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MASS-SPECTRUM AND EVAPORATION MECHANISM OF AS-S GLASSES

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In the process of preparing amorphous chalcogenide films by thermal evaporation in vacuum there may be distinguished several main stages: a) forming different activated atomic particles on the evaporation surface and their transition into the gas-vapour phase; b) flying away of particles from the evaporation surface into vacuum; c) the evolution of their proper spatial structure and atomic structure by polyatomic particles of the vapor that is equivalent to their self-organization into separate clusters of one or other isomorphic form. However, taking into account the very low concentration of vapour phase particles under standard conditions of evaporation into vacuum the processes of their interaction with each other during movement of the vapour flow from the evaporation surface can be considered insignificant.

To our mind, the determining factor is just the first stage of evaporation, which sets the peculiarities of all subsequent stages. For this stage, there are two limiting mechanisms of forming the activated atomic particles possible. According to the first of these mechanisms the formation of such atomic particles occurs through gradual adjunction to the centre of nucleation of new individual atoms. This mechanism is very complicated to be analyzed. One should take into account the fact that the adjunction of each new atom may be accompanied by a complete restructuring of already existing activated complex into the new formation (including the collapse of the already existing nucleus) on the evaporation surface. The second limiting mechanism is much simpler: a certain activated cluster of corresponding chemical composition is formed in one physical act with a certain number of atoms on the surface. For As-S materials this fact is responsible for the appearance on the evaporation surface of a certain localized atomic formation with As_nS_m chemical composition. These clusters will be weakly bound to the evaporation surface and to some extent they will be isolated from the bulk of the substance.

Taking into account the nature of the investigated mass-spectra and calculated structures of the vapour phase clusters, we believe that the first phase of evaporation of As-S materials follows the second mechanism. Here are the main reasons for this conclusion.

1. The composition of the vapour phase of As-S materials is very complicated because it includes about three dozen of different As_nS_m clusters according to their chemical composition. To form such a broad spectrum of different activated particles by a step-by-step assembly from separate atoms the considerable time intervals are required that do not correspond to the real high rate of evaporation of As-S materials at a relatively low evaporation temperatures from 400 to 800 K.

2. For As_nS_m clusters the formations, quite large by the absolute value of formation energy, are typical that presets the high probabilities of their self-organization into separate atomic formations on the evaporation surface.

3. Strong covalent bonds are acting between atoms in the clusters formed on the surface. It defines small probabilities of their disintegration into separate fragments and, accordingly, sufficiently long lifetimes. Thus the formed clusters have significant probabilities of evaporation in the already finished form.

4. First principle calculations show that the lowest energies of formation are typical for those As_nS_m clusters, which do not have free valences. Others, quite stable clusters, have only one or, exceptionally, two unsaturated chemical bonds. These clusters interact weakly with the evaporation surface and can easily leave it while obtaining small activation energies of the evaporation process.