Spatially Confined High-Temperature Blackbody Radiation from C_{60} Films

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Under green laser irradiation, intense white light emission was observed from C_{60} films into which Si thin layers were intercalated by sputtering. The emission spectra extending from the visible to mid-infrared region fit quite well with the Planck's formula for blackbody radiation, yielding surprisingly high blackbody temperatures of 1100−2140 K. Optical micrographs, AFM, microscopic FT-IR, and microscopic Raman measurements show that the emission comes from small domains scattered over C_{60} films that are morphologically and chemically modified to some extent by the combined effects of the sputtering process and laser irradiation.

1. Introduction

The optical properties of C_{60} have been attracting much attention because of its unique, spherical π-conjugated electrons. The photoluminescence of C_{60} extending from 1.4 to 1.8 eV is usually weak and dominated by the vibronic couplings with the higher energy levels which have appropriate symmetry.[1,2] Under high-intensity laser excitation, on the other hand, some groups reported that C_{60} powder or small crystallites emit strong and broad luminescence extending to the near-infrared region.[3,4] Although it seems to represent some intriguing, still unknown aspects in the optical properties of C_{60}, the phenomenon is not yet well understood mainly because important features such as the entire emission spectrum and spatial distribution remain to be revealed.

In the process of research aimed at developing new photonic composite materials comprising fullerenes and other materials, we found that C_{60}/Si layered films emit intense white light under green laser irradiation.[5] Also revealed was that the emission comes from modified solid C_{60} itself but not from Si or its specific interaction with C_{60}. In this paper we report that the spectra of the white light emission from the (modified) C_{60} almost perfectly fit the Planck's formula for blackbody radiation in the wavelength range 700−4500 nm, yielding surprisingly high blackbody temperatures of 1100−2140 K, although the samples were kept at far lower temperatures (23−300 K). To understand this unique emission behavior, attempts were made to analyze the emissive spots in the films by optical microscopy, AFM, microscopic FT-IR, and microscopic Raman measurements.

2. Experimental Section

Composite films were fabricated by alternate deposition of C_{60} and Si layers; the former by vapor deposition and the latter by radio frequency (rf) sputtering using a vacuum chamber in an Ar atmosphere of 10^{-3} torr. Fused quartz plates were used as substrates unless otherwise specified. The thickness of each layer was 30−50 nm for C_{60} and 0.2−0.6 nm for Si, and the deposition was repeated 30−100 times, resulting in a total film thickness of 1−5 μm.

For the measurement of light emission spectra, two detection systems were used: one for the visible and the other for the infrared regions. The excitation source was a CW Ar ion laser for both cases. The wavelength of the laser is 514.5 nm unless otherwise specified. For the former, the emission from the samples was passed through a monochromator and detected by a GaAs photomultiplier under the illumination density of 0.8−2.5 W/cm^2. For the latter, the emission from the samples was collimated with a CaF_2 lens and fed into a Fourier transform infrared (FT-IR) spectrophotometer equipped with a liquid nitrogen cooled InSb detector and the power density of Ar laser used was 4−14 W/cm^2. Spectral calibration of the detection systems was done using a tungsten−halogen standard lamp. The samples were kept in a vacuum chamber without cooling unless otherwise specified. For measurements done at low temperature, a vacuum cryostat was used.

For the analysis of the emissive spots in the composite films, microscopic FT-IR transmission spectra from the samples were measured in air with KBr plates used as substrates. The probed region was a 50 μm square. Microscopic Raman spectra were measured in a N_2 atmosphere by using the 488 nm line of an Ar ion laser whose beam size was 10 μm.

