Numerical modeling of the structure of strongly disordered films

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A structural model is formulated in the zero energy approximation of condensing atoms. The mean density, number of nearest neighbors, and the characteristic cavity dimensions are determined and the pair correlation function is calculated. Calculation results are compared with results from an experimental investigation of the properties of metallic films deposited through a liquid helium layer [I. L. Landau *et al.*, JETP Lett. **38**, 545 (1983); S. É. Kubatkin and I. L. Landau, *ibid.* **46**, 102 (1987)].

Experiments on the deposition of metallic films through a superfluid helium layer[1,2] served as the impetus for these calculations. The electrical properties of such films were quite unusual, and obtaining at least some representation of their possible structure was of indisputable interest. It turned out that numerical modeling based on the simple interaction mechanisms of the atoms would help understand certain aspects of the phenomena occurring in actual experiments.

Before proceeding to an examination of the mathematical model we will first discuss the physical processess occurring on condensation of the atoms through a substrate below the liquid helium layer. An atom beam impacting the liquid helium[1)] decelerates to the critical superfluid velocity $v_s \approx 60$ m/s over a length of a few tens of angstroms, and the atoms will then travel nearly as free particles in a vacuum, since at such low temperatures it is possible to ignore their interaction with the quasi-particles in the helium.[3,4] These properties of a superfluid liquid make it possible, on the one hand, to sharply reduce the kinetic energy of the atoms and, on the other, to avoid mutual collisions and clustering of the metal atoms on the path from the fluid surface to the substrate. These experiments therefore have implemented conditions where atoms were condensed on the substrate and the kinetic energy of the atoms was hundreds of times smaller than in regular vacuum deposition. In this case it is important to remember that the interaction energy of the metal atoms exceeds by several orders of magnitude their interaction energy with the helium atoms; this fact suggests that the helium had no substantial influence on the formation of the film structure, and the presence of helium was not taken into account in the formulation of the model.

MODEL

We employed the hard-sphere model which is commonly used for such calculations. It was assumed in this case that each new atom traveling with random coordinates in the direction of the substrate is trapped upon first contact with any of the previously deposited atoms and can then move by maintaining contact with only this first atom. Such motion occurs up to the nearest point where the new atom makes contact with at least three other atoms already in the substrate.[2)] Such a position is stable, and its coordinates were fixed. Trapping of the atom upon first contact and the absence of film rearrangement with increasing film thickness are rather natural assumptions at low energy of the incoming atoms. Analogous calculations were carried out by Hensel *et al.*,[5] although they reported only the picture of the resulting structure, while we are also interested in its quantitative characteristics.

The "deposition" process was done through a substrate in the xy plane; here the initial random coordinates lay in the $0 \leqslant x \leqslant 20d$ and $0 \leqslant y \leqslant 20d$ planes (d is the atomic diameter) and moved along the normal to the substrate. There was a total of 11,000 atoms impacting this area. To avoid singularities from appearing along the boundaries of this region, a periodic continuation was used so that a point with the coordinate $\pm x_0$ corresponded identically to a point with the coordinate $20d \pm x_0$ (and analogously along the y axis).

The primary results were obtained when the first atoms impacted a smooth substrate, and if an atom landed in an empty position it became fixed at that point. If the atom impacted one of the preceding atoms, it then "rolled off" the atom towards the substrate along the shortest path; the atom either impinged on the substrate by making contact with one or two atoms, or stopped under the substrate if it turned out that the atom was suspended on three other atoms. Figure 1 shows all atoms ending up directly on the substrate as a result of this procedure.

The case where the first layer was a regular square lattice with a period a was also analyzed. Three variants were examined: $a = d$, $a = 2d/\sqrt{3}$ which corresponds to a body-centered-cubic structure, and $a = 1.25d$. Substantial deviations of all these cases from a smooth substrate were observed only in the first layers, while at distances greater than $3d$ from the substrate all the characteristics of the atom distribution were virtually identical (see Fig. 2).

