

Is It Possible to Make Materials With Ionic Super Conductivity? Part II

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Abstract

The basic idea is that we try to find some materials in which bosonic ions with sufficiently small *effective mass* are used as charge carriers instead of Cooper's pairs in order to provide high temperature *ionic* superconductivity. Ionic crystals LiCl, LiF, LiBr and LiI were considered with lithium isotope Li⁶. Calculations show that Bose condensation temperature for lithium ions in these crystals is of the order of 10⁻³⁴-10⁻⁴³K. If, however, the crystal is compressed so that the wave functions of neighboring lithium ions are sufficiently overlapped, then Bose-condensation temperature of Li⁶-ions can be increased significantly. Our estimates show that compressing the crystals by 20-22% in all three directions one can raise the Bose-condensation temperature in all crystals considered to *above room temperature*. To realize materials with room temperature superconductivity in practice the use of molecular beam epitaxy is proposed for the formation of heterostructures from thin and thick layers of thoughtfully-chosen composition.

I. INTRODUCTION

We consider ionic crystals LiF, LiCl, LiBr and LiI with NaCl crystal structure. The distribution of lithium ions in the crystal lattice is governed by Schrödinger equation with appropriate translational symmetry. Eq. differs from well-known equation for electrons in a periodic potential only by the mass of the particle considered. As a consequence, from well known analysis of this equation one can conclude that ion energy in ionic crystals is a function of wave vector and solutions for real-valued ion eigenenergies exist only for specific energy intervals. Note, that naturally occurring ionic crystals composed from single-charged ions are formed from fermions. The only exception is the ion of Li⁶ isotope which consists from even number of fermions i.e. 3 neutrons, 3 protons and 2 electrons. Let us estimate the possible characteristics of ionic crystals LiF, LiCl, LiBr and LiI with Li⁶ isotope using tight-binding approximation (an analysis is quite similar to that for electrons).

II. PROBLEM DEFINITION AND THEORY

If $U_k(\vec{r}) - U_i(\vec{r})$ is the difference of the ion potential energy in the crystal and in the corresponding mole-

cule (the molecule is composed from positive lithium ion and negative halogen ion) and E_i is the ground state energy of lithium ion in the molecule, then for the rock salt cubic structure we obtain taking into account only nearest neighbor interaction

$$E(\vec{k}) = E_i - E(0) + 2E(100)(\cos ak_1 + \cos ak_2 + \cos ak_3) \quad (1)$$

$$E(0) = -\int \psi_i^*(\vec{r}) \{U_k(\vec{r}) - U_i(\vec{r})\} \psi_i(\vec{r}) d\vec{r} \quad (2)$$

$$E(100) = -\int \psi_i^*(\vec{r} + \vec{a}) \{U_k(\vec{r}) - U_i(\vec{r})\} \psi_i(\vec{r}) d\vec{r} \quad (3)$$

Here, k_1, k_2, k_3 are wave vector projections on different crystallographic axes, a is lattice constant and ψ_i is normalized Li⁶ ion wavefunction. For the calculation of Li⁶ ion effective mass in the crystal we can use a well-known (in tight-binding approximation) expression for NaCl-type lattice

$$m^* = \hbar^2 / (2E(100)a^2) \quad (4)$$

The form of molecular potential (U_i) used in the calculations is fitted to experimental molecular dissociation energies, as well as their size, and elastic properties and lattice constants of ionic crystals [1]. In a paper [1] the calculation of wave functions is fulfilled.

