

CENTENARY OF YA I FRENKEL'S BIRTH

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Ya I Frenkel' — man, scientist, teacher

V E Golant

This issue of *Uspekhi Fizicheskikh Nauk* celebrates the centenary of Yakov Il'ich Frenkel', an outstanding theoretical physicist of our country. His name is known to every physicist, and not just in Russia alone, because of his work in practically all branches of theoretical physics, and also because of his text books and monographs. Therefore, everyone, or almost everyone can say something about his or her personal meeting, face-to-face or otherwise, with Yakov Il'ich. I take this opportunity to do the same in this brief introductory note celebrating his centenary.

My contacts with Yakov Il'ich have come about because in 1944 I became a student of the Physicomechanical Faculty of the Leningrad Polytechnic Institute where Frenkel' gave all his courses of theoretical physics and held a chair. I was fortunate to be able to prepare a diploma thesis under Frenkel's supervision. It was concerned with a theory of vibrational – rotational spectra of nuclei. After graduation from the Leningrad Polytechnic Institute I worked at the 'Svetlana' factory on microwave electronics and gas discharges, so that I was able to appreciate Frenkel's classical work on the theory of microwave resonators. Finally, from 1958 I was at the A F Ioffe Physicotechnical Institute, with which the creative activities of Yakov Il'ich were linked for thirty years, I realised then the enormous influence that he had on the development of modern physics, particularly those fields which had been and are being studied at the Ioffe Institute, namely the physics of semiconductors, condensed matter physics, and nuclear physics. On visits abroad I have been frequently reminded of the high regard for Frenkel's work among his foreign colleagues.

Much of the work of Yakov Il'ich had become classical during his own lifetime. This includes studies of real crystals (Frenkel defects), kinetic theory of liquids, semiconductors, quantum-mechanical theory of electrical conduction in metals, and physics of magnetic phenomena.

There are however in Yakov Il'ich's heritage some contributions which have reached the status of fundamental work after his death, which is often the fate of pioneering studies. This applies to his 'soliton' paper of 1939 on the motion of dislocations (Frenkel solitons). Another example is the work on viscous flow in crystals (1945), which had become the scientific basis of powder metallurgy. I need not mention Frenkel excitons (1931). However, in the course of preparations to celebrate the centenary of Frenkel's birth we have learnt much new about how his work carried out in the twenties, thirties, and forties has gained a 'second wind' in our time. This is true of astrophysical research, including a theory of white dwarfs (1928), a theory of formation of real surfaces of crystals (1945), the work on the tunnel effect as applied to contact phenomena (1930) and to the physics of nuclei (1946).

My meetings with Yakov Il'ich in the late forties and early fifties gave me an insight not only into his professional capacity at lectures, seminars, and consultations which he readily provided to anybody who asked him at the Ioffe Institute. I have visited the hospitable home of the Frenkel' family and there, in an unpretentious and unaffected atmosphere, I have been able to appreciate the human qualities of Yakov Il'ich, his high intellect, artistic talent, bright wit, sensitivity, kindness, and readiness to help those in need.

At this uneasy time the name of Yakov Il'ich Frenkel', his selfless devotion to science, his civic courage, and high mental faculties give us an encouraging and inspiring example.

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Scientific session of the Division of General Physics and Astronomy of the Russian Academy of Sciences celebrating the centenary of Ya I Frenkel's birth

A scientific session of the Division of General Physics of Astronomy of the Russian Academy of Sciences, held on 23 February 1994 at the P L Kapitza Institute of Physics Problems of the Russian Academy of Sciences, celebrated the centenary of Ya I Frenkel's birthday. The speakers were as follows:

(1) A S Borovik-Romanov introduced the session;

(2) B P Zakharchenya spoke on "Discovery of excitons";

(3) R A Suris presented a paper "Ya I Frenkel' on real surfaces of crystals";

(4) V Ya Frenkel' described "The work of Ya I Frenkel' on nuclear physics."

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Discovery of excitons

B P Zakharchenya

Yakov Il'ich Frenkel' was an outstanding creator of models of physical phenomena in various branches of physics. Innovation was a distinguishing feature of his creative work. I became convinced of this as a student of the Physics Faculty of the Leningrad University when I had attended several of this lectures in the second half of the forties and the early fifties at the Polytechnic Institute. When I had compared him with my former professors at the Leningrad State University, I became sharply aware of the difference between the innovator and the archaists. Alas! Far from all the students have appreciated the difference and many of them were not happy with lectures by Yakov Il'ich.

One of the pinnacles of Frenkel's creativity has been the development of a theoretical model of an exciton, which is a quasiparticle that transports energy but not charge in a crystal. The existence of excitons has been confirmed experimentally for semiconductors, as well as for molecular, insulating, and ionic crystals. The concept of excitons is used to account for various physical processes (photoelectric phenomena, formation of radiation defects, luminescence, etc.) in crystals and polymers, including biological materials.

In 1931 and 1936 Frenkel' published his famous papers in which he introduced for the first time the concept of an exciton as an electron excitation wave in a crystal [1, 2]. In the thirties the description of the band states in crystals has been based on the Bloch scheme, derived by the Hartree – Fock method, which has no room for electron correlations. Frenkel' has realised this and chose the Heitler – London approximation to develop his theory of excitons. Therefore, his first papers have been concerned with crystals formed from rare gases and consisting of weakly interacting atoms. The wave function of an exciton regarded as an excitation wave (known as the translational wave function) used by Frenkel' is

$$\chi_p = \sum_k \frac{1}{n^{1/2}} \, \mathrm{e}^{\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{k}} \Phi_k \,, \tag{1}$$

where $\boldsymbol{p}$ is the quasimomentum, n is the number of atoms in a crystal, Φ_k is a many-electron wave function of an exciton†, which (if the exchange at the kth site of the localisation of the excitation is neglected) is

$$\Phi_k = \psi_1 \psi_2 \ldots \psi_k \ldots \psi_n \,, \tag{2}$$

where ψ_i ($i = 1, 2, \ldots, k-1, k+1, \ldots, n$) and ψ_k are the wave functions of the unexcited and excited states, respectively.

The selection rules for exciton optical transitions are derived in Ref. [2]: they specify that the exciton quasimomentum $\boldsymbol{p}$ is equal to the photon quasimomentum $\boldsymbol{q}$. This seems trivial to a modern physicist accustomed to the law of conservation of momentum in a solid, but it has not been simple to demonstrate this at the time that the exciton model was being developed. In view of the smallness of q, excitons form with a momentum close to zero, i.e. at the bottom of the exciton energy band. The corresponding lines in the optical spectra should be narrow. In the case of an electron and a hole formed by the absorption of a photon and moving independently with the quasimomenta $\boldsymbol{p}_\mathrm{e}$ and $\boldsymbol{p}_\mathrm{h}$ the selection rule is

$$\boldsymbol{p}_\mathrm{e} + \boldsymbol{p}_\mathrm{h} = \boldsymbol{q}$$

and the optical spectra consist of wide bands. For example, semiconductors (whose absorption coefficient is high) have a 'boring' (feature-free) edge of the continuous absorption if there are no excitons.

