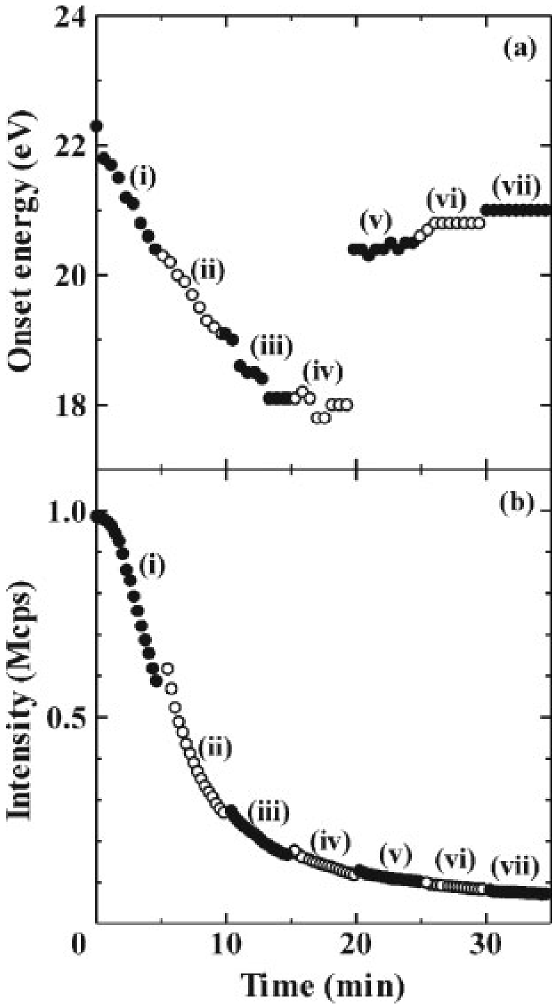

Fig. 3. (a) Onset energy and (b) maximum of 1 keV He^+ IISE spectra as function of ion irradiation time. (i) to (vii) correspond to those during the first to seventh ion irradiations.

spectra at 0 and 5 min of each round of stopping ion irradiation does not change, indicating that charging is sustained during non-ion irradiation for 5 min. This fact is consistent with the profiles of the IISE spectra at 5 min of the *i*-th ion irradiation and at 0 min of the (*i*+1)-th ion irradiation shown in Fig. 2(a) are very close. Note that charging is sustained during non-ion irradiation even though the FE was induced. This is due to the fact that the number of emitted electrons is only approximately 1/100 of that emitted during ion irradiation, which is considered to be very low to cause the change in charging conditions.

Since the profiles of the electron spectra obtained during non-ion irradiation for 5 min does not change, for more quantitative investigation of the energy distribution of electrons emitted during non-ion irradiation, all the spectra obtained during each period of non-ion irradiation were summed up. Figure 2(c) shows plots of the resultant spectra. A comparison between the electron spectra shown in Fig. 2(c) and the IISE spectra shown in Fig. 2(a) revealed that the profile of the electron spectra obtained during non-ion irradiation is close to the IISE spectra obtained just before and immediately after non-ion irradiation. The onset energy of the IISE spectrum corresponds to the vacuum level at the MgO surface, indicating that the energies of the electrons emitted during non-ion irradiation is above the vacuum level at the MgO surface. The most well known mechanisms of the electron emission from insulators due to the FE induced by the accumulation of the positive charge during charged particle irradiation are the Malter and Townsend-Avalanche effects [6-9]. The energies of electrons emitted by the Malter and Townsend-Avalanche effects could be roughly estimated from their mechanisms. In the case of the Malter effect [6,7], the energy of electrons is almost the same as the bias voltage applied to the substrate since electrons were emitted directly from the substrate by the FE. In contrast, electrons emitted by the Townsend-Avalanche effect [8,9] have an energy below the bias voltage because electrons lose their energy in the MgO film by producing SEs. These considerations suggest that the peaks at the higher- and lower-energy sides observed during non-ion irradiation could be attributed to the Malter and Townsend-Avalanche effects, respectively.