3. Results

The light emission varied tremendously depending on the position of the laser spot. When viewed by human eyes through laser protection glasses, some parts demonstrated very bright, white emission, while others emitted no appreciable light. In the latter case, only with a photomultiplier could we observe weak photoluminescence peaking around 730 nm as is usual for pristine solid C_{60}.[1] Figure 1 shows the visible spectra of the white light emission from a C_{60}/Si composite film at 20 K under four different excitation powers (0.8−2.5 W/cm^2), which are dominated by a broad, structureless feature. Peaks around 730 nm originating from the pristine C_{60} can still be recognized but are progressively buried under the broad feature as the excitation intensity increases. Moreover, if the sample was not cooled, these peaks around 730 nm disappeared while the broad emission remained strong. From the spectral shape shown in Figure 1, one sees that the measurement must be extended to the longer wavelength region for a better understanding of this peculiar phenomenon. By using a FT-IR spectrophotometer

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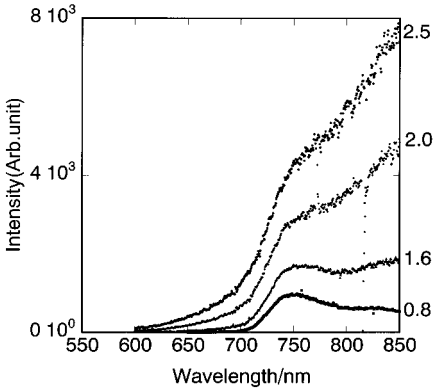

Figure 1. The white emission spectra of a C_{60}/Si composite film in the visible range at 20 K measured by a GaAs photomultiplier under the indicated excitation powers (0.8−2.5 W/cm^2). The peak around 730 nm is attributed to the pristine C_{60}.

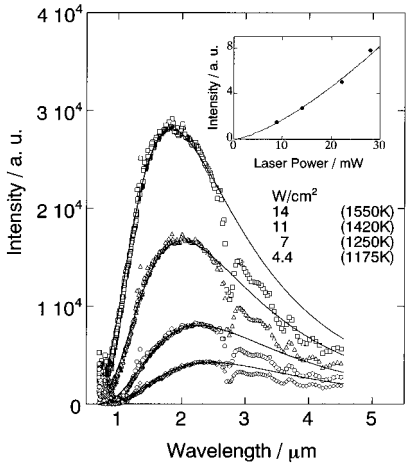

Figure 2. Emission spectra of a C_{60}/Si composite film on a fused quartz substrate at room temperature under Ar ion laser irradiation (514.5 nm) at four different power densities (4.4−14W/cm^2). Solid curves are the best fits based on the Planck's blackbody distribution, yielding the effective blackbody temperatures as indicated. The inset is the integrated emission intensity (on the basis of the best fit curves) as a function of the excitation power. The size of the laser beam was approximately 0.2 mm^2. Note that the structures observed at longer wavelengths (>2.6 μm) are due to the halogen lamp used for calibration.

equipped with an InSb detector, we obtained the whole spectra of the white light emission. Figure 2 shows the emission spectra under four different excitation powers (4.4−14 W/cm^2) measured at room temperature. The peak shifts to the shorter wavelength as the laser power increases. It is striking that the spectral shapes almost perfectly fit the Planck's formula for blackbody radiation as indicated by the solid lines, unequivocally demonstrating that the white light emission has a thermal origin. Spectral structures observed at longer wavelengths (>2.6 μm) are attributed to the tungsten−halogen standard lamp used for the calibration; it was confirmed by separate measurements that the lamp's quartz envelope inevitably absorbs light in this wavelength range due to −OH groups, etc. Curve fitting yielded the effective blackbody temperatures of 1175−1550 K depending on the excitation power of the laser (I_{in}). At such high temperatures in a vacuum, C_{60} would immediately evaporate;[6] nevertheless, the emission was stable at least over a few hours. As shown in the inset of Figure 2, the integrated intensity of the emission (on the basis of the fit curves over the wavelength range 0.7−10 μm) was found to be superlinear and approximately fit to the power of 1.4 of I_{in} for the excitation density 4.4−14 W/cm^2.

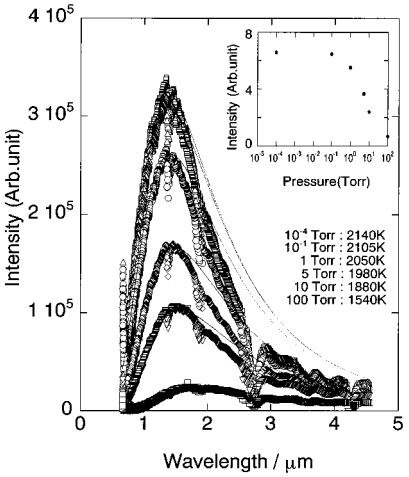

Figure 3. White light emission spectra of a C_{60}/Si composite film in different N_2 gas pressures ranging 10^{-4}−10^2 torr under Ar ion laser excitation (488 nm, 12.5 W/cm^2). Curve fitting yields the effective blackbody temperatures of 1540−2140 K. The inset shows the integrated emission intensity as a function of the N_2 gas pressure.

