



Isomerization structure of photochromic diarylethene film based on electrical carrier injection

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ABSTRACT

Photochromic films have attracted interest as an organic semiconductive memory material. Isomerization structures of a photochromic diarylethene (DAE) film induced by electrical carrier injection were investigated and were made clear for the coloring-reaction mode (CRM) and decoloring-reaction mode (DRM). Filaments consisting of the colored molecules in the colorless matrix were generated in the CRM, while random distribution of isomerized molecules occurred in the DRM. The ON-OFF ratio in current in the CRM was larger than that in the DRM. These results would be important for constructing organic semiconductor memory device using DAE.

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1. Introduction

Such organic electronics devices as light emitting devices, thin-film transistors, and solar cells have attracted interest due to their, light weight, large area and low cost process ability [1–4]. Organic semiconductor memory is one of essential organic devices and various memory principles have been proposed [5,6]: charge traps on floating metal electrodes in an organic layer [7] or generation of conductive filaments consisted of metal nanoparticles [8].

Photochromic diarylethenes (DAEs) are well known photo-switchable molecules having fatigue resistance and thermal stability in both isomerization states [9]. Various properties of DAE changed according to photoisomerization, including color, dipole moment, glass transition temperature, and metal-vapor deposition property [10,11]. Electrical property with a photoswitching capability of DAEs has attracted interest [12,13] and has been investigated for applying to such electronic devices as organic light emitting diodes [14], thin film transistors [15–17], and light-electric conversion devices [18]. Almost all studies, however, utilizes on light irradiation for changing the electrical property. A typical property undergone photoswitching is molecular electronic states; the colored (closed-ring) state of DAE has an ionization potential (I_p) of 5.7–5.8 eV and the colorless (open-ring) molecule over 6.2 eV, and the difference of I_p induces a change of current injection into the DAE layer from the electrode [12].

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An electrical isomerization principle of DAEs based on the carrier injection has been proposed and the principle indicates potential application to organic semiconductor memory devices [19–21]. The excited state, in which an electron and a hole are on the LUMO and HOMO levels, respectively, is generated by both-carrier injection from electrodes and induces the isomerization reaction. In such electrical isomerization, there are two reaction modes: coloring reaction mode (CRM) and decoloring reaction mode (DRM). Only a few study on the isomerization structure of the DAE film in the CRM has been attained [20,21], while the difference of the isomerization structures between the CRM and DRM has not been studied so far. In this paper, we report a detailed study on the isomerization structure of the DAE film based on these two modes by observation of injected current, photon emission and absorption.

2. Results and discussion

The DAE molecule isomerizes to the colored (closed-ring) state upon UV-light irradiation ($\lambda_s=365$ nm) from the colorless (open-ring) state and reverts to the colorless state upon visible-light irradiation. Fig. 1 shows a device structure with a DAE layer. The DAE molecule has benzothiazole bases for achieving well electron-affinity and is expected to have both-carrier (hole and electron) acceptance, which means easy to make an excited state for isomerization reaction. Each layer was prepared by a conventional vacuum evaporation method under 2×10^{-5} Torr at 25 °C in turn

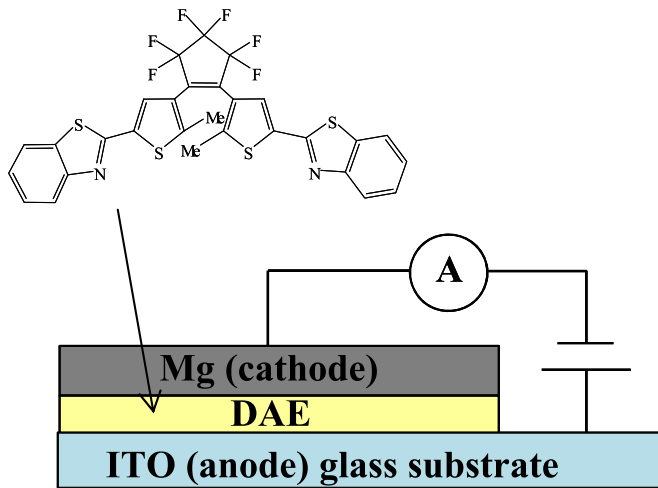


Fig. 1. Device structure with DAE layer. Cell area was $2 \times 2 \text{ mm}^2$. Thicknesses of DAE amorphous and Mg layers were 50 nm and 70 nm, respectively.

on a ITO substrate. Isomerization state of the DAE film as deposition was in the colorless state.

When both carriers were injected into the DAE film, meeting of a hole and an electron on the DAE molecule makes the excited state. Three processes for the excited molecule are possible: isomerization, photon emission, or thermal quenching. A fluorescence quantum yield ϕ_f , which was measured using the quantum efficiency measurement system (QE-2000, Otsuka Electronics Co., Ltd.), of the colorless DAE was very small in a film state ($\phi_f \sim 10^{-3}$), whereas the colored molecule did not emit photons ($\phi_f=0$). Intensity of photon emission from the excited colorless state, therefore, refers the quantity of colorless molecules in the film. The very weak emission was observed using a photon counting method.

