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Transient absorption measurement of organic crystals with femtosecond-laser scanning microscopes

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Abstract

Two types of femtosecond-laser scanning transient absorption microscopes have been developed as tools for observing ultrafast exciton dynamics in organic solids, which have recently received much attention for their roles as organic photofunctional devices, such as solar cells and light-emitting diodes. One microscope is a multiphoton excitation transient absorption microscope, and the other is an apertured probe transient absorption microscope. Their performance was evaluated by measuring transient absorption spectra, kinetics and images of perylene crystals.

Keywords: Transient absorption; Organic crystal; Laser scanning microscope; Exciton; Perylene; Ultrafast spectroscopy

1. Introduction

Recently, organic solids, such as fullerene, carbon nanotubes, phthalocyanine, perylene derivatives and some metal complexes (e.g., Alq3 [1] and Ir(ppy)$_3$ [2]), have received much attention because they can be used in such photofunctional devices as solar cells [3,4] and light-emitting diodes [1]. Dye-sensitized solar cells are also attracting much interest as organic–inorganic hybrid solar cells [5]. Understanding the dynamic behaviors of excitons and charge carriers generated by photoexcitation in solar cells or by electric current injection in light-emitting diodes is important for the design of efficient devices. The size of the devices is on the millimeter scale, and that of the device components (e.g., the metal and transparent electrodes and the light-absorbing, light-emitting and charge transport layers) that form the layer structure, is on the micro- or nanometer scale. The inner structures of each component are on the nanometer scale. Such structures range from nano- to millimeters in size and are important in realizing high performance of the devices. Compared with inorganic devices based on silicon processing technology, the organic devices have uncontrolled structure; therefore, inhomogeneous reactions of photogenerated excitons and charge carriers are expected to occur. Namely, position dependent reaction rates and yields are expected to observe. This fact makes the mechanism of these devices difficult to understand.

To observe ultrafast dynamics of excitons and charge carriers as a function of their position in these devices, time- and space-resolved spectroscopic techniques need to improve. Masuhara and coworkers have already reported transient absorption microscopic systems [6–9]. Recently, another group reported femtosecond fluorescence up-conversion microscopy [10]. However, application of such techniques in photofunctional devices has not been achieved.

Because multiphoton absorption occurs only at the focus point of an intense laser, it is possible to excite only a desired point in a three-dimensional space, whereas single-photon absorption occurs from the irradiated surface to a penetration depth determined by the absorption coefficient of the material. Therefore, measurement of transient absorption using multiphoton excitation would be useful for examining actual photofunctional devices. Ultrafast reactions in selected device components or at interfaces between components can be measured, when the size of the components is on the micrometer scale. Also, by scanning the sample using a three dimensional translation stage, transient absorption images can be obtained, which will enable us to discuss inhomogeneous nature of photo-generated spices within the devices. However, such a technique has not yet been reported.

As also mentioned above, smaller device structure – to nanometer scale – is important. The technologies of scanning probe microscopes, such as scanning-tunnelling microscopy

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(STM) and atomic force microscopy (AFM), have been developed to achieve nanometer-level or even atomic-level resolution of the surface topography. Scanning near-field optical microscopes (SNOM) are also well-known tools for obtaining optical information with <100-nm spatial resolution, which is better than the diffraction limit of light. The combination of SNOM and ultrashort pulse laser techniques to measure transient absorption, with temporal resolution of ∼100 fs and spatial resolution of ∼100 nm, has been reported by several groups [11–18].

This method would be useful for measuring ultrafast reactions at the surface of photofunctional devices. However, owing to the metal-coated sharpened optical fiber probes used to guide the pump and probe lasers in reported systems, which limit the available wavelength range, application of the time-resolved SNOM to photofunctional devices is difficult. Instead of such conventional optical fiber probes, an apertured AFM cantilever tip will be convenient because of the following advantages [19]. The high throughput of light through the aperture makes it possible for large and precise transient signals to be obtained. The fiber-free optical set-up minimizes the expansion of the temporal pulse width of the femtosecond laser arising from the group velocity dispersion. It is also easy to use the pump and probe lasers with widely separated wavelengths. To obtain widely tunable wavelengths of femtosecond laser, amplified Ti:sapphire laser systems combined with optical parametric amplifiers (OPA) and a white-light continuum generation system will be useful. In earlier reports, however, Ti:sapphire oscillators at around 800 nm were used; therefore, tuning the pump wavelength has not yet been realized, although a tunable probe has been realized very recently using photonic crystal optical fibers [17].

