results show that the beam waist is located between the mirror and 5 μm inside of the mirror up to 20 mW. Figure 6 shows the results of cw aging test at the constant output power of 20 mW/mirror and the heat-sink temperature of 50 °C.

In summary, we have achieved high output cw operation by the newly developed window VSIS laser. Window structure is easily made because the window region and the stimulated region are grown at the same time. The beam waist exists almost at the mirror surface because the built-in optical waveguide is also formed in the window region. As a result, low threshold current, fundamental transverse operation, and high cw output power were obtained. Furthermore, the reproducibility and the reliability of the device were also as high as the ordinary VSIS laser.

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Ultraviolet photodecomposition for metal deposition:
Gas versus surface phase processes

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Photodecomposition of organometallic gases has been shown to be potentially useful for high resolution direct metal deposition. However, a number of problems, particularly the low writing rates, must first be solved. An understanding of the deposition mechanism is essential to this task. In particular, the decomposition must be shown to occur either in the gas phase or in the adsorbed layer. We show that this question can be resolved by the dependence of the writing rate on spot size. Measurements of the process show that the decomposition occurs in the gas phase. This result is shown to cause inhomogeneous deposition for exposures through masks with varying feature sizes, and effect will be important to future applications.

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Recently there has been a great deal of interest in ultraviolet photodecomposition for localized metal deposition and semiconductor etching.1–5 Metal deposits with submicron features have been generated.1 This "direct writing" method has applications to custom large scale integration (LSI) fabrication, mask and wafer repair, etc. However, despite considerable work, very little is known about the mechanism of the deposition. This is an important problem, since many potential applications are currently being prohibited because of the slow writing rate (typically roughly 20 Å/s in a 50-μ spot). Furthermore, as we will show, the method exhibits an unexpected undesirable aspect: nonuniform growth rates in features of different sizes. This result can be understood in the context of the mechanism presented here.

In a typical experiment, a substrate is placed in a few Torr of an organometallic gas such as dimethyl cadmium, and exposed to a focused UV, cw laser beam. Deposited metal is observed in the illuminated region. This metal may have been deposited by two different mechanisms. Organometallic molecules adsorbed on the surface may absorb a photon and decompose, leaving the metal behind on the substrate. Conversely, the decomposition may occur in the gas phase, followed by metal migration to and sticking on the substrate. As shown below, the number of molecules participating in these two reactions is comparable, so it is not possible to differentiate between them a priori without accurate measurements of the adsorbed and gas phase photodecomposition cross sections. Although gas phase decomposition has been suggested as the dominant factor,6 no previous experiments directly address this question. Differentiation between these two processes is necessary to an understanding of the mechanisms of deposit localization, and attempts to increase deposition rates.

Ehrlich et al. have proposed a two-step model for writing in which the deposit is first localized by decomposition of adsorbed organometallic to form critical nuclei, followed by gas phase decomposition to form the bulk of the deposit.7 Although this model successfully explains observation of localized deposits on prenucleated regions, the experimental observations do not indicate whether the bulk of the final deposited spot is due to decomposition in the gas or adsorbed phase.

In this letter we derive a technique for determining whether the decomposition occurs in the adsorbed or gas phase, by measuring the dependence of the writing rate on laser spot size. We apply this technique to a commonly used reaction, the decomposition of cadmium from dimethyl cadmium, and show that, for this system, the bulk of the writing
on the substrate and \(l\) is the average distance a metal atom travels in the gas phase before it is removed by a gas phase reaction. Measurements on gas phase population decay indicate that \(l \approx 10\) cm for typical reaction conditions.\(^1\) Since \(h\bar{\nu}w_0\) for all cases studied here, we make the approximation \(e^{-\bar{\nu}/l} \approx 1\) in Eq. (4).

Equation (4) now becomes

\[
R_s(r_0, w_0) = \frac{\alpha N_s \sigma_s P_L}{2\pi \hbar w_0} \int d\phi \int dz \int dr \frac{z}{d(r_0)^3} \exp \left( -\frac{2r^2}{w_0^2} \right) .
\]  

(5)

For \(r_0 \neq 0\), Fig. 1 indicates that the symmetry is very low, so this integral is difficult to evaluate. Fortunately, we do not need to do so, since we are only interested in the dependence of \(R_s\) on \(w_0\). We observe by direct substitution in the integral that for an arbitrary scaling factor \(\gamma\),

\[
R_s(r_0, \gamma w_0) = \gamma^{-1} R_s(r_0, w_0)
\]  

(6)

or

\[
R_s(r_0 w_0) = \frac{1}{w_0} R_s \left( \frac{r_0}{w_0} \right).
\]  

(7)

We observe that Eq. (2), for adsorbed decomposition, is in the form

\[
R_s(r_0, w_0) = \frac{1}{\eta w_0^2} f_s \left( \frac{r_0}{w_0} \right).
\]  

(8)

Our technique for differentiating between adsorbed or gas phase decomposition is based on observing the deposition rate \(R\)

\[
R(r_0, w_0) = \frac{1}{w_0^2} f(r_0, w_0).
\]  

(9)

If \(n\) is observed to be 2, Eq. (8) is valid and the decomposition is surface phase. If \(n\) is observed to be 1, Eq. (7) is valid, and the decomposition gas phase.