The first paper of Frenkel' on the subject, consisting of two parts, was called "On the transformation of light into heat in solids" because the principal aim of his theoretical investigation has been the search for a universal mechanism of nonradiative transitions in crystals excited by light. The

†Here, k denotes the position of an exciton in space, i.e. it is the radius (position) vector in Eqn (1). This is not a fortunate selection of the symbol, because k is usually employed to denote the wave vector.

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problem has clearly arisen from the emission of weak secondary radiation by crystals with a poorly controlled (at the time) impurity compositions and concentration and because of Frenkel's desire to develop a rigorous theory of nonradiative dissipation of the photon energy, similar to the existing (at the time) in physics of isolated atoms when the electronic excitation is transformed by inelastic collisions into the kinetic energy of particles. Recognising the difficulties encountered in the attempt to convert the energy of an optical photon into smaller portions carried by phonons, Frenkel' began to develop the idea of a strong interaction of an exciton with phonons on the assumption that the vibration frequencies and the equilibrium positions are different for the lattice oscillators in the ground and excited electron states. This idea, together with the concept of a 'stuck' or self-trapped exciton, considered also by Peierls [3], who participated in the development of the ideas under dissipation of energy via excitons, has served as a stimulus of many subsequent theoretical investigations.

As we now know, physicists have mastered the methods of controlled doping and reduction of the surface and bulk sources of nonradiative recombination, which has enables them to construct powerful sources of radiation and converters of light from crystals. It might then seem that Frenkel' has based his exciton concept on incorrect (in the thirties) information on weak secondary radiation emitted by crystals. This is not true, since in his work Frenkel' fully recognises the importance of excitons for the optical spectroscopy of crystals. According to Frenkel', the exciton energy bands are located within the band gap separating the ground state of a crystal from the continuous spectrum and the optical transitions obeying the selection rule stated above should give rise to narrow lines in the spectra of crystals.

It should be mentioned that Frenkel' also established the selection rules for one-phonon processes, so that the pattern of the optical spectra of crystals suggested by Frenkel' (narrow lines and their phonon replicas) has remained the most universal one at the present time.

We have mentioned earlier that Yakov Il'ich has criticised the Bloch scheme in which, according to Frenkel', there is no room for exciton states [2, 4]. However, there is a case in which the Bloch energy band scheme can be supplemented by exciton states. The relevant model had been proposed by Wannier [5] and Mott [6]. A Wannier–Mott exciton resembles a hydrogen atom or, more closely, a positronium. It is formed by an electron with an effective mass m_e and a hole with a mass m_h, which are bound by the Coulomb interaction in a medium whose relative permittivity is ε. The binding energy of such a quasiparticle is

$$E_n = \frac{\mu e^4}{2\hbar^2 \varepsilon^2 n^2}\,, \qquad n = 1, 2, \ldots, \tag{3}$$

where

$$\mu = \frac{m_e m_h}{m_e + m_h}$$

is the reduced effective mass of an exciton. The exciton radius is

$$a_{ex} = \frac{\hbar^2 \varepsilon}{\mu e^2}\,. \tag{4}$$

Eqn (3) is valid if $a_{ex} \gg a$, where a is the crystal lattice constant. This inequality is satisfied best in semiconductors. Excitons in semiconductor crystals are usually called large or large-radius excitons. However, this is only a terminological jargon and not an attempt to provide a fundamental distinction between the Frenkel' exciton, in which the excitation is localised within one unit cell, and the Wannier–Mott exciton.

A hydrogen-like spectrum, consisting of narrow absorption lines and described fairly well by Eqn (3), was first discovered by E F Gross working at the Leningrad Physicotechnical Institute, Academy of Sciences of the USSR. He made this discovery in 1951 [7, 8] when investigating a crystal of cuprous oxide (Fig. 1). A happy combination of the parameters of this semiconductor enabled Gross to find up to 11 terms (!) of a series of lines in the spectra of high- quality crystalline plates studied at 4.2 K. Yakov Il'ich knew from Gross of this outstanding experiment, but he did not manage to go to Gross's laboratory and to feast his eyes on the spectrum of his quasiparticle. He was already in poor health and he died in January 1952.

The observation of narrow lines in the spectra of semiconductors has been completely unexpected, because nothing like this has been observed for this class of crystals. However, even before the theoretical work of Frenkel', narrow lines have been observed by J Becquerel [9] in the spectra of crystals of rare-element compounds and I V Obreimov (Obreimow) has found such lines in the spectra of molecular crystals [10]. Wider bands, representing the structure of the fundamental absorption edge, have been reported by Hilsch and Pohl for alkali halide crystals [11]. Much later, years after the work of Frenkel', the narrow lines in crystals of the first type have been found to exhibit what is known as the Davydov splitting [12], demonstrating their exciton nature. The exciton origin of the bands in the spectra of alkali halide crystals has been proved by elegant experiments of Apker and Taft [13]. All these experiments, including that reported by Gross, have been carried out at approximately the same

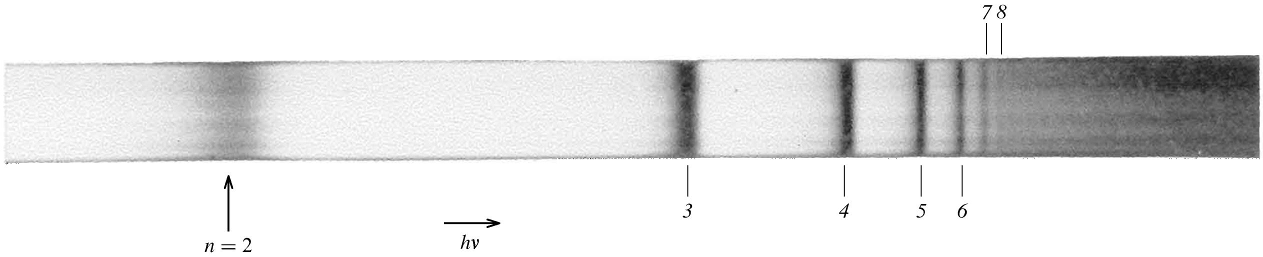

Figure 1. Optical spectrum of an exciton in cuprous oxide, showing the yellow–orange part of the visible spectrum.

time. However, the exciton discovered experimentally by Gross has become the most popular because of the colossal interest in semiconductors as materials used widely in electronics, electrical engineering, and power studies. The progress made in both theoretical and experimental investigations of the exciton effects in semiconductors has overshadowed the advances made in the study of excitons in insulating and molecular crystals. It should also be pointed out that the experimentalists who have observed the Davydov splitting in molecular crystals [14] have not identified directly the observed lines as the exciton lines, but have attributed them to hypothetical 'crystalline states'. This circumstance is usually delicately avoided, but we must remember the saying ''I have disliked the oval from my childhood and from that time I drew an angle instead''. Gross however believed firmly in the exciton nature of the hydrogen-like series of lines he discovered and he interpreted the experimental results accurately in the very first paper on the subject and his second paper was entitled ''The optical spectrum of an exciton'' [7]. The discovery of excitons in semiconductors and Gross's defence of the exciton interpretation of the experiments have been described in detail [8]. Here I would like to say that the main argument used by Gross in support of the exciton origin of his lines has been their narrowness, which follows from Frenkel's theory. This has not been grasped by many of the opponents, since in the early fifties few have understood that the band – impurity optical transitions do not usually give rise to narrow lines because the wave functions of the impurities are smeared out in the momentum space. Numerous experiments have proved the exciton origin of narrow lines at the edge of the continuous absorption by semiconductors.