We have developed two types of femtosecond-laser scanning transient absorption microscopes for application to actual photofunctional devices. One is the multiphoton excitation transient absorption microscope, and the other is the apertured probe transient absorption microscope. In the former system, multiphoton excitation is induced by intense femtosecond-laser pulses at 800 nm, and the white-light continuum in the visible and near-IR wavelength region is used to monitor transient absorption at the excited point. In the latter system, frequency-doubled femtosecond-laser pulses at 400 nm and the white-light continuum pulses were introduced into a small aperture with ∼1 μm diameter as pump and probe light, respectively. In this paper, we describe the optical set-ups of these systems, and then we evaluate the performance of the microscopes on the bases of measured transient absorption spectra, kinetics and images of well-known perylene crystals.

2. Experimental

Optical set-ups for multiphoton excitation and apertured probe transient absorption microscopes are shown (Fig. 1, systems A and B, respectively). The light source was an amplified Ti:sapphire system (800-nm center wavelength, 50- or 150-fs full width at half-maximum (FWHM) pulse width, ∼1 mJ/pulse intensity, 1-kHz repetition; Spectra Physics, Super Spitfire or Hurricane). For exciting the samples, the fundamental light at 800 nm was used in multiphoton excitation transient absorption microscopy, and the second harmonic at 400 nm was used in the apertured probe transient absorption microscopy. The pump beams were modulated at 500 Hz by a mechanical chopper (New Focus, 3501). The white-light continuum, generated by focusing the fundamental light into a sapphire plate (2 mm thick), was used as the probe light for both microscopic systems. Pump and probe beams were collinearly aligned after the pump–probe delay time was adjusted by a translational stage, and then the beams were introduced into the entrance port of the microscopes.

As shown in Fig. 1 (system A), in the multiphoton excitation transient absorption microscope, pump and probe beams with a 4-mm waist were introduced into the microscope and were focused by an objective lens (4× magnification, 30-mm working distance) set under the sample. Light transmitted upwards through the sample was collected by another objective lens (same specification) placed above the sample. The collimated beams were focused again at a pinhole (100 μm diameter) to form a confocal microscope alignment. The pump beam was blocked by using an interference filter. The probe light through the pinhole was detected by a silicon or InGaAs PIN photodiode after passage through a monochromator (Acton Research, SpectraPro-150). The photodiode signals were acquired into a computer on a pulse-to-pulse basis. The intensity of the probe light entering the microscope was monitored to compensate for the pulse intensity fluctuation. Transient absorption intensity was calculated as $\log(I_0/I)$, where I_0 and I are (fluctuation compensated) probe pulse intensities, without and with excitation, respectively. Typically, several thousands of pulses were averaged to gain a transient absorption accuracy of 10^{-3}. The sample can also be scanned by an *XYZ* piezo stage (Physik Instrumente, P-562.3CD) to obtain transient absorption images.

In the apertured probe transient absorption microscope, a commercial scanning near-field optical microscope (Alpha-SNOM, WITec, Ulm, Germany) was used. The pump and probe beams (Fig. 1, system B) were directed through a small aperture at the tip on the bottom side of the cantilever probe. The laser beam was focused around the aperture on the top side of the cantilever by an objective lens (8× magnification), and the light transmitted through the aperture and a thin sample was collected by another objective lens (20× or 60×), collimated and directed into the entrance of an optical fiber that was connected to a silicon PIN photodiode or an avalanche photodiode (APD). Two CCD cameras were used to monitor the positions of the focused beams on the cantilever from both the top and the bottom sides. The AFM was operated in the contact mode using a 980-nm feedback laser. An *XYZ* piezo stage was used to scan the sample. Although an aperture diameter <100 nm was available for the commercial system, the aperture was expanded to about 1 μm by laser ablation in order to obtain enough light intensity for the pump–probe measurements.