In the IISE spectra obtained during ion irradiation, two peaks are observed as well. Since the two peaks observed in the spectra obtained during non-ion irradiation could be attributed to the Malter and Townsend-Avalanche effects, the appearance of two peaks during ion irradiation might be attributed basically to the effect of the FE. Taking into account that profiles of the IISE spectra are close to those of the spectra of electrons emitted during non-ion irradiation, the IISE spectrum consists of not only electrons emitted by the kinetic and potential emission processes induced by the interaction of primary ions and the solid surface [24,25] but also electrons emitted by FE induced by charging. In addition, the similarity of the profiles of the IISE spectra obtained during ion irradiation and those obtained during non-ion irradiation, which consists of electrons emitted only by FE, implies that the field affects the energy distribution of SEs emitted by the kinetic and potential emission processes, resulting in the appearance of two peaks in the IISE spectrum. Note that, since the field emission strongly depends on the physical properties of the MgO film, e.g., thickness, conductivity, porosity, and the measurement condition, these factors may introduce uncertainty into the evaluation of the MgO film by the γ measurement as the protective layer in the PDP cell.

3-2. Effects of Electron Irradiation

Following the measurement of the IISE spectra shown in Fig. 2, electron irradiation was performed to investigate the effects of electron irradiation on the IISE emission. 10 keV electrons at ~2 nA were irradiated for 5 min immediately after the experimental procedure shown in Fig. 1. After the irradiation of 10 keV electrons, the recovery of the charging condition is confirmed. Figures 4(a) and (b) show the electron spectra obtained during the three repetitions of 5-min ion irradiation and 5-min non-ion irradiation after irradiation of 10 keV electrons. It is clearly seen that the intensity of the peak at ~23 eV increases after electron irradiation as confirmed by the comparison of the curves (i) in Figs. 4(a) and (b) with that before electron irradiation shown by the curves (vii) in Figs. 2(a) and (c). The peak at the lower-energy side appears again. In addition, the intensity of the peaks decreases upon ion irradiation. The peak at the lower-energy side shifts toward the lower-energy side and disappears. These behaviors are the same as those observed before electron irradiation.

Since the first set of measurements of the IISE spectrum, of which the procedure is shown in Fig. 1, was performed after 10 keV electron irradiation for the AES measurement in order to confirm the cleanliness of the sample surface, the two peaks observed in Figs. 2 and 4 might appear as a result of the electron irradiation. In order to confirm the effects of the irradiation of electrons before the measurement of the IISE spectra, the procedure shown in Fig 1 was performed without electron irradiation in advance. The resultant spectra are shown in Fig. 5. It is clearly seen that the electron spectrum consists of two peaks at the initial stage of ion irradiation. All the behaviors of two peaks upon ion irradiation observed in Fig. 5 are the same as those shown in Figs. 2(a) and (c), indicating that two peaks did not appear by electron irradiation but by ion irradiation. The same behavior of the recovery of the charging condition by 10 keV electron irradiation was repeatedly observed.

The range of 10 keV electron is much larger than that of 1 keV He^+. The number of SEs excited kinetically within the MgO film by 10 keV electrons is also much larger than that by He^+. In addition, it is well known that irradiation of electrons induces the so-called electron-beam-induced conduction [26,27]. These electron-beam-induced phenomena might contribute to the recovery of the charging condition. Although the beam diameter of electrons is ~10 μm and much smaller than that of ions of ~1 mm, the re-