Figure 3 shows the white light emission measured in a N_2 gas atmosphere at room temperature under the excitation of an Ar laser operating at 488 nm with a power of 12.5 W/cm^2. Depending on the gas pressure ranging from 10^{-4} to 10^2 torr, the emission behavior changed drastically. Curve fitting based on Planck's formula yielded the effective blackbody temperatures of 1540−2140 K which are much higher than those shown in Figure 2. These high effective blackbody temperatures may be attributed to the increase in the absorption at 488 nm or the specific local condition of the sample. The inset of Figure 3 shows the dependence of the integrated emission intensity on the gas pressure. It can be seen that the emission intensity was almost constant for the pressure <0.1 torr but decreased dramatically in the range 10^{-1}−10^2 torr. When the sample chamber was evacuated again, the emission was restored, showing that the change is reversible and the N_2 gas enhances the heat dissipation from the hot spots involving no chemical reaction. This is in contrast with O_2 gas, the introduction of which was found to irreversibly weaken and quench the white light emission. It is conceivable that at such high temperatures carbon reacts with O_2 to produce CO_2.

To see the spatial detail of the light emission, we microscopically observed C_{60}/Si thin films under laser irradiation using a combination of an optical microscope (×5 objective lens), a CCD camera, and a monitor. A filter that rejects the laser line was placed in front of the objective lens. This system enables the real-time observation of the events that occur while the laser beam is scanned over the sample surface. Figure 4a shows a microphotographic view of a C_{60}/Si composite film, which is rather inhomogeneous and contains several cracks. It principally consists of dark islands and reddish background; the latter is shown as dark gray in the micrograph and the whitish parts represent the bare quartz surface. As might be expected from its appearance, microscope spectroscopic measurements showed that the dark islands transmitted no detectable light in the range of 300−900 nm, while the transmittance of the reddish background was almost the same as that of pristine C_{60}. It should be noted that, while the cracks were formed in the process of sample preparation, the dark islands were thought to be created by laser irradiation itself, because they could not be seen for nonirradiated samples. Indeed, monitoring by the CCD camera, we did observe that the darkening occurred almost instantaneously as the laser beam hit the area. An important

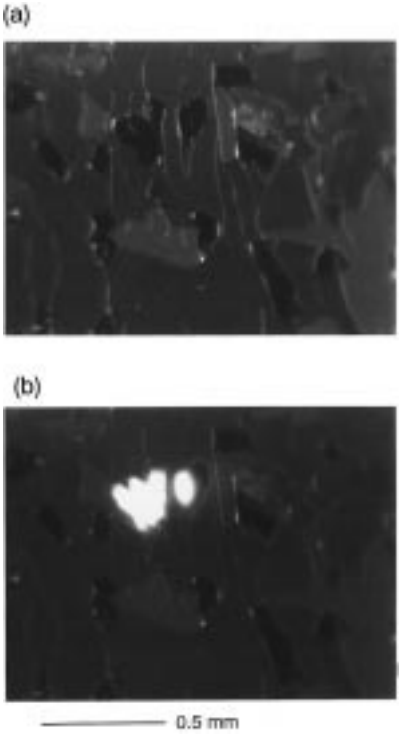

Figure 4. (a) Optical micrograph of a C_{60}/Si composite film showing inhomogeneous textures; (b) the same spot as (a) under laser irradiation (514.5 nm, 14W/cm^2) with a dark island emitting white light. Note that the dark island itself was formed by laser irradiation.

finding here is that the dark islands always emit strong white light under laser irradiation (the beam size was approximately 0.7×0.35 mm^2 and had an elliptical shape), while other parts remain nonemissive (see Figure 4b). The shining parts are clearly outlined against the background, meaning that the high temperature area is effectively confined in the islands and the heat does not seem to spread to the surroundings.