A single crystal of α-perylene was grown by the Bridgman method from extensively zone-refined material. Plate-like crystals, which were fragments from the ingot, were chosen for transient absorption measurement. In the multiphoton excitation transient absorption microscope, a single crystal with dimensions of about 3 mm × 3 mm × 0.1 mm was placed on an

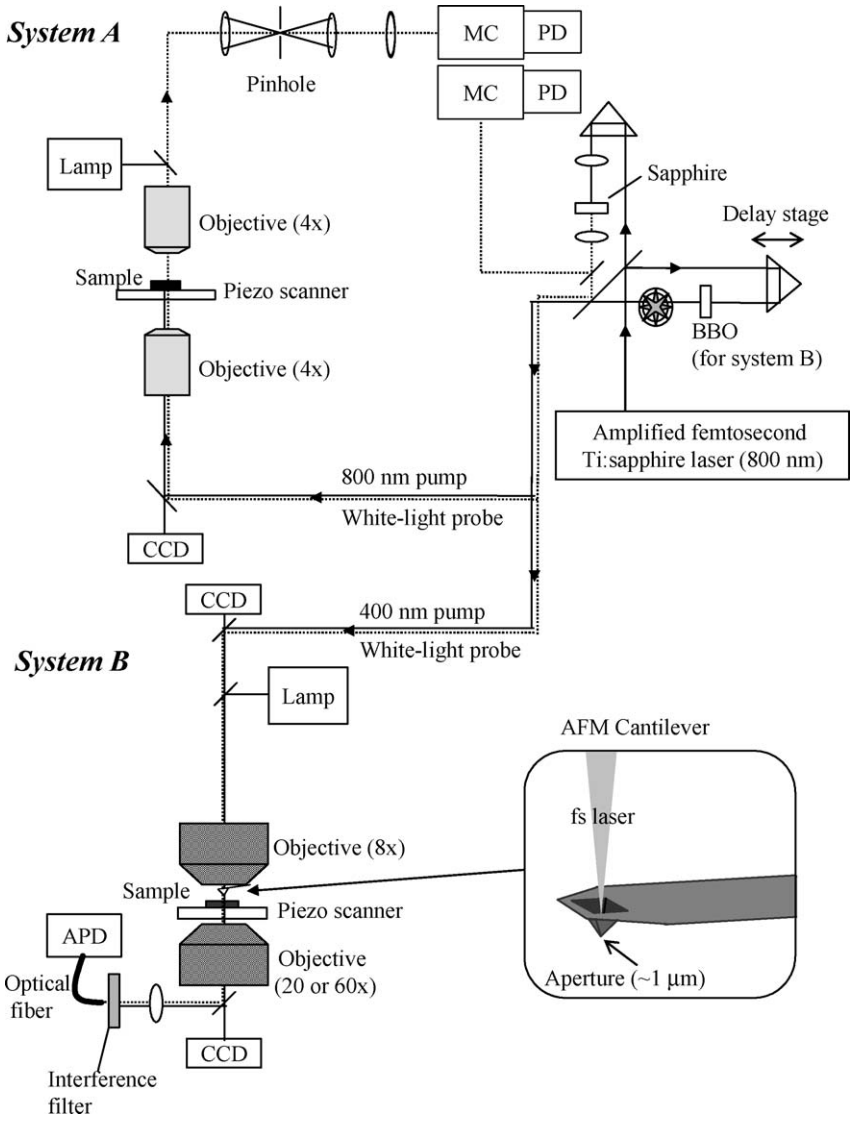

Fig. 1. Optical set-ups for multiphoton excitation and apertured probe transient absorption microscopes (systems A and B, respectively).

acrylic substrate, in which a small hole with 1-mm diameter was drilled just below the crystal to avoid multiphoton excitation of the substrate. In the apertured probe transient absorption microscope experiment, crystals of dimensions smaller than 1 mm × 1 mm × 0.1 mm were placed sparsely over a cover glass on the sample stage of the microscope. The cleaved surface of the crystal is usually the *ab* plane, so that under the microscopes the incident light was normal to the crystal *ab* plane.

3. Results and discussion

3.1. Multiphoton excitation transient absorption microscopy

Transient absorption spectra and kinetics of a single perylene crystal were measured to evaluate the performance of the multiphoton excitation transient absorption microscope described here. Transient absorption spectra at a delay time of 3 ps in the spectral range from 550 to 750 nm (closed circles) and transient absorption kinetics up to 15-ps delay time are shown (Fig. 2a and b, respectively). The solid line in Fig. 2a indicates the transient absorption spectra of a large perylene crystal measured by a nanosecond-laser transient absorption system [20]. This line can be regarded as reference data for the present study using microscopes. Because the absorption onset of the perylene crystal is around 530 nm (2.34 eV), the 800-nm excitation laser (1.55 eV) cannot be absorbed by a single-photon process, and multiphoton absorption – higher than a two-photon process – is necessary to electronically excite the perylene crystal. To confirm whether a multiphoton absorption process takes place under the present experimental conditions, the fluorescence intensity was measured as a function of the 800-nm laser intensity. In the weaker excitation intensity region than transient absorption could be measured, the fluorescence intensity was proportional to the square of the laser intensity, indicating that two-photon absorption took place. Transient absorption was measured at an excitation photon density that was about three orders of magnitude higher than this condition; thus the observed transient

