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A hollow-cathode plasma sputter source for production of metal cluster beams

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Abstract. We have developed a source for the production of clusters of refractory metals by plasma sputtering. One aim is to produce metal clusters with a density of $\sim 10^{12}$ atoms/cm^3 in a size range of 10^2–10^5 atoms per cluster for the purpose of performing X-ray diffraction studies. Net sputtering rates have been measured for a Cu cathode, as a function of pressure, flow velocity, current, and cathode geometry. Computer simulations of the mass loss from the cathode reproduce the experimental results with reasonable agreement.

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1. Introduction

The production of clusters from refractory metals demands the use of either an extremely hot oven or a laser for the evaporation of the material. Alternatively, a primary ion source or an electric discharge may be used to sputter atoms of the material into the gas phase. This type of source has the crucial advantage of a very high yield. While most measurements of cluster properties do not require particularly intense beams, the highest possible cluster beam intensity becomes a determining factor in the investigation of the crystalline structure of freely suspended metal clusters via X-ray or electron diffraction.

Sputtering ion sources have already had some impact in the investigation of cluster properties, see e.g. refs. [1–4]. Here we report on the development of a metal cluster ion source, based on a dc plasma discharge in conjunction with aggregation in an argon gas.

2. Experimental setup

A schematic diagram of the apparatus is shown in Fig. 1. Ar at $\sim$ 1 mbar flows through the hollow cathode at a adjustable rate (1–80 m/s). When a potential difference between anode and cathode of some 300–500 V is applied an electric discharge is ignited. The Ar$^+$ ions, accelerated towards the inner walls of the cathode, sputter atoms from the cathode into the gas. Typical currents are 100–1000 mA.

The inert gas serves two purposes apart from maintaining the plasma: On the one hand collisions with the small clusters provide cooling to remove the heat of condensation, thus facilitating formation of larger clusters. On the other hand the flowing gas sweeps the atoms and clusters away from the hot plasma region.

The source has been investigated in several geometries. In the one described here, the inert gas enters the apparatus through the cathode and then proceeds through the anode, which is at ground potential.

The cathode is manufactured from the desired cluster metal (designed to allow a minimum of material), and has the geometrical form of a cone. External cooling of the cathode is required, since typically 200–400 watts are deposited on it during the discharge.

It is mounted in an oil cooling unit and the conical shape allows good thermal an electrical contact to the cooling unit which is connected to a HV power supply. The cathode can easily be removed for inspection or replacement.

3. Results and discussion

The rate with which atoms are removed and escape from the cathode depends on two competing processes: sputtering and redeposition. Backscattering of sputtered atoms contribute to redeposition [5], but is here considered not to be significant. The main contribution to redeposition comes from diffusion of atoms back to the wall. The data presented here, of the weight lost from the cathode under various discharge conditions, monitor the combined result of sputtering and redeposition.

Fig. 2 shows that the apparent sputtering rate from a 5.0 mm diameter 40.0 mm long hollow cathode significantly decreases for argon pressures both above and below about 2.0 mbar. The filled circles in the figure represent numerical simulations of the mass loss. The

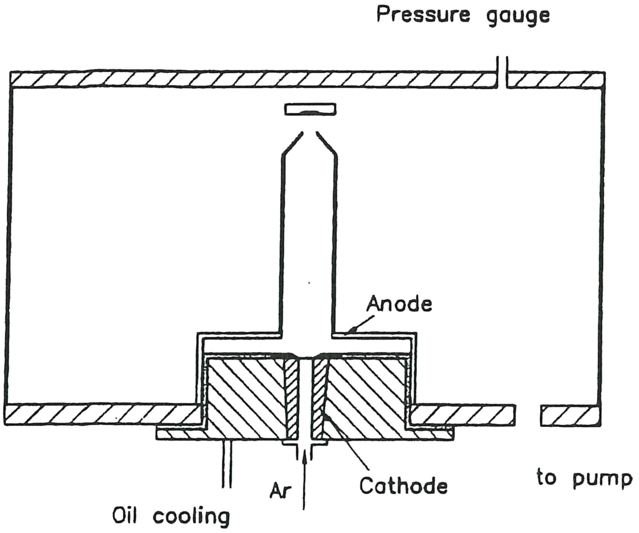

Fig. 1. Schematic of the experimental setup

calculations uses Fick's first law for flux of particles A in a gas of particles B. The total flux is the sum of diffusion and collective flow. The curve can be understood qualitatively as follows.

Cathode atoms leave the surfaces with energies from a few eV up to several hundred eV, but the majority will have energies below 10 eV [6]. When secondary atoms have thermalized within the carrier gas, they will obtain the flow velocity determined by the velocity profile. In the low pressure range the mean free path is so long that diffusion to the opposite cathode wall becomes important. For decreasing pressures this effect will result in decreasing net sputter yields in terms of mass loss from the cathode, see Fig. 2. In the high pressure domain, on the other hand, the mean free path is so short that secondary atoms are confined close to the surface. Due to the low flow velocity close to the wall, the atoms will have a long residence time in the cathode. This leads to a high redeposition probability. The important role of redeposition phenomena is furthermore supported by the considerable reduction observed in the apparent sputtering rate when the flow velocity was decreased.

With an applied current of ~ 500 mA a density of sputtered atoms up to 3×10^{14} atoms/cm^3 is obtained. At this current, the temperature close to the exit of the cathode was measured 800 K. For these conditions the Cu vapor has a partial pressure of 4×10^{-2} mbar, which is far beyond supersaturation. Thus conditions facilitating aggregation are present.

The cluster size distribution has been examined with a transmission electron microscope, which has a resolution of approximately 8 Å. At a distance of ~ 15 cm from the cathode exit samples have been taken to probe the beam. The size distribution was observed to be in the range of 10–100 Å, which correspond to Cu$_n$ with $30 < n < 30 \times 10^3$.

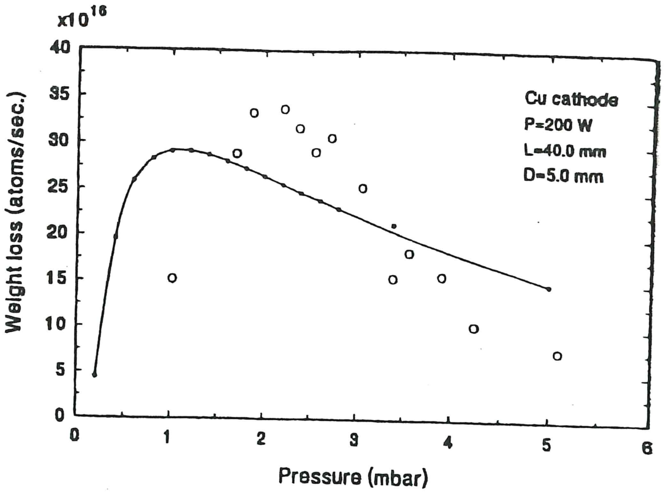

Fig. 2. The dependence of mass loss from the cathode versus pressure. The filled circles represent the numerical result

4. Summary

We have presented the state of a newly developed sputter source. The source promises very high beam density and a very high mass distribution. Also, the source shows very stable running conditions and is easy to handle. The principle allows many different materials, which can easily be exchanged within minutes, and promises a broad spectrum of changeable parameters to control the mass distribution. The source at the present stage has a higher density than required, but this is a the exit of the cathode only. The beam must be guided into high vacuum before diffraction is possible. This problem is yet to be solved.

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