To see the role played by Si in the phenomenon, a C_{60}/TiO_2 composite film was prepared, TiO_2 layers rf sputtered in a similar way as Si. This fabricated composite demonstrated the same kind of white light emission, proving that the essential component for this phenomenon to occur is C_{60} itself but not the sputtered materials (Si or TiO_2). Moreover, the white light emission was never observed for a pure C_{60} film under the same excitation condition. For the as-deposited composite film, namely, before laser irradiation, macroscopic FT-IR and Raman spectra showed no characteristic peaks of amorphous carbon (a-C) or graphite, confirming that the C_{60} molecules were kept intact during the sample preparation. We suspect that the sputtering process in combination with the subsequent laser irradiation facilitates the formation of some appropriate modification of C_{60} films.

The microscopic texture of the films was examined by the measurement of AFM, showing that both dark islands and reddish backgrounds are made up of particles. However, as shown in Figure 5, their surface roughness was considerably different. The dark islands generally had a rougher surface than did the background. Such roughness may be indicative of porous structures formed by the partial evaporation of C_{60} under laser irradiation.

Microscopic FT-IR transmittance measurements were performed to identify the dark islands. The reddish background showed no obvious differences from pure C_{60} films (Figure 6a; note that the feature around 2300 cm^{-1} comes from CO_2 in the atmosphere). For the dark islands, on the other hand, broad

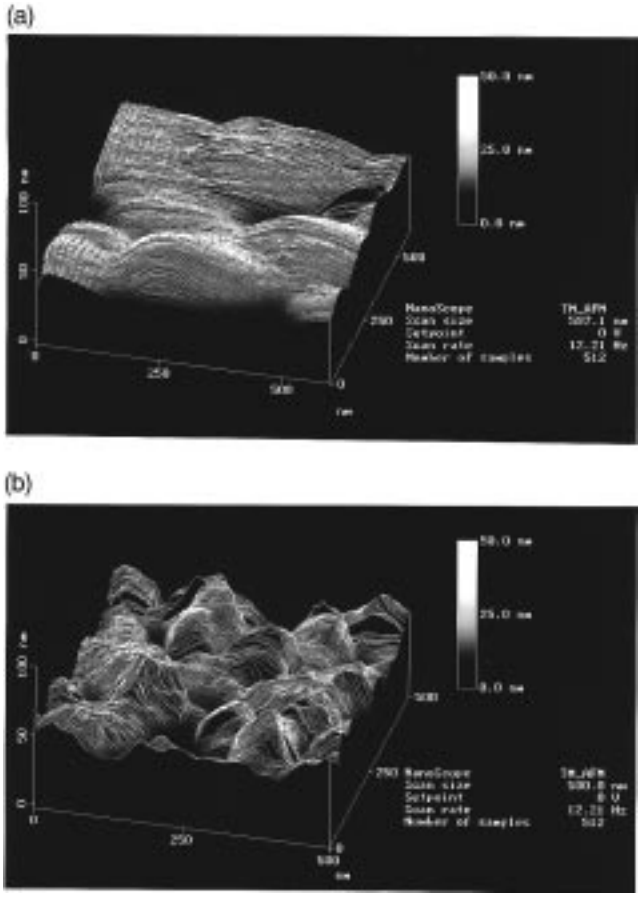

Figure 5. Typical surface images by AFM of (a) the reddish background and (b) a dark island.

