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Abstract

Full Text

Physical Chemistry

Yu. I. Petrov, B. A. Rusin

Dispersed Condensates of Aluminum Vapor on Glass

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The formation of shadow images of an object placed in the path of a molecular beam may be due to various causes: the geometric dimensions of the source, scattering of molecules by residual gases, and surface migration of the molecules of the deposit.

We observed very curious phenomena during the condensation of aluminum vapor, placing a diaphragm with various apertures near a glass plate. The apparatus, shown in Fig. 1, was located under the glass bell jar of a vacuum system. After preliminary heating and prolonged degassing in vacuum, $\sim 10^{-4}$ mm Hg, an aluminum charge was slowly evaporated from a coil of tungsten wire inside a closed brass shield cooled by running water (4°). In the upper part of the shield there were replaceable diaphragms made of copper foil 0.1 mm thick with various apertures. Above the diaphragm was a metal shutter, which was released by a magnet after the operating evaporation conditions had been established. A glass plate, previously degreased by washing in alcohol and firing in the mild flame of a gas burner, was installed at various distances from the diaphragm. The principal experiments were carried out with the plate placed at a distance of 10 mm from the diaphragm with circular apertures 10 mm in diameter. The geometry of the experiments was such that the linear dimensions of the evaporating droplet could not give the observed penumbra of the apertures.

Fig. 1. Schematic of the evaporator:
1 —tungsten wire with an aluminum charge;
2 —brass shield cooled by running water;
3 —diaphragm with various apertures;

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The gas pressure, measured by thermocouple and magnetic manometers, was maintained at 10^{-3} mm Hg by regulating the rates of evaporation and pumping. The average thickness of the deposit, according to weighing, was $\sim 10^{-5}$ cm. Complete evaporation of a charge of ~ 100 mg was carried out in ~ 20 min, so that the flux of atoms onto the plate surface averaged $\sim 1.5 \cdot 10^{15}$ $\mathrm{cm}^{-2} \cdot \mathrm{s}^{-1}$. The density of the condensate was measured with an MF-4 microphotometer.

The character of the deposit changed from a sharply outlined, mirrorlike coating in the shape of the aperture, when the plate was pressed against the diaphragm, to an increasingly blurred image as the plate was moved away. The deposit did not appear immediately, but after a certain time following the release of the shutter. At first it had the appearance of a small dark spot, which rapidly increased to a limiting size. A deposit also appeared on the reverse side of the diaphragm, which indicates reflection of atoms from the glass. The presence of substantial reflection of atoms was revealed by an experiment with two parallel plates (Fig. 2). In this case the density of the mirror-

of its condensate on one plate exceeded the density on the other by only a few times. Hollind (1) calculated that, under ordinary conditions for preparing an aluminum mirror, of 1000 atoms colliding with the glass only one is deposited, while the rest are reflected. The most curious result consists in the appearance, around the central mirror deposit, of a narrow concentric ring (Fig. 3). The structure of the deposit was studied under an electron microscope. For this purpose the condensate was deposited on a collodion film covering the glass surface and separable from the latter in water. The mirror part of the deposit consists of separate islands, each of which is made up of many small particles of a size at the limit of the microscope resolution, $\lesssim 30$ Å. The peripheral ring is formed by approximately the same particles, far removed from one another. Figure 4a gives a comparison of the observed deposit density with that expected according to rectilinear propagation of the atoms. As can be seen, the diameter of the condensate is approximately 1.5 times greater than the diameter of the aperture. Figures 4b–d demonstrate the change in the density distribution of the deposit as two identical apertures of the diaphragm are brought closer together. In the region of overlap, the measured density is not equal to the sum of the densities given by the separate apertures (Figs. 4b, c), and the configuration of the deposit is deformed in such a way as if two beams of atoms repel one another behind the diaphragm. In order to clarify the possibility of electrical interaction of the atom streams, an electrically isolated tungsten filament of diameter 0.05 mm was stretched along the diameter of the aperture; a voltage of 300 V, of different polarity relative to the evaporator, was applied to it. No change in the shape or density of the deposit was detected.