Inserting the ground state wavefunction into (3) one can calculate the overlap integral $E(100)$ and then the effective mass of lithium ion from (4). Finally, the temperature of Bose-condensation (T_b) can be estimated [2] from

$$T_b = \frac{2\pi\hbar^2}{m^*k_B} \left(\frac{N}{2.612 \times V} \right)^{\frac{2}{3}} = \frac{2\pi\hbar^2}{m^*k_B} \left(\frac{0.5}{2.612 \times \left(\frac{a}{2}\right)^3} \right)^{\frac{2}{3}} \quad (5)$$

where k_b is Boltzmann constant, N/V is boson number density. The calculations have shown, that $m^* > 10^{38} m_e, T_b < 10^{-33}$ K, where m_e is a free electron mass [1]. Due to a huge value of lithium ion effective mass all ionic crystals are dielectrics at any real temperature. Indeed, if we assume that in a defect-free crystal the dominant scattering mechanism for Li

ions is scattering by acoustical phonons, we can estimate the specific resistivity of ionic crystal ρ_c in comparison with that of metal ρ_m as $\rho_c \approx \rho_m (m^*/m_e)^{5/2}$, where $\rho_m = 10^{-5}$ Ohm cm is a typical metal resistivity. No contradiction with experiment is obtained since real ionic conductivity in available crystals is completely dominated by defects and is many orders of magnitude higher. If, however, the ionic crystal is compressed so that Li wavefunction overlap becomes significant, than Bose-condensation temperature for Li^6 ions can be increased significantly. In earlier work [1] the following expression has been used for overlap integral

$$E(100) \approx -\int \psi_i^*(r+a_s) \left\{ U_k \left(\frac{a}{2} \right) - U_i \left(\frac{a}{2} \right) \right\} \psi_i(r) dr \quad (6)$$

where a_s is lattice constant of compressed crystal. A more consisting approach would be to use

$$E(100) \approx -\int \psi_i^*(r+a_s) \left\{ U_k \left(\frac{a_s}{2} \right) - U_i \left(\frac{a_s}{2} \right) \right\} \psi_i(r) dr \quad (7)$$

In this case the periodic potential is replaced by its minimal value and pressure-induced modification is properly taken into account. Since the volume of one molecule in the crystal is $2r^3$ the ion binding energy in the crystal under pressure is given by

$$U_k = -\frac{\alpha q^2}{4\pi\epsilon_0 r} [1 - (r\rho/r_0^2) \exp((r_0 - r)/\rho)] + 2P(r^3 - r_0^3) \quad (8)$$

where P is pressure, $\alpha = 1.748$ is Madelung's constant and $r_0 = a/2$. The connection between pressure (P) and inter-ion distance ($r = a_s/2$) in NaCl lattice is given by $dU_k/dr = 0$. Parameters ρ were taken from [3] or [1]. The calculated pressure dependence of lattice constant is shown in Figure 1.

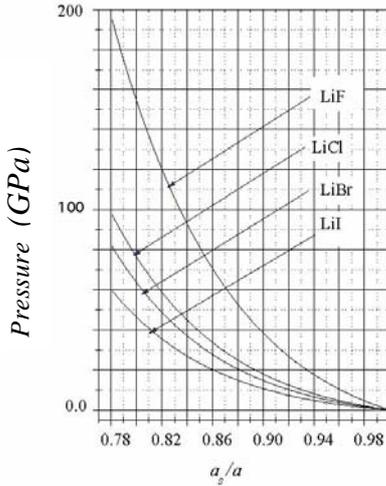


Fig.1. The pressure required to decrease lattice constant from a to a_s .

Inserting the ground state wavefunction into (7) one can calculate the overlap integral $E(100)$ and then to obtain the effective mass of lithium ion and the temperature of Bose-condensation (T_b) from (4),(5), when $a = a_s$. The results are shown in Figure 2 and Figure 3. Our calculations have shown that in order to make Bose condensation temperature greater than 300K one should compress LiF by 22%, LiCl by 21,3%, LiBr by 21%, LiI by 20,3%. The pressures required to achieve these compressions are 200, 100, 70 and 50 GPa for LiF, LiCl, LiBr and LiI respectively. Since exact results are presently unavailable it is difficult to estimate the errors involved due to approximations used in the calculations. However, even if the calculated effective mass of lithium ions is increased by a factor of 30, the Bose condensation temperature of Li ions in crystals compressed by 21-22% is greater than 300K.