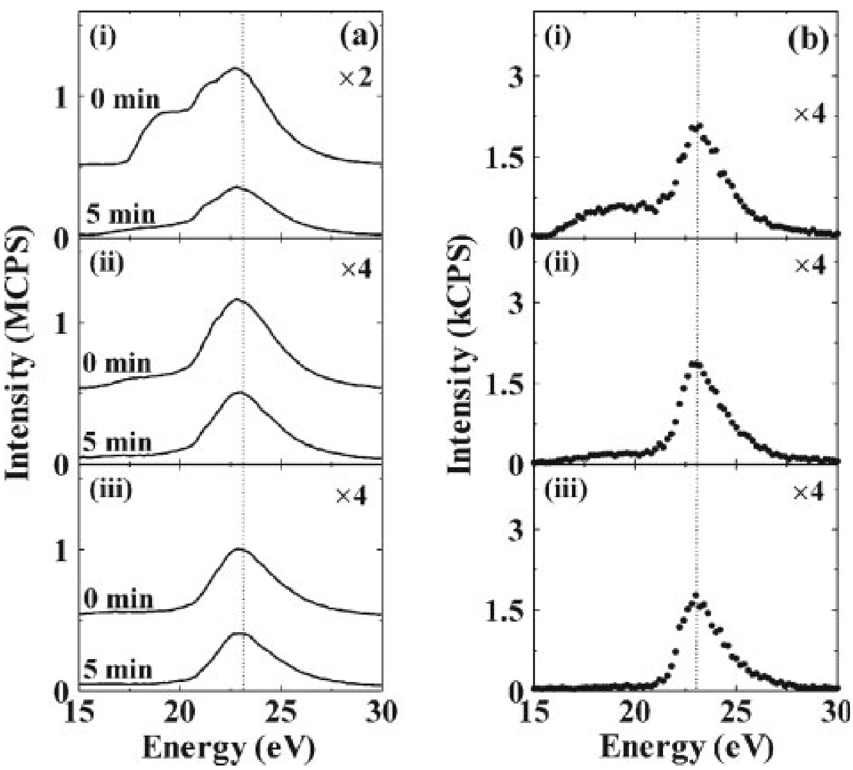

Fig. 4. (a) IISE spectra obtained during ion irradiation and (b) electron spectra obtained during non-ion irradiation. The spectra are obtained after the measurement shown in Fig. 2 followed by 5-min electron irradiation. (i) to (iii) correspond to those obtained during the first to third irradiations. In (a), the spectra obtained at 0- and 5-min ion irradiations are shown. In (b), all the spectra obtained during non-ion irradiation are summed up. The intensity is multiplied by the factor as labeled.

