Selected results and open problems in a semiclassical theory of dense matter

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Abstract. Studies of the behavior of materials under high external pressure have started in Serbia shortly after the middle of the last century. The aim of this lecture is to review the theoretical foundations of this work, present a selection of the results, and indicate some open problems within the theory.

INTRODUCTION

Systematic studies of the behavior of materials under high external pressure have started near the end of the XIX century. The main "driving force" behind the rise of interest in this field was a professor of physics at Harvard, P.W. Bridgman (1882-1961). In 1946, he was awarded the Nobel prize in physics for "for the invention of an apparatus to produce extremely high pressures, and for the discoveries he made therewith in the field of high pressure physics". Presses used by Bridgman were large, expensive to build and use, and once enclosed in them specimen were no longer visible. Experimental work in the field was later facilitated with the discovery of the diamond anvil cell (DAC). The DAC fits into a hand, it is much cheaper, simple to operate, the upper limit of pressure is much higher, and the specimen is visible throughout the experiment. Modern theoretical interest in the field started in the thirties of the last century when Fermi hinted that increased pressure leads to changes in the atomic structure. The basic aim of these studies is to solve the eigen-problem of a general, non-relativistic many-body Hamiltonian, defined as follows:

$$H = -\frac{\hbar^2}{2m} \sum_{i=1}^{N} \nabla_i^2 + \sum_{i=1}^{N} V(|\vec{x}_i|) + \sum_{i,j=1}^{N} v(|\vec{x}_i - \vec{x}_j|)$$

(1)

According to rigorous quantum mechanics, the energy of the system having a Hamiltonian $H$ is given by

$$E = \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle}$$

(2)

All the symbols in these equations have their standard meaning. Sums in eq. (1) go over the number of particles which, in real physical situations, is of the order of Avogadro’s number. This implies that these sums can not be calculated exactly. The calculation of such sums is a serious problem in many-body physics. Once the eigenproblem of
$H$ defined in eq.(1) is somehow solved, the thermodynamic potentials, phase transition points (or regions) and the phase diagram in general follow by application of standard "prescriptions" of statistical mechanics.

This lecture is devoted to a review of a simple theory of dense matter, whose "‘founding fathers’” are two professors of the University of Belgrade: Pavle Savić and Radivoje Kašanin. As a consequence of the first letters of their family names it is often called "the SK theory". It is also often called "the semiclassical theory" because it is founded on laws of classical physics and just a few ideas of atomic physics. The next section contains a review of the basic physical ideas of their theory, while selected results of various applications are outlined in the third part. The fourth part contains a discussion of some of the open possibilities for future work on this theory, and the lecture ends with the final comments.

**THE BASIC IDEAS**

The development of what later became the SK theory started in 1961 when P. Savić published a short paper [1] presenting an unusual idea: the mean planetary densities, as calculated from the masses and radii known at the time, could be linked to the mean solar density by an extremely simple expression:

$$\rho = \rho_0 2^\phi$$

(3)

In this expression $\rho_0$ denotes the mean solar density, which at the time was estimated at $4/3$ g cm$^{-3}$. By choosing integral values of the exponent $\phi$ in the interval $\phi \in (-2, 2)$ Savić managed to fit the numerical values of the densities of the major planets. No hint of any possible physical explanation of this simple fit was given. In the following 4 years, collaborating with Radivoje Kašanin, [2] he managed to develop a theory of the behavior of materials under high pressure. In later years, it was nicknamed "the SK theory" after the first letters in their family names.

The basic physical idea of their theory is simple. SK assumed that sufficiently high values of external pressure lead to changes of the electronic structure of atoms and/or molecules. The possibility of such an interaction was for the first time hinted by Fermi several decades before SK. However, according to the available literature, he simply noted the possibility of such an influence, but did not find it interesting enough so as to explore it in detail [3]. The possibility of such an influence has also been invoked around that time by P.W. Bridgman [14]. For the special case of a one-dimensional finite potential well this problem has recently been discussed by the present author [4]. An analytical expression for the first pressure derivative of the energy of a massive particle in such a well has been obtained and discussed in that paper.