At the very beginning of the exciton research, Abram Fedorovich Ioffe answering Gross's critics at one of the scientific sessions of the Division of Physicomathematical Sciences, said that even if the series of lines discovered by Gross and his colleagues are not due to excitons, their experiments are the starting point of a new branch of research which is the optics and spectroscopy of semiconductors.

In fact, both experimental and theoretical investigations of excitons in semiconductors carried out in the fifties and sixties have been the 'Sturm and Drang' (storm and stress) period in the spectroscopy of semiconductors, if we use the words of Goethe and Schiller. Gross and his colleagues in Leningrad, theoreticians in Kiev, American scientists J J Hopfield and D G Thomas, soon discovered a number of unusual properties of excitons in electric and magnetic fields, observed spatial dispersion effects in the exciton spectra, predicted and discovered an exciton polariton and exciton – impurity complexes, and determined the role of the excitons in the formation of the luminescence and photoconductivity spectra.

Attempts to form a boson (exciton) condensate have led to studies of the process in semiconductors under the conditions of intense laser excitation. An interesting physical problem of an electron – hole liquid in crystals has arisen in this connection. Under the conditions of relatively strong excitation of indirect-gap semiconductors it has been possible to observe many-exciton complexes with a structure surprisingly close to that of atoms with shells occupied by electrons and holes. The atomic-like nature of excitons in semiconductors has suggested the possibility of observation, in semiconductor crystals, of such phenomena as the optical orientation, the Hanle effect, interference between quantum states, and anticrossing of levels, usually found for isolated atoms. All these effects have been observed successfully in semiconductors revealing new properties specific to crystals.

Finally, the exciton states have played an enormous role in the spectroscopy of quantum-well structures fabricated by modern technological methods. In these structures the binding energy and oscillator strength of excitons are increased so much that the exciton features can be observed in room-temperature spectra.

Yakov Il'ich Frenkel' not only had discovered theoretically the exciton, but he also gave the name to his creation deriving it from the Latin word *exitare* which means 'to excite'. He probably did not expect the child of his mind to grow into a giant. When Frenkel's work was reported at Pauli's seminar, he characterised it as 'falsch' (wrong), since he frequently summarised the work of others by the simple summary: ''Das ist entweder falsch oder trivial'' (it is either wrong or trivial). Yakov Il'ich was happy with this assessment, because the later ('trivial') would have been more offensive [15]. Indeed ''None ... could divine to which side the conquest would incline'' (Samuel Butler, 1612 – 1980).

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Ya I Frenkel' on real surfaces of crystals

R A Suris

Throughout the years of his scientific activity Ya I Frenkel' has been investigating intensively the subject of molecular physics. I think that the range of problems in this field attracted Frenkel' because of its complexity and opportunities for almost artistic exploration: formulation and solution of problems requires creation of beautiful three-dimensional images.

I am unable to review here fully the whole creative inheritance of Ya I Frenkel' in this field. I even think that nobody would dare to attempt to give such a review. My aim is to recall one paper of Frenkel' in which he develops a model which is playing an important role in modern science of crystals and technology of semiconductor nanostructures.

We are speaking here of the paper "On the surface creep of particles on crystals and natural roughness of crystal faces" written during the time of evacuation to Kazan in December 1944. The English version of this paper was published in 1945 (*J. Phys. USSR* **9** 392) and the Russian version appeared in 1946 (*Zh. Eksp. Teor. Fiz.* **16** 39).

In this paper Ya I Frenkel' drew a very striking and literally three-dimensional picture of the shapes of surface bounding real crystals. The main feature of this picture is the representation of crystal faces not as solidified surfaces but as varying continuously and transforming under the influence of the thermal motion in the crystal. According to Frenkel', a crystal face is multitiered stepped surface with a relief that 'breathes' as a result of thermal displacements of atoms adsorbed on it.

Ya I Frenkel' started from the picture of a vicinal surface developed first by Ehrenfest in 1915 [1] and then by Yamada [2]. In a two-dimensional situation a vicinal face with the indices $(n, 1)$ (where n is a fairly large integer), i.e. a face close to the $(1, 0)$ face, consists of segments of $(1, 0)$ faces separated by steps (kinks) at intervals of n lattice periods (Fig. 1). Clearly, the energy of such a face exceeds the energy of the $(1, 0)$ face by an amount equal to the additional energy used in the formation of the kinks. If the energy of one kink is denoted by w, then the energy of the $(n, 1)$ face per unit length is

$$\sigma = \sigma_0 \cos\varphi + \frac{w}{a}|\sin\varphi| .$$

Here, σ_0 is the surface energy of the $(1, 0)$ face; a is the lattice period; $\varphi = \arctan(1/n)$ is the angle of tilt of the $(n, 1)$ face relative to the $(1, 0)$ face. The linear density N of the kinks, equal to the reciprocal of the distance λ between them, is related to the angle by the self-evident expression

$$N = \frac{1}{\lambda} = \frac{1}{an} = \frac{1}{a}\tan\varphi .$$

Obviously, the energy of the $(n, 1)$ face is independent of the direction of its tilt and, therefore, the expression for the energy contains the modulus of the sine function.

The energy of a vicinal face differs little, because N is small, from the energy of the base face. Hence, Frenkel' reaches a natural conclusion that thermal fluctuations give rise to random surface irregularities (Fig. 2). Here Frenkel' introduces in fact the concept of an elementary excitation, a kink, responsible for the surface roughness.

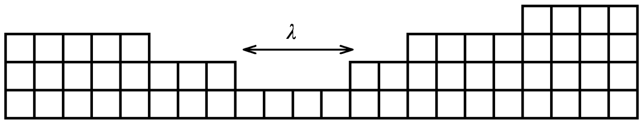

Figure 2.

Simple statistical considerations lead to the following expression for the average linear density of kinks on the $(1, 0)$ face:

$$N = \frac{2}{a}\exp\left(-\frac{w}{T}\right) . \qquad (1)$$

The factor 2 appears in the above expression because both positive and negative kinks appear with the same probability. For $w = 0.4$ eV and $T = 900$ K, we have $Na \approx 1/100$.

