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VACUUM ULTRAVIOLET SPECTRA OF A FREE PLASMA FILAMENT IN A MICROWAVE HIGH-PRESSURE DISCHARGE

D. B. DIATROPTOV and V. D. PESKOV

Institute of Physics Problems, USSR Academy of Sciences

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The spectra of a free plasma filament in a high-pressure microwave discharge were analyzed in the 1100–4000 Å range. A continuous background was observed in the 1230–1400 Å range when the discharge took place in mixtures of helium and deuterium. The intensities of the Balmer lines were proportional to the concentrations of deuterium in these mixtures, whereas the intensities of the wings of the 1215 Å line were independent of the deuterium concentration. The experiments provided insufficient data for unambiguous interpretation of the results.

P. L. KAPITZA[1] described a free plasma filament which he observed in a high-pressure microwave discharge. The experimental data suggested a model, according to which the discharge consisted of an inner cylindrical region filled with hot plasma, in which the electron temperature was $T_e \sim 10^2$ eV, and a surrounding cloud of partially ionized plasma with $T_e \sim 0.5$ eV. The present communication reports some results of an investigation of the spectra of Kapitza's plasma[1] which were observed in the vacuum ultraviolet region.

1. APPARATUS

The experiments were carried out using a DFS-29 spectrograph and a VM-1 monochromator in the wavelength range 1100–4000 Å. The radiation was detected with SFM-1 and SFM-3 photon counters, fitted with lithium fluoride windows, or with an FÉU-38 photomultiplier with a sodium salicylate phosphor. The counting rate was determined with an ICh-6 frequency meter. The output voltage, which was proportional to the counting rate, was applied to the Y terminal of an XY automatic recorder. When the photomultiplier was used, the Y terminal was fed with a signal from a U1-2 electrometer and the X terminal with a signal from a variable resistor, which was connected by a velocity linkage to the wavelength drum of a diffraction grating.

The spectrographic unit was within the same gas system as the plasma unit (Fig. 1). The gas in the system was circulated via zeolite traps, which were cooled with liquid nitrogen. The absorption of vacuum ultraviolet radiation of wavelengths $\lambda > 1100$ Å by impurities in the deuterium and the helium (which were used to fill the system) did not exceed a few percent.

The auxiliary source of ultraviolet was in the form of a high-frequency low-pressure (of the order of 1 torr) discharge, which was excited in a quartz tube by means of external cylindrical electrodes. These electrodes were connected to a 3-MHz oscillator which supplied voltages up to 1 kV and whose power output was ~100 W. The low-pressure discharge was separated from the plasma system by a lithium fluoride window. Normally, a lithium fluoride window of a source of this type would rapidly lose its transparency under the influence of the discharge. We avoided this difficulty by removing the impurities, formed in the discharge tube, with a zeolite adsorber cooled with liquid nitrogen (Fig. 1). When this was done, the lithium fluoride window remained transparent even after hundreds of hours of operation of the low-pressure discharge tube.

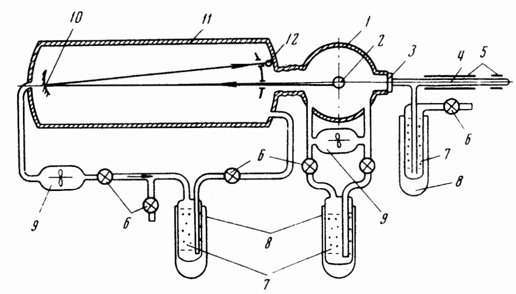

FIG. 1. Schematic diagram of the apparatus: 1) plasma unit resonator; 2) filamentary discharge; 3) lithium fluoride window; 4) low-pressure discharge tube with electrodes; 5, 6) valves; 7) zeolite traps; 8) dewars with liquid nitrogen; 9) fans of the EV50/100 type; 10) diffraction grating; 11) spectrograph; 12) radiation detector. The arrows show the beam path.