The SK theory is in a way similar to the "jelium model" from classical solid state physics. It represents a material as a uniform distribution of particles with the mean interparticle separation $a$ defined by

$$N_A (2a)^3 \rho = A$$

(4)
In this expression \( N_A \) denotes Avogadro’s number \( \rho \) is the mass density and \( A \) is the mean atomic mass of the material. Having thus introduced \( a \), one can define the "accumulated" energy per electron as

\[
E = \frac{e^2}{a}
\]  

(5)

Logically, one might expect that a relation such as the last one should contain the ionic charge \( Z \). It can be shown, (for example [5]) that \( a \) as defined in eq.(4) is a multiple of the radius of a Wigner-Seitz cell, which actually contains \( Z \). The radius of a Wigner-Seitz (WS) cell is defined by

\[
\frac{\text{Am}_\rho}{\rho} = \frac{4}{3} \pi r_{WS}^3 = \frac{Z}{n}
\]  

(6)

It follows from eq.(6) that

\[
r_{WS} = \left( \frac{3}{4\pi} \right)^{1/3} \left( \frac{A}{\rho N_A} \right)^{1/3} = 2a \left( \frac{3}{4\pi} \right)^{1/3} = \left( \frac{3}{4\pi} \right)^{1/3} \left( \frac{Z}{n} \right)^{1/3}
\]  

(7)

The basic premisses on which the SK theory is based are the following statements [6]

1. The density of a material is an increasing function of the pressure to which it is exposed.
2. With increasing density, every material undergoes a sequence of first order phase transitions. Phases are numbered by an index \( i \) and the phase ending at the critical point is denoted as the zeroth phase.
   In any given phase (i.e., for any value of \( i \)) there exist two limiting values of the density \( \rho \) such that

\[
\rho^0_i \leq \rho_i \leq \rho^*_i \Rightarrow \left( \frac{1}{\alpha_i} \right) \rho^*_i \leq \rho_i \leq \rho^*_i
\]  

(8)

where \( \alpha_i > 1 \)

3. The maximal densities of two successive phases are related by

\[
\rho^*_i = 2 \rho^*_i
\]  

(9)

This expression follows directly from eq.(2) by assuming that \( \phi_{i+1} - \phi_i = 1 \).

4. It is assumed that

\[
\frac{E^*_i}{E^0_i} = \frac{E^0_{i+1}}{E^*_i}
\]  

(10)

Some form of a link between the accumulated energies in two successive phases was needed in order to render the calculations tractable, and this form was accepted because of its simplicity. After some algebra, it follows that \( \alpha_i \alpha_{i+1} = 2 \) and that

\[
\alpha_i = \begin{cases} 6/5, & i = 1, 3, 5, \ldots \\ 5/3, & i = 2, 4, 6, \ldots \end{cases}
\]  

(11)
5. The final density of the zeroth phase is
\[ \rho_0^* = \frac{A}{3\bar{V}} \tag{12} \]

which is approximately equal to the critical density in the van der Waals theory. \( \bar{V} \) denotes the molar volume of the material at \( T = 0K \). In the terminology of the van der Waals theory \( \bar{V} = b \).

6. Using assumption 3., it can be shown that
\[ \frac{A}{\bar{\rho}} = \frac{A}{2} \left( \frac{1}{\rho_0^*} + \frac{1}{\rho_2^*} \right) \tag{13} \]

In this expression \( \bar{\rho} \) denotes the density at the zero-point, defined as \( \bar{\rho} = A/\bar{V} \).