Therefore, under thermal equilibrium conditions the $(1, 0)$ face is straight only on the average. It represents a stepped line with randomly distributed kinks which in our example are separated on the average by a distance of 100 lattice periods. In view of the approximate statistical independence of the distribution of the different kinks, Frenkel' calculated the mean-square transverse displacement of the line representing the face. The result is very clear: in a section of length s the mean-square displacement

$$\overline{(\delta y)^2} = 2a^2Ns \qquad (2)$$

is proportional to s, in the same way as the mean-square displacement of a diffusing particle is proportional to time, $\overline{(\delta y)^2} = 2Dt$, and the role of the diffusion coefficient D is played by a^2N. For the parameters given above such a 'diffusive' displacement of the line per 1 mm of its length is considerable and amounts to about 10^{-5} cm.

Let us now consider what is a two-dimensional face with kinks? Frenkel points out that the above picture describes a step with kinks on a crystal face (Fig. 3). The concept of such a surface structure had been introduced earlier by Kossel [3] and Stranski [4] but Frenkel' was the first to draw attention to the need to regard thermal fluctuations as the source of the appearance of kinks and estimated their density, which varies because of thermal fluctuations of the surface of a crystal.

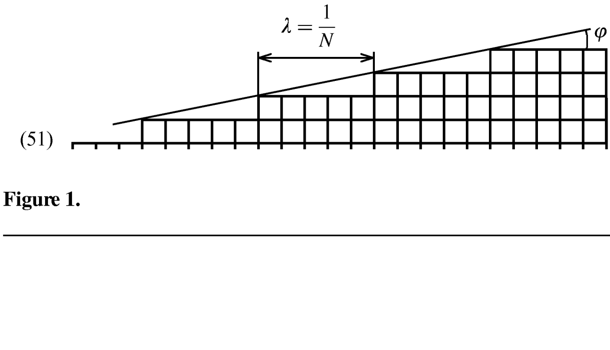

Figure 1.

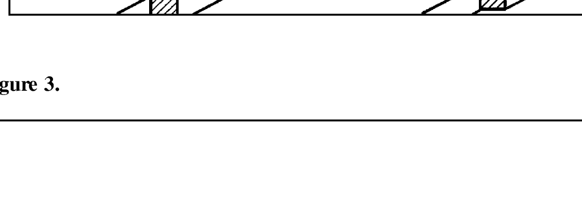

Figure 3.

The next extremely important point of Frenkel's paper is the question of the processes that determine the dynamics of changes of the shape of the surface. Frenkel' gives an extremely clear and convincing justification of the proposed picture: he assumes that changes in the surface roughness with time are determined by the processes of diffusion of atoms bound to the surface of a crystal (nowadays they are called adsorbed atoms or adatoms).

Frenkel' introduces the concept of one- and two-dimensional gases of adatoms. A one-dimensional gas is formed by those adatoms which are captured by a 'potential trench' near a step on the crystal surface (Fig. 4). These adatoms diffuse along the step and can either join a kink or 'evaporate' into a two-dimensional gas of adatoms moving along the surface of a terrace. The atoms in this gas can either drop into a trench near a step or evaporate into three-dimensional gas above a crystal. In the simplest model of Kossel, in which a crystal is formed by atoms of cubic shape bound to one another along their faces, the activation energies for the transitions

kink → trench,

trench → terrace,

terrace → three-dimensional gas,

are equal to the same binding energy w. However, the energies of the kink → terrace and trench → three-dimensional gas transitions are $2w$. The energy 'price' of the kink → three-dimensional gas and step → terrace transitions is $3w$. The energy needed to detach atoms from a step into the three-dimensional gas and from a layer forming a terrace onto the surface of the terrace is $4w$.

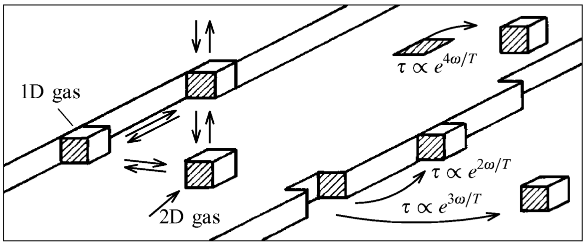

Figure 4.

Since the lowest activation energy is that of the three processes listed in the column above and since the activation energy of the diffusion of adatoms is known to be less than the detachment energy w, Frenkel' draws the conclusion that it is these processes together with surface diffusion that determine both the kinetics of fluctuation of the shape of a crystal as well as the growth processes and that the 'active centres' in both cases are the kinks and the steps. Kossel also regards kinks as the growth centres. However, he assumes that there is a direct exchange of atoms between the kinks and the three-dimensional phase. It follows from Frenkel's reasoning that, at least in the case of growth of a crystal from the gaseous phase, this is invalid.

Let us quote from Yakov Il'ich's article under discussion: "The proposed generalisation of the Kossel theory not only gives a more correct representation of the processes that alter the *volume* of a crystal, but also accounts for the possibility of changes in its *shape*, which are unrelated even to temporary changes in the volume, i.e. which do not occur directly via evaporation (or dissolution) and crystallisation, but by surface creep or surface diffusion of atoms in a crystal. Such a surface mechanism of a change in the shape of crystalline bodies has recently been proposed by P I Lukirskii to account for his experiments on multistage formation of faces on the surface of a rocksalt crystal initially machined to form a sphere."

It should be explained that on 6 October 1944 P I Lukirskii sent this paper "Experiments on rocksalt single crystals" for publication (it appeared in 1945 [5]). Lukirskii showed that a sphere, machined from a rocksalt crystal, became faceted as a result of annealing at temperatures 720 – 760 °C for several hours, changing into a 48-facet figure. Weighing and annealing under equilibrium vapour pressure conditions and in the absence of such conditions led Lukirskii to the conclusion that evaporation did not play a significant role. He concluded that the main process was a diffusive creep of atoms which minimised the surface energy.

Lukirskii's paper also attracted the interest of L D Landau. In 1950 in a collection of papers celebrating the 70th birthday of A F Ioffe [6], Landau published a paper "On the equilibrium shape of crystals" in which, leaving aside the problems of kinetics, he used a thermodynamic analysis allowing for the interaction of steps with one another to show that the equilibrium shape of a crystal should consist of a small number of low-index faces. In conclusion, Landau thanked Lukirskii for drawing his attention to this problem.

In 1951, Burton, Cabrera, and Frank developed, in their famous and continuously cited paper [7], a detailed theory of the equilibrium structure of the surface and growth of crystals. Naturally there are several references to the paper of Ya I Frenkel' discussed here and then frequently in a disputatious manner. There is no need to consider the details of this dispute. It is important to stress that the paper of Ya I Frenkel' which we are discussing here provides a picture of a 'live' and continuously varying structure.

I shall conclude with a few comments on the current state of the problem. The availability of the ultrahigh vacuum technology, molecular-beam methods for the growth of crystals, and refined methods for the investigation of surfaces with atomic resolution have been responsible for the colossal progress made in this field. It is now possible to monitor the growth of crystals to within a small fraction of an atomic layer and to observe the state of the surface directly during growth. Remarkable opportunities have been opened up by the methods of electron diffraction [8] and scanning tunnelling microscopy [9].

