Optimization of a laser-plasma x-ray source for contact x-ray microscopy

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Presented are the detailed characteristics of a laser-plasma x-ray source that is successfully used for producing x-ray images of biological specimens in water by contact microscopy. The conversion efficiencies of laser energy to soft x rays are measured for various target materials using a 0.53 μm, 500 ps laser pulse. Yttrium target showed the highest conversion efficiency of around 2.5%/sr to the “water window” region x rays at 3×10^12 W/cm^2. Angular distributions of x rays and ions are studied to find the best angular position of specimen for reducing the target debris. The effective x-ray spectrum contributing to making an image of a specimen in water is discussed. It is shown that an extremely small laser could be employed in high-resolution contact microscopy.

I. INTRODUCTION

A laser-produced plasma can emit strong x rays with high conversion efficiency. Some applications of the laser-produced plasma x-ray source have been reported in the fields of microscopy, lithography, and so on. We studied laser-produced plasma extensively as an x-ray source for contact x-ray microscopy. X-ray microscopy has the potential of observing living specimens in water at a high spatial resolution. For the observation of biological specimens in water, the wavelength of x rays must be within the so-called “water window” region (λ = 2.33–4.37 nm). For high-resolution imaging with a high signal-to-noise ratio, quite high flux exposure is required. Furthermore, extremely short duration exposure is essentially important for high-resolution x-ray imaging because of the thermal expansion of the specimen heated by the x-ray absorption. Allowable x-ray exposure time is considered to be shorter than 1 ns to observe specimens at a high resolution by x-ray microscopy. From these points of view, a laser-produced plasma can be the most suitable flash x-ray source for x-ray microscopy.

We have already reported the observation of a sperm of a sea urchin in water by contact x-ray microscopy using a laser plasma x-ray source. In this article, we report the study on laser-plasma x-ray source performed prior to the work reported in Ref. 5. Detailed characteristics of the x-ray emissions from laser plasmas, which allowed us to choose the target material, are reported. Angular distributions of x rays and ions, which suggested to us the best angular position of specimen, are also described. In Sec. IV, how the x rays outside the water window region affect the x-ray imaging, and what are the optimum laser irradiation conditions are discussed.

II. EXPERIMENTAL PROCEDURE

In this section we describe the experimental configuration for spectral measurement, how the absolute x-ray conversion efficiency was estimated, and how the dissolution rate of poly(methyl methacrylate) (PMMA) resist was calibrated.

The experimental arrangement is shown in Fig. 1. The second harmonic of the Nd:glass laser (λ = 0.53 μm) was focused on a flat target by an aspherical lens (f/7, focal length of 500 mm) under vacuum of lower than 0.1 Torr. The beam diameter was about 70 mm and the beam divergence was better than 0.1 mrad. The pulse width was 500 ps. The energy of the frequency-doubled pulse was about 8 J at the maximum. The x-ray image of the plasma was observed by an x-ray pinhole camera with a commercial charge-coupled device (CCD) camera as a detector placed at 45° from target normal. X rays were spectrally resolved by a transmission grating (of gold substrate of 0.35 μm thick, 1000 lines/mm, Heidenhain) placed at 45° from the target normal and recorded on an x-ray film (Kodak 101-07). Spectral resolution with a slit of 50 μm width was limited by the plasma size of 50 μm and was 0.5 nm. The laser power density on target was changed by changing the laser energy fixing the focal spot diameter to 50 μm for power density higher than 10^12 W/cm^2. For power density lower than 10^12 W/cm^2, the focal spot size was increased.

To estimate the absolute x-ray conversion efficiency, we need to know the absolute x-ray intensity on film and the diffraction efficiency of the transmission grating. The absolute x-ray intensity on film was estimated by using the film sensitivity reported by Henke, Fujiwara, and Tester. Films were developed under the same procedure as in Ref. 7 and the temperature of all solutions was controlled at 20±1°C.