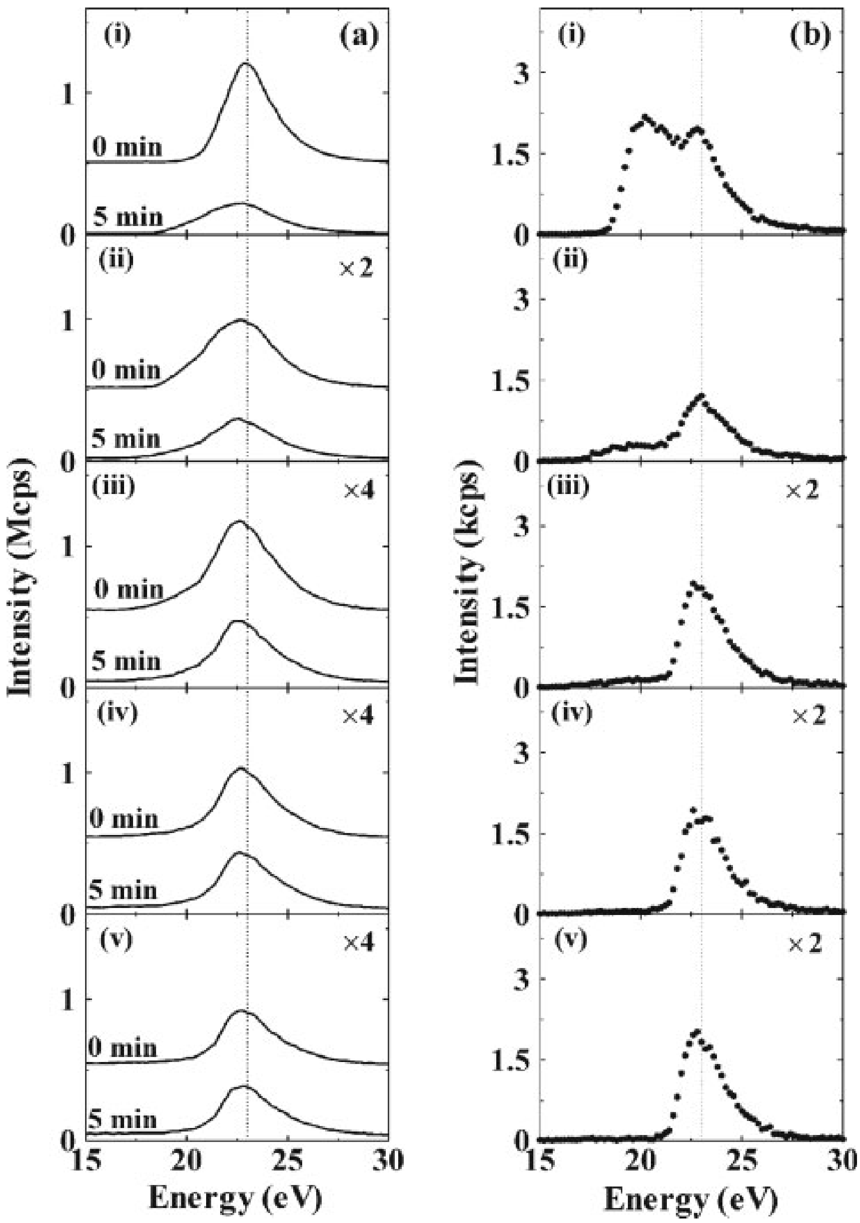

Fig. 5. (a) IISE spectra obtained during 5-min ion irradiation and (b) the electron spectra obtained during 5-min non-ion irradiation. The spectra were measured without electron irradiation in advance. (i) to (v) correspond to those obtained during the first to fifth ion irradiations. The spectra obtained at 0- and 5-min ion irradiations are shown in (a). In (b), the spectra obtained by summing up all the electron spectra measured during non-ion irradiation are shown. The intensity is multiplied by the factor as labeled.

covery of the charging condition could be expected. The area irradiated by ions is positively charged. The SE emitted from the surface by 10 keV electrons could be returned to the positively charged area by the electric field between the point of the SE emission and the surrounding positively charged area on the sample surface [28-30]. This redistribution of electron-induced SEs might also contribute to the recovery of the charging condition. It should also be noted that the electron energy used for the neutralizer in the surface analysis equipment is much lower than 10 keV.

3-3. Dependence on Ion Species

In order to investigate the effects of ion species on IISE emission, electron emission under 1 keV Ar^+ irradiation was performed. All the measurement conditions were the same as those for 1 keV He^+ irradiation. The beam current was ~70 nA and the bias voltage was -23 V. The AES measurement was not performed before the measurement of IISE. The resultant spectra are shown in Fig. 6. As observed for 1 keV He^+ irradiation, the two peaks are confirmed as indicated by arrows in the figure. Other features are also the same, indicating that the appearance of the two peaks does not depend on the ion species. The recovery of the charging condition by 10 keV electron irradiation was also confirmed. These findings suggest that the appearance of the two peaks is caused purely by the ion irradiation of the MgO surface.

3-4. Bias Voltage Dependence

For investigating the bias voltage dependence of the behavior of the electron emission, the electron spectra were measured under He^+ irradiation by changing only the bias voltage from -23 V to -28 V. The experimental procedure is the same as those shown in Fig. 1 except for the bias voltage.

Figures 7(a) and (b) show the resultant spectra obtained during He^+ irradiation and during non-ion irradiation, respectively. The two peaks are confirmed. One hardly shifts during ion irradiation. The other peak shifts toward the lower-energy side and disappears upon the ion irradiation. In addition, the recovery of the charging condition by 10 keV electron irradiation was also confirmed. All the features are the same as those observed in the case of the bias voltage of -23 V except for the peak position. The position of the peak at the higher-energy side is ~28 eV correspond-

