method also includes b) introducing a metal-bearing precursor gas having a metal M to the chamber interior. The metal-bearing precursor gas reacts with and remains on the substrate. The method also includes c) purging the chamber interior of excess metal-bearing precursor gas. The method also includes d) introducing a chalcogenide precursor gas into the chamber interior using a plasma. The chalcogenide precursor gas reacts with the metal-bearing precursor gas that remains on the substrate, to produce an amorphous film of MX or MX_2. The method also includes e) purging the chamber interior. The method also includes f) scanning a laser beam over the amorphous film to heat the amorphous film to a temperature of between 650 °C and 1200 °C to produce the substantially 2D film of either MX or MX_2 on the surface of the substrate. The substantially 2D film is substantially crystalline.

[0027] Another aspect of the disclosure is the method described above, wherein the metal M is one of Mo and W.

[0028] Another aspect of the disclosure is the method described above, wherein the chalcogenide X is one of S, Se and Te.

[0029] Another aspect of the disclosure is the method described above, wherein the plasma includes X-bearing radicals.

[0030] Another aspect of the disclosure is the method described above, wherein the X-bearing radicals include H_2S*.

[0031] Another aspect of the disclosure is the method described above, the method further includes processing the substrate to remove the substantially crystalline and substantially 2D film from the surface of the substrate.

[0032] Another aspect of the disclosure is the method described above, wherein the laser beam has a nominal wavelength of 532 nm.

[0033] Another aspect of the disclosure is the method described above, wherein in act d), the providing of the chalcogenide precursor gas is performed in a continuous manner or a pulsed manner.

[0034] Another aspect of the disclosure is the method described above, wherein the 2D film has dimensions of 25mm x 25mm or greater.

end 34, wherein the narrow open end 32 is operably arranged closest to plasma source 25 and the wide open end 34 is operably arranged at the upper wall 22 at the aperture 23. In an example, the conical wall 30 supports a transparent window 36.

[0077] The system 10 also includes a chuck 38 operably disposed within the interior 18. The chuck 38 operably supports a substrate 40 that has an upper surface 42 on which the laser-assisted ALD methods are carried out. In an example, the substrate 40 is silicon, or sapphire. Other substrates can be used, and silicon or sapphire substrates 40 may be preferred only because they happen to be widely available and because semiconductor processing equipment and apparatus (e.g., the chuck 38) are typically designed to handle silicon or sapphire substrates 40. It is noted that the upper surface 42 of the substrate 40 typically includes OH^- molecules when exposed to air, and that these molecules can play a role the ALD process as is known in the art and as described below.

[0078] The substrates 40 are used in the methods disclosed herein to form large-area 2D films of metal chalcogenides. In many of the prior art methods, the 2D materials are in the form of very small (e.g., few millimeter diameter) flakes. In an example, a large-area 2D film has dimensions of greater than 25 mm x 25 mm or 50 mm x 50 mm or 100 mm x 100 mm. Note that a large-area film of 300 mm diameter can be divided up into a number of smaller but still large-area 2D films.

[0079] The system 10 also includes a laser source 50 operably arranged relative to the ALD process chamber 14 so that it can be selectively activated to generate a laser beam 52. The laser beam 52 passes through the transparent window 36 and into the interior 18 and then onto the upper surface 42 of the substrate 40 or more specifically onto the particular film formed thereon as described below. In an example, the chuck 38 can be configured to heat the substrate 40 to an initial temperature for processing. In an example, the laser source 50 is a YAG laser that emits the laser beam 52 having a wavelength of 532 nm. Other types of the laser sources 50 can be used wherein the laser beam 52 can heat the upper surface 42 of substrate 40 or the film formed thereon. In an example, the chuck 38 is movable in the x-y plane, as indicated by double-arrow AR, to effectuate the scanning of laser beam 52 over upper surface 42 of the substrate 40. In an example, the chuck 38 is also adjustable (e.g., movable) in the z-direction. Alternatively, the laser beam 52 can be scanned over the

What is claimed is:

1. A method of forming a substantially two-dimensional (2D) film of a metal chalcogenide on a surface of a substrate, the method comprising:

a) adhering a layer of metal-bearing molecules to the surface of a heated substrate using an atomic layer deposition (ALD) process;

b) causing an oxidant precursor gas to react with the layer of metal-bearing molecules to form a layer of MO_3;

c) repeating acts a) and b) to form an MO_3 film having multiple layers of MO_3;

d) laser annealing the MO_3 film to form therefrom an MO_2 film;

e) causing a chalcogenide-bearing radicalized precursor gas to react with the MO_2 film to form an amorphous and substantially 2D film of the metal chalcogenide; and

f) laser annealing the amorphous and substantially 2D film to form therefrom a substantially crystalline and substantially 2D film of the metal chalcogenide, wherein the metal chalcogenide can have the form MX or MX_2, where M is a metal and X is a chalcogenide.

2. The method according to claim 1, wherein the metal M is one of Mo and W and wherein the chalcogenide X is one of S, Se and Te.

3. The method according to claim 1, wherein act e) includes providing the chalcogenide-bearing radicalized precursor gas using a plasma.

4. The method according to claim 3, wherein the chalcogenide-bearing radicalized precursor gas comprises H_2S*.

5. The method according to claim 1, further comprising processing the substrate to remove the substantially crystalline and substantially 2D film from the surface of the substrate.

6. The method according to claim 1, wherein the substantially crystalline and substantially 2D film of the metal chalcogenide has dimensions of 25 mm x 25 mm or greater.

7. A method of forming a two-dimensional (2D) film of either a metal monochalcogenide (MX) or a metal dichalcogenide (MX_2) on a surface of a substrate using an atomic layer deposition process, the method comprising:

a) providing the substrate in a chamber interior having a pressure in the range from 0.1 Torr to 0.5 Torr and heating the substrate to an initial temperature of between 150 °C and 500 °C;

b) providing a metal-bearing precursor gas having a metal M to the chamber interior, including purging any excess metal-bearing precursor gas, wherein the metal M is one of Mo and W;

c) providing an oxidant precursor gas into the chamber interior to form a layer of MO_3, and purging any excess oxidant gas;

d) repeating acts b) and c) to form an MO_3 film having multiple layers of MO_3;

e) laser annealing the MO_3 film to form an MO_2 film;

f) introducing a chalcogenide precursor gas into the chamber interior using a plasma, wherein the chalcogenide precursor gas reacts with the MO_2 film to produce a film of amorphous MX or MX_2, and purging the chamber interior; and

g) scanning a laser beam over the amorphous film of MX or MX_2 to heat the amorphous film of MX or MX_2 to a temperature of between 650 °C and 1200 °C to produce a substantially crystalline film of either MX or MX_2.

8. The method according to claim 7, wherein the oxidant precursor gas is one of H_2O, O_3, O* and O_2.

9. The method according to claim 7, wherein the chalcogenide precursor gas includes sulfur.

10. The method according to claim 7, wherein the metal-bearing precursor gas is selected from the group of precursor gases consisting of:

Bis(tert-butylimido)bis(dimethylamido)Molybdenum, $MoCl_5$, Molybdenum hexacarbonyl, bis(tert-butylimido)bis(dimethylamido)Tungsten, $WH_2(iPrCp)_2$ and WF_6.

11. The method according to claim 7, wherein the laser beam has a nominal wavelength of 532 nm.

12. The method according to claim 7, wherein in act e), the providing of the chalcogenide precursor gas is performed in either a continuous manner or a pulsed manner.

13. The method according to claim 7, further comprising removing the substantially crystalline film of either MX or MX_2 from the surface of the substrate.

14. The method according to claim 7, wherein the laser scanning is performed in a raster scan.

15. The method according to claim 7, wherein the substrate is made of silicon or sapphire.

16. The method according to claim 7, wherein the substrate is supported by a heated chuck, and in act a), the substrate is heated to the initial temperature by the heated chuck.

17. The method according to claim 7, wherein the MO_3 film has between 3 and 8 layers of MO_3.