The theoretical value of the diffraction efficiency of the transmission grating to the first order on one side is 10%–11% in the range of 1.3–7 μm wavelength, when the transmission of supporting bars is neglected. Because some possible defects of the grating could change the diffraction efficiency, we estimated the diffraction efficiency experimentally as follows. The diffraction efficiency was evaluated from the ratio of the number of photons into the first order to the total number of photons on the film. X-rays in this experiment must be monochromatic or quasi-monochromatic. As shown in Fig. 2, an Fe plasma emits x rays...
at 1.3 nm with a sharp profile and at 6 nm with a broad profile. This spectrum was obtained at the laser power density of $7 \times 10^{13}$ W/cm$^2$. If x rays at 6 nm can be eliminated, x rays at 1.3 nm from an Fe plasma can be a good quasimonochromatx-ray source for the calibration of diffraction efficiency. If zeroth order of the spectrum contains radiations of other wavelengths, the estimated diffraction efficiency will be inaccurate. Because the resolution of the spectrometer was 0.5 nm, x rays shorter than 0.5 nm can be included in the zeroth-order spectrum. However, it is quite implausible in our case, because the power density of $7 \times 10^{13}$ W/cm$^2$ is low to generate such-short wavelength x rays. X-rays at 6 nm and longer x rays were eliminated by the use of a Be filter 3.5 $\mu$m thick. Hence, only the x ray at 1.3 nm can reach the film. Thus, the evaluated diffraction efficiency into the first order on one side was $\sim 6\%$. The transmission of supporting bars of the grating was 65% which was observed under an optical microscope. Thus, our grating was found to have a diffraction efficiency quite close to the theoretical value.

A laser-produced plasma emits not only x rays but also debris. The debris can damage the resist that records an image. To find the optimum angular position to place the specimen where the amount of debris is as small as possible but with sufficient x-ray flux, we have to know the angular distributions of x rays and debris from a laser-produced plasma. We used a PMMA resist for the measurement of x-ray angular distribution. PMMA does not have a sensitivity to visible light and can be treated under the light and many resists of small pieces can be placed at the same time. When the dissolution rate of the resist is known as a function of x-ray flux, x-ray flux on resist can be obtained from the dissolution rate. The relation of the dissolution rate to the x-ray flux was experimentally calibrated as follows. PMMA resists were located at various distances from a target at 67.5° from the target normal. They were exposed to 2.8 nm x rays in the water window region, which was emitted from a Y plasma. Besides 2.8 nm x rays, an yttrium plasma emits 9 nm x rays and longer wavelength radiations (as shown later in Fig. 6). However, we can eliminate the effect of those radiations of 9 to 100 nm wavelength by the use of 0.1 $\mu$m thick Si$_3$N$_4$ membrane in front of the resist. A PMMA resist covered by a 6-mm-thick MgF$_2$ crystal, which blocks x rays but is transparent to the radiation longer than 120 nm, did not dissolve after the laser shot. Thus, that the effect of radiations longer than 120 nm is negligible to the dissolution of a PMMA resist was confirmed. The x-ray flux on the resist was estimated using the x-ray conversion efficiency observed in this experiment which will be described later. The observed dissolution rate is shown in Fig. 3. The relation between

![FIG. 1. Experimental setup for spectral measurement.](image1)

![FIG. 2. X-ray spectrum from an Fe plasma.](image2)

![FIG. 3. Dissolution rate $R$ (nm/min) of a PMMA resist for 2.8 nm x rays as a function of x-ray flux $E$ (mJ/cm$^2$) on resist.](image3)
the dissolution rate \( R \) (nm/min) and the 2.8 nm x-ray flux on the resist \( E \) (J/cm\(^2\)) was \( R \propto E^{1.5} \).

As seen in Fig. 3, x-ray flux in our experiment was 20 mJ/cm\(^2\) at the maximum. This value looks like quite low compared to the exposure of around 1 J/cm\(^2\) for the proximity x-ray lithography using 0.8 nm x rays. However, the dissolution rate of resist should be a function of the absorbed energy per unit volume, and the dissolution rate is expected to be independent of x-ray wavelength if the absorbed energy density per unit volume is the same. 1 J/cm\(^2\) flux of 0.8 nm x rays corresponds to 1 kJ/cm\(^3\). The upper scale in Fig. 3 shows the absorbed energy per unit volume for 2.8 nm x rays. 50 mJ/cm\(^2\) of 2.8 nm x-ray flux corresponds to about 1 kJ/cm\(^3\). 0.8 nm x rays from a synchrotron source is reported to give the same dissolution rate shown in Fig. 3 when plotted as a function of absorbed energy per unit volume. Thus, we know the experiment was done at the energy density similar to the best condition in x-ray lithography. This big difference of flux for 2.8 and 0.8 nm comes from the big difference in the absorption coefficients.