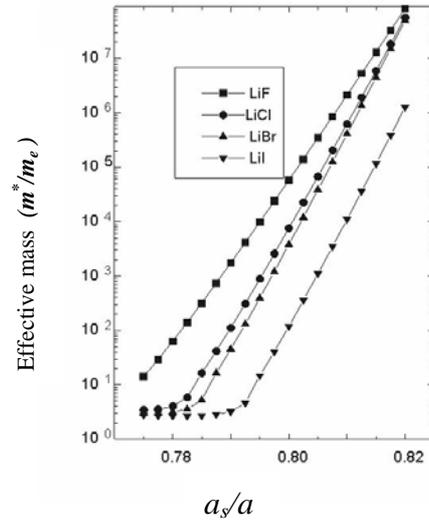


Fig. 2. Effective mass of Li^6 ions m^* as a function of crystal compression, a_s is the lattice constant of compressed crystal, a is lattice constant of strain-free ionic crystal, m_e is free electron mass.

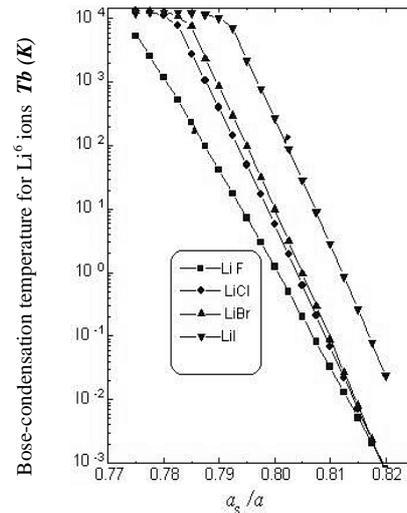


Fig.3. Bose-condensation temperature for Li^6 ions as a function of crystal compression.

And the tight-binding approximation, when applied to electrons, does not lead to errors in the order of magnitude of the calculated effective mass. As can be seen from Figure 3 the compressions greater than 18% are of interest for performing practical measurements. For these large compressions the notion of lattice site as a point where lithium ion energy is at minimum become meaningless. The lithium ion binding energy in the crystal (E_c) can be easily calculated.

$$E_c = -\frac{\alpha q^2}{4\pi\epsilon_0 r} [1 - (r\rho/r_0^2)\exp((r_0 - r)/\rho)];$$

$\alpha = 1.748$ – is Madelung's constant, $r_0 = a/2$, $r = a_s/2$

As long as this energy is less than binding energy of the ion in a molecule the ions occupy lattice sites. In opposite case, which is realized already at 18-20% compression, the potential reach a minimum value not at a point but on the surface similar to spherical surface in a molecule. The problem then becomes quite similar to that of electron in the solid and application of tight-binding approximation would appear more appropriate. See Figure 4, which exemplifies the dependence of ion binding energy on the distance between Li and Br ions in the crystal and in the molecule. Parameters U_i , ρ were taken from [1,3]

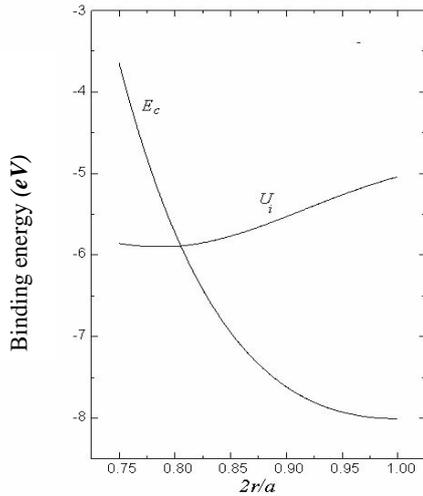


Fig. 4. Binding energy as a function of distance between Li and Br ions in the crystal (E_c) and in the molecule (U_i).