2. SPECTRA OF A PLASMA COLUMN IN THE 1400–4000 Å RANGE

The spectrum of a plasma column in deuterium, recorded with the photomultiplier, is shown in Fig. 2. Only the radiation of wavelengths $\lambda > 1400$ Å could be recorded because of the low sensitivity of the photomultiplier and the low value of the signal/noise ratio of the plasma. The peak in the region of 1600 Å was attributed to a many-line emission of the deuterium molecule (transitions $2p^1\Sigma$-$1s^1\Sigma$, $2p^1\Pi$-$1s^1\Sigma$). A strong continuous background and high-order Balmer lines were observed in the 3000–4000 Å region. Following the model of a plasma column given in [1], we attributed the radiation emitted in this region to the external cloud of the discharge.

The number of resolved Balmer lines enabled us to estimate the average electron density in the cloud by the method developed by Inglis and Teller.[2] According to Fig. 2, the electron density was $n_e \sim 2$

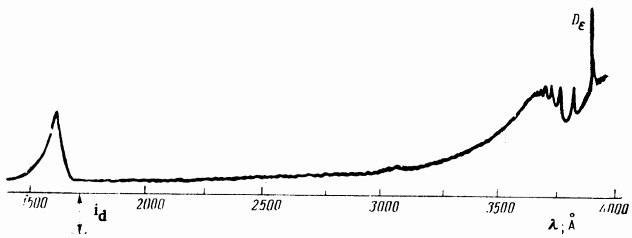

FIG. 2. Spectrum of deuterium plasma recorded with a photomultiplier. Power input 6.6 kW; pressure 1.3 atm; resolution 4Å (i_d is the dark current). A quartz filter was used to remove the second-order spectrum in the $\lambda > 2000$Å range. The dip at $\lambda = 3850$Å was due to a minimum in the transfer function of the VM-1 monochromator.

$\times 10^{14}$ cm^{-3}. This value was in agreement with the results given in [1] when allowance was made for the errors in the calculation and in the measurements. The intensity of the continuum per unit volume of the plasma, per unit time, and per unit frequency interval was calculated in [3] and [4]:

$$I = D_0^+ + D_+^+ + D_-^0 + D_0^0. \quad (1)$$

The term D_0^+ describes the free-bound transitions in the field of an ion (Balmer continuum), D_+^+ represents the free-free transitions in the field of an ion, D_-^0 corresponds to the free-bound transitions in the field of a neutral deuterium atom ("H^- continuum"), and D_0^0 describes the free-free transitions in the field of a neutral atom.

In a narrow spectral range, $\lambda = 3200-3600$ Å, the dependence $I \propto \exp(h\nu/T_e)$ —which applies to any of the terms in the sum of Eq. (1)—yields the electron temperature of the cloud $T_e = 0.5 \pm 0.1$ eV for the spectrum in Fig. 2. This temperature is in agreement with the results reported in [1]. The measured values of n_e and T_e are in agreement with Saha's formula.

Knowing the values of T_e and n_e in the cloud and using the results reported in [3,4], we estimated that $D_-^0 \sim 10D_0^+ \sim 2 \times 10^3 D_+^+ \sim 10^2 D_0^0$. Consequently, that the main contribution to the continuous radiation in the range $\lambda \gtrsim 3000$ Å was made by the term D_-^0. This continuum was observed also in hydrogen arcs struck at atmospheric pressure.

3. SPECTRA OF A PLASMA FILAMENT IN THE 1100–1600 Å RANGE

The radiation of wavelengths shorter than 1600 Å was recorded by means of photon counters. Figure 3 shows the spectrum of a deuterium plasma recorded in the range 1180–1350 Å. The dip in the region of 1215 Å was due to a self-reversed line L_α, and that near 1295 Å was due to the resonance absorption in xenon, the filler gas in the counters. The regions of insensitivity of the counters, corresponding to the Xe lines at 1192, 1295, and 1469 Å, enabled us to eliminate the effect of scattered light and were used as the reference level in the measurements of the intensity. The rise of the intensity in the range $\lambda > 1300$ Å was caused by the wing of the molecular peak at 1600 Å, shown in Fig. 2.