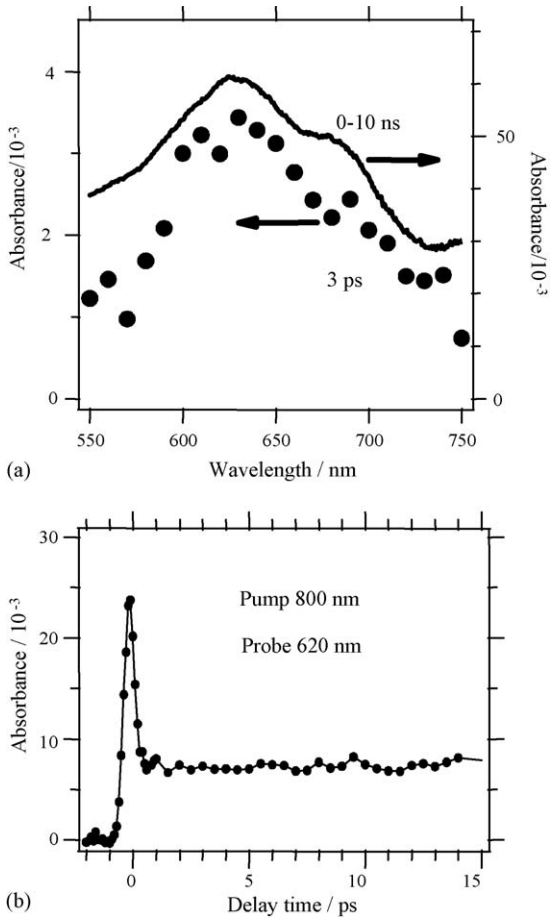

Fig. 2. (a) Transient absorption spectra of a perylene crystal photoexcited at 800 nm at a delay time of 3 ps in the spectral range from 550 to 750 nm (closed circles), and transient absorption spectra of a large perylene crystal measured by a nanosecond-laser transient absorption system (solid line). (b) Transient absorption kinetics at 620 nm, with a delay time of up to 15 ps.

absorption signals were surely induced by a multiphoton process.

In our recent study of the photoexcited perylene crystal, we observed that self-trapped excitons (excimer-like state) formed within 100 fs for the major component, accompanied by a minor, slow (∼2 ps) process [20], and its lifetime was roughly 80 ns. Therefore, the solid-line spectrum (Fig. 2a) corresponds to the absorption band of self-trapped excitons in a perylene crystal. A characteristic peak is seen at 620 nm. This absorption band is identical to the transient absorption spectrum measured under the microscope. This result indicates that the high photon density under the microscope did not induce any sample damage or even ionization of perylene molecules. Ionization due to high photon density effect has been reported for a transient absorption measurement of a perylene crystal under a microscope in a previous study [8]. In the present study, the generated exciton density was calculated to be about 10^{-6} mol/L, which corresponds to a molecular ratio of only 10^{-7}.

The transient absorption kinetics at 620 nm, the absorption peak, shows a spike at 0 ps and constant intensity after this spike (Fig. 2b). The spike is due to the nonlinear processes of the pump and probe pulses, which is probably two-photon absorption involving one photon of the pump pulse and another of the probe pulse, because the spike intensity was proportional to the pump intensity. The width of the spike can be considered to be the time resolution of the system, which was determined to be 320 fs in the FWHM. The constant transient absorption is due to the dominant rapid self-trapping process and to the absence of exciton–exciton annihilation decay.