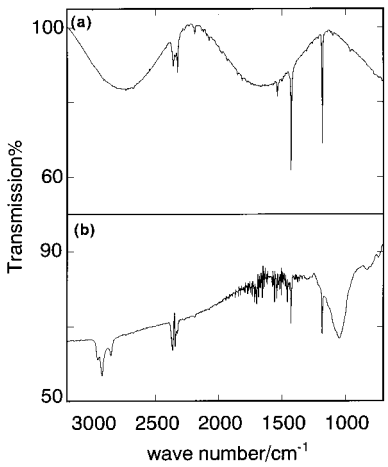

Figure 6. Microscopic FT-IR spectra of (a) the reddish background and (b) a dark island both in a C_{60}/Si composite film on a KBr substrate. The features around 1000 and 2900 cm^{-1} originate from oxygen and hydrogen-related modification of the C_{60} cage, respectively. The peaks around 2300−2400 cm^{-1} are due to CO_2 in the atmosphere and the fine structures in the region of 1500−2000 cm^{-1} are due to the vibration of OH groups in the adsorbed water.

bands around 1000 and 2900 cm^{-1} were observed in addition to the characteristic peaks of C_{60} at 1182 and 1428 cm^{-1} (Figure 6b). The 1000 cm^{-1} band could be attributed to CCO bending originating from oxidized C_{60} as reported for light-induced dicarbonyl C_{60} O_2.[7] In our case, however, there is not a strong signal due to CO stretching at about 1700 cm^{-1}. The peaks around 2900 cm^{-1} are attributable to CH stretching, although hydrogen was not introduced in our experiments; they should

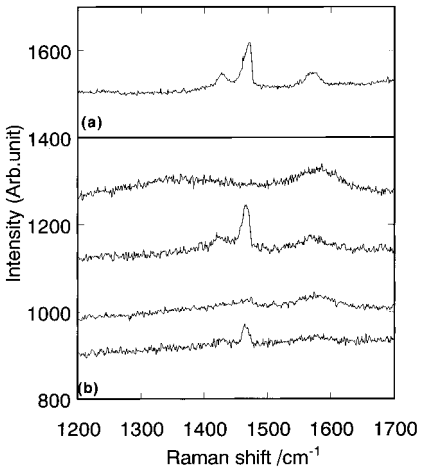

Figure 7. Microscopic Raman spectra of points (a) in the reddish background and (b) in a dark island.

be originated from water adsorbed in the sample or KBr substrate. Weak and sharp structures in the region of 1500−2000 cm^{-1} are attributed to the vibration of OH groups in the adsorbed water. While the results of the microscopic FT-IR measurements indicated partial modification of the C_{60} cage, no significant spectral features were detected around 1100−1700 cm^{-1}, at which wavenumber graphitic structures normally have strong and broad peaks. These are suggestive of minor graphitization, if any. More detailed analysis was performed by microscopic Raman spectroscopy.

As shown in Figure 7a, all the measured spots in the reddish background gave almost identical Raman spectra that indicated little difference from that of pure C_{60} thin films for which the peak at 1469 cm^{-1} is assigned to the pentagonal pinch mode of C_{60}. This is consistent with the results of the FT-IR measurements mentioned above. On the other hand, the Raman spectra of the dark island differed from spot to spot as shown in Figure 7b. While some spots showed nearly intact C_{60} spectra, others gave completely different features. Amorphous carbon made by the photothermal oxidation of fullerite was reported to have two major features; peaks at about 1600 cm^{-1} (G band) and 1400 cm^{-1} (D band).[8] As shown in Figure 7b, some of the spectra have broad peaks corresponding to G band and D band. The broadness of these bands implies the highly disordered structure of a-C. The difference of these spectra should reflect various degrees of carbonization. These observations demonstrate that the transformation of the C_{60} cage is quite inhomogeneous depending on local conditions such as the content of impurity, the morphology of a spot, etc. and that the transformed carbon is in a high degree of disorder. From the results of the Raman and FT-IR measurements, we can conclude that, in the black island which emits white light, some parts of C_{60} solids undergo oxidative transformation to amorphous carbon.

Knowing that the white light emission is almost unquestionably due to high-temperature blackbody radiation, we measured its dependence on the temperature at which a sample was set in a vacuum cryostat; the result was striking as shown in Figure 8. Contrary to what is expected for thermal radiation, the emission intensity increased considerably when the sample was cooled from 300 to 23 K. Moreover, the effective blackbody temperature as determined by the curve fitting to the Planck's formula increased by a few hundreds of degree, meaning that the temperature rose when the sample was cooled. It seems that cooling enhances the heat confinement and heat conduction across the border between the dark islands and the surroundings