The transparency of the external cloud was estimated by illuminating the plasma filament and the region around it with light from an external source (a

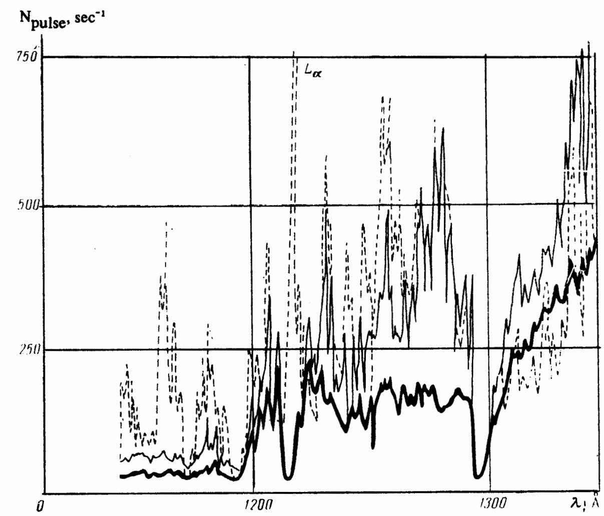

FIG. 3. Spectrum of deuterium plasma (thick curve) recorded with an SFM-3 counter. Power input 9.3 kW; pressure 1 atm; resolution 1Å. The dashed curve is the spectrum of a low-pressure discharge in hydrogen, and the thin curve is the spectrum of a plasma and a low-pressure discharge.

low-pressure discharge tube). This made it possible to determine whether the radiation generated in the central part of the filamentary discharge emerged outside. Figure 3 shows the results obtained when a plasma filament was illuminated (the visible diameter of the column was ~1 cm and the width of the auxiliary beam was ~0.5 cm). The plasma column could be shifted a distance of ~2 cm below the resonator axis by altering the discharge parameters. This made it possible to illuminate the nonluminous region outside the plasma filament. It was found that this nonluminous region (representing the external cloud) yielded the same transmission spectrum as that obtained by illuminating the filament itself with the low-pressure discharge radiation. In view of this, we concluded that the absorption in the external cloud surrounding the filament was less than 0.1 of the total radiation in the $\lambda > 1220$ Å range. The absorption in the nonluminous cloud was 2–3 and the radius of this cloud exceeded 2 cm. Thus, the radiation was absorbed mainly by excited deuterium molecules in the cloud. It is evident from Fig. 3 that this absorption was very strong in the range $\lambda < 1200$ Å.

The filamentary discharge was completely transparent in the 1600–4000 Å region.

4. SPECTRA OF DISCHARGES IN HELIUM-DEUTERIUM MIXTURES AND IN PURE HELIUM

The spectra of the type shown in Fig. 3 could not be used to distinguish the bremsstrahlung of the central part of the discharge from the molecular emission of the cloud because the separation between the molecular lines in the range covered in Fig. 3 was less than 0.3 Å,[5] and these lines could not be resolved by the spectroscopic instruments used in our study. The molecular emission of the cloud was weakened by the addition of helium to the deuterium present in the system. This gave the following results: 1) the gradual reduction of the concentration of deuterium (by a factor of

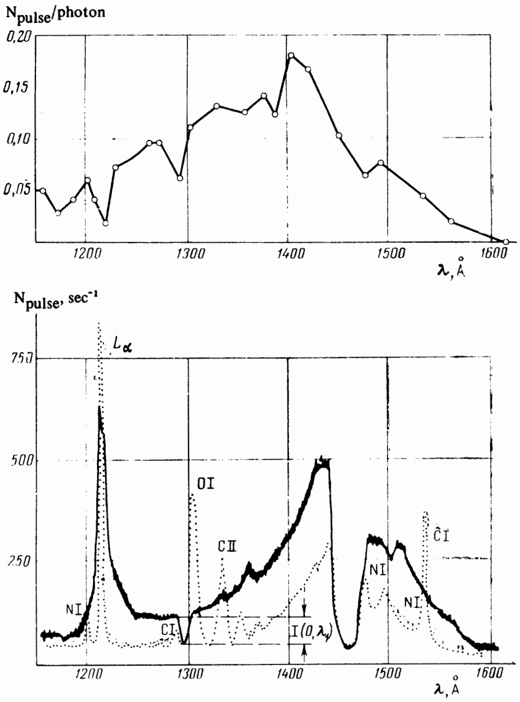

FIG. 4. Plasma spectra of an He + 2% D_2 mixture (continuous curve) at a pressure of 2 atm (filamentary discharge) and of helium (dotted curve) at a pressure of 2 atm (diffuse discharge). Power input 1.8 kW; resolution 2Å. The arrows show the continuum $I(0, \lambda_1)$. The top part of the figure shows the sensitivity characteristic of our SFM-3 counter, as supplied by the factory. This characteristic was recorded at fairly wide intervals and does not show insensitivity regions at λ = 1192, 1295, and 1469Å.