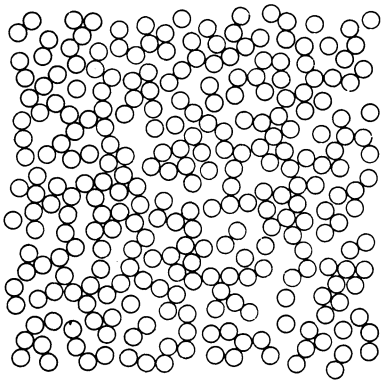

FIG. 1. The first layer of atoms on a smooth substrate. A total of 276 atom centers impacted a $20d \times 20d$ square.

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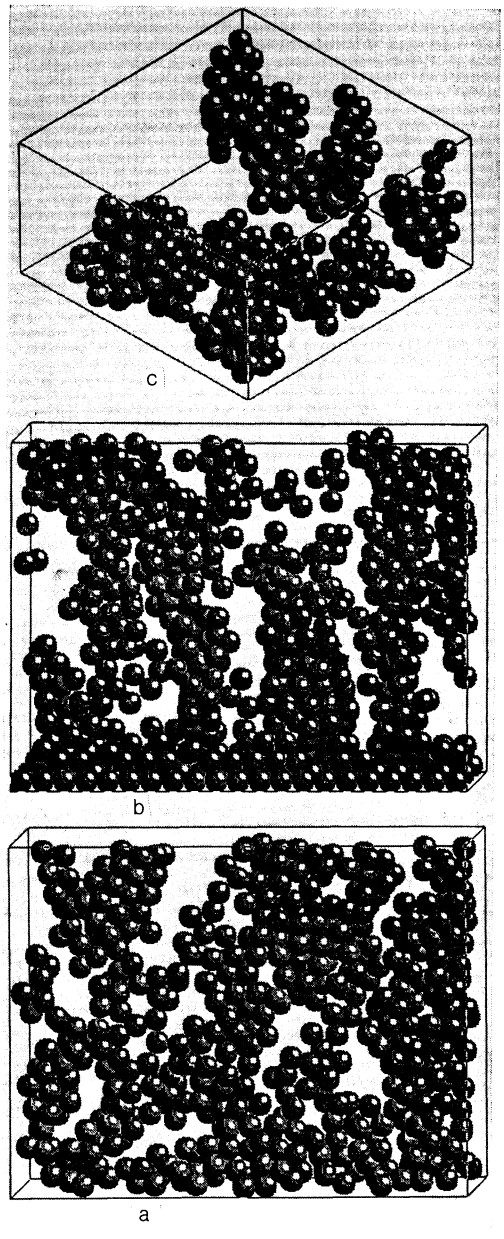

FIG. 2. The form of the structure (a, b) near the substrate: $0 \leqslant x \leqslant 20d$, $0 \leqslant y \leqslant 2.5d$, $0 \leqslant z \leqslant 15$ for the case of a smooth substrate (a), a square lattice with $a = d$ in the first layer (b), and the upper section of the film (c): $0 \leqslant x \leqslant 12d$, $0 \leqslant y \leqslant 12d$, $43d \leqslant z \leqslant 49d$. The figures only show atoms whose centers fall within the indicated parallelepipeds; hence certain atoms are supported on atoms that are not shown in the figures.

CALCULATION RESULTS

After the technique described above was employed to find the coordinates of all atoms, a number of characteristics of the resulting structures were determined. The results of a determination of the number of nearest neighbors are given below (the nearest neighbors are the atoms with a distance between the surfaces less than $10^{-3}d$).

Numbers of neighbors	3	4	5	6	7	8	9	10	11
Percent of atoms	5.7	12.1	19	24.3	20.2	12	5.2	1.3	0.2

The calculations were carried out for 20,000 atoms in three different implementations. The atoms from layers adjacent to the substrate and from the upper film layers were ignored. Only three atoms had 12 neighbors each from the sample, i.e., corresponded to close packing. We note that both the mean and largest probable number of nearest neighbors were equal to six.