Starting from these premisses, the following set of simple analytical relations can be derived, which are used in all subsequent calculations within this theory. Second order phase transitions can be considered as a special case for which \( \bar{V}_i^* - \bar{V}_i^0 \to 0 \).

\[
\begin{align*}
\rho_i^* &= 2^i \rho_0^*; \rho_i^0 = \frac{\rho_i^*}{\alpha_i} \\
\bar{V}_0^* &= 3\bar{V}; \bar{V}_i^0 = \alpha_i \bar{V}_i^* 2^{-i}; \bar{V}_i^* = 2^{-i} \bar{V}_0^* \\
r_0^* &= \left( \frac{15}{4N_A 10^{-23}} \right)^{1/3} \bar{V}^{1/3}; r_i^* = 2^{-i/3} r_0^*; r_i^0 = r_i^* \alpha_i^{1/3} \tag{14}
\end{align*}
\]

The following section of this lecture contains a selection of results obtained in various applications of this theory. The algorithm for the calculation of phase transition pressure is relatively short, so it will be presented in detail. The set of equations needed for modelling the internal structure of celestial objects is much longer, so the reader interested in its full details should consult [2].

**SELECTED RESULTS**

**Laboratory applications**

The application of the SK theory which is most easily verifiable in laboratory experiments is the calculation of the value of pressure on which a first order phase transition occurs in a given material. This value of pressure can be calculated by considering the work done by the external pressure in compressing the material,
\[ \Delta W = p_i^*(\bar{V}_i^* - \bar{V}_{i+1}^0) = p_i^* V_i^* \left( 1 - \frac{1}{\alpha_i} \right) \tag{15} \]

and equating it to the change of the accumulated energy
\[ \Delta W = \Delta E = N_A (E_{i+1}^0 - E_i^*) \tag{16} \]
Starting from eqs. (11)-(13) it can be shown that the maximal pressure in a phase $i$ of a material is given by

$$p_i^* \approx 1.8077\beta_i \tilde{V}^{-4/3}2^{4i/3}MBar$$

(17)

with

$$\beta_i = 3\frac{\alpha_i^{1/3} - 1}{1 - 1/\alpha_i}$$

(18)

Values of $\alpha$ for various values of the index $i$ are given in eq.(8). Finally, the value of the external pressure needed to "provoke" a first order phase transition from phase $i$ to the phase $i+1$ in a material is given by

$$p_{tr} = p_i^* - p_0^i = p_i^*(1 - 2^{-4/3}\frac{\beta_i^{-1}}{\beta_i})$$

(19)

This expression gives a simple mathematical procedure for the calculation of the sequence of possible values of a phase transition pressure in a given material. Those values of pressure at which a first order phase transition is physically possible are selected by the following criterion:

$$E_0^* + E_I = E_i^*$$

(20)

The symbol $E_I$ denotes the ionisation potential, $E_0^*$ and $E_i^*$ can be calculated from eqs. (5) and (12) with $a = 10 \times r$ nm. Applying the procedure described above, an analysis of the applicability of the SK theory to real materials under high pressure was made [7]. A set of 19 materials for which experimental data on phase transitions under high pressure were easily available was analyzed. The aim was to calculate within the SK theory values of pressure at which first order phase transitions could be expected, and then compare the results with the experimental data and analyze possible causes of the discrepancies. It was shown in [7] that the relative discrepancies between the measured values of phase transition pressure and those calculated within the SK theory are material and pressure dependent. Two basic causes of the discrepancies were identified:

one is due to that fact that the SK theory takes into account only the simplest form of the electrostatic potential, while in reality in atoms and molecules one "deals" with charge distributions;

the second "source of problems" is represented by the fact that the SK theory neglects the contribution of various non-electrostatic components to the overall intermolecular potential. This is expectable for a semiclassical theory, but it clearly increases the discrepancy between the measured and calculated values of the phase transition pressure. More details on these two problems are available in [7].

Another interesting result of the SK theory concerns the establishment of the thermal equation of state of solids under pressure. Namely, the temperature is not explicitly present in the original formulation of this theory. All its calculations are performed in the $P - \rho$ plane. However, the temperature can be introduced in a simple way: by equating the internal energy densities of a solid, as expressed within the SK theory and in standard solid state physics.