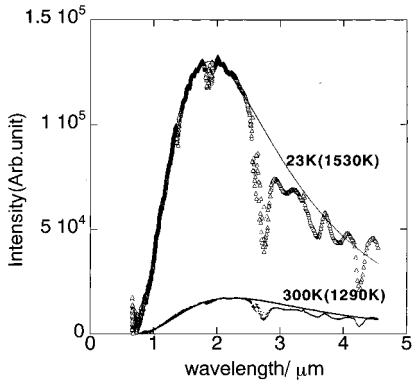

Figure 8. Emission spectra of the same spot of a C_{60}/Si composite film set at 23 K and 300 K under Ar ion laser excitation (514.5 nm, 14 W/cm^2). The effective blackbody temperatures obtained by curve fitting are indicated in the parentheses.

is strictly forbidden. While this unusual behavior is rather puzzling, possible mechanisms will be discussed in the following section.

4. Discussion

Because the white light emission conforms almost perfectly to Plank's theory for blackbody radiation, undoubtedly it has a thermal origin. The emission occurred only from those parts in the composite film that turned black immediately when the laser beam hit them. This phenomenon could be connected to the transformation of the solid C_{60} as indicated by the results of the microscopic FT-IR and Raman measurements (Figures 6 and 7). Manfredini et al. reported that solid C_{60} transformed to a-C under high power laser (514.5 nm) in the presence of oxygen and it depended strongly on the absorption and dissipation rate of the incident power, suggesting a thermochemical mechanism.[8,9] According to their results, if films were too thin, the reaction could not proceed because the amount of accumulated energy was insufficient. This appears to show some similarity to our finding that some thickness (>1 μm) was necessary for the darkening in the composite films to occur. However, our experimental conditions were drastically different from theirs (laser intensity 4.4−14 vs >4000 W/cm^2 and in a vacuum vs in air). They reported that, in Ar atmosphere, no transformation to a-C proceeded even under 4000 W/cm^2 of laser illumination. Moreover, while the transformation of their films took at least a few minutes, the darkening demonstrated in Figure 4 occurred in less than one second. These facts suggest that, although it seems to involve the same basic mechanism as theirs, the very fast and efficient transformation found here was made possible by the presence of some specific and localized morphology in solid C_{60}. Such morphology may be formed during the fabrication of the composite films, contributing to the substantial lowering of the threshold laser power for the transformation.

The optical absorption spectra of the C_{60}/Si composite film do not show an obviously enhanced absorption at 514.5 nm as compare with pure C_{60}, which is reasonable because only a small amount of Si was intercalated into the film. Therefore, Si itself seems not to play an important role in the formation of the black spots. Moreover, the use of TiO_2 instead of Si in the preparation of composite films also resulted in the white light emission under the same irradiation condition. Thus we suspect that the sputtering process itself facilitates the formation of some appropriate modification of the film's morphology. Observing the composite films microscopically, we found that they consist

of islands isolated from each other by cracks. These cracks were formed probably due to stress in the composite film as a result of Si (or TiO_2) intercalation. We consider that such cracking of the film will introduce some "premodification" such as microopening or porous structures, leading to symmetry lowering that enhances the optical absorption and to the prohibition of the heat conduction to the substrate and neighboring islands. Under laser irradiation, such premodified parts will be heated considerably and eventually undergo the transformation (darkening) as manifested in Figure 4b. The fact that no such transformation could be observed when irradiation was done in air demonstrates the importance of gas pressure in the microopening for the heat dissipation between the island and surroundings.

Our results of the FT-IR measurement suggest the involvement of oxygen and hydrogen during the modification of C_{60} cage, although the sample preparation and the laser irradiation were all performed in inert atmosphere (in Ar or vacuum). Possibly ambient water and oxygen absorbed in samples during their transfer or storage are involved in the laser-induced reactions. This seems to be consistent with the results by Manfredini et al. that oxygen was necessary for the transformation of solid C_{60}.[8,9] However, our results indicate that even a small amount of oxygen was enough for the oxidative transformation of C_{60}. This suggests that the premodification of our sample together with the vacuum condition brought about a much higher temperature under the laser excitation, resulting in the transformation even if no oxygen was supplied intentionally.