The distinction between the two cases can be qualitatively understood by considering the point \(r_0 = 0\). There \(R\) can be evaluated analytically in both cases:

\[
R_s(0, w_0) = \frac{2N_s \sigma_s P_L}{\pi \hbar w_0^2} \frac{1}{w_0},
\]  

(10)

\[
R_s(0, w_0) = \frac{\alpha N_s \sigma_s P_L}{2\sqrt{2\pi \hbar w_0^2}} \frac{1}{w_0}.
\]  

(11)

We see that \(R_s \sim w_0^{-2}\) and \(R_s \sim w_0^{-1}\). The additional factor of \(w_0\) in \(R_s\) occurs because atoms created within a distance of order \(w_0\) from the surface contribute to the deposition. We note that for the conditions used here, the gas and surface phase decomposition rates are of the same order of magnitude, assuming equal decomposition cross sections.\(^7\)

The experimental layout is shown schematically in Fig. 2. Light at 5145 Å from a Spectra-Physics 171 argon-ion laser is frequency doubled by a temperature regulated ammonium dihydrate phosphate (ADP) crystal. The UV light is separated from the fundamental, filtered, and monitored before it is focused on the sample. Approximately 2 mW of UV light was obtained at the sample, and this was maintained constant within 5%. The light was focused on the quartz front window of a gas cell; deposition took place on this...
FIG. 2. Optical schematic. Green cw light from an argon-ion laser is frequency doubled in a 90° phase matched ADP crystal, and the resulting UV light is filtered, monitored, and focused on the front window of the gas cell. The attenuation of the UV beam is used as a monitor of the deposition.

window’s inner surface. Although some deposition did take place on the rear window, gas phase attenuation and the larger laser spot size meant this was not a significant contributor to the attenuation rate. Deposited spot sizes were adjusted by translating the focusing lens in the 2 direction. Gases were introduced into the cell through an all metal gas handling system. For this experiment, 1 Torr of electronic grade dimethyl cadmium from Alfa was buffered with 750 Torr of research grade helium. Pressures were read from a MKS model 220 capacitor manometer or a United Instruments diaphragm pressure gauge. After deposition, spot sizes were observed directly in a Zeiss light microscope.

Deposition rates were monitored by the rate of the attenuation of the UV beam. The insert to Fig. 3 shows a typical transmission plot as a function of time. After a sharp rise at time t = 0, an exponential attenuation is observed. For each deposition, a time constant, τ(ω₀), is calculated for the attenuation by measuring the initial slope of the transmission decay. The time constant so defined measures the deposition rate in a average weighted over the laser power density, Eq. (1). It can be shown that the dependence of this time constant on ω₀ is

τ(ω₀) = ω₀τ₀,

where τ is defined in Eq. (9) and τ₀ is an arbitrary constant.

These time constants are plotted as a function of spot size, ω₀, in Fig. 3. The slope of the log-log plot gives the value of the exponent n in Eq. (9). The observed value, n = 0.73 ± 0.15, is most consistent with n = 1, which, as Eq. (7) shows, implies gas phase decomposition. The fact that the observed value of n is slightly less than 1.0 implies that there is some error in the simplifying assumptions used in our analysis. These assumptions include neglect of nonuniformities of the beam intensity profile due to absorption as the deposit develops, and the residual deposition on the rear window. Nevertheless, the analysis is sufficient to decide un-

ambiguously between n = 1 and n = 2, and thus between gas and surface phase decomposition.

Our data have been analyzed in terms of either pure gas phase or surface phase decomposition. In the context of the two stage writing model by Ehrlich et al., we show that the bulk of the decomposition does indeed take place in the gas phase, and that this decomposition is the rate limiting step.

We have applied our technique for isolating adsorbed and gas base decomposition to one particular reaction, dimethyl cadmium decomposition. The fact that the bulk of the decomposition contributing to the deposit occurs in the gas phase has important implications for improving deposition rates. Molecules and laser wavelengths should be chosen for maximum gas phase decomposition cross sections. Within limits, writing rates should be linear with reactant gas pressure. Finally, Eq. (11) shows, at constant laser power density (such as would be achieved by illumination through a mask), the writing rates should be lower for small features than for large ones. This property, which is similar to the proximity effect observed in electron beam lithography, will be particularly important to the understanding of growth of complex patterns, where all features are not the same size.

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7. This statement is based on α = 1, N₀ = 2 × 10¹⁴ cm⁻² (approximately one monolayer), 1 Torr reactant gas pressure, and ω₀ = 50 μ.
8. This has been observed in Ref. 2, but the small pressure range available and the approximately linear dependence of N₀ on pressure make this a poor tool for deciding between adsorbed and gas decomposition.