The probability $w(r)$ of finding an atom at a distance less than r from any given atom (the pair correlation function) was also determined. Figure 3 is a plot of

$$f = \frac{d^3}{2^{5/2}\pi r^2}\frac{dw}{dr}$$

against r. The normalization was selected so that at large distances, when f is independent of r, the value of f yielded the film density with respect to close packing of the atoms. Thus the mean density of such a structure was quite small, only 0.44.

The peaks observed in the $f(r)$ relation in the range $d \leqslant r \leqslant 2d$ characterize the short-range order in the distribution of the atoms (the calculation results were averaged over 4000–6000 atoms with a step $\Delta r = 0.01d$). Peak 2 corresponds to the close packing defect shown in Fig. 4a, with $r_{AB} = 2d/3^{1/2}$ representing the distance between atoms A and B, and the low amplitude of the peak can be attributed to the fact that the stable cluster corresponding to this defect should consist of at least 10 atoms and its formation in a random structure has a low probability. The distribution of the atoms corresponding to peak 5 is shown in Fig. 4b, with $r_{AB} = 3^{1/2}d$. A more complex cluster is shown in Fig. 4c. Atoms A, B, and a (one of the atoms is not shown in the figure) form the double tetrahedron characteristic of close packing, while atoms C and D are stacking faults in this cluster; in this case the distances $r_{AB} = r_{aC} = r_{aD} = 2^{3/2}d/3^{1/2}$ correspond to peak 3 while the distances $r_{AD} = r_{BC} = 5d/3$ correspond to peak 4 while the distance $r_{CD} = 2^{5/2}d/3^{3/2}$ corresponds to peak 1. Three atoms along a straight line correspond to peak 6. We also note the specific form of peaks 5 and 6: an abrupt right edge and a gently-sloping left edge; such a form is due to the fact that the large distances for these combinations of atoms have small probabilities, while short distances are easily obtained by rotation of the outermost atoms about the symmetry axis (see Fig. 4b).

It is clear in Fig. 2 that the film contains a large number of cavities of various size; we estimated their maximum dimensions and it turned out that virtually none of the cavities in the film could contain a sphere of diameter $D > 2d$. Since the cavity dimensions were small, electrons could, evidently, easily tunnel through them and thus these cavities could not be considered dielectric inclusions in the metallic phase.

It is possible to determine the sizes of the film inhomogeneities from Fig. 2c, which shows one section of the upper part of the film.

DISCUSSION OF RESULTS

The most significant calculation result is the very low density (see Fig. 3) and the high degree of disorder of the resulting structures. It is important to note that this model is a limiting case of the maximum disorder that is possible in a hard-sphere system. In actual conditions the final binding energy released upon condensation of the atom can cause a rearrangement of an already existing structure. Such a rearrangement will naturally serve to increase the density, al-

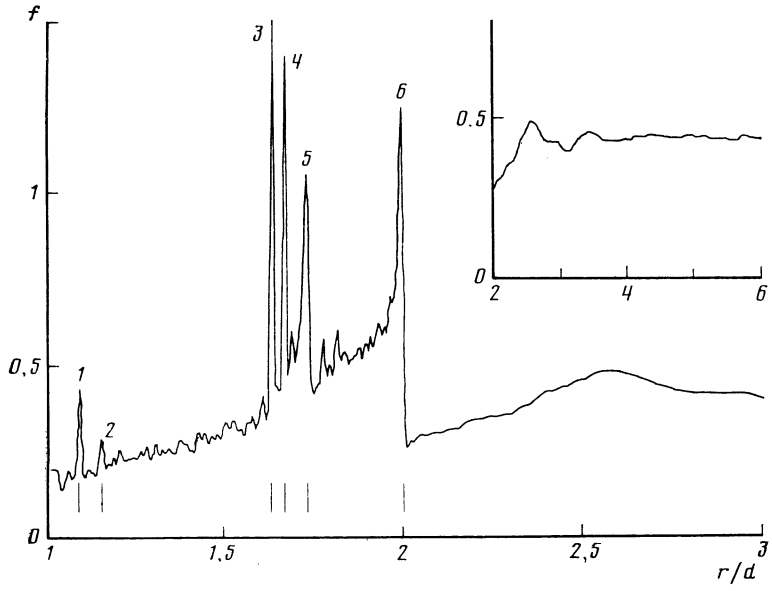

FIG. 3. Pair correlation function. The vertical strokes mark the calculated peaks. The inset shows the range of distances $r > 2d$ to a smaller scale.