All this has been stimulated by the development of semiconductor microelectronics and optoelectronics. Quantum semiconductor heterostructures, consisting of regions of nanometric size built into one crystal and characterised by different electron and hole spectra, are being used in the fabrication of ultrafast electronic circuits and semiconductor injection lasers for fibre-optic communication lines which are revolutionising the information technology. These structures utilise the special features of the wave functions and energy spectrum of carriers, which are established because of their interaction with heterojunctions that separate the parts of semiconductors consisting of chemically different components. In structures made by the methods of molecular beam epitaxy these heterojunctions play the role of 'instantaneous photographs' of the surface with all its irregularities at the moment when the atomic composition of the beam is altered. Since carriers move in these structures in regions of dimensions amounting to several atomic layers, the irregularities of their surfaces have an extremely strong influence on their electrical and optical properties. Carriers can 'see' the surface of a crystal frozen into the structure, the properties of which have been described by Ya I Frenkel'.

Real crystals used to grow such structures — mainly the compounds $A^{III}B^{V}$, $A^{IV}B^{VI}$, $A^{II}B^{VI}$, as well as silicon and germanium — cannot be described by the simple Kossel model. The (1, 0, 0) surface is used most frequently for growth. There is an important feature due to the predominantly covalent nature of these crystals and the strong directionality of their chemical bonds. Two steps on the (1, 0, 0) surface are inequivalent. It is evident from Fig. 5 that all the covalent bonds at the edge of a [110] stop are directed along the step and, consequently, the formation of kinks requires breaking of chemical bonds. Therefore, the energy of a kink is approximately equal to the energy of one bond. On the other hand, all the bonds at the edge of a $[1\bar{1}0]$ step are already broken and the formation of a kink does not require additional bond breaking, so that its energy is much less than for steps of the first type. Therefore, the nucleus of the next layer, which is an island bounded by a closed step, should assume a shape strongly elongated along [110] so as to minimise its own energy [10].

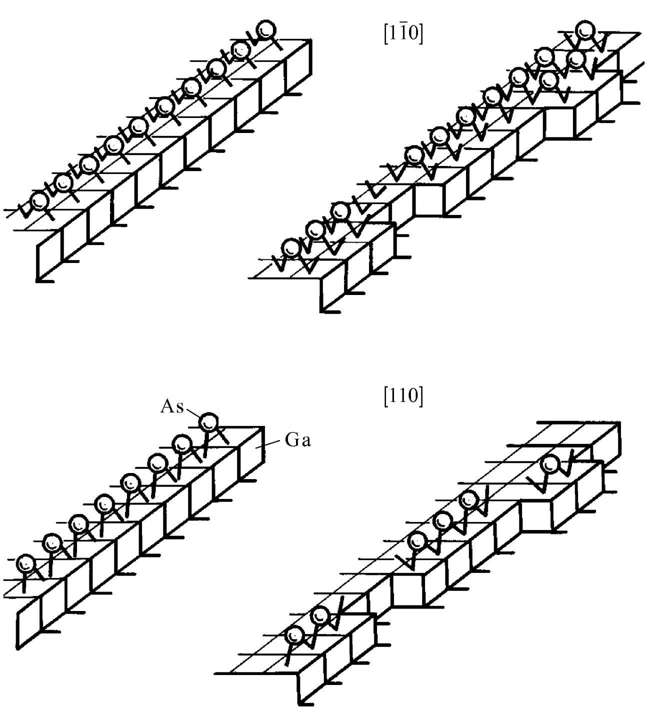

Figure 5.

This directionality of the bonds leads to a strong anisotropy of the irregularities of the steps predicted by Frenkel' and described by Eqn (2). Since the energy of formation of kinks on a $[1\bar{1}0]$ step is considerably less than on a [110] step, the average density of kinks given by Eqn (1) on $[1\bar{1}0]$ is much higher than on [110]. Consequently, in accordance with Eqn (2), a $[1\bar{1}0]$ step should be much more broken up because of thermal fluctuations. All this is supported by direct observations carried out with a scanning tunnelling microscope [11].

Finally, I shall give an example of the nontrivial consequences that may result from Frenkel's model of crystal growth because of the surface diffusion of atoms. Let us consider a vicinal surface on which an atomic beam is incident (Fig. 6). The atoms captured by this surface diffuse through the steps, are captured by kinks, set each step in motion, and cause a crystal to grow. If we assume that an atom is captured by a step on approach to it along the lower terrace with a greater probability than in the case of approach along the upper terrace (and there are physical reasons to assume why this is correct), then a system of steps form periodic structure which is stable against deviations from periodicity [12]. In fact, if one of the terraces is smaller than its neighbour, then because fewer atoms reach it from the gaseous phase than those arriving on other terraces, the step rising above it (shown on the right in Fig. 7) moves at a velocity less than the left-hand terrace, because the motion of the latter is due to a high diffusive flux collected from the adjacent (Fig. 7) left-hand terrace whose length is greater. Consequently, the length of this shortened terrace increases until it becomes comparable with the other terraces. Such 'self-organisation' of the surface has been suggested for the growth of structures with a one-dimensional electron gas (quantum wires). However, it has been shown [13] that steps are unstable under flexural fluctuations. This can be avoided if there is a surface of the type (n, m, 1), where n and m are fairly large numbers [14]. On a surface of this kind the steps have a nearly periodic system of kinks of one kind which — because of the asymmetry of the capture of atoms by kinks are similar to the asymmetry of the capture by steps described above — is stable against departures from periodicity and this means that the distribution of steps is also stable.

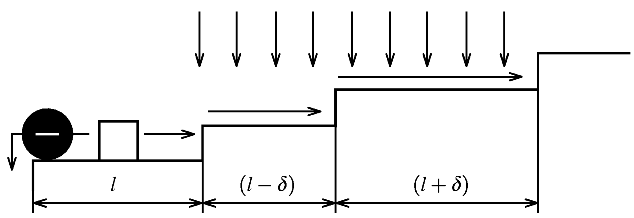

Figure 6.

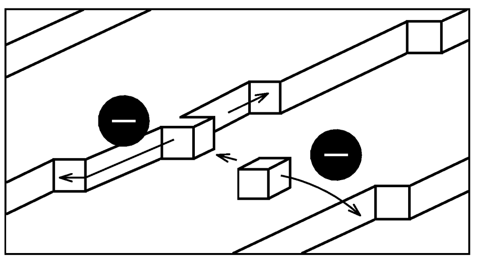

Figure 7.