### III. RESULTS

It is known that the wavelength of x rays, the conversion efficiency to x rays, and the spectral profile depends strongly on target materials and on the irradiating laser intensity. However, there are not so many studies on laser-plasma x-ray source for the application to contact x-ray microscopy. To find the best target material, irradiation conditions for contact x-ray microscopy, we did a detailed study on the laser-plasma x-ray source.

In contact x-ray microscopy, the x-ray source needs to satisfy the following conditions.

(i) To take clear images of living specimens, x rays should be in the so-called water window region \([\lambda=2.33 \text{ (oxygen K edge)}-4.37 \text{ nm (carbon K edge)}]\) to produce a high contrast of carbon absorption against oxygen. X rays outside the water window region reduce the contrast of carbon against oxygen.

(ii) To make an x-ray microscopy system small, the x-ray conversion efficiency should be as high as possible.

(iii) A laser-produced plasma emits not only x rays but also debris from a target. To protect the resist from the damage by debris, the sample (or resist) must be placed at the position where the amount of the debris is small and x rays are sufficiently strong.

To find a suitable target material for contact x-ray microscopy, x-ray spectral profiles and x-ray conversion efficiencies of various target materials were observed. The results are described in Sec. III A. To find the optimum angular position of a specimen, angular distributions of x rays and debris were observed. The results are described in Sec. III B.

#### A. Spectra of x rays emitted from various target materials

To understand detailed characteristics of the laser-plasma x-ray source, spectra of x rays from various target materials (C, F, Si, Sc, Ti, Fe, Cu, Zn, Y, Mo, and Ta, \( Z=6-73 \)) were observed. The laser intensity in this experiment was \( 10^{12}-10^{13} \) W/cm\(^2\), since the conversion efficiency to x rays in the water window region was high at these laser intensities, as described later in this subsection.

All materials emitted a few band spectra, as typically shown in Fig. 2. The spectral resolution was 0.5 nm. According to Ref. 11, Fe\(^{18+} \) ion emits 20–30 strong lines in the range of 1–1.5 nm caused by transitions of 3s-2p, 3d-2p, and 4d-2p. Fe\(^{15+} \)-Fe\(^{22+} \) ions also emit strong lines between 1 and 1.5 nm wavelength. Therefore, the x-ray band at 1.3 nm in Fig. 2 is considered to consist of many bright lines of a quite narrow bandwidth emitted from various kinds of Fe ions. All emissions are caused by transitions to the L shell having the principal quantum number \( n=2 \). Thus, we call this band emission the “L-shell emission.” X rays at 6 nm consist of line emissions caused by the transitions to the M shell \( (n=3) \). Therefore, we call this band emission the “M-shell emission.” According to Ref. 11, the average wavelength of x rays is shorter for an ion of higher ionization stage. Yttrium emitted M-shell emission at 2.8 nm. The peak wavelength of 2.8 nm was observed to shift to shorter wavelength slightly (a few tenths of nm), when the laser intensity was increased in the range of \( 10^{12}-10^{13} \) W/cm\(^2\). This was probably caused by the increase of the average charge number of ions with increasing laser intensity.

The observed peak wavelengths of band emissions from various target materials are plotted in Fig. 4. The peak wavelength \( \lambda \) decreases with the atomic number, \( Z \), of the target material as \( \lambda \propto Z^{-2} \). From this relation, we can guess peak wavelengths of band emissions of all materials, although we studied only 12 materials. This relation is the same as the Moseley’s law, and is also reported in previous works on laser plasma. From Fig. 4, we can say that the water window x ray can be generated by C \( (Z=6) \) and N \( (Z=7) \) plasmas for K-shell emission, P-Ca \( (Z=15-20) \)
for L-shell emission, Ga–Mo (Z=31–40) for M-shell emission, and Ce–Re (Z=55–75) for N-shell emission.