First, changes of the injected current and photon emission into/from the colorless DAE layer were investigated at a constant voltage of 12 V (Fig. 2a). The current increased rapidly from 360 μA at the colorless state to 2400 μA after 600 s, which was the current stationary state (CSS), and the photon emission decreased. The current and photon emission characteristics of the current-injected sample did not change after storage in the dark for one hour and were recovered to the initial state upon visible-light ($\lambda > 480 \text{ nm}$) irradiation. This means that the changes in current and photon emission originate in the isomerization from the colorless state to the colored state (coloring-reaction mode, CRM).

Next, changes of current and photon emission into/from the colored DAE layer were investigated at a constant voltage of 12 V (Fig. 2b). The initial colored state was the photostationary state (PSS) obtained by the sufficient UV irradiation. The current decreased from 11,400 μA to 4400 μA with time and became the CSS. On the other hand, the photon emission increased from zero to 440 count/s. The excited colored DAE molecules does not emit photon. It is, however, possible to meet some holes and electrons on the colored molecules, generating colorless molecules electrically and the excited colorless molecule can emit a photon. Increase of photon emission, therefore, indicates an increase of the colorless molecules. The current and the photon emission characteristics of the current-injected sample did not change after storage in the dark for one hour and was recovered to the initial state by UV-light irradiation. This means that the changes in current and photon emission originate in the isomerization from the colored state to the colorless state (decoloring-reaction mode, DRM). The ON-OFF ratio in current, which was defined by the ratio of the CSS to the initial state, obtained by the DRM was smaller than that by the CRM and, therefore, the CRM is desirable as a

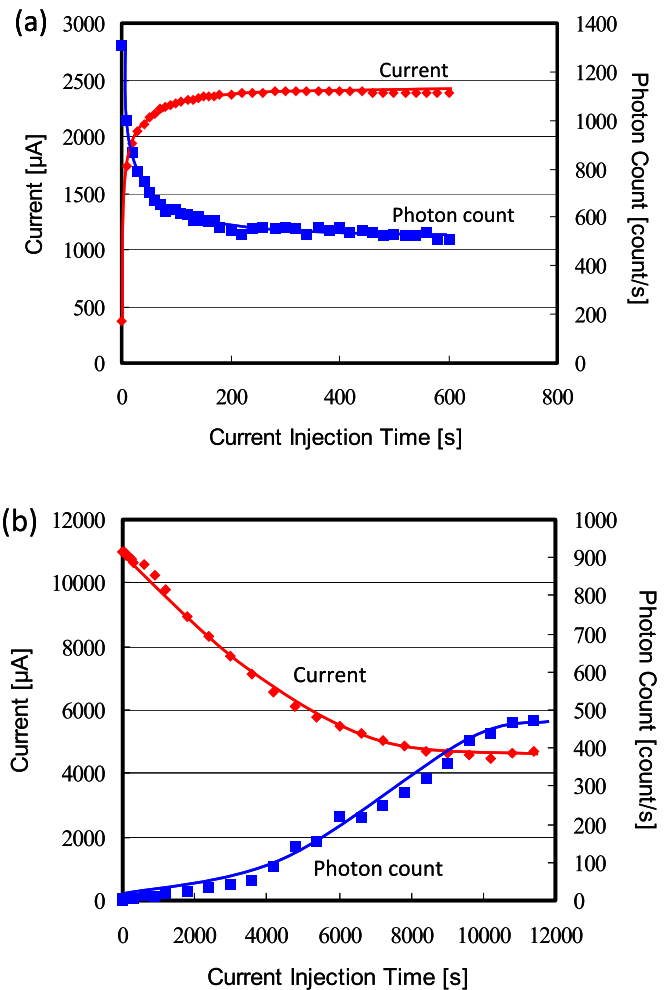


Fig. 2. Changes of injected current and photon emission in (a) CRM and (b) DRM.

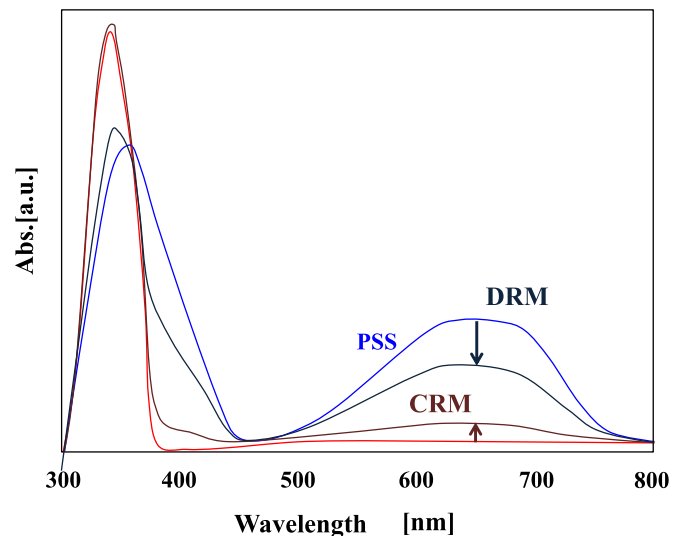


Fig. 3. Absorption changes in CRM and DRM.

memory device.

Fig. 3 shows the absorption spectrum changes of the DAE films in the electrical carrier injection. The absorption changes confirmed directly the isomerization reaction by the electrical carrier injection. The CSS levels generated by the CRM and DRM did not coincide each other. The CSS in the CRM was a considerable lower

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