20) weakened the intensity of the emission in the 1230–1300 Å range by a factor of 2 and the intensity of the molecular emission of deuterium near λ = 1600 Å by a factor of 50–80; 2) the intensity of the continuum [given by Eq. (1)] in the 3000–4000 Å range decreased by a factor of about 80; 3) the intensity of the Balmer lines decreased by a factor greater than 20; 4) no HeI lines were observed in any part of the spectrum (at the photomultiplier sensitivity level). The HeI lines were also not observed in the 5000–7000 Å range, which was investigated by means of a prism spectrograph.

The illumination of the plasma filament with light of wavelengths 1150–1350 Å, generated by a low-pressure discharge, indicated that—beginning from a deuterium concentration of 50%—the nonluminous cloud became completely transparent.

Figure 4 shows the spectrum obtained for a mixture of He and 5% D_2 and that for helium of maximum purity. The discharges in the helium-deuterium mixture and in the pure helium differed even in their appearance. The discharge in helium produced a dim uniform region. The addition of deuterium (2% in our experiments) gave rise to a thin red column at the center of the discharge. This was manifested spectrally by the appearance of a continuum in the 1200–1400 Å range [denoted by $I(0, \lambda_1)$ in Fig. 4] and by a strong increase in the intensity of the wings of the 1215 Å line.

The spectrum of the helium discharge included the impurity lines L_α, C I, C II, NI, O I, the molecular spectrum of hydrogen, and—at longer wavelengths—the He I lines and the continuum given by Eq. (1) [the dominant term in Eq. (1) was not that representing the free-free transitions in the field of neutral helium atoms]. The spectrum obtained by means of the prism spectrograph show clearly the He I line at 5875 Å. The spectra of a discharge in deuterium and of discharges in mixtures of helium with deuterium included the impurity lines only if the gases were not very pure. The higher purity of the mixtures with deuterium and of deuterium itself was due to the formation of compounds between carbon, nitrogen, and oxygen on one side, and deuterium on the other. These compounds were adsorbed strongly in the zeolites.

An analysis of the edge and the intensity of the continuum of Eq. (1), carried out in the range 3000–4000 Å, yielded the electron temperature T_e of the plasma clouds in mixtures of helium and deuterium. This temperature was 0.5±0.1 eV and it was independent of the concentration of deuterium (to within 0.1 eV). The same analysis yielded the electron density to within a factor of 2–3 and showed that this density varied as $\sqrt{n_D}$ (n_D is the number of deuterium atoms per 1 cm^3), i.e., approximately in accordance with Saha's formula. The values of T_e and n_e found in the same way for discharges in helium were, respectively, 0.7 ± 0.1 eV and 2×10^{13} cm^{-3}. Although the concentration of the hydrogen impurity in the helium discharges was very low, the higher electron temperature made the intensity of the hydrogen radiation comparable with the intensity of the deuterium radiation in a discharge occurring in a mixture of He and 2% D_2 (Fig. 4). Moreover, the electron densities in discharges in pure helium and in helium with 2% D_2 differed by a factor of 2–3.

5. CONTINUOUS RADIATION IN THE 1200–1400 Å RANGE

The emission spectra of hydrogen and deuterium have been investigated by many workers[6] (p. 21), who have discovered that the variation of the current, the pressure, or the method of excitation, and the presence of impurities (in particular, helium) in the discharge, did not alter the distribution of the intensity in the spectrum corresponding to singlet transitions. Therefore, the many-line spectrum of hydrogen has long been used as a standard.