Next, we discuss the difference of the characteristics of each shell emission. First, the brightness at the peak wavelength of each band emission is compared. We take 0.5 nm as the unit bandwidth, because our spectral resolution was 0.5 nm. Figure 5 shows the x-ray energy into the 0.5 nm bandwidth normalized by the laser energy. In Fig. 5, solid lines are for L-shell emissions and dashed lines for M-shell emissions. Among L-shell emissions, the laser intensity giving the maximum value was higher for shorter-wavelength x rays, as is expected. The x-ray energy into the 0.5 nm bandwidth normalized by the laser energy was larger for L-shell emissions than for M-shell emissions, as seen in Fig. 5. Both shell emissions consist of many lines. However, a larger number of lines is expected for the emission to the M shell which has a larger principal quantum number than the L shell. Therefore, the ratio of the brightness of each single line in the L-shell emission to that of M-shell emission will be larger than seen in Fig. 5.

Next, we compare the bandwidth of L-shell and M-shell emissions by taking Sc and Y as examples, because both materials emit emissions of nearly the same peak wavelength. The bandwidth of M-shell emission (3 nm) was four times that of L-shell emission (0.8 nm).

When the emitted x-ray energy integrated over the whole band emission is compared, it was larger for M shell.

From this difference of L-shell and M-shell emissions, we can expect that K-shell emission will consist of fewer bright lines in a narrow bandwidth with less total energy, and for N-shell emission, weaker lines in a broader bandwidth with larger total energy.

We now know the general characteristics of each shell emission. Next, we discuss which shell emission is more suitable as an x-ray source for contact x-ray microscopy. In the water window region, by taking Cl, Y, and Ta as examples, the bandwidths of L (Cl), M (Y), and N (Ta) -shell emissions were 1, 3, and 6 nm, respectively, as shown in Fig. 6. In contact x-ray microscopy, the x-ray energy integrated over the whole water window region should be large. Because the bandwidth of M-shell emission is closer to the bandwidth of the water window region, M-shell emission will be more suitable than other band emissions. A quantitative comparison is shown in Fig. 7 which shows the conversion efficiency only into the water window region for Cl, Y, and Ta plasmas. The conversion efficiency for M-shell (Y) emission was 2.5%/sr at $3 \times 10^{12}$ W/cm$^2$ which was twice of that for N-shell (Ta) emission.

Thus, Ga–Mo (Z=31–40) which have M-shell emissions in the water window region will be the best target materials for contact x-ray microscopy.
From the angular distribution of x rays and ions shown in Fig. 8, damage caused by debris will be greatly reduced without sacrificing the x-ray flux so seriously by placing a sample at an angle of about 60°–70° from the target normal.

IV. DISCUSSION

From these experimental results, we now know that, for contact x-ray microscopy, one of the best target materials is yttrium, the laser intensity giving the maximum conversion efficiency (2.5%/sr) into x rays within the water window region is about 3×10^{12} W/cm², and the optimum angular position of a sample is about 60°–70° from the target normal.

In contact x-ray microscopy, specimens floating in water will be held between a Si₃N₄ membrane as an x-ray window and a PMMA resist as a recording medium, as reported in Ref. 5. Separation between the Si₃N₄ membrane and the PMMA resist will be less than a few μm. X rays reach a PMMA resist after penetrating through the Si₃N₄ membrane and water. Therefore, the spectral distribution of x rays reaching the resist can be quite different from that of x rays emitted from a plasma. The calculated “effective” x-ray spectrum contributing to make an image is described in Sec. IV A.

The finite size of a plasma causes penumbral blurring, which limits spatial resolution in contact x-ray microscopy. When the penumbral blurring is taken into consideration, the optimum laser intensity might be different from that for maximizing x-ray conversion efficiency. The optimum irradiation condition to minimize the penumbral blurring and the possibility of miniaturizing a contact x-ray microscopy system are discussed in Sec. IV B.

A. Effective x-ray spectrum in contact x-ray microscopy

X rays inside the water window region contribute to make high-contrast images. X rays outside the water window region deteriorate the contrast of the image of living cells, because the difference between the absorption coefficients of carbon and oxygen is quite small for these x rays. Moreover, radiations of wavelength longer than carbon K edge produce larger diffraction blurring and worsen the spatial resolution. Here, we discuss the effective x-ray spectrum contributing to imaging in contact x-ray microscopy, and the influence of radiations outside the water window region to an x-ray image.