It has been amply demonstrated in semiconductor technology that during epitaxial growth of one semiconductor (e.g., Ge) on the substrate of another semiconductor (e.g., Si) the pseudomorphic film can be obtained [4] if the grown film thickness is less than the so called critical thickness (which is about several monolayers (ML) for the Ge/Si heterosystem with a lattice mismatch $\sim 4\%$). Often a mixed alloy ($\text{Si}_x\text{Ge}_{1-x}$) buffer layer is provided on Si substrate in order to reduce the dislocation density. Similar behaviour is observed in epitaxial growth of ionic materials [5]. Epitaxy is greatly assisted by similarity of the structure and chemical bonding in the substrate and overlayer materials. In our case it means that desired compression can be accomplished if the ionic crystal with larger lattice constant is grown pseudo-

morphically on the substrate from another ionic crystal with smaller lattice period. In fact, a thin (1-3 ML) working layer can be overgrown by a thick (10-100 ML) layer of substrate material forming thus a double heterostructure or even a multi-quantum well structure. An epitaxial route to highly compressed ionic crystals seems to be worthy to attempt at using, for example, the molecular beam epitaxy. Evidently, very thin layers (in the 2-3 monolayer range) are desirable since compression of thick layers to the targeted 20% is practically impossible. Assuming lattice constant of the compressed crystal is equal to that of the substrate we estimated through (4)-(5) when $a = a_s$, the effective mass of lithium ion and Bose condensation temperature for some combinations. Results are shown in Tab. I. In fact, a thin layer of the crystal to be compressed could be patterned and overgrown by a thick layer of substrate material. In that case it will be compressed along all three axes.

TABLE I

Calculated Bose-condensation temperature for epitaxially- compressed ionic crystals

Substrate and thick overlayer material	Thin layer material	Li ⁶ ions effective mass m^*/m_e	Bose-condensation temperature for Li ⁶ ions (K)	Lattice compression a_s/a
LiF	Li ⁶ Cl	9.7	4700	0.784
LiF	Li ⁶ Br	3.1	14200	0.732
LiF	Li ⁶ I	3.3	13600	0.671
NaF	Li ⁶ I	2.7	13100	0.772
LiH	Li ⁶ Cl	510	86	0.793
LiH	Li ⁶ Br	3.3	14500	0.741
LiH	Li ⁶ I	2.4	13000	0.680

In order to reduce the layer/substrate lattice mismatch even further, both members of heteroepitaxial composition can be made from solid alloys. As an illustration let us take the following example. In the Li⁶I / NaF system the lattice mismatch is about 22,8% ($a_s/a = 0.772$). Admixing Li⁶F we get the solid solution $(\text{Li}^6\text{I})_{1-x}(\text{Li}^6\text{F})_x$. For $x < 0,5$ the electronic properties of the alloy will not, probably, differ substantially from that of pure Li⁶I in a sense of wave function overlap between neighboring lithium ions. But the average distance between nearest- neighbor ions (a_c) in the $(\text{Li}^6\text{I})_{1-x}(\text{Li}^6\text{F})_x$ could be made much closer to that of the NaF substrate (a_s). As a result the dislocation density in epilayer could be reduced. Due to Vegard's law $a_c = (a(1-x) + xb)$, where b is the lattice constant of admixed Li⁶F, a is the lattice constant of Li⁶I. For $x = 0,3$ we have $a_c = 2,7042 \text{ \AA}$, $a_s/a_c = 0,9014$. Under condition of equality of the lattice parameters of the compressed $(\text{Li}^6\text{I})_{1-x}(\text{Li}^6\text{F})_x$ film and substrate, compressing $(\text{Li}^6\text{I})_{0,7}(\text{Li}^6\text{F})_{0,3}$ by only 9%, we will compress the Li⁶I by 22,8%! Note also that ionic crystals can be compressed much more easily than semiconductors since their bulk moduli (B) are smaller (for example, $B_{\text{Ge}}/B_{\text{LiI}} \approx 4.4$). In any case, the pressures to be applied are in excess of 50 GPa.. It is, however, possible to reduce the magnitude of this pressure a few times. To this end it is sufficient to fabricate the heterostructures with less than 10% lat-

tice mismatch, which are more easily realizable in practice, and subject them to additional hydrostatic pressure. As an example, one can consider a thin layer of LiI on LiBr substrate or thin layer of LiBr on LiCl substrate. It is possible to use $(\text{Li}^6\text{I})_x (\text{Li}^6\text{Br})_{1-x}$ instead of Li^6I and $(\text{Li}^6\text{Br})_x (\text{Li}^6\text{Cl})_{1-x}$ instead of Li^6Br . Current technology allows to obtain hydrostatic pressures in the range 8-100 GPa only in very small volumes using diamond anvils. For pressures less than 8 GPa the volume of the working chamber is almost unlimited. Therefore, it is reasonable to try first to fabricate the $\text{Li}^6\text{I}/\text{LiCl}$ or $(\text{Li}^6\text{I})_{1-x} \text{LiCl}_x/\text{LiCl}$ heterostructure (LiCl is a substrate) and then to subject it to hydrostatic compression with $P < 8$ GPa.