In our experiments on mixtures, the changes in the molecular spectrum obeyed the empirical formula (deduced by a more careful analysis of the results presented in Sec. 4):

$$I_{obs}(p, \lambda) - I(0, \lambda_1) = pa(p_1) I_{stand}(p_1, \lambda) \qquad (2)$$

which was valid at pressures 0.02 atm $\le p \le$ 0.3 atm and for wavelengths 1230 Å $\le \lambda \le$ 1400 Å and 1230 Å $\le \lambda_1 \le$ 1300 Å. [In the pressure range $p > 0.3$ atm, we found that $pI_{stand}(p, \lambda) > I(0, \lambda_1)$ and $I_{obs}(p, \lambda) \sim pI_{stand}(\lambda)$.] Here, $I_{obs}(p, \lambda)$ is the observed intensity in the range 1230–1400 Å; p is the partial pressure of deuterium; $I_{stand}(p_1, \lambda)$ is the characteristic distribution of the intensity in the molecular spectrum of deuterium in the 1230–1400 Å range (singlet transitions); $I(0, \lambda_1)$ is the constant found by the extrapolation of $I_{obs}(p, \lambda)$ to $p \to 0$; $a(p_1)$ is the coefficient of proportionality. The standard distribution $I_{stand}(p_1, \lambda)$ can be, for example, the distribution of intensity in the molecular spectrum of deuterium obtained by means of a low-pressure discharge (0.1 torr $< p_1 <$ 10 torr), or the distribution of intensity in the spectrum of a deuterium plasma (0.85 atm $\leq p_1 \leq$ 2 atm) or of a plasma in mixtures of helium and deuterium with a partial deuterium pressure p_1. The presence of the term $I_{stand}(p_1, \lambda)$ in Eq. (2) makes it possible to interpret the constant term $I(0, \lambda_1)$ as a continuum which is not associated with the singlet transitions. Now the question arises of the physical nature of the continuum $I(0, \lambda_1)$: is this continuum due to the bremsstrahlung emitted by the core of the discharge or does it represent the emission of the cold cloud?

6. LINE AT 1215 Å IN HELIUM-DEUTERIUM MIXTURES

The line at 1215 Å is an additional feature which was noted in the spectra of discharges in helium-deuterium mixtures (the equivalent width of the self-reversed resonance line of deuterium was proportional to the deuterium concentration n_D and, therefore, a decrease in n_D resulted in the transformation of the absorption band at λ = 1215 Å—Fig. 3—to a line—Fig. 4). It was known[3] that the distribution of intensity in the far wing of the L_α line was asymptotic to the dispersion profile: $I(\Delta\lambda) \propto \gamma/(\Delta\lambda)^2$. In our case, the L_α line could be broadened by the electron mechanism ($\gamma \propto n_e$), the resonance mechanism ($\gamma \propto n_D$), or the van der Waals mechanism ($\gamma \propto n_{He}$, where n_{He} is the number of helium atoms per 1 cm^3). Using the measured values of the electron density, we estimated[3] that the principal contribution to the broadening of the L_α line in the cloud of the deuterium discharge was made by the resonance mechanism. When the concentration of deuterium decreased below a few percent, the van der Waals mechanism became more prominent. Therefore, when the partial pressure of the deuterium was reduced from 0.5 to 0.05 atm, but the electron temperature T_e was kept fixed, the intensity of the wing should have decreased tens of times. In our experiments, the dilution of the deuterium with helium had practically no effect on the intensity of the wings of the 1215 Å line (we recall that the experiments in which an additional ultraviolet illumination was used demonstrated that the wings of the 1215 Å line were transparent beginning from a deuterium concentration of ~50% and from $\Delta\lambda > 5$ Å). On the other hand, the intensities of the Balmer lines recorded by the photomultiplier (Sec. 4) decreased strongly when the concentration of the deuterium was reduced. The intensity and the half-width of the H_β line, recorded in [1], also decreased rapidly when n_D was reduced.

7. DISCUSSION OF RESULTS

The continuum $I(0, \lambda_1)$ cannot yet be interpreted unambiguously. It is not the continuum of Eq. (1) because its intensity calculated for λ = 1200 Å and $T_e \sim$ 0.5 eV is 5–6 orders of magnitude lower than the observed intensity. The continuum $I(0, \lambda_1)$ cannot be the usual helium continuum or the continuum of the HeD quasimolecules because they are not excited at the temperatures obtaining in the discharge ([6] p. 36;[7]). In general, the assumption that $I(0, \lambda_1)$ is the continuum of the HeD quasimolecules or the HeD ions, or that it is the continuum of D_2,[8] is in conflict with the observation that the intensity of this continuum is not affected when the partial pressure of deuterium is made several times higher or lower, whereas the intensity of the molecular spectrum of deuterium is affected very greatly. Such precise "self-tuning" of the intensity of the continuum is unlikely when the discharge conditions are greatly altered.