The internal energy per unit volume of a solid is, in the SK theory, given by

$$E = 2e^2Z\left(\frac{N_A\rho}{A}\right)^{4/3}$$

(21)
and in standard solid state physics, it is given by

\[ E = \frac{\pi^2 (k_B T)^4}{10(h\bar{u})^3} \]  

(22)

The symbol \( \bar{u} \) in the last expression denotes the mean value of the velocity of sound, and all the other symbols have their standard meanings.

Details are available in [8] and the final result is that the equation of state of a solid in the \( T - \rho \) plane has the following form:

\[ T [K] = 1.4217 \times 10^5 \left( \frac{\rho}{A} \right)^{7/12} \left( \frac{m_e}{M} \right)^{3/8} Z^{7/8} \]  

(23)

The symbol \( m_e \) denotes the bare electron mass, \( M \) is the ionic mass, \( A \) denotes the mass number of the material and \( Z \) is the charge of the ions. A result of an important astronomical application of this equation of state is presented in the following subsection of this contribution.

Last, but certainly not the least, we come to the problem of hydrogen under high pressure, and its possible metallization. The SK theory has been applied to this problem [9], and the result obtained at the time was encouraging. Metallisation was predicted to occur at \( P = 300 \) GPa, which agreed with predictions by various other theoretical methods. Theoretical predictions of the metallisation pressure of hydrogen have been of this order of magnitude for the last 70 years [10].

The experimental situation in work on hydrogen under high pressure is much less well defined. There have been claims at the beginning of the nineties that metallisation occurs at a pressure \( P = 150 \) GPa. A short time after these results were shown to be incorrect and due to a chemical reaction of ruby with hydrogen [11]. Static experiments performed at values of pressure \( P \leq 342 \) GPa have not shown signs of metallization of hydrogen [12], so the problem seems (at present) to be completely open. The literature on this topic is huge, but for examples of interesting recent papers see [13] and [14]. One of these papers [13], invokes the influence of disorder as a possible reason why metallization was not observed at the theoretically predicted value of the pressure.

Applications in astronomy

The theory we are discussing in this lecture can be applied to modellization of the internal structure of celestial objects. To be precise, the interest of Savić and Kašanin in this problem was the initial "grain of salt" which led to its development. As the calculations in this theory do not contain any mention of internal energy generation in celestial objects (i.e., nuclear reactions), it is inapplicable to work on stellar structure.

The complete calculational scheme for astronomical applications of the SK theory is available in [2]. Unfortunately, due to various reasons, it has not been reformulated in a modern way. The only input data needed for making the model of the internal structure of a celestial object within this theory are the mass and the radius of the object.

Starting from these data, it gives the number and thickness of layers which exist in the interior of the object, the distribution of the values \( P, \rho, T \) with depth under the surface, the
strength of the magnetic field and the interval of the physically allowed values of the 
speed of rotation. The theory also gives as a result the mean atomic (or molecular) mass of 
the chemical mixture that the object under study is made of.

The first celestial body to be modelled was logically the Earth and the main characteristics of this model are shown in the following table.

TABLE 1. The interior of the Earth according to SK

| depth (km)     | 0 - 39 | 39 - 2900 | 2900 - 4980 | 4980 - 6371 |
|----------------|--------|-----------|-------------|-------------|
| \(\rho_{\text{max}}\) [kg/m\(^3\)] | 3000   | 6000      | 12000       | 19740       |
| \(P_{\text{max}}\) [GPa]      | 25     | 129       | 289         | 370         |
| \(T_{\text{max}}\) [K]        | 1300   | 2700      | 4100        | 7000        |

The mean mass number of the material which makes up the Earth is \(A = 26.56\). Taking into account the simplicity of the theory, this model is in remarkable agreement with modern knowledge. For a discussion of the temperature in the interior of the Earth, see for example [15].

Note also that a current model of the Earth’s crust called CRUST 5.1, [16] gives around 70 km as the maximal value. Here in Holland, the thickness of the Earth’s crust according to this model is around 35 km, which is in excellent agreement with the value calculated within SK.