III. TECHNICAL APPLICATIONS

Let us assume that ionic superconductor is created. The question then is what can be done with that. Ionic superconductivity has its own peculiarities. The discussion of the full range of differences between ionic and electronic superconductivity must be postponed until experimental validation. Some points, however, can be mentioned. The usage of ionic superconductors in direct current electrical circuits seems to be problematical, because one must be able to compensate the loss of ions at the one end of ionic superconductor and fight with their excess at the other end. This peculiarity can be used in advantage for the fabrication of capacitors. The metal-ionic superconductor-metal structure forms the capacitance. Obviously, the magnitude of the capacitance will be large since electric field is concentrated in thin near-contact regions (electrons from the metal can not penetrate into ionic superconductor, whereas ions from the former are unable to diffuse into metal). The author does not see any obstacles for the application of ionic superconductors in alternating current circuits. Low frequency alternating current can be transmitted practically lossless through transformer coupling or through the system composed from ionic superconductor with ohmic contacts, which can be fabricated from a saturated water solution of some electrolyte (e.g. LiI) involving Li^6 ions. Ohmic contacts can also be made from pure Li^6 metal since in that case the Li^6 ions could probably tunnel from the metal into ionic superconductor and vice versa. A closed frame made from ionic superconductor could probably replace the bulky windings of electric generators and motors. A cavity inside ionic superconductor will be an ideal lossless cavity for ultra-high frequency applications. When ionic Bose condensate is placed in the magnetic field the current is generated which opposes the penetration of the field deep into superconductor material and which persists after the field is switched off. It is thus possible to generate the ultra-high magnetic fields on the premise that Meissner effect exists in ionic superconductors.

In this connection I recollected a funny idea that came into my mind some 15-20 years ago (at that time no one from my friends wished to check it). An idea can be described as follows. If a spinning ring made from superconductor is stopped the charge carriers

responsible for superconductivity keep on spinning and generate magnetic field. In case of Bose particles remaining at rest during spinning the source of the magnetic field will be a rotating superconductor. In its turn the expulsion of magnetic field from superconductor volume (Meissner effect) will make some amendments. For usual superconductors the topic can be developed in many directions – one can, for example, to spin the superconductor above critical temperature, next cool it down and only then to stop in order to see what magnetic field will be produced. All this is somewhat unusual but for the generation of ultra-high magnetic fields using ionic superconductors the idea has an added bonus because the ion number density in ionic superconductors is many orders of magnitude higher than that of Cooper's pairs in traditional superconductors. Moreover, the magnetic field will, probably, be generated even if the Meissner effect is absent in ionic superconductors.

IV. DISCUSSION OF RESULTS

Below I am going to enlist some objections to my work that have been received in discussions and correspondence with different specialists adding short comments as to why the arguments appear not very convincing for the author.

1. *It can be argued that in ionic crystals there are no such things as positive or negative ions. The notion of ions is merely a simplification with unclear applicability range.* However, the measured conductivity of ionic crystals at room temperature is determined by ion movement through crystal vacancies and is of the order of 10^{-7} - 10^{-6} (Ohm cm) $^{-1}$ at RT.

2. *Some doubts are cast upon the possibility to make the effective mass of the ion many orders of magnitude smaller than its gravitational mass since in metals the electron effective mass is of the order of m_e .* Similar doubts would probably come also to my mind before the work was started if I did not know that in several semiconductors the effective electron mass is much smaller than m_e (a factor of 20 in GaAs and a factor of 70 in InSb).