The observed relationships cannot be explained by assuming that T_e at the center of a discharge is somewhat higher than T_e of the external cloud, or by assuming that T_e increases when the concentration of deuterium is reduced. Numerous investigations of high-pressure discharges in hydrogen and in mixtures of hydrogen with inert gases (arc, torch, high-frequency, and microwave discharges) have demonstrated that the deviation of n_e from the value calculated by means of Saha's formula is not more than a factor of 2 or 3. In our case, the electron density n_e corresponding to T_e = 0.5 eV does not deviate from Saha's formula by a factor greater than that just quoted and the agreement with this formula should not deteriorate with increasing T_e. This is supported also by simple estimates of the rates of ionization and recombination.[3] However, it then follows from the criterion (6.55) given in [3] that the Saha equilibrium will be established in respect of the density of electrons located at higher levels. Therefore, at a fixed pressure, the intensities of the Balmer lines and of the continuum given by Eq. (1) should vary rapidly with temperature. For example, it follows from calculations that the intensities of the Balmer lines (per unit volume) corresponding to T_e = 0.7 eV should be 2–3 orders of magnitude higher than the intensities of the Balmer lines at T_e = 0.5 eV. Moreover, the half-width of the D_β line should be 10 times higher and the intensity of the continuum of Eq. (1) in the region of 3000 Å should be 40 times higher when the electron temperature is increased from 0.5 to 0.7 eV. Therefore, the presence of a central core of the cloud with 0.6 eV $< T_e <$ 50 eV would have affected the intensities and the half-widths of the Balmer lines, as well as the slope and the intensity of the continuum of Eq. (1). Moreover, the He I lines should be observed in the spectra.

The 1215 Å line cannot be explained by assuming that a change in the concentration of deuterium should increase the transparency of the wings, which would compensate the reduction in the intensity of the L_α line. Our experiments involving illumination with the additional ultraviolet source and an estimate of the transparency region based on the formulas given in [9] show that, at deuterium concentrations below 50%, the far wings of the L_α line become completely transparent.

It must be stressed that the strong wings of the 1215 Å line and the $I(0, \lambda_1)$ continuum appear only in a column discharge (Fig. 4). If the discharge is diffuse, the continuum $I(0, \lambda_1)$ practically vanishes and the wings of the L_α line (in this case, $\gamma \propto n_{He}$) are much weaker than the wings of the 1215 Å line in a filamentary discharge, although other features of the spectra do not differ greatly (except that the He I lines are visible in the helium discharge, as reported in Sec. 4).

According to the plasma filament model suggested in [1], the continuum $I(0, \lambda_1)$ should correspond to the bremsstrahlung of electrons interacting with ions and the 1215 Å line should correspond to ionized helium.

In order to confirm the supposition that the 1215 Å line was associated with helium ions, we had to record other He II lines. Here, however, we encountered a basic difficulty which was associated with the fact that the radiation emitted by the cloud was much stronger than the radiation emitted by the hot core (if such a core did exist). The 1215 Å line was hardly visible in the spectra recorded with the photomultiplier. We were unable to observe the He II lines at 1640 and 3203 Å (the intensity of the second line should be seven times weaker than the He II line at 1215 Å) at long wavelengths where the continuum of Eq. (1) was strong and much of the light was scattered. Therefore, it is our intention to carry out a more careful search for the long-wavelength lines of He II and to record the spectrum of discharges in deuterium-neon mixtures, which should not have lines in the region of 1215 Å.

The authors are deeply grateful to P. L. Kapitza for his active participation in the formulation and discussion of each experiment, and to A. V. Mitrofanov, who adjusted the monochromator and recorded several spectra. We also thank L. S. Sorokin for supplying us with photon counters, and N. I. Kondrat'ev, M. S. Tyurin, A. V. Lebedev, and all the staff of the plasma laboratory for their technical help with this investigation.

Note added in proof (July 12, 1971) Experiments on Ne + D_2 mixtures that the 1215Å line was indeed the L_α line. The HeII line at 1640Å was absent, irrespective of the concentration of helium, in the plasma spectra of He + D_2 mixtures recorded with a counter sensitive to radiation in the range λ = 1100-2000Å (mean quantum efficiency ~10%).

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