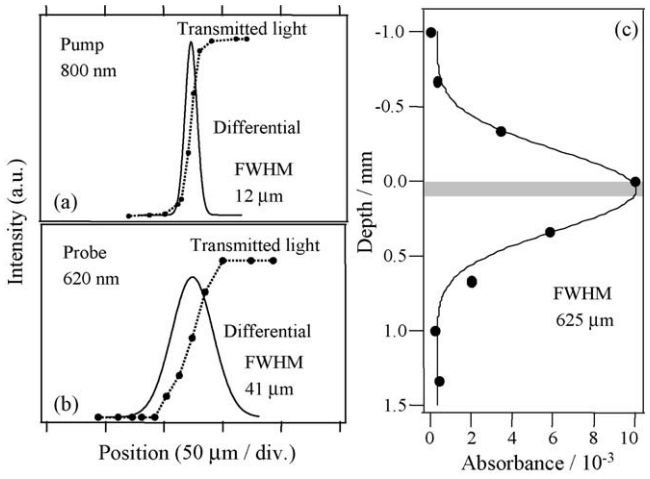

Fig. 3. Results of a knife-edge experiment to examine the focus sizes of the pump (a) and probe (b) beams. (c) Transient absorption profile of a thin perylene crystal at 950 nm when the crystal was scanned along the optical axis. See text for details.

These spectral and kinetic data demonstrated that this multiphoton excitation transient absorption microscope is useful for studying exciton dynamics in organic crystals without the interference of the effects of high exciton density.

We now discuss the three-dimensional spatial resolution of this system. To evaluate the lateral resolution, the focus sizes of the pump and probe beams were measured by scanning a knife edge across the focus spots. The results are shown for pump and probe lasers (Fig. 3a and b, respectively). The intensities of the transmitted light increased as the knife edge moved. Their differential functions were fitted to Gaussian functions with FWHM values of 12 and 41 μm, respectively. Because the overlapped area of both the pump and the probe lasers contributes to transient absorption signals, the lateral resolution is determined by the pump laser in this configuration. When two-photon excitation is considered, the lateral resolution will be $12/2^{0.5} = 8.5$ μm. Note that the larger area of the probe spot relative to that of the pump spot decreases the overall transient absorption intensity, but this does not degrade the lateral resolution.

The axial resolution was evaluated by scanning a thin perylene crystal (<100 μm) along its optical axis. The result of setting the probe wavelength at another peak of the self-trapped exciton, at 950 nm [20], is shown (Fig. 3c). When the depth was zero, the laser focus was on the top surface of the perylene crystal. Because the travel range of the piezo stage in the axial direction was 200 μm, the two objective lenses were shifted vertically in the same direction. In the figure, the crystal thickness is indicated by the grey area. Compared with this area, the range in which transient absorption signals were obtained is larger, and the width was analyzed to be 625 μm (FWHM). This can

be regarded as the axial resolution. Hence 8.5-μm lateral and 625-μm axial resolutions of this system using 4× objective lenses were determined. Based on the theory of two-photon fluorescence microscopy [21], 7.3-μm lateral and 630-μm axial resolutions were estimated using 800-nm wavelength and 0.04 N.A., showing good agreement with the experimental estimations. Therefore, if we replace the present objective lenses with, for example, ones with 0.5 N.A. (20× magnification), about 0.6-μm lateral and 4-μm axial resolutions can be expected. We are planning such an experiment now.

The temporal and spatial resolutions of the multiphoton excitation transient absorption microscope were evaluated. This microscope could be applied to actual photofunctional devices, for instance, dye-sensitized solar cells [5]. Because of the metal electrode and the colored electrolyte in the device, conventional transmittance-mode transient absorption techniques cannot be applied. We measured transient absorption of this device without the electrode and the electrolyte in our previous study [22–24]. Because the dye-sensitized semiconductor film electrode, which is the most essential part of the device to convert sunlight into electricity, is about 10 μm thick with an area of 5 mm × 5 mm, a selected part of the dye-sensitized semiconductor electrode can be measured when laser beams are in-plane of the film, and position-dependent ultrafast reactions would be revealed. Such a study is now in progress.