As discussed above, the pre-modified parts of the sample underwent the transformation of C_{60} under laser excitation. Judging from the microscopic Raman and FT-IR spectra the transformed spots should be a mixed phase of C_{60} and a-C. Combining these results with the AFM image (Figure 5b), as the entity of the transformation, we propose carbonaceous shell structures entrapping C_{60} molecules. As an example of such shell structures, we might think of a carbon aerogel,[10] a highly porous and thermally insulating phase of a-C. Such a picture may explain why the tremendous thermal nonequilibrium is retained between the emissive spots and the surrounding. At such high temperatures as 1200−2140 K, intact C_{60} molecules should be vaporized. However, they will not disappear because they are confined in the shell structures. It is interesting to note that gas-phase C_{60} desorbed by giant laser pulses emitted white light that could be well described by blackbody radiation with effective temperature of 2300−3000 K.[11]

The temperature of the emissive spots has been shown to be affected by several factors such as illumination intensity, the ambient pressure, and the temperature of the substrate. We note that there is one energy input and two energy outputs for the emissive islands to be considerd here. The energy input is the absorption of the laser excitation which is expected to be proportional to the laser intensity. The energy outputs are the heat dissipation to the surroundings through the solid or by the ambient gas and the heat dissipation by the blackbody radiation. In the steady state, the energy absorbed from the laser must be equal to the sum of the heat dissipation by heat conduction and radiation. It is known that the heat transfer (Q) between the hot spot and the ambient can be expressed by the heat transfer coefficient (h) and the temperature difference (δT) between the hot spot and the ambient as $Q = h(\delta T)$. Here, the heat transfer between the islands and the surroundings is considered in two ways. One is the heat conduction through the solid such as the transformed material or substrate. The other is the heat conduction through the ambient gas on the surface of the islands or in the microopening and porous structures. The heat transfer coefficient for the former is determined by the thickness and thermal conductivity of those solids. Similarly, that of the latter is determined by the size of the openings and the thermal conductivity of the gas while in the high-pressure range. In both cases the heat transfer coefficient is independent of the gas pressure. However, when the pressure is low enough and the gas opening or thickness is much smaller than the mean free path of the ambient gas, the heat transfer coefficient of the gas is no longer dependent on the size of the openings and could be expressed by the free molecular heat transfer coefficient which is dependent linearly on the ambient gas pressure.[12,13]

With the picture described above we can understand the dependence of the white light emission on the laser intensity. If the temperature of the hot spot is high enough and the radiation is the main process to lose heat, the light emission will increase linearly with the excitation power. On the other hand, if another dissipation (i.e., heat conduction) is important the dependence of the blackbody radiation on excitation intensity should be superlinear. The inset of Figure 2 shows somewhat superlinear dependence of white light emission on the illumination density, implying still nonnegligible heat conduction with the surroundings. In the low-pressure condition ($<10^{-3}$ torr) as employed in most of the present experiments, only the heat conduction through the solid porous structures should be important, because the dissipation through ambient gas can be neglected. The superlinear dependence is derived from such a situation.

As the gas pressure increases, the thermal conduction through the ambient gas becomes nonnegligible as compared with other heat dissipation processes. The emission intensity decreased substantially when the pressure rose from 10^{-1} to 100 torr (Figure 3). The curve seems to level off above 100 torr, probably corresponding to the fact that the mean free path of the N_2 molecules has decreased and become smaller than the size of the gaps separating hot spots and the surroundings. From these results, we can estimate the size of the micropores surrounding the hot spots at the order of micrometers. Indeed, gaps of this size separating the hot spots from the surroundings could be seen by SEM.