Apart the Earth, the theory was applied to all the other planets except Saturn and Pluto, the Moon, the Galileian satellites of Jupiter, the satellites of Uranus, Neptune’s satellite Triton and the asteroids 1 Ceres and 10 Hygiea.

The results are scattered in the literature, but a "safe" general comment is that the agreement with the consequences of observations and with theoretical work of other authors is good [6].

The following table contains the values of the mean atomic masses of various objects in the Solar System, calculated according to the SK theory.

TABLE 2. The composition of the Solar System according to SK

| object     | A  | satellite | A  |
|------------|----|-----------|----|
| Sun        | 1.4| Moon      | 71 |
| Mercury    | 113| J1        | 70 |
| Venus      | 28.12| J2 | 71 |
| Earth      | 26.56| J3 | 18 |
| Mars       | 69 | J4        | 19 |
| 1 Ceres    | 96 | U1        | 38 |
| Jupiter    | 1.55| U2 | 43 |
| Saturn     | /  | U3        | 44 |
| Uranus     | 6.5 | U4 | 32 |
| Neptune    | 7.26| U5 | 32 |
| Pluto      | /  | Triton 67 |    |

It can easily be seen from the preceding table that our planetary system is far from being chemically homogenous; at first sight, the well known division on the terrestrial and jovian planets is clearly visible. These differences are obviously a "remnant" of various transport and mixing processes which have been active in the formation epochs of the
planetary system. Note also that similar differences are visible in the satellite systems which were modeled.

Various conclusions can be drawn from data in Table 2. For example, asteroid 1 Ceres is currently orbiting the Sun between the orbits of Mars and Jupiter. However, by its chemical composition it is similar to the planet Mercury [6]. As chemically similar bodies are expected to have been formed close to each other, this similarity implies that "once upon a time" Ceres and Mercury originated in the same region of the protoplanetary system, but that their orbits later diverged. The physical process(es) which have led to this diverging of their orbits can be a subject of further studies.

Concerning asteroids, using the value of $A$ calculated for 1 Ceres, the mass of the asteroid 10 Hygiea was calculated, and the result turned out to be in excellent agreement with the result known in celestial mechanics [17].

Two cases of applications of the SK theory are especially worth mentioning. The composition of the Galilean satellites was determined within this theory in 1987. In 1996 some results of this calculation were confirmed by measurements from the Galileo space probe. Details are given in [18] which contains the reference to the original publication of 1987.

Another interesting application of this theory concerns Neptune’s satellite Triton [19]. It was shown in that paper that Triton has a composition similar to Mars, which was interpreted as implying that Triton is a captured body, in perfect agreement with earlier work based purely on celestial mechanics. This conclusion is of interest for cosmogony, as it can be regarded as an independent proof that collisions were important in the early phases of existence of our planetary system. For a recent example of observational work on the structure of a protoplanetary cloud see [20].

SOME POSSIBILITIES FOR FUTURE WORK

In this lecture we have so far reviewed the basic ideas and a number of results of applications of the SK theory. However, in spite of successes, there are open possibilities for future work related to the theory we are discussing, and this section is devoted to an outline of these topics.

Paper [1] was devoted to a possible relation tying the mean planetary densities (derived from their masses and radii) with the mean solar density. Eq. (1) was the result, and it was applied in various calculations in its original form ever since. The important detail in that paper was not only the simple form of eq. (1), but also the fact that the exponent $\phi$ in eq. (1) had integral values. However, as nearly 40 years have elapsed since [1], it seemed appropriate to undertake a verification of eq. (1) but with modern data, and on a broader set of objects. Such a check has recently been performed [21] and the results are shown in table 3.

Modern data on densities of 22 planets and their major satellites were used. It was shown that eq. (1) is still valid, but that the exponent $\phi$ can also take non-integral values. The immediate question is of course the interpretation of these non-integral values. Namely, the integral values of $\phi$ which appeared in [1] were interpreted there and in later publications as a qualitative analogy with atomic quantisation, implying that high
external pressure leads to changes in the atomic structure. The problem is open, and one of the possibilities would be to try to cautiously pursue the analogies between the atomic structure and the structure of the planetary system.

