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AN EXPLANATION FOR THE OSCILLATIONS IN THE PROBABILITY OF NEGATIVE MUON CAPTURE BY ATOMS BASED ON THE PERIODIC OSCILLATIONS OF STOPPING POWER

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The oscillations in the probability of atomic capture of stopped negative muons $W(Z_1)/W(Z_2)$ with the cation Z_1 atomic number discovered for the first time in oxides^{/1/} and presently observed in some other compounds (see refs.^{/2-6/} presenting the summary data) are regarded as the most characteristic property of muon localization in matter.

Since ref.^{/1/}, there has been established a consistent tradition of interpreting the oscillations of functions $W(Z_p)/W(Z_p)$ via the modulating influence of chemical bond factors against the background of a monotonously changing with Z_1 total number of electrons in the atom. This assumption has been stimulated, evidently, by a well known initial formulation of the Fermi-Teller "Z-law" ^{/7/}. In conformity with this law the relative probability of negative muon capture by atoms (per one atom) in chemical compounds and mechanical mixtures is equal to the ratio of atomic number values components.

The "Z-law" appeared as a result of basically correct (but approximated) Fermi-Teller calculation of Z dependence for the rate of energy loss by muons penetrating through media.

Starting from the very first researches of the Coulomb capture of negative muon - practically all experimentalists and theorists used an approximated finite relation

$$W(Z_{1})/W(Z_{2}) = Z_{1}/Z_{2}$$
 (1)

instead of a more general relation binding the probability of negative muon capture by atoms with stopping power assumed as a basis for the Fermi-Teller model^{/8/}

$$W(Z_1) / W(Z_0) = S_e(Z_1) / S_e(Z_2),$$
 (2)

where $S_{e} = \frac{1}{N} \frac{dE}{dx}$ is stopping power of the atom (of its electron part, to be exact); $\frac{dE}{dx}$, linear energy losses by charged particles in the element target; N, the number of atoms per cm⁸.

Further development of the theory of muon capture by atoms in a number of papers (see for example ref. $^{9/}$) has brought to more complex but always smooth dependences of $W(Z_2)/W(Z_2)$ on Z_1 which are in better agreement with the experimental data averaged by the big interval Z_1 , than those of relation (1). Since the Fermi-Teller theory (and its subsequent modifications, see ref. $^{\prime 9\prime}$) does not distinguish between chemical compounds and mechanical mixtures of atoms, and the obvious taking into account of chemical binding (when the capture probability changes proportionally to the relative number of remaining electrons) does not reproduce the oscillations, there have appeared phenomenological models. The authors attempted within the framework of these models to interpret the deviations $W(Z_1)/W(Z_2)$ from the smooth dependence on the basis of different assumptions $^{\prime 4,6,10,11\prime}$

By the present paper we would like to attract attention to the fact that the oscillations $W(Z_2)/W(Z_2)$ can be explained in a natural way basing on the main provisions of the Fermi-Teller model expressed through relation (2).

This relation binds the two measurable physical quantities which are presently studied independently. For those dealing with physics of negative muon capture by atoms remain unknown the impressive achievements (see for example 12) obtained in the last decades in the experimental and theoretical investigation of the process of stopping the low energy charged particle and related directly to the problem of negative meson capture by atoms, since by recent theoretical estimations $^{13\cdot15}$ muons are captured at $10^1:10^2$ eV energies.

The most characteristic property of low energy particles stopping (at the velocities $v \leq v_0$, where $v = 2.2 \times 10^8$ cm/s the velocity of an electron in the atom of hydrogen) is, like in the case of muon capture by atoms, the presence of function $S_e(Z_1)$ periodic oscillations depending on the atomic number Z_1 of the stopping atom of the element target, measured at a constant for all stopping atoms velocity of incident particles (12). It has been discovered 10 years before the observation of the oscillations of muon Coulomb capture probability.

Figure 1a presents the values S_e for the electronic stopping cross section of single charged positive elementary particles and ions on the element targets at the velocity $v = 2 \times 10^8 \text{ cm/s}$, obtained on the basis of compiled in ref.^{12'} experimental data on stopping of protons and alpha particles of different energies (the data for boron are based on the experiments on stopping the ions of Lithium ^{12,16'}). The values S_e for Nu , Zn , As , Tl , U are deduced through interpolation, with taking into account $S_{e^-}v$ at $v < v_0$. This statement is checked experimentally up to proton velocities $v = 10^7 \text{ cm/s}^{-12'}$ which is equivalent to the muon energy of the order of several eV. For each element we took the mean values from the proton and alpha particle measurements (S_e values for alpha particles in the wide energy scale are twice as large as for pro-





tons, that is why at such averaging the values obtained through alpha particles, have diminished twofold) with an error depending on the deviation of these two values from the mean value (the mean value of the deviation was +11%). In cases when there were data either for protons or alpha particles, along with the experimental error we took into account the systematic error +11%. Reference^{/12/} points out that the systematic errors at measuring S_e, are as a rule, considerably larger than the statistical one. That is why at averaging of data from different papers, with each of them measuring with high statistical accuracy the amplitudes of oscillations of $\rm S_e$ for a certain set of elements (see for example ^{16 \prime}), the total error increases.