It is appropriate to end with a quotation of a biographical sketch written by A F Ioffe and included in the second volume of *Sobraniya Izbrannykh Trudov* (Selected Works) of Ya I Frenkel' (1958): ''It is not easy to select what is most valuable from the rich scientific heritage of Ya I Frenkel'. Some of the results have joined the 'golden treasury' of science; about many others the last word has not yet been said, although undoubtedly they have played an important role in the history of physics. The significance of many of the papers has become evident only after Frenkel's death and these have predicted the development of the field in question, but have not been recognised when they appeared in print.''

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The work of Ya I Frenkel' on nuclear physics

V Ya Frenkel'

The history of nuclear physics in our country can be divided arbitrarily into several stages. The first and the longest, with few important events in Russia, began in 1896 (which is the year of the discovery of radioactivity and the first work done on the subject in Russia) and extended up to the 'miracle year' 1932, so rich in outstanding events. In the USSR this year was marked above all by the development of a proton–neutron model of the atomic nucleus (D D Ivachenko), the beginning of the work on the construction of a cyclotron (L V Myovskii, I V Kurchatov, et al.), and the first studies of the physics of nuclei at the Leningrad and Kharkov Physicotechnical Institutes. The second stage began in 1932 and lasted to the beginning of 1936. In February of 1936 Niels Bohr put forward the idea of a compound nucleus, which provided a fresh impetus to theoretical studies of nuclear physics; the third stage therefore covered 1936–1938. The fourth stage began with the publication, in January 1939, of the famous paper of O Hahn and F Strassmann on the fission of uranium. It basically ended in the USSR, in June 1941 or — bearing in mind that during the first year and a half of the Great Patriotic War there was practically no work in nuclear physics — in the last months of 1942. Under the direction of I V Kurchatov the USSR began organisational and then scientific and technical work on the development of atomic weapons. This was also the starting point of the fifth stage of investigations. In August 1945, after the atomic bombs fell on Hiroshima and Nagasaki, these investigations were greatly intensified. This stage ended in 1946, the year of successful commissioning of the first Soviet nuclear reactor. The sixth stage lasted from 1946 to 1949, the latter being the year in which the Soviet atomic bomb was constructed and tested.

Ya I Frenkel' participated in the research in this physics field during the first, third, fourth, and fifth stages. His participation is summarised here only briefly, for lack of space.

1. In 1916, by the time Yakov Il'ich Frenkel' graduated from the Physicomathematical Faculty of the Petrograd University, he had already written and sent for publication a paper on an electrical double layer on the surfaces of solids and liquids. In this paper, presented at a seminar on new physics led by A F Ioffe (at the Polytechnic Institute) Frenkel' was able to account correctly, on the basis of the Rutherford–Bohr planetary model of an atom, for the characteristics of the contact phenomena that have been investigated experimentally by A Volta. It would seem natural to expect Yakov Il'ich to present this paper (published simultaneously in Russia and in England) as his diploma work for a Master's degree. However, there was a tradition in the former Physicomathematical Faculty at the time (mentioned by A F Ioffe and also by I E Tamm [1]) that the students graduating from the faculty were recommended to write review-type diploma theses.

Frenkel' followed this unwritten rule. In three papers published in 1917 [2] he gave a detailed review of the state of the atomic (nuclear) physics at the time, particularly the physics of radioactivity. Two other reviews published in the same journal [3, 4] are not comparable with Frenkel's work either in respect of completeness or the depth of presentation of the material. I remember that when the collected works of Ya I Frenkel' were being prepared for publication both Ya G Dorfman and A G Samoilovich proposed, on the basis of the merits of Frenkel's review [2], to include it in the second volume (selected papers). The editorial board did not agree because this volume already had 600 pages.

Yakov Il'ich had a remarkable professional memory so that the material which he organised in a systematic manner was remembered by him permanently. This probably helped him later to rapidly join as a participant in the relevant research.

In Petrograd, in the first half of the twenties, Frenkel' found himself immediately the only breadwinner for a big family (wife, son, his own and wife's parents, and aunts). I think this was the reason why he was then quite active in the science popularisation field, particularly in editing a number of books. They included the book of a well-known German engineer Hans Gunter *Technical Dreams* [5]. Yakov Il'ich wrote a fairly long appendix to this book. Gunter discussed energy sources and it is appropriate to quote here a passage from Frenkel's appendix to this book [5] headed ''Does interatomic [nuclear] energy exist and can it be utilised?'': ''Fusion of hydrogen to form helium should release the excess of the hydrogen atoms. Obviously if we were able to induce such fusion, we could forget all our energy troubles. Although the energy we are speaking of represents only 0.8% of the virtual energy [mc^2] which has been the subject of many dreams, this small energy is real and we can at least hope to master it.''

This was written in 1925. This was seven years before the discovery of the neutron and four years before the first work (by F G Houtermans and R d'E Atkinson) on the nuclear origin of the stellar energy. However, the atomic masses of elements in the Mendeleev periodic table were already well known.

Let us now devote a few lines to Frenkel's attempt to explain the nature of the strong interaction of particles in a nucleus which prevents its decay. He based his explanation on the magnetic interaction of particles in a nucleus which prevents its decay. He based his explanation on the magnetic interaction of forces between protons and (intranuclear) electrons [6]. This work is now only of historical interest, but it had been favourably noted in the famous Bakerian lecture by Rutherford in 1927.

2. Frenkel' began to investigate systematically the physics of nuclei under the stimulus of the brilliant paper of N Bohr on the theory of the compound nuclei (published in *Nature* on 29 February 1936 [7]). In March of the same year I E Tamm summarised this work in a review which he presented at a meeting of the Physics Group of the USSR Academy of Sciences in Moscow [8]. Directly during the discussion after Tamm's paper, on the same day, Yakov Il'ich proposed to extend, to the behaviour of excited (because of the absorption of a neutron) nuclei, the ideas of statistical physics, noting that the number of nucleons in heavy nuclei is sufficiently large to justify this approach [9]. Speaking in this discussion Frenkel' also introduced the concepts of the temperature of a nucleus, and of the evaporation and condensation of neutrons in nuclei. His contribution to the March 1936 session was later included in a collective book *Neitron* (Neutron) [10]. His ideas were accepted immediately by the scientific community: one can mention here particularly the well-known paper by Bohr and Kalckar [11]. The correspondence between Frenkel' and Bohr, which dealt with these topics, is reproduced in Refs [12, 13]. A detailed statistical theory of the atomic nuclei was developed by Frenkel' a year later [14]. It is worth mentioning the elegant analogy used between the process of alpha decay and sublimation of NaCl molecules from the rocksalt lattice that does not contain ready-made molecules of this kind. The statistical approach to the behaviour of atomic nuclei and their properties has been subsequently developed by L D Landau [15] and V Weisskopf [16].

3. The first issue of *Naturwissenschaften* for 1939 saw the publication of the classic work of O Hahn and F Strassmann [17]. They discovered fission of uranium by neutrons into two parts approximately equal in respect of the atomic masses. There has been controversy on the subject when and how the results of Hahn and Strassmann had become known to Soviet physicists, particularly those working in Leningrad. A personal letter of F Joliot-Curie to A F Ioffe, received in Leningrad at the end of 1938 has been mentioned (in his reminiscences F Strassmann reacted ironically to this premonition ahead of the date of publication of Ref. [17]). A recent discovery, in the Niels Bohr Archive in Copenhagen, of a letter to Bohr from Frenkel'† gives a clear answer as to when the results of the German scientists became known at the Physicotechnical Institute, which at that time was the centre of nuclear physics research.