Another "open" astronomical application of the SK theory is the calculation of the angular speed of rotation of a celestial object. Instead of proposing an algorithm for this calculation and giving a unique number at the end, this theory gives a physically allowed interval in which the speed of rotation can be.

It was applied to the Earth and several other planets. The results are very promising, but it would be useful to reduce this allowed interval to a unique number.

A possibility for future work concerns laboratory applications of the SK theory. We have discussed to some extent the applicability of this theory to the calculation of phase transition pressure in various solid materials, and to the establishment of a thermal equation of state of solids. The discrepancies between the predicted values of phase transition pressure and experimental results are material dependent. Work in this direction could in the future advance along several different lines:

At first it would be useful to apply the theory to more materials for which first order phase transition pressures are known. In this way one would obtain a more precise empirical estimate of the systematic trends (if there are any) in the discrepancies between the calculated and measured values of the phase transition pressure.

On the purely theoretical side, it would be necessary to refine the theory with the inclusion of the contribution of more components to the interparticle potential energy.

| object   | $\rho_{mean}$ | $\phi$ | $\rho_{calc}$ |
|----------|---------------|--------|---------------|
| Sun      | 1408          | 0      | 1408          |
| Mercury  | 5427          | 2      | 5632          |
| Venus    | 5243          | 2      | 5632          |
| Earth    | 5515          | 2      | 5632          |
| Moon     | 3350          | 5/4    | 3349          |
| Mars     | 3933          | 3/2    | 3982          |
| Phobos   | 1900          | 2/5    | 1858          |
| Deimos   | 1750          | 1/3    | 1774          |
| Jupiter  | 1326          | -1/10  | 1314          |
| JI       | 3530          | 7/5    | 3716          |
| JII      | 3010          | 1      | 2816          |
| JIII     | 1940          | 1/2    | 1991          |
| JIV      | 1840          | 2/5    | 1858          |
| Saturn   | 687           | -1     | 704           |
| Titan    | 1881          | 2/5    | 1858          |
| Uranus   | 1270          | -1/7   | 1275          |
| Ariel    | 1670          | 1/4    | 1674          |
| Neptune | 1638          | 1/4    | 1674          |
| Triton  | 2050          | 1/2    | 1991          |
| Pluto   | 1750          | 1/3    | 1774          |
| Charon  | 2000          | 1/2    | 1991          |
An even more interesting problem would be to link the SK theory to the well-established theoretical framework of statistical physics. Although the SK theory has given good results in a range of applications, its unusual formulation hinders its wider spread in the research community. A preliminary attempt in that direction has recently been performed in [21], where it was shown how the parameters appearing in the SK theory can be connected with those of the Landau theory. It would be interesting trying to reformulate the SK theory along the lines of present day theories of quantum phase transitions (for example, [22]).

A note concerning the equation of state of solids under pressure in $T - \rho$ plane:

This calculation, described in detail in [8], gives physically acceptable numerical values, which is good. However, the calculation invokes the speed of sound in a solid, and (as well known) this invokes the knowledge of $\partial P/\partial \rho$ - that is the equation of state.

In the calculation which has led to the proposed form of thermal equation of state, an approximation called the "Bohm-Staver" formula was used [23]. It is planned in the future to repeat this calculation, but using some more refined form of the equation of state.

**FINAL COMMENTS**

This lecture was an attempt to present a "balanced" review of a theory of dense matter proposed a little less than half a century ago by P. Savić and R. Kašanin. The adjective "balanced" simply means that the basic ideas, successful applications, but also problems for future work were discussed. In the past, work within the SK theory has been more oriented towards various applications and to a lesser extent towards its modernization. In future work, efforts will concentrate more on the refinement of this theory and extension of its range of applicability. It is hoped that as result of this workshop some "joint venture" in that direction will be started.

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