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Abstract

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ON THE TRANSFORMATION OF CLOUD DROPLETS INTO ICE CRYSTALS

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Experimental verification of the possibility that supercooled cloud droplets can transform into well-faceted individual ice crystallites is of essential importance for resolving the question of the secondary character of the process of formation of the ice phase in the atmosphere. A theoretical consideration of this question leads to the conclusion that the process of condensation in clouds and subsequent crystallization of the liquid phase is highly probable (1), and calculations of the freezing of supercooled aqueous aerosols agree well with this assumption and make it possible to assert that in the atmosphere the solid phase is formed, as a rule, as a result of freezing of droplet-liquid moisture (2). Numerous studies of the crystallization of water as a whole also provide a basis for the hypothesis of ice formation in the atmosphere through the intermediary of the liquid phase (3).

However, this point of view is still called into question, since it is based only on statistical considerations and has no direct experimental confirmation (4). Indeed, direct observations of the process of formation of ice particles in clouds and fogs have so far not been possible with a sufficient degree of accuracy (5). In this connection we have carried out laboratory experiments to elucidate the nature of the freezing of water droplets close in order of magnitude to the sizes of cloud droplets, and to refine their behavior in a medium supersaturated with water vapor.

The investigations were carried out with droplets of distilled water several tens of microns in diameter. Such droplets are usually easily formed on a glass thread when, in the chamber in which this thread is located, there is strongly supersaturated vapor. The experiments showed that if these small droplets freeze, then within 2–3 min they transform into well-faceted ice crystallites. Figure 1a shows part of a glass thread with supercooled water droplets condensed on it. As can be seen from Fig. 1b, which shows the appearance of the same thread 4–5 min after the droplets froze, instead of droplets there are faceted ice crystals situated on the thread. In shape these crystals are strongly elongated prisms, the dimensions of whose minor axes are close to the diameters of the original droplets, although the major axes considerably exceed them.

In order to follow all the stages of such a transformation of frozen droplets into ice crystals, we filmed the entire course of this rapid process. Examination of the frames of the film makes it possible to establish that immediately after the droplets freeze (Fig. 3a), flat faces appear on their surface (Fig. 3b), separated by barely noticeable rounded regions; with time, as the size of the frozen droplets increases as a result of the subsequent direct "deposition" (sublimation) (6) of water vapor on them, the faces on their surface grow (Fig. 3c) and, on coming into contact, form dihedral angles (Fig. 3d). The growing crystallites thereby exhibit all the principal features of a sublimation origin.

Thus, under laboratory conditions it is possible to carry out the direct transformation of supercooled water droplets into ice crystals and experimentally to confirm the previously proposed (7) scheme of the secondary formation of ice crystals in the atmosphere.

It should be noted that the microscopic ice granules formed in our experiments during the freezing of supercooled "cloud" droplets (Fig. 3a), as additional investigations in polarized light have shown, are usually all single crystals. This also predetermines their inevitable subsequent transformation (under conditions of supersaturation) into well-faceted ice crystallites, since their transformation should in principle proceed in exactly the same way as usually occurs in the crystallization of spheres (8), composed of individual crystals or obtained by some other method, and should lead to the formation of a faceted single crystal.

A distinctive feature of the crystallization of small droplets, however, is that in its course rough regions (7), which usually develop at the initial stage of crystallization on larger ice spheres and hinder their complete faceting, in fact do not appear. This is explained by the fact that, during growth, changes in the surface structure of extremely small crystalline spheres have a somewhat different character. The flat growth layers nucleating on the surface of these spheres (Fig. 2), as they spread further, have limited (owing to the small size of the sphere) possibility for thickening (7) through coagulation with the underlying concentrically arranged steps. Therefore, up to the moment of their contact with neighboring layers, they retain high rates of tangential growth, differing little from the initial ones. At the same time, the increase in roughness in the intermediate regions lags substantially behind the spreading of the flat regions, and the intermediate regions become buried beneath the overhanging edges of the flat layers. As a result, the faces developing on the surface of small frozen droplets practically close up completely with one another, forming angles with sharp edges.

The data of our experiments are of interest to consider in connection with recent achievements in the study of the mechanism of ice formation in clouds under the action on them of particles of ice-forming substances. It has recently been experimentally established (9) that particles of silver iodide must mainly act as freezing nuclei. Noting that ice nucleation in this case occurs as a result of the collision of such particles with cloud droplets, the authors, however, do not indicate the specific mechanism of transformation of water droplets into faceted ice crystallites. The present work fills this gap and shows that the most important processes in this variant of the occurrence of ice in the atmosphere are the freezing of a supercooled cloud droplet with the formation of a single-crystal ice granule (^{10}) and the subsequent faceting of this frozen droplet.

The problem of the formation of the smallest single crystals from melt droplets has recently attracted the attention of a wide circle of investigators. Thus, for the purpose of studying the kinetics of martensitic transformations in Fe—Ni alloys (^{11}), a special method has been developed for producing very small ($< 37\mu$) spherical single crystals of an Fe—Ni alloy; for carrying out precise measurements of the intensities of diffraction maxima necessary for X-ray structural investigations (^{12}), a method has been proposed for growing small (100—1500μ), almost ideally spherical single crystals of beryllium, vanadium, zirconium, the intermetallic compound nickel—aluminum, and others. The idea of the formation from vapor of a solid phase through an intermediate liquid phase underlies the explanation of the mechanism of formation of a crystalline condensate on a neutral (amorphous) substrate during the deposition of molecular beams (^{13}); it is also extremely important for interpreting the results of investigations of the crystallization of melts of alkali halides (^{14}), especially in the many cases in which extremely small single crystals in the form of cubes are formed from droplets of molten salts.

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Fig. 1

Fig. 2

Fig. 3

Fig. 1. a —view of small supercooled water droplets condensed on a glass filament; filament thickness $\sim 30\,\mu$; b —prismatic ice crystallites formed from these droplets 4–5 min after the moment of their freezing.

Fig. 2. Example of a flat growth layer (right) formed on the surface of a frozen water droplet under supersaturation conditions; droplet diameter $\sim 550\,\mu$; the face is in the reflecting position under side illumination.

Fig. 3. Enlarged frames from a motion picture. The C-axis of the crystallite is situated almost perpendicular to the plane of the figure.

The experiments we have carried out give a clear picture of the mechanism of crystal formation in all such cases.

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