In contact x-ray microscopy, x rays reach onto the resist after penetrating through a Si₃N₄ membrane x-ray window and water in which a specimen is floating. Thus, a modified x-ray spectrum reaching the resist is shown by the broken line in Fig. 9, in which the observed spectrum is shown by the thin solid line. In the calculation, the thick-

B. Angular distribution of x rays and ions

A laser-produced plasma emits x rays and debris, and debris could present a serious problem. To avoid the damage of the recording medium of x-ray images, it should be placed where the amount of debris is quite small. However, at that position, there should be plenty of x-ray flux to record clear x-ray images. To find the optimized position, we need to know angular distributions of x rays and ions.

First, we studied the angular distribution of the water window x-ray using a PMMA resist. PMMA resists were placed at various angles, and x-ray fluxes on resists were estimated from the dissolution rate of resists. The relation of the dissolution rate against the x-ray flux, as shown in Fig. 3, was obtained as described in Sec. II. The observed angular distribution of x-ray emission is shown in Fig. 8, normalized to unity at 67.5°. When the x-ray angular distribution was fitted by a function of (cos θ)^n, where θ is the angle from the target normal, good fitting was obtained for n = 1. Similar angular dependence is reported in earlier works.10,14 (n = 0.95 in Ref. 10 and n = 1 in Ref. 14). Figure 8 shows that the x-ray angular distribution varies only slightly from 0° to 70°, and decreases rapidly at greater than about 70°.

Next, we must measure the angular distribution of debris. Debris consists of ions and neutral particles. It is difficult to detect neutral particles. Because of the ease of detection, the ion angular distribution from a plasma was observed using charge collectors,15 expecting a similar distribution for neutral particles. The charge collectors were located at 20°, 35°, 47°, and 74° from the target normal. The broken line in Fig. 8 shows the angular distribution of ions from a copper plasma for the spot size of 70 μm, and the dashed-dotted line for the case of 250-μm-diam spot size. The signals are normalized to unity at 20°.

Fitting a function (cos θ)^n to the angular distribution of ions gives n = 8 for the spot size of 250 μm diameter (dashed-dotted line), and n = 4 for 70 μm diameter (broken line). The experimental result shown in Fig. 8 reveals that the ion emission has a sharper directivity to target normal.

FIG. 8. Angular distribution of x rays and ions. The x-ray flux is normalized to unity at 67.5°. Ion currents are normalized to unity at 20°. The broken curve shows the angular distribution for the spot diameter of 70 μm, and the dashed-dotted curve shows the angular distribution for the spot diameter of 250 μm.

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The finite size of a plasma causes penumbral blurring, which limits spatial resolution in contact x-ray microscopy. When the penumbral blurring is taken into consideration, the optimum laser intensity might be different from that for maximizing x-ray conversion efficiency. The optimum irradiation condition to minimize the penumbral blurring and the possibility of miniaturizing a contact x-ray microscopy system are discussed in Sec. IV B.

A. Effective x-ray spectrum in contact x-ray microscopy

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In contact x-ray microscopy, x rays reach onto the resist after penetrating through a Si₃N₄ membrane x-ray window and water in which a specimen is floating. Thus, a modified x-ray spectrum reaching the resist is shown by the broken line in Fig. 9, in which the observed spectrum is shown by the thin solid line. In the calculation, the thick-
FIG. 9. Effective x-ray spectra contributing to making x-ray images. The thin solid line is the experimentally observed x-ray spectrum from a yttrium plasma. The broken line is the calculated spectrum of x rays passing through a Si₃N₄ membrane and 1 μm water layer. The solid line is the spectrum contributing to x-ray imaging when the sensitivity spectrum of a PMMA resist is considered.

ness of the Si₃N₄ membrane is 0.1 μm, and the thickness of the water layer is 1 μm.

In the wavelength region longer than the oxygen K edge, longer-wavelength x rays are much attenuated by the absorption of water and Si₃N₄ membrane. The calculated spectrum is shown by the broken line. More important is the effective spectrum after taking into account the resist sensitivity. Just outside the carbon K edge, the sensitivity of the PMMA resist is quite low. The calculated effective spectrum is shown by the solid line. The contribution of long-wavelength x rays outside the water window region is negligibly small owing to the low sensitivity of resist and to the absorption by the x-ray window and water.