3. *Objection consists in that the relevance of tight-binding approximation is called in question.*

In the situation when exact solution is impossible the appropriateness of any approximation could be tested only by experiment. It is known, however, that tight-binding approximation gives qualitatively correct results in the calculations of electronic band structure for both metals and semiconductors. This approximation does not lead to errors in the order of magnitude of calculated values for electron and hole effective masses. And the smaller the magnitude of the overlap integral is, the better (more exact) the calculated results are. In considering the ionic crystals we have exactly the case of small overlap integrals when the size of compressed lattice constant is larger than $2r_m$ (r_m is the distance between ions in a molecule). However, even if the calculated effective mass of lithium ions is increased by a factor of 30-40, the Bose condensation temperature of Li ions in the crystals compressed by 21-22% is greater than 300K.

For more exact solution of the problem it is necessary to find a periodic potential. Inserting this potential into Schrodinger equation and using plane wave method one can obtain the secular equation for exact dispersion $E(\vec{k})$ in single-ion approximation. To find the lithium ion periodic potential in compressed crystal it seems to be necessary to solve self-consistently the quantum mechanical problem for the crystal containing sufficiently large number of molecules (greater than $100 \times 100 \times 100$). As a preliminary, a complete system of lithium ion molecular wave functions must be constructed and a method similar to that of Hartree-Fock is to be used. In contrast to HF, however, the solution must be represented not as the Slater determinant, but as a sum of the products for different permutations of the lithium ion molecular wave functions. The problem at hand is far from being simple and even if somebody will be able to get a solution it is unlikely to enjoy more confidence than estimates obtained in the present work.

4. *For some readers it seems that in order to get the useful result the pressures required should provide the sufficient overlap of lithium nuclei wave functions. These pressures are orders of magnitude higher than those presented in my work.*

The argument is relevant if we compress pure Li^6 . But for compression of ionic crystals the situation is different. In a molecule composed from lithium and halogen the wave function of lithium ion have a maximum near the surface of the sphere with radius r_m . Beyond this distance the wave function is sharply diminished. Consequently, upon crystal compression the surfaces of neighboring spheres come close together and overlap integral steeply increases during compression as long as the distance between oppositely charges ions in crystal lattice is greater than corresponding distance in a molecule (r_m). When the distances mentioned above become equal the spheres are in touch and further compression does not lead to substantial increase of the overlap integral. Numerical results for the lithium ion effective mass (which is inversely proportional to the overlap integral) confirm this conclusion. These results can be interpreted qualitatively if one notes that in undeformed crystals the binding energy of ion in the crystal is lower than corresponding binding energy of ion in a molecule. Consequently, lithium ions occupy lattice sites. But already in crystals compressed by 18-20% the binding energy of ion in the crystal becomes greater than that in a molecule. Due to disappearance of potential barrier the lithium ions can now move through the crystal.¹

5. *For some people it seems that reasonable results could be obtained only if correlational interaction between lithium ions is taken into account. Their ar-*

gument goes as follows. Since lithium ion is firmly sitting in the lattice site of ionic crystal and do not allow to occupy this site by other lithium ions then correlational effects must be accounted for.

In my opinion it is not necessary. The fact that lithium ions arrange themselves over crystal lattice sites is already accounted for by the potential used in the calculations. The potential is specifically constructed to agree with observed lattice parameter, elastic properties, dissociation energy and molecular size. The fact that ion occupy the lattice site means only the high probability to find it in that place. The notion of lattice site for Li^6 ions in compressed crystals can become meaningless. The binding energy of Li ion at the site can be easily calculated. As long as this energy is lower than binding energy of the ion in a molecule the ions can be thought of as occupying lattice sites. In opposite case, which is realized already at 18-20% compression, the potential reach a minimum value not at a point but on the surface similar to spherical surface in a molecule.