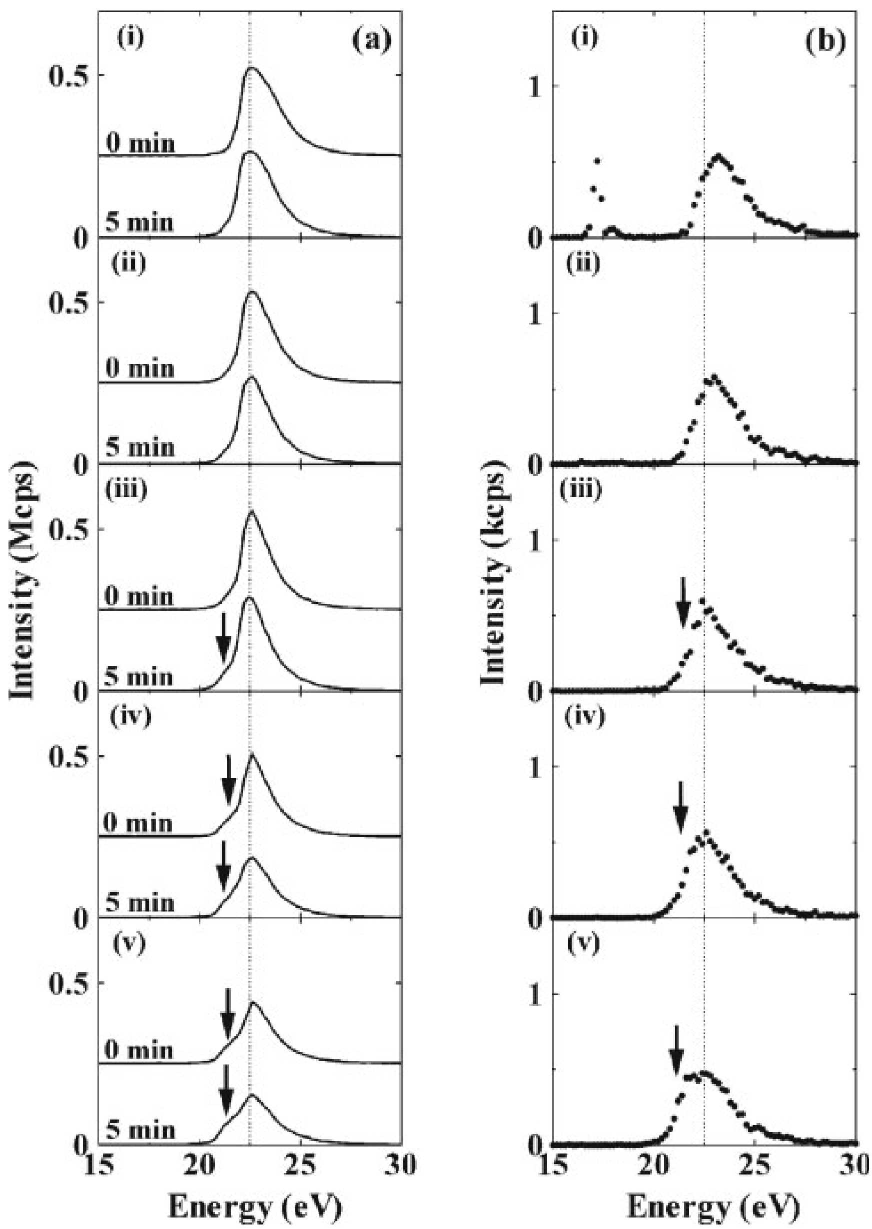

Fig. 6. (a) Ar^+ IISE spectra obtained during 5-min Ar^+ irradiation and (b) electron spectra obtained during 5-min non-ion irradiation. (i) to (v) are those obtained during the first to fifth ion irradiations. In (b), the spectra obtained by summing up all the spectra are shown.

ing to the bias voltage. This bias voltage dependence of the spectrum is consistent with the appearance of the two peaks, which could be attributed to the Malter and Townsend-Avalanche effects, since the electrons emitted by the Malter and Townsend-Avalanche effects have energies close to and below the bias voltage applied to the substrate, respectively.

4. Summaries

In the present study, the change in the energy distribution of IISEs emitted from the MgO surface upon ion irradiation and the effects of electron irradiation on the IISE emission were investigated. The present results are summarized as follows.

(i) The 1 keV He^+ IISE spectrum consists of two peaks at the initial stage of ion irradiation. The intensity of the two peaks decreases upon ion irradiation. One peak hardly shifts. The other peak shifts toward the lower-energy side and disappears. The behaviors of the two peaks upon ion irradiation are different, suggesting that the two peaks appear through different mechanisms of electron emission.

(ii) The electron emission induced by the FE due to positive charging of the surface was observed during non-ion irradiation. The profile of the spectra is very close to that observed during ion irradiation.