though even estimating the magnitude of this effect is very difficult; all the more since in experimental conditions a substantial fraction of this energy may be transferred not to the film but to the surrounding helium. Below we will attempt to examine experimental results[1,2] by assuming that the structure of metallic films deposited through liquid helium is close to the model outlined above.

The low density and the extremely high degree of microscopic structural inhomogeneity of the films should enhance the electron-localization effects. The fact that a noticeable conduction in films deposited through helium sets in at thicknesses an order of magnitude greater than in vacuum deposition becomes clear from this viewpoint; this same fact should cause their anomalously high resistivity as well.

It is clear from Fig. 2 that certain atoms are not in very stable positions, as is also indicated by the significant percentage of atoms having only three nearest neighbors. It would be natural to expect structural changes for such atoms even with a slight rise in temperature; in the experiments irreversible changes in the resistance of cadium films began upon heating to 1.5–2 K, while in bismuth films such changes were already observed at temperatures near 1 K.

Annealing should cause a significant rise in density and a corresponding increase in conductivity, as was in fact observed in the experiments. It is important to bear in mind that the large structural inhomogeneities of the films evidently can not be eliminated by annealing and are more likely to increase, and be converted into dense and comparatively thick metallic granules distributed in regions where the film thickness was substantially smaller. This fact, which has already been noted in Ref. 1, was in all probability also responsible for the fact that even after heating to room temperature the resistance of the films remained significantly greater than the resistance of vacuum-deposited films of identical thickness.

In discussing the development of the concepts presented in the present study it would be very interesting to calculate the electrical conductivity of the film within the framework of this model, although, unfortunately, such calculations lie outside the scope of our capabilities. On the other hand in experiments on films deposited through liquid helium it would be necessary to change from resistance measurements to investigating their actual structure.

I wish to express deep gratitude to Yu. V. Sharvin, S. É. Kubatkin and E. G. Astrakharchik for discussion of the issues touched on in the present article.

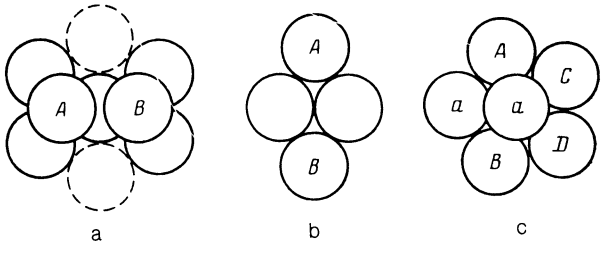

FIG. 4. Combinations of atoms that produce peaks in the correlation function (there is some inaccuracy in this figure: Atoms C and D are not in contact in reality and the distance between their surfaces is $\approx 0.075d$).

[1)]The experiments were carried out at temperatures $T < 0.5$ K; in this case the vapor pressure over the helium was negligible and it was possible to assume that the liquid helium bordered directly on the vacuum.

[2)]Physically such behavior corresponds to attraction between the atoms, which rapidly diminishes with increasing distance.

[1]I. L. Landau, S. E. Kubatkin, and Yu. V. Sharvin, Pis'ma Zh. Eksp. Teor. Fiz. **38**, 449 (1983) [JETP Letters, **38**, 545 (1983)].
[2]S. E. Kubatkin and I. L. Landau, Pis'ma Zh. Eksp. Teor. Fiz. **46**, 84 (1987) (JETP Letters **46**, 102 (1987)].
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[5]B. Hensel, M. Kippert, and H. Adrain, Jpn. J. Appl. Phys. **26**, 132, Suppl. 26-3 (1987).

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