The data plotted in Fig.1a demonstrate the character of periodic oscillations $S_{e}(Z_{1})$ which show up against the background of a smooth grouth of S_{e} values with increasing Z_{1} at small Z_{1} , with S_{e} reaching the plateau at large Z_{1} . There have been suggested several formulae for a phenomenological description of oscillations $S_{e}(Z_{1})$. The arrows above the axis of abscissae of Fig.1a indicate the location of function $S_{e}(Z_{1})$ minima and maxima for low energy protons (following from the formula of ref.^{12/}). But this formula is not accurate enough in reproducing the dependence of oscillation amplitude S_{e} on Z_{1} and v. Due to this the periodic curve in Fig.1a is drawn via the experimental values with taking into account the extremes relative depth following from ref.^{12/} in the places where the experimental data are absent.

The amplitude of function $S_e(Z_1)$ oscillations tends to zero at the increase of particle velocity (at highly accurate measurements the oscillations are still visible at $v = 10^9 \text{ cm/s}^{17}$) and at $v < v_0$ it is independent of v^{-12} . The oscillations $S_e(Z_1)$ are explained $^{(12,18,19)}$ by the peculiarities of the structure of the electron shells. $S_e(Z_1)$ is independent of orbitals dimensions and of the number of electrons on these orbitals. As a first approximation $S_e^{-\sum_i \pi} \pi n_i r_i^2$, where r_i

is the radius of i-orbital; n_1 -number of electrons on the orbital. According to the phenomenological formula from ref.¹² the S_e dependence on Z_1 manifests periodicity of two types: with periods approaching $\Delta Z_1 = 18$ and 9. The period with $\Delta Z_{\pm} = 18$ corresponds approximately to the periodicity of element properties in two big periods. The period with $\Delta Z_1 = 9$ corresponds to the periodicity of properties of the main group elements. These elements contain completely closed d and f shells (or they are stripped of all electrons on d and f shells).

Basing on relation (2) one can expect the oscillation $S_e(Z_1)$ to manifest itselves in the dependence of $W(Z_1)/W(Z_2)$ on Z_1 . And the character of oscillations $W(Z_1)/W(Z_2)$ really has much in common with those observed for $S_e(Z_1)$. Figure 1b gives the values of $W(Z_1)/W(0)$ for oxides. We have obtained these values by averaging the data from different papers (the experimental data summary tables are presented in refs.^(2-6/)). The broken curve in Fig.1b is not drawn via the experimental points, it repeats the curve of Fig.1a preserving the relative amplitude of oscillations and the location of extremes. The experimental data on $W(Z_1)/W(Z_2)$ do not contradict and in the region $Z_{1}=35,87$ even correspond to the character of oscillations S_{e} . Thus, it is necessary to assume that the probability of atomic capture and the effective stopping are defined by the same atomic structure factor - the structure of electron shells.

But the smooth dependence of the averaged values $W(Z_1)/W(Z_2)$ and $S_e(Z_1)$ is not the same in the region of small Z_1 ; the values $W(Z_1)/W(0)$ drop stronger with decreasing Z_1 than the values $S_e(Z_1)$. This, along with the fact that the chemical binding of elements in oxides preserves the character of $S_e(Z_1)$ oscillations measured for element targets, can be understood within the framework of the concepts used for explaining the oscillations $S_e(Z_1)$.

The bulk of experimental data on the values of S_e refers to element targets. But lately there started to appear more and more frequently the investigations of stopping power of chemical compounds. These investigations (see for example/16/) are aimed at verifying the Bragg rule by which the stopping power of a chemical compound is a sum of stopping powers of the atoms it comprises. It turned out/16/, that at low particle energies the Bragg rule is more often than not violated due to the influence of the chemical bond on the value of atomic S_e in the compound. When the atoms are coupled by a chemical bond there takes place a redistribution of electrons, followed by changes in the radius of their orbitals and subsequently by a change in S_e .

For quantitative comparison of the data on S_e and $W(Z_1)/W(0)$ in oxides it is necessary to correct the value S_e by taking into account the redistribution of electrons at chemical binding of atoms; i.e., to obtain the values S'_e for the atoms contained in oxides. We defined this correction in the same way for all elements on the basis of the data of ref.^{19/} which gives (see Fig.1 of ref.^{19/}) the results of calculating the contribution of different shells into the stopping power of atoms with $Z_1 \leq 54$. It was assumed that an atom of an element transfers (depending on the valency of the atom in the oxide) to oxygen different integral number of electrons equal to the formal valency of this atom. The essence of this assumption is discussed further below.