The increase in the effective blackbody temperature and radiation intensity that accompanied the cooling of the sample seems still puzzling. To understand this seemingly strange behavior, one has to invoke the decrease in thermal conductivity of the sample or its surroundings accompanying the cooling. However, C_{60} solid showed very little change in thermal conductivity in the temperature range 23−300 K.[14] In addition, those of Si and SiO_2, both used as substrate materials in the present study and giving similar results, have opposite dependence on temperature, excluding the involvement of the heat conduction through substrates. We also attempted to explain the results by invoking the cryopumping effect, where the cooling leads to the decrease in the pressure of the residual gas surrounding the sample, making the heat confinement more effective. However, as shown in Figure 3, the range of the gas pressure for which the emission intensity most drastically changed was 10^{-1}−100 torr. This is completely different from the change of the gas pressure induced by the operation of the cryostat, which turned out to be 10^{-3} torr at 300 K and 10^{-6} torr at 23 K, discarding this explanation. The only possibility left is the decrease in thermal conductivity of a certain carbon phase connecting the hot spots and cooled parts. Assuming that the heat conduction through such phase is suppressed as the temperature is lowered, we would be able to explain the

observation. It may be related to the fact that graphite shows considerable decrease in thermal conductivity in the temperature range 100−20 K. However, since the connecting carbon phase presumed here has not been characterized yet, we still remain at a speculative stage about its temperature dependence.

Next, we discuss the relationship of our results with the previously reported (so called) white photoluminescence from solid C_{60}.[3,4] Their results, which were obtained on pristine solid C_{60} (powder or microcrystals), were characterized by strong nonlinear dependence on the excitation intensity, while our results on modified C_{60} indicated only slight nonlinearity (the inset of Figure 2). These different sample conditions and contradictory results might lead to a presumption that our white light emission is different in nature from theirs.

However, we suspect that the discrepancy in the dependence on the excitation intensity can be reconciled by examining the method for analyzing the light emission spectra data. As demonstrated in Figure 2, the increase in the excitation intensity causes a strong blue-shift of the emission peak. Therefore, if a wavelength range covered by a detection system used is limited to some narrow width in the shorter wavelength region as compared with the entire emission range, a seemingly highly superlinear dependence may arise, while the integrated intensity over the entire wavelength range should show less nonlinear dependence as substantiated by the inset of Figure 2. Indeed, our preliminary measurements that used a photomultiplier with a sharp cut off at 850 nm, thus practically limiting the measurement range to 600−850 nm, falsely showed a superlinear dependence that was proportional to the power of 2.8 of the excitation intensity. These considerations suggest that, if the measurements, calibration, and integration of the emission spectra are properly performed, all the white light emission from solid C_{60} reported so far will conform to the Planck distribution for blackbody radiation, that is, confirming that the emission is of a thermal origin.

The difference in the sample condition (pristine vs modified C_{60}) may not be so difficult to reconcile. Even if no intentional treatment is done on solid C_{60}, it is quite probable that it contains some defects or sites with lowered symmetry as compared with the perfect crystalline lattice. We can expect that such a modified structure in pristine solid C_{60} will work in a similar way as do the defects introduced by sputtering in our experiments. In fact, the white light emission from pristine microcrystalline C_{60} was observed to be inhomogeneous,[15] being consistent with the importance of defective structures. Our results and their interpretation are in one aspect supported by a recent work by Palstra et al. that reported white light emission from C_{60} single crystals induced by electric current; by the measurements of microscopic Raman spectra, they revealed a-C like structures in emissive spots in their samples.[16]

5. Conclusion

We have observed intense white light emission from C_{60}-based composite thin films under green laser irradiation and revealed its entire emission spectra in the visible to infrared region. The spectral shape that conforms excellently to Planck's formula has established that it originates from high-temperature blackbody radiation with effective temperatures of 1100−2140 K, depending on the excitation intensity and sample conditions. Such high-temperature is shown to be confined in the dark islands, which are formed by the laser irradiation of the premodified parts in the composite films. The premodification seems to be induced by the intercalation of Si or TiO_2 into C_{60} films by sputtering. On the basis of the measurements of AFM, microscopic FT-IR, and Raman spectra, we propose that the dark islands consist of mixture of C_{60} and a-C, the latter forming the carbonaceous shell structures entrapping C_{60} molecules. We suggest that the previously reported white light emission[3,4,16] from fullerite under intense laser irradiation have essentially the same origin as that found in this work.

From the present results, the approach adopted here, that is, rational modification of fullerene thin films, shows promise as a new route toward the development of optically functional materials, because neither intact C_{60} nor a-C solid itself seems to show such intense white light emission.

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