3.2. Apertured probe transient absorption microscopy

We evaluated the performance of the apertured probe transient absorption microscope constructed by measuring the transient absorption of perylene microcrystals. Time-resolution was examined by a two-photon absorption method that we used in our previous research [19]. We showed that the apertured probe transient absorption microscopes had a time resolution of ca. 200 fs when using 660-nm laser pulses from an OPA pumped by a 160-fs Ti:sapphire laser at 800 nm, where the autocorrelation function was measured by the two-photon absorption method. To examine whether better time resolution could be achieved, shorter pulses of 50-fs FWHM at 800 nm were introduced to the microscope, and the auto-correlation function was measured by the same method. A GaAsP photodiode was placed at the sample position and was contacted by the probe aperture. Two-photon induced photocurrents were measured as a function of the time interval between two pulses of equal intensity. The experimental result is shown in Fig. 4, and the width was determined to be 63 fs. Note that, in this experiment, a negative chirp was provided to the pulse to compensate for the group velocity dispersions of the optical components in front of the aperture in the microscope. The degree of the negative chirp was adjusted in such a way that the shortest autocorrelation width (89 fs) could be obtained without the cantilever probe. Therefore, the apertured cantilever probe was proved to maintain the pulse width, so that, regarding time-resolution, transient absorption experiments using this probe would be possible as conventional transient absorption experiments.

Perylene microcrystals were put on a cover glass, and an AFM image was measured for one of the crystals (Fig. 5a); an

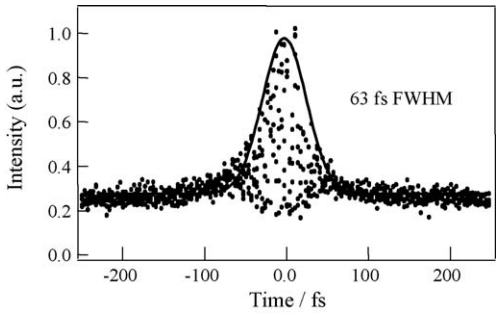

Fig. 4. Autocorrelation function of 50-fs laser pulses at 800 nm through the aperture of a cantilever probe, measured by the two-photon absorption method.

80 μm × 80 μm area was scanned, and several raised portions, with heights from 250 to 600 nm, were seen. The flat part of the surface is considered to be the *ab* plane of the crystal, but the origin of the raised portions is not clear. Transient absorption was measured at several positions, and the results are discussed below.

The excitation wavelength was 400 nm, and the probe wavelength was 700 nm, where both the quasifree and the trapped excitons can be observed [20]. Although the details of the exciton dynamics are still under consideration, at least it is certain that, at 700 nm, transient absorption decay with a time constant of about 2 ps can be observed, corresponding to self-trapping of quasifree excitons, and that long-lived absorption due to the generated self-trapped exciton, whose absorption peak is located at 620 nm (Fig. 2a), can be observed. For 400-nm excitation, the skin depth can be expected to be on the order of 1 μm, owing to the large absorption coefficient [25]. The aperture diameter of the cantilever probe was about 1 μm. This dimension (1-μm diameter and depth) can be regarded as the observed volume in this experiment.

First, to judge whether tip–sample interaction affected the observed transient absorption, the tip–sample distance was changed from zero to 350 ± 50 nm at tip position A. Energy transfer or electron transfer from the excited perylene crystal to metal (aluminium) coated on the probe might take place (see Fig. 5b). No appreciable difference was seen between the two time profiles, indicating there is no tip–sample interaction in the picosecond time range.

When the exciton density in organic crystals is high, exciton–exciton interaction induces the annihilation process and transient absorption decay becomes faster. This process is undesired because our aim in the future is to apply this microscopic system to actual photofunctional devices that operate under weak light, such as sunlight. Transient absorption decay profiles at 700 nm were measured as a function of excitation laser intensity. The result is shown in Fig. 5c, where the laser intensity was doubled between the two profiles. The normalized profiles are identical, so that the exciton–exciton annihilation process is negligible and the observed decay corresponds simply to monomolecular relaxation of the quasifree exciton. The decay time was 2.2 ± 0.5 ps. Absence of exciton–exciton annihilation is further confirmed by the measured transient absorption time profile at 600 nm, where constant absorbance can be