I shall now cite a passage from this letter of Frenkel' dated 12 March 1939:

''Dear Professor Bohr! Near the end of February we first became aware of the discovery of a new type of fission of uranium nuclei (from a paper by Joliot in *Comptes Rendus* [18] and somewhat later from the American *Science News Letters* [19]). Several days later I developed a theory of this process which seems identical in its main features with that proposed by Frisch and Meitner (briefly, we are speaking here of a reduction of the surface tension due to an electric charge) and particularly with your letters in *Nature* and *Physical Review*.

''Since I developed mainly the quantitative aspects of the problem, which might be missing in the work of other authors, I am sending my paper to a new Soviet journal [the *Journal of Physics of the USSR*], which is intended to replace *Physikalische Zeitschrift der Sowjetunion* and *Technical Physics of the USSR*. This journal will be published by the Academy of Sciences. However, I am afraid that my paper will appear in print after a great delay (although I was told that the first two issues for the year are already in press). In any case I think you will be interested to read my paper before its publication. I am therefore sending you the text of this paper. I would be glad if it were possible to publish it, or some parts of it, in *Physical Review* (as a Letter to the Editor).''

On the same day Yakov Il'ich sent one copy of the English version of his paper to Prof. E Hill (who was his assistant at the time when Frenkel' taught theoretical physics at Minnesota University in the USA). Hill produced a summary of the first part of Frenkel's paper, which contained a simple calculation of the fission reaction energy.

In accordance with Frenkel's wish, this part of the paper was published in *Physical Review* [20] and the date of submission of the paper was given as 12 March 1939. Another copy of the same paper was sent by Frenkel' to Niels Bohr; he probably knew that Bohr was away in the USA from 16 January. We can see later that Bohr received in time the manuscript of Yakov Il'ich's paper.

It is worth mentioning here that the first calculations of the energy of the fission of uranium by slow neutrons were carried out independently by different physicists as they learnt of the paper of Hahn and Strassmann. Table 1 gives the relevant information on the publication of these calculations.

Frenkel's paper in *Physical Review* [20] was subsequently included in a collection published on the fortieth anniversary [24] of the paper by Hahn and Strassmann.

An undoubtedly more important step forward was made by the mathematical development of the theory of fission. Its description was given by Peierls [25]. I shall therefore limit myself to brief comments. The electrocapillary theory of fission of heavy nuclei was developed independently by

†I take this opportunity to express my gratitude to Dr F Ocerude, Director of this Archive for hospitality during my stay in Copenhagen, and to Dr H Levi for acquainting me with two letters from Frenkel' to Bohr (dated 1939 and 1946), which up to that time had not been included in the general catalogue of Bohr's correspondence.

Table 1.

Author	Journal	Received	Published
L Meitner and O Frisch	*Nature* (*London*) [21]	16 January 1939	11 February 1939
C F von Weizsacker	*Naturwissenschaften* [22]	9 February 1939	24 February 1939
E Feenberg	*Phys.Rev.* [23]	14 February 1939	11 March 1939
J Frenkel	*Phys.Rev.* [20]	12 March 1939	15 May 1939

Ya I Frenkel' [26, 27], on the one hand, and by N Bohr and J A Wheeler [28], on the other. The absorption of a neutron by a uranium nucleus gives rise to oscillations of its shape which grow and can result in fission. A calculation of such fission was carried out by Frenkel' and in greater detail by Bohr and Wheeler. The latter [28] point out that the results given in the relevant part agree with those obtained by Frenkel', which were available to these authors from the manuscript which Frenkel' sent them. The chronological sequence of publications is given in Table 2.

Table 2.

Author	Journal	Received	Published
Ya I Frenkel'	*Zh. Eksp. Teor. Fiz* [26]	14 April 1939	No. 6 (June) 1939
J Frenkel	*J. Phys. USSR* [27]	15 March 1939	No. 2 (March – April) 1939
N Bohr and J A Wheeler	*Phys. Rev.* [28]	28 June 1939	1 September 1939

It should be pointed out that in 1939 Frenkel' together with V Cherdyntsev of the Radium Institute published one more paper on the theory of nuclei (statics of the nuclei) [29]. The paper is entitled "On the gas model of an atomic nucleus". In particular, this paper gives a theoretical dependence — in very good agreement with the experimental results — of the isotopic number $I = A - 2Z$ of a nucleus, with an atomic mass A and a charge Z, on the value of Z. Moreover, the numbers of isobars and isotopes are determined and the problems of the density and thermal expansion of atomic nuclei, etc are covered in this paper. All these results follow from the concept of a nucleus as a gas of its component particles at absolute zero (this demonstrates a direct genetic relationship between this paper and Ref. [30]). In 1936, Yakov Il'ich called his paper "On the solid-body model of heavy nuclei" [31] because he used the Einstein formula for the quantum theory of specific heat of solids (1907) to describe the energy of particles in nuclei. Frenkel's 1939 work on nuclear-liquid oscillations [26, 27] was based on the liquid-drop model (the history of this model, including the contributions of G A Gamow and P Ehrenfest, is given in Ref. [32]). Therefore, Frenkel', as he has done in his other work on theoretical physics, described nuclei employing three different (and, at first sight, mutually exclusive) approaches, which in fact are in no way contradictory. In this case we can speak, by analogy, of these approaches as three projections of an object which is an atomic nucleus and which make it possible to get a relief representation of the object. Finally, let us mention that the development of his work [26, 27] led Frenkel' to a study of the spectroscopy of atomic nuclei [33] in which he made progress in a mathematical theory of electrocapillary oscillations of a charged nuclear liquid.

4. In the history of science, and in science itself, the very formulation of specific problems may be important and meaningful. In this connection it is worth mentioning the question of what considerations and principles were followed by I V Kurchatov when he was forming his team at the beginning of 1943. At the time under consideration among the several theoretical physicists who have made important contributions to nuclear physics, it seems that the most significant results have been obtained by Frenkel', as demonstrated by the above list of his investigations in 1936, 1937, and 1939. Therefore, it is at least strange that he has not been involved in the work on the atomic bomb, in contrast to Ya B Zel'dovich, who began working from the very start in 1943, and L D Landau and I E Tamm, who became involved later in the late forties or early fifties.

This question becomes even more interesting in the light of a document obtained from the Russian Scientific Centre 'Kurchatov Institute' [34]. A copy of this document was supplied to me with the kind help of Prof. I N Golovin. It is a letter from Ya I Frenkel' to I V Kurchatov dated 22 September 1945. By this time the work on the construction of atomic weapons was going on full blast in the Soviet Union and the first nuclear reactor had already begun working. The Americans had dropped atomic bombs on the cities of Japan.