(iii) The charging condition is recovered by the irradiation

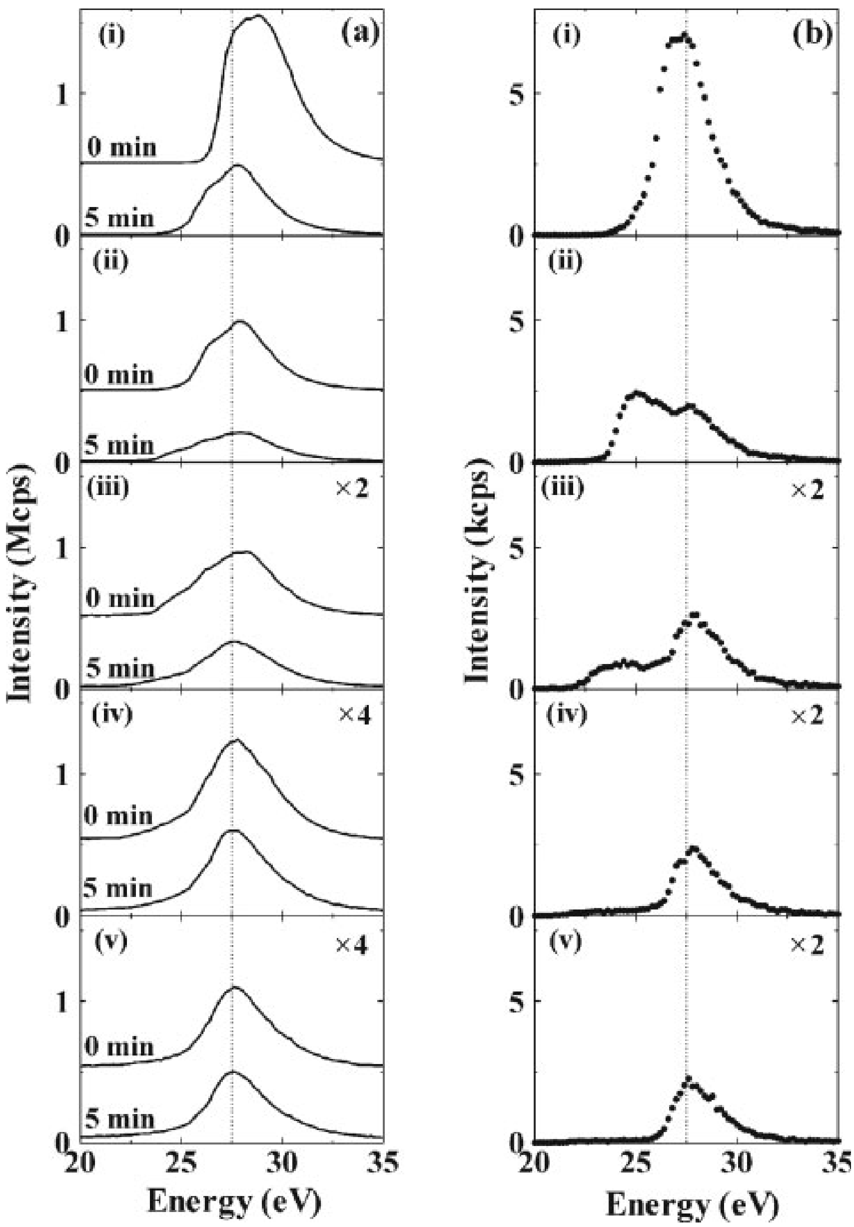

Fig. 7. (a) He^+ IISE spectra obtained during ion irradiation and (b) electron spectra obtained during non-ion irradiation. The bias voltage applied to the sample during the measurement was -28 V. (i) to (v) are those obtained during the first to fifth ion irradiations. In (b), the spectra obtained by summing up all the spectra are shown. The intensity is multiplied by the factor as labeled.

of 10 keV electrons.

(iv) The behavior of the electron spectra upon ion irradiation does not depend on both the ion species and bias voltage. These results revealed that the appearance of two peaks is purely caused by the ion irradiation of the MgO surface.

(iv) The peak staying at the same energy and that shifting towards the lower-energy side might be attributed to the Malter and Townsend-Avalanche effects, respectively.

The present results revealed that there is a contribution of the FE to the SE yield during γ measurement for the evaluation of the MgO film as a protective layer in the PDP cell. Since the FE strongly depends on the physical properties of the MgO film and the condition of the γ measurement, the FE may introduce the uncertainty into the evaluation of the MgO film by the γ measurement. Further detailed investigation of electron spectra obtained during ion irradiation and non-ion irradiation is underway and will be reported shortly.