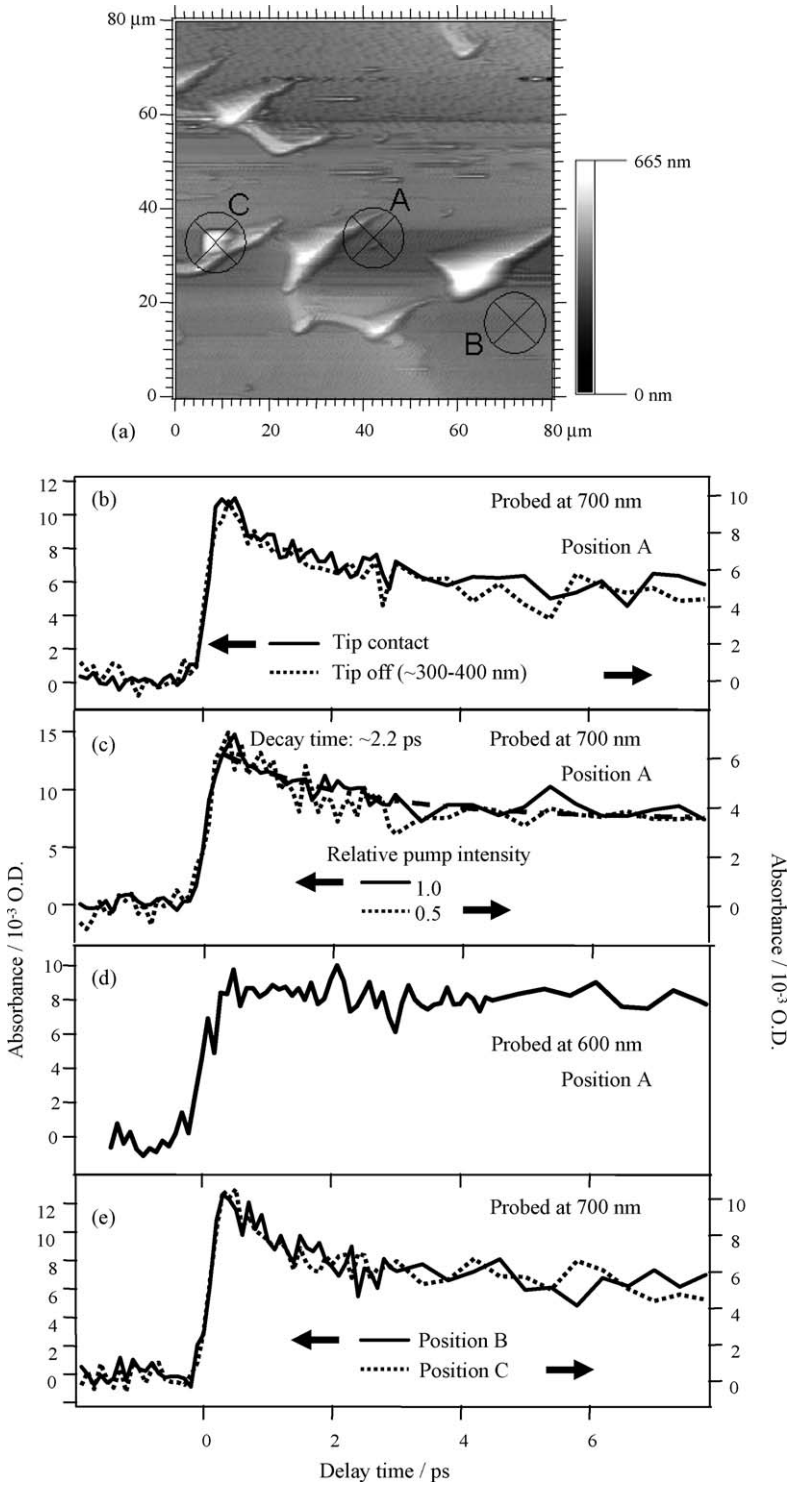

Fig. 5. (a) AFM image of a perylene microcrystal. (b) Tip–sample distance dependence of transient absorption kinetics at 700 nm measured at position A in the AFM image. (c) Excitation intensity dependence of transient absorption kinetics at 700 nm measured at position A. (d) Transient absorption kinetics at 600 nm measured at position A. (e) Transient absorption kinetics at 700 nm measured at positions B and C.

expected (see Fig. 2b) and actually no decay was observed (see Fig. 5d).