There is one more date with which one should compare the time of the letter form Frenkel' to Kurchatov. In the second half of June 1945 the 220th anniversary of the USSR Academy of Sciences was celebrated solemnly in Moscow. It is quite obvious that this was not a 'round' date and was selected to demonstrate the importance attached by the USSR Government to science. The anniversary celebrations were timed to coincide with the victory over Fascism. This was the first international meeting after many years and it seemed to signify restoration of international scientific contacts, broken off basically in 1937 (when the Third All-Union Conference on the Physics of Nuclei took place in Moscow and at which foreign physicists were present for the last time before World War II).

The participants of the anniversary session of the Academy were invited to the historical Victory Parade in Red Square (24 June 1945). Among dozens of foreign guests there were also outstanding physicists from France: the husband and wife F and I Joliot-Curie, P Auger, and F Perrin.

This document shows that a confidential meeting took place between Frenkel' and Joliot-Curie (they knew each other from the time of the foreign trips of Yakov Il'ich, and they met later in Leningrad where Joliot-Curie came in 1933 and 1936). The topic discussed is the subject of a detailed note sent by Frenkel' to Kurchatov. It begins as follows:

"In his talk with me Prof. Joliot told me the following about the method used by Americans to make atomic bombs. Instead of separating the light isotope of uranium, it has proved simpler and more practical to prepare uranium-239 by neutron irradiation of the usual uranium. An important role in the preparation of this isotope is played by heavy water and graphite (of a special kind). The explosion is ignited by spontaneous decay of uranium. The 'waste' energy released by uranium during bomb manufacture is used to drive machines of 15 000 horsepower.

''I found it awkward to ask Joliot for details, since I assumed that he himself will tell all he knows in the course of the help he proposed to give Soviet physicists working on the problem of uranium by way of consultation or joint work.

''Since there has been a delay in getting permission to involve Joliot in this work, I feel it is appropriate to present briefly the ideas and considerations which are based on the brief information obtained from Joliot, and also on what has been reported by foreign newspapers and radio'' (Ref. [34], p. 1).

The information at Joliot's disposal was obtained by him back in the war years. In a paper on ''Atomic energy in France'' [35] he wrote: ''During the occupation of France I was told of the progress made [in the USA and England] by one of the Free-France fighters who came to France with instructions from the French Government in London'' (Ref. [36], p. 500). Joliot-Curie knew of German scientists on atomic weapons [36] back in 1940 when his laboratory in Paris was visited by high-ranking German physicists. Their questions readily revealed that some work on the development of atomic weapons was going on in Germany. It follows from this paper of Joliot-Curie [35] that several weeks after the liberation of Paris he went to London and met there the French physicists who worked in the military establishments of the Allies (Ref. [35], p. 500) and obviously told him in general terms about their research.

It is still not clear whether Joliot-Curie met any of the physicists working directly on the uranium project in the Soviet Union. Let us cite a passage of the memoirs of B G Kuznetsov about Joliot-Curie: 'I first saw Frederic Joliot-Curie on a summer morning in 1945 on the stairs at the Presidium of the Academy of Sciences Building. He was looking for me to pass on the best regards from Ya I Frenkel' from Leningrad and also to tell me something on the advice of Yakov Il'ich'' (Ref. [37], p. 74). (Kuznetsov, a well-known historian of science, held in 1941 – 1945 a responsible position in the Presidium of the USSR Academy of Sciences.)

In his letter (detailed note) Frenkel' tells Kurchatov about his ideas on significant (from his point of view) potential ways of developing the work on the bomb construction. They include the proposal to use plutonium instead of uranium-235 and then the ideas on the construction of reactors (heterogeneous structure, choice of moderators, particularly for the preparation of plutonium and industrial energy generation). These are, at least very approximately, ways along which scientists and engineers were proceeding at this time in the USA and in the Soviet Union. This detailed note includes other suggestions and opinions which are evidently incorrect (for example, Yakov Il'ich suggests on the basis of some considerations that the explosion in a uranium bomb with a plutonium detonator is not of chain but of thermal nature). It is worth noting particularly the last (tenth) of the suggestions put forward by Frenkel':

''10. It would be interesting to use high (thousands of millions of degrees) temperatures induced in the explosion of an atomic bomb in order to carry out fusion reactions (for example, the formation of helium from hydrogen), which are the source of stellar energy and which can increase even further the energy released as a result of the explosion of the base material'' (Ref. 34, p 4).

This suggestion is worth noting, apart from the intrinsic interest, for two reasons. First, in an evident manner, it harks back to what Frenkel' wrote in the appendix to H Gunter's book in 1925 [5]. Second, it is interesting to recall that this prediction was made well before the well-known suggestion of Edward Teller (see, for example, the paper of Yu B Khariton [38] reminiscing about Ya B Zel'dovich).

These citations from the detailed 1945 note of Frenkel' provides a further confirmation that the idea of the hydrogen bomb did not come to us from the West and particularly not by spying, as has been put about recently in our press (i.e. through K Fuchs). I should mention also that these summary ideas were quite obvious to Yakov Il'ich and were not secret. They were presented in one of the first (if not the first) popular science articles on the release of atomic energy published in our country [39]. However, it is remarkable that these ideas had been excluded from both books on the same subjects published in 1946 and 1950 [40, 41].

It seems to me very surprising that to the best of my knowledge, there had been absolutely no reaction by I V Kurchatov to this letter of Frenkel'. I was told about this by my mother S I Frenkel', after my father's death. Naturally, she did not know the main content of the note, but she mentioned that Yakov Il'ich was surprised and hurt by the absence of any response to this note. This throws into even sharper relief my earlier reference to Kurchatov's selection of theoreticians for working on the bomb. My attempts to find any logical reason for his choice have been unsuccessful.

In conclusion, I must mention that in 1946 Ya I Frenkel' developed an interesting quantum-mechanical theory of the mechanism of the fission of heavy nuclei [42]. In his view, this theory accounts for the asymmetric nature of such fission associated with the tunnelling nature of this process (as noted by Frenkel', the ordinary α-decay is the limiting case of such asymmetry).

Frenkel' sent the reprints of his articles [42, 43] to his colleagues both in the Soviet Union and abroad. I know of two responses to these reprints. One came form Max Born (in which Born mentions especially the clear language of Frenkel's paper which can be understood by nonspecialists) (Ref. [12], p. 436). Niels Bohr also commented on the papers. His letter, in response to one from Frenkel' of 1 July 1946, (which accompanied the reprints) did not reach Yakov Il'ich (both letters, from Frenkel' and from Bohr, were shown to me by Dr H Levi — see footnote on p. 8). Bohr made some critical comments on Frenkel's work. These comments are given in a somewhat modified form also in a letter from Bohr to J A Wheeler of 13 July 1949 (Ref. [25], p. 666).

I hope to present the documents referred to above and to comment on them in detail in a separate publication.

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