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References

[1] H. Uchiike, K. Miura, N. Nakayama, T. Shinoda and Y. Fukushima, IEEE Trans. Electron Dev. **ED-23**, 1211 (1976).
[2] K. S. Moon, J. Lee and K. W. Whang, J. Appl. Phys. **86**, 4049 (2004).
[3] H. S. Uhm, E. H. Choi and J. Y. Lim, Appl. Phys. Lett. **80**, 737 (2002).
[4] T. Hirakawa, S. Goto, S. Zhang and H. Uchiike, *Proc. Int. Display Workshop (IDW) '02* (2002) p. 749.
[5] V. van Elsbergen, P. K. Bachman and G. Xhong, *Proc. Int. Display Workshops (IDW) '00* (2000) p. 687.
[6] L. Malter, Phys. Rev. **49**, 478 (1936).
[7] L. Malter, Phys. Rev. **50**, 48 (1936).
[8] H. Jacobs, J. Freely and F. A. Brand, Phys. Rev. **88**, 492 (1952).
[9] D. Dobischek, H. Jacobs and J. Freely, Phys. Rev. **91**, 804 (1953).
[10] J. B. Johnson and K. G. Mckay, Phys. Rev. **91**, 804 (1953).
[11] J. Drenser, J. Appl. Phys. **48**, 4760 (1977).
[12] N. J. Chou, J. Vac. Sci. & Technol. **14**, 307 (1977).
[13] W. Yi, S. Yu, W. Lee, T. Han, T. Jeong, Y. Woo, J. Lee, S. Jim, W. Choi, J. Heo, D. Heon and J. M. Kim, J. Appl. Phys. **89**, 4091 (2001).
[14] W. S. Kim, W. Yi, S. Yu, J. Heo, T. Jeong, J. Lee, C. S. Lee, H. J. Heong, Y. M. Shin and Y. H. Lee, Appl. Phys. Lett. **81**, 1098 (2002).
[15] D. M. Taylor, J. Phys. D **11**, 2443 (1978).
[16] Y. Mizuhara, J. Kato, T. Nagatomi, Y. Takai and M. Inoue, J. Appl. Phys. **92**, 6128 (2002).
[17] T. Tsujita, T. Nagatomi, Y. Takai, Y. Morita, M. Nishitani, M. Kitagawa and T. Uenoyama, Jpn. J. Appl. Phys. **43**, L753 (2004).
[18] T. Tsujita, T. Nagatomi and Y. Takai, Surf. Interface Anal. **37**, 137 (2005).
[19] Y. Mizuhara, T. Bungo, T. Nagatomi, Y. Takai, S. Suzuki, K. Kikuchi, T. Sato and K. Uta, Surf. Interface Anal. **37**, 171 (2005).
[20 T. Bungo, Y. Mizuhara, T. Nagatomi and Y. Takai, Jpn. J. Appl. Phys. **42**, 7580 (2003).
[21] Y. Mizuhara, T. Bungo, T. Nagatomi and Y. Takai, Surf. Interface Anal. **37**, 343 (2005).
[22] K. G. Eyink, B. C. Lamartine and T. W. Haas, Appl. Surf. Sci. **21**, 29 (1985).
[23] I. Ogoh, R. Shimizu and H. Hashimoto, Jpn. J. Appl. Phys. **24**, 1145 (1985).
[24] H. D. Hungstram, Phys. Rev **96**, 336 (1954).
[25] W. O. Hofer, *in Fundamental Electron and Ion Beam Interactions with Solids for Microscopy, Microanalysis and Microlithography*, eds. J. Schou, P. Kruit and D. E. Newbury (1990) Scanning Microscopy Suppl. 4.
[26] K. Ura, Electron Microsc. **35**, 254 (2000) (in Japanese).
[27] K. Ura and H. Fujioka, Adv. Electronics Electron Phys. **73**, 233 (1989).
[28] K. Ura, J. Electron Microsc. **47**, 143 (1998).
[29] S. Aoyagi and K. Ura, J. Electron Microsc. **48**, 555 (1999).
[30] K. Ura and S. Aoyagi, J. Electron Microsc. **49**, 157 (2000).