The absence of tip–sample and exciton–exciton interactions indicates that apertured probe transient absorption microscopy can be used to measure exciton dynamics in perylene under conditions similar to sunlight irradiation. The position dependence of the exciton dynamics was investigated by measuring transient absorption decay at several points of the apertured probe: positions A, B and C. In Fig. 5e, the decay profiles at positions B (on a flat area) and C (at a raised portion) are overlaid after normalization, showing no difference in the decay dynamics. A slight decrease (∼20%) in the transient absorption intensity for

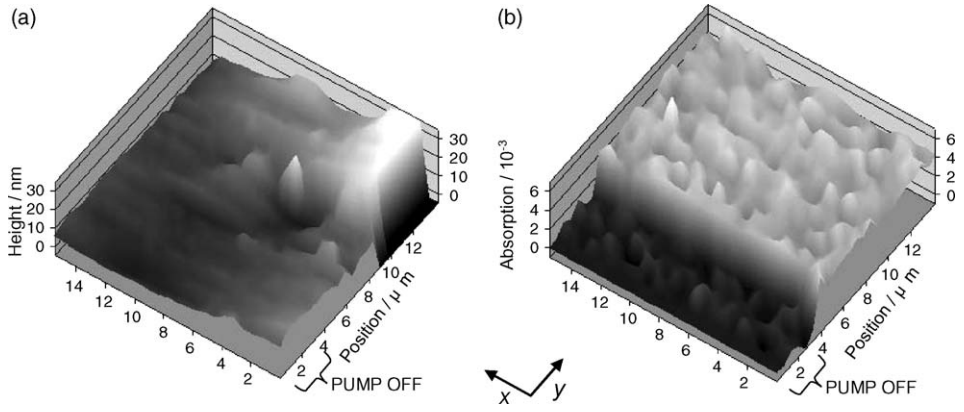

Fig. 6. AFM and transient absorption images of a perylene microcrystal. An area of 15 μm × 15 μm area (24 × 24 pixels) was scanned. The delay time was 12 ps, and the probe wavelength was 700 nm.

position C may arise from less efficient pump-light absorption due to different optical condition at the raised portion. Therefore, it is likely that exciton dynamics in the measured perylene crystal were homogeneous under measurement with lateral and axial resolutions of 1 μm.

Measurement of the transient absorption image will be a more efficient way to confirm the homogeneous nature of photogenerated excitons. At a fixed delay time, 12 ps, and at a fixed probe wavelength, 700 nm, a 15 μm × 15 μm area (24 × 24 pixels) was scanned to observe the exciton density in the crystal. If quasifree excitons are quenched by some reason, for example, effect of impurities or defects, weak transient absorption at 12 ps is expected to detect. AFM and transient absorption images were obtained (see Fig. 6a and b, respectively, for three-dimensional views). In the AFM image, near $(x, y) = (2, 12)$, there is a portion that is slightly (∼20 nm) higher than other positions, whereas in the transient absorption image the observed signals are homogeneous over the whole scanned area with an optical density of $(4.1 \pm 0.6) \times 10^{-3}$. Note that in the y range from 0 to 3 μm, the pump light was not incident to check the baseline of transient absorption measurement. From the experiments described above, exciton dynamics in the perylene crystal were clearly homogeneous, which is reasonable because the crystals used have high purity and high crystallinity [26].

It has been reported that when a perylene microcrystal is slightly damaged by irradiation of a high laser power, the decay dynamics changed slightly more rapidly than before the photodamage [10], indicating that some defects in the crystal affect exciton dynamics. In the present study using the apertured probe transient absorption microscopy, however, no position-dependent reaction dynamics were observed. To clarify the effect of defects in the crystal, improvement of the apparatus to obtain better spatial resolution (∼100 nm) and examination of sample-preparation dependence will be necessary. These studies are being planned, and the results will be published elsewhere.

4. Conclusion

Two types of femtosecond-laser scanning transient absorption microscopes have been developed as tools for observing ultrafast exciton dynamics in organic solids: one is the multiphoton excitation transient absorption microscope, and the other is the apertured probe transient absorption microscope. We evaluated their performance by measuring transient absorption spectra, kinetics and images of perylene crystals. In the former system, multiphoton excitation is induced by intense femtosecond-laser pulses at 800 nm, and a white-light continuum in the visible and near-IR wavelength region is used to monitor transient absorption at the excited point. Using 4× objective lenses, lateral resolution of about 10 μm and axial resolution of about 600 μm were estimated. The time resolution was about 300 fs. In the latter system, frequency-doubled femtosecond-laser pulses at 400 nm and white-light continuum pulses were introduced into a small aperture with ∼1 μm diameter as pump and probe lights, respectively. Relaxation dynamics of quasifree excitons to self-trapped excitons with a time constant of ∼2 ps was observed, and the exciton dynamics near the surface of the perylene crystal were found to be homogeneous.

Acknowledgements

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