At present it is difficult to state with any certainty about the technical possibility for the fabrication of the proposed heterostructures since the author is unaware of any experiments on epitaxial growth of very thin lithium halogenides films on ionic crystals substrates. Published experiments with other ionic materials [5] have shown that pseudomorphic films (several monolayers thick) can be produced for layer/substrate mismatch up to 10%. It is hoped that using solid alloys between LiI (or LiBr) and other ionic compounds in order to reduce their misfit with corresponding substrates can lead to success especially if one takes into account a very high compressibility (almost record-high among binary chemical compounds) of LiI. It is quite possible that for the fabrication of materials with high temperature ionic superconductivity a combination of epitaxy and hydrostatic compression will be required.

As can be seen from Figure 3 very high pressures (> 50 GPa) are required in order to compress bulk crystals. In real experiments at such a high pressures a substantial disordering of crystalline structure is always observed. And one can suspect that due to avalanche growth of lattice irregularities at very high pressures the crystal lattice as a periodic ensemble cease to exist.

Heterostructures could be a remedy in that respect. To obtain the necessary compression in a heterostructure (for example in LiI/LiBr with $\sim 9\%$ misfit) the pressures required are several times lower than 50 GPa, which must be applied for bulk LiI. The use of perfect substrates and growth of dislocation-free pseudomorphic layers can be regarded as additional factors for preservation of crystalline order upon subsequent compression.

It may be of interest to consider a natural question: "Why the wave-like ionic conductivity is absent in common ionic crystals?" Let's consider for an example Li^7I crystal. Since there is spin-degeneracy for Li^7 ions the number of available states in ionic valence band must be much higher than ion number density. The absence of wave-like ionic conductivity

¹For example, it has been reported in [6] that in Na halides the type of crystal structure changes from NaCl to CsCl in the pressure range of 20-30 GPa. If similar structural changes do occur in Li halides then ion movements in crystals is possible at least in the conditions of structural transformation and ionic superconductivity should probably be looked for at these conditions.

is then possible only if the spin-degeneracy is somehow removed. It is easy to see, however, that only one spin state is energetically favorable for ions in ionic crystals. Indeed, in Heisenberg approximation (used in treatments of ferromagnetism) it has been shown that for a system consisting of two atoms or ions the energy change due to spin moment reversal is given by $U_{exc} = -2A(\vec{S}_i \vec{S}_j)$ where U_{exc} is the exchange energy and \vec{S}_i, \vec{S}_j are the spin moments of interacting atoms. All known calculations of exchange integral (A) indicate that it has a positive sign (the only exception being exchange interaction between the atoms with incompletely filled internal electronic shells) and the magnitude of A is about several eV. Under these circumstances the antiparallel spin orientation of lithium and halogen ions will provide the minimum of the system energy and the difference of exchange energy due to spin change by 1 for any ion in the pair will be $2A\vec{S}_i\vec{S}_j - 2A\vec{S}_i(\vec{S}_j - \vec{1}) \gg kT$. Then all lithium ions in the crystal (or its part) are forced to have the same spin value, corresponding to the minimum of system energy. That is almost equivalent to completely filled ionic valence band. In our opinion, this is a reason for absence of wave-like ionic conductivity in common ionic crystals. Another reason, however, can be a small ionic wave function overlap in those crystals.

V. CONCLUSION

It is far from being clear to what extent the band-structure theory can be applied to the ions in ionic crystals. As to electrons, the theory already has a multiple experimental confirmations. Unfortun-

nately, for ions there is presently no experimental facts supporting or denying its applicability, simply because it is impossible to study magnetic and electrical properties of any substance at 10^{-40} K. It seems also that nobody has ever compressed ionic crystals with lithium isotope by 20% and more. Only experiment is able to definitively answer the question about the adequacy of our calculations to reality. The author hopes that some of the readers will be conducive in doing experiments and discovering ionic superconductivity. Some additional information concerning ionic superconductivity can be found on the site www.v-ioffe.ru.

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Valery M Ioffe, the writer of a number of nonstandard scientific works and inventions, has completed institute of electrical engineering in 1974 in Novosibirsk, then worked in institute of physics of semiconductors and institute of electrical engineering the engineer and scientific employee, built houses in the Kemerovo area, has struck out some in essence new semiconductor devices and has offered idea of other superconductivity, site www.v-ioffe.ru