Fig. 2. Experiment demonstrating mirror reflection of an aluminum vapor beam

Figure 2 diagram

Figure 2: Figure 2 diagram

Fig. 3. Photo prints of deposits on glass. The formation of thin rings around the central specular deposit is clearly visible

Figure 3: Fig. 3. Photo prints of deposits on glass. The formation of thin rings around the central specular deposit is clearly visible

The results obtained are most simply explained if one assumes that near the evaporator the aluminum atoms combine into small aggregates which, colliding with molecules of the residual gas, are deflected into the region of the geometrical shadow behind the diaphragm aperture. It is also possible that complexes of atoms are emitted during evaporation of aluminum. The aggregates may be reflected several times from the plate and the diaphragm before they settle on the glass, forming a dispersed condensate. In this way a bell-shaped distribution of the deposit density arises. When two apertures of the diaphragm are brought closer together, aggregates entering through one of them, after reflection from the glass, partially fly out through the other, which leads to deformation of the deposit in the interval between the apertures. Aggregates flying near the edge of an aperture and not having undergone collisions with molecules of the residual gas may be specularly reflected twice from the plate and the diaphragm, and then be deposited in the form of a narrow ring on the glass. The following experiment convinces us of this. A tube 15 mm long, rolled from thin aluminum foil, was inserted into the aperture so that its lower end almost coincided with the surface of the diaphragm. The glass plate was placed at the same distance from the upper end of the tube as it had previously been from the diaphragm aperture. In this case the ring disappeared because of the removal of the plane of the diaphragm, while the deposit itself became more uniform, with fairly sharp boundaries.

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The formation of a narrow ring due to double specular reflection of atoms from the glass and copper foil is practically excluded, in view of the diffuse nature of their scattering by the surface of rough materials. The possibility of aggregation of atoms near the evaporator, where their concentration is high, is quite probable, since increasing the residual-gas pressure only twofold, from 10^{-3} to $\sim 5 \cdot 10^{-2}$ mm Hg, led to the formation of a highly dispersed aerosol powder of black color, which uniformly covered the diaphragm. At the same time, a specular deposit arose on the distant glass plate, with a diameter smaller than the diameter of the aperture. If, under the same conditions, a cooled (4°) copper foil is installed instead of the glass plate, then a faint spot of black aerosol

Fig. 4 schematic profiles

Figure 4: Fig. 4 schematic profiles

powder is obtained on it. In the presence of residual gas, aluminum atoms lose energy by inelastic collisions, and the probability of the formation of aggregates increases ([2]).

Fig. 4. Photometric density profile of deposits. *s* is the optical density; *d* is the distance on the glass plate; *a* is the density distribution of a circular deposit along its diameter; *b*–*d* show the interaction of deposits when two identical diaphragm apertures are brought closer together; *d* shows that the bridge between the diaphragm apertures has been removed (a common oval aperture). 1 is the expected pattern in the case of rectilinear propagation of atoms. 2 is the total density from overlapping portions of two curves corresponding to the separate apertures.

On the polished surface of glass, aggregates may pack into a crystal lattice with a characteristic metallic luster. An analogous phenomenon was observed earlier during condensation of Ge and Si vapor in a hydrogen atmosphere, when thin films were obtained with better properties of crystal structure and better electrical properties than vacuum condensates ([3]).

It should be expected that the probability of deposition or reflection of aggregates will depend on their size, on the temperature and state of the surface, and also on a number of other factors. Allowing for the possibility of the formation of atomic aggregates in the volume of metallic vapor, it is easy to explain the critical temperature of vapor condensation on a solid surface, its dependence on beam density and observation time ([4,5]), the sudden appearance of primary particles on the substrate and the disperse structure of deposits ([6–8]), as well as the differences in density and structure of deposits on a nonuniformly heated glass surface ([9]).

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