The values of the correcting factor a, which should be multiplied by the value S_e estimated on the element target in order to obtain the value S'_e for the stopping power of an atom in the given compound, have been defined in the following way.

They have been taken equal to a=1-p, where p - the relative contribution into S_e of those electrons which are

transferred by the atom to oxygen. If an atom transfers a part of its electrons from one of its shells, the contri-

bution of the transferred elect-1.0 rons into S_e is taken propor- α tional to their contribution into the initial number of electrons in the given shell. 0,5 Fig.2. The dependence of corrective factor α on Z₁. The calculated value a, through which smooth curves are drawn. Z is presented by indexes(\Box ,o, Δ) 30 50 for cations 1^+ , 3^+ and 5^+ , 6^+ . 70 90 10 $W(Z_1)W(0)$ W(Z1)/W(OH) $W(Z_1)/W(J)$ 1,5 a) b) c) 5 5 1,0 0,5 $S'_{e}(Z_{i})$ $S'_{e}(Z_{i})$ $S'_{e}(Z_{1})$ 10 20 30 30 20 10 $W(Z_1)/W(F)$ $W(Z_{\mu})/W(S)$ 3 d) e) f) 5 2 2 1 $S'_{e}(Z_{1})$ $S'_{e}(Z_{1})$ Se (Z

Fig.3. The relation of the probabilities of atomic capture W(Z₁)/W(Z₂) (per atom) depending on the stopping power S'_e(Z₁)for different Z₂: a) oxides (χ^2 per point 1.); b) hydroxides (the values for H₂SeO₃ and H₂SeO₄ are averaged; χ^2 per point 0.9); c) iodides (χ^2 per point 1.0); d) fluorides (χ^2 per point 0.2); e) chlorides (without AgCl; ; χ^2 per point 0.3); f) sulphides (the values for FeS and FeS₂ are averaged; χ^2 per point 0.5).

We defined factor a by the data of ref.⁽¹⁹⁾ individually for each atom with $Z_1 \leq 54$. For heavier atoms such data are not available. But the values for elements with different valencies depend smoothly on Z_1 and reach the plateau at large Z_1 (see Fig.2). That is why we have taken this extrapolated bound values a for heavy elements.

Figure 3 presents the data on $W(Z_1)/W(0)$ for oxides and S'_e, whence it appears that there is a linear dependence between these quantities. χ^2 per point is close to 1 (calculating χ^2 we have taken into account the error for both $W(Z_1)/W(0)$ and S'_e: the relative errors S'_e and S_e have been assumed to be equal). This means that the spread of values $W(Z_1)/W(0)$ versus S'_e depends only on the error of measuring these quantities. It is important to note that the deviations of points in fig.3a from the straight line corresponding to the minimum χ^2 are of accidental character and the systematic grouping of points depending on such parameters as Z_1 , formal valency of the element in the given compound, the position of the element versus the function extremes S_e(Z₁) is not observed.

The linear dependence of $W(Z_1)/W(Z_2)$ on S'_e , similar to the one for oxides in Fig.3a also shows itself in the case of atomic captures in hydroxides, fluorides, chlorides, sulphides and iodides (Figs.3b-e). The values S'_e for the same cation have been assumed to be the same in all the compounds. The character of the bond $W(Z_1)/W(Z_2)$ and S'_e is in good agreement with relation (2) if one understands S'_e as a value corrected by the chemical bond.

For cations with equal charges the dependence of a on Z_1 is of smooth character (Fig.2) and the value of a is not correlated with positions of the function extremes $S_e(Z_1)$ (the mean deviation of a from the curves +3.5%). In reality, there is a change of a within the limits of each period of the Mendeleev table, since a depends on the cation charge. This explains the difference of curves $W(Z_1)/W(Z_2)$ and $S_e(Z_1)$ in the region of $Z_1 < 20$ (Fig.1). The amplitude of a-changes in the range $Z_1 > 20$ is significantly smaller than the amplitude of oscillations $S_e(Z_1)$. That is why chemical binding of elements does not change significantly (for $Z_1 > 20$) the position and amplitude of $S'_e(Z)$ value extremes with respect to $S_e(Z)$.

It is desirable to stress one more circumstance. The mean value of the oscillation amplitude for stopping of nonchanneled particles by the data of numerous investigations (see ref. $^{12/}$) is equal to $\pm 25\%$ like in the case of oscillations in the probability of atomic capture in all the discussed compounds. In the experiments studying the stopping of channeled

particles^{/20/} when the stopping occurs mainly on valence electrons, the mean amplitude of oscillations S_e is approximately twice as high.