Next, we consider the effect of shorter-wavelength x rays. As shown by the solid line, there remains a finite effect of short-wavelength x rays on the resist dissolution, contrary to the case of long-wavelength x rays. Therefore, to have clear x-ray images of living cells, short-wavelength x rays should be reduced as much as possible. From this point of view, a Ta plasma, which emits a larger amount of shorter-wavelength x rays than Y plasma, might produce images of living cells with slightly lowered contrast.

We also notice that the peak x-ray wavelength of the effective spectrum changes slightly depending on the thickness of the water layer. It is 2.3 nm for a 5-μm-thick water layer and 3.1 nm for a 1-μm-thick water layer.

B. Optimum irradiation condition

We discuss here the optimum irradiation condition to minimize the penumbral blurring and possibility of realizing a table-top contact x-ray microscopy system.

Resolution of contact x-ray microscopy can be limited by (i) diffraction blurring [Fig. 10(a)], (ii) penumbral blurring [Fig. 10(b)], and (iii) other factors including the resolution of the resist. Here, we discuss the optimum irradiation condition. The diffraction blurring δ₁ and the penumbral blurring δₚ are given by

\[ \delta_1 = \frac{\sqrt{\hbar \lambda}}{d}, \]
\[ \delta_p = \frac{D h}{d}, \]

where \( h \) is the distance between the specimen and the resist, \( \lambda \) is the x-ray wavelength, \( D \) is the diameter of the plasma, and \( d \) is the distance between the plasma and the specimen, with usually \( d \gg h \). The laser energy \( E_L \) is given by

\[ E_L = \frac{\rho_E}{\mu \eta(I)} d^2, \]

where \( \rho_E \) (J/cm³) is the energy density on resist, \( \mu \) is the linear absorption coefficient (cm⁻¹) of the resist, and \( \eta(I) \) is the x-ray conversion efficiency at the laser intensity \( I \) (W/cm²). Assuming that the plasma diameter is the same as the laser spot diameter on target, the penumbral blurring \( \delta_p \) is given by

\[ \delta_p = 2h \left( \frac{\rho_E}{\mu \pi \tau_p \eta(I)} \right)^{1/2}, \]

where \( \tau_p \) is the pulse width of the laser light. Equation (4) shows that when the required exposure on resist is specified, the penumbral blurring does not depend on \( d \), the
FIG. 11. Calculated penumbral blurring as a function of the laser intensity on target for the case when yttrium is used as a target material; required exposure of the resist $E_r = 1 \text{ kJ/cm}^3$, the distance between specimen and the resist is 1 $\mu$m, the thickness of water layer is 1 $\mu$m, and the thickness of $\text{Si}_3\text{N}_4$ membrane is 0.1 $\mu$m. Diffraction blurring $\delta_d$ is 0.05 $\mu$m (broken line). Minimum penumbral blurring will be 0.03 $\mu$m at $3 \times 10^{12} \text{ W/cm}^2$.

The penumbral blurring remains unchanged for the same exposure on the resist, when the laser intensity is kept constant. In principle, an extremely small laser can be used if the distance between the plasma and resist can be reduced infinitesimally. For example, we may need only about 50 mJ for the laser energy, when the distance between a plasma and specimens is 1 mm. However, in the calculation of Fig. 12, we assumed the same diameter for the x-ray source and the focused laser beam and this can be invalid for small focal spot size irradiation. Furthermore, in a practical configuration, there will be a limit in the distance of the x-ray source from the specimen. Hence, some more studies are required to determine whether 50 mJ energy is large enough to have good images, and whether a smaller energy will do.

V. CONCLUSION

We have measured spectra of x rays emitted from a laser-produced plasma of various kinds of target materials. Yttrium was found to be one of the best target materials for contact x-ray microscopy. Calculated effective spectra showed that the contribution of x rays outside the water window region to making x-ray images is very small owing to the absorption by the x-ray window and water and to the sensitivity spectrum of the resist.

We discussed the optimum laser irradiation conditions for taking an x-ray image. We showed that the laser system can be miniaturized by reducing the distance of x-ray source and resist.

We observed various living cells by contact x-ray microscopy with a laser-plasma x-ray source. Biological components of 40 nm diameter are now observed.\textsuperscript{16}
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12 G. Herzberg, Atomic Spectra and Atomic Structure (Dover, New York, 1944), Chap. 1, p. 62.