The assumption that the oscillations $W(Z_1)/W(Z_2)$ depend only on the oscillations $S_e(Z_1)$ means that the contribution from valence electrons into the process of atomic capture is equal, like in the stopping process on which it depends, to approximately a half, i.e., the contribution from one valence electron exceeds considerably the contribution from the core electron. This fact has been first established in our paper $^{/21}$ on the basis of processing the data on negative muon capture by atoms in chloralcyls. This deduction is verified by the results of recent calculations of the contribution from different atomic shells into the stopping power (the calculations make use of the atomic wave functions by the Hartree-Fock-Slater model $^{/18,19/}$). And really the total contribution of valence shell electrons for different shells (and with shells filled to different extents) is 35+65%.

From the fact that the mean values of the oscillation amplitudes $W(Z_1)/W(Z_2)$ and $S_0(Z_1)$ are approximately equal it follows (assuming that the oscillations $W(Z_1)/W(Z_2)$ depend upon the oscillations $S_0(Z_1)$) that the capture of muons by atoms in the compounds under discussion occurs at $v \leq v_0$.

On the face of it the assumption of the overall electron transfer made at calculating the factor a contradicts to the known values of the ionic charge for oxides $^{22/}$ which do not reach their limiting values equal to the formal valency. Nevertheless when considering both the stopping of charged particles and the meson capture by atoms in chemical compounds it is necessary to take into account along with electron transfer the Debye shielding factor $^{12,23/}$. In the case of atomic capture when the meson is close to the cation it repels the valence electron cloud which assists in creating the situation when the cation displays its formal valency.

It is important to take into account the Debye shielding factor only for light elements of the I:III groups. For intermediate and heavy element with large formal ionic charge a single muon charge is certainly not enough to provide for the multiple electron transfer from cation to anion. But in the latter case the values of a are, as a rule, little different from unity and it is senseless presently to take into account (within its large errors when measuring $S_e(\pm 17\%)$) the true value instead of the formal charge.

In future, with more accurate measurements of S_{θ} it will be necessary to refine the procedure of introducing the correction for electron redistribution. Note that for fluorides (Fig.3) when the ionic charges are approaching the limiting values $^{/22/}$ defined by valency without taking into account the Debye shielding factor the linear relation between $W(Z_1)/W(Z_2)$ and S'_{e} is expressed most clearly.

It is possible to maintain the periodic character of oscillations W(Z₁)/W(Z₂) (caused by S_e oscillations) only when the mean velocity of mesons in atomic capture \bar{V}_{capt} is a smooth function of Z and there are no sharp changes in \bar{V}_{capt} for elements close to Z. If the premises leading to the linear dependence W(Z₁)/W(Z₂) on S'_e are correct one should assume that at $Z \ge 3$ within the error of measurements W(Z₁)/W(Z₂) and S_e the value of \bar{V}_{capt} depends weakly on Z or that the dependence of \bar{V}_{capt} on Z has the same form as the dependence of α on Z.

The process of atomic capture, speaking generally, is not reduced to stopping though it is a part of the latter. In literature two more factors are discussed which are characteristic only for the process of atomic capture. First of all, it is a factor taking into account the relative probability of meson transfer into the bound state during stopping. This factor depends primarily on the density of bound states at the mesic atom formation. According to estimations '24' this factor depends weakly on Z. Reference $\frac{725}{}$ (see also ref. $\frac{10}{}$) discusses the influence of ionic charge on the probability of atomic capture in oxides. Judging by the fact that in Fig.3 the points for differently charged ions are located symmetrically versus the straight line one can conclude that in oxides and other ionic compounds this factor for positive ions depends weakly on the ionic charge. Nevertheless we think that for F⁻, the oxygen ion (as well as for the ions of OH-, Cl-, I^- , S^-) both the above-mentioned factors can be different from the value which is characteristic, on the average, for positive ions. Due to this the relation of $W(Z_1)/W(Z_2)$ and S'_2 represented in Fig.3 with straight lines can be written in the following form,

 $W(Z_1)/W(Z_2) = K(Z_2)S'_{e}(Z_1),$

where $K(Z_2) = a(Z_2)/S_{\theta}(Z_2)$; $a(Z_2)$ is the coefficient which takes into account the ratio of the above-mentioned factors for positive ions (averaged as a first approximation for all $Z \ge 3$) and anions O⁻, OH⁻, F⁻, Cl⁻; S'_{\theta}(Z_2), the stopping cross section of these ions. The values of coefficient K for oxides, hydroxides, fluorides, chlorides, sulphides and iodides are equal respectively to 0.190+0.025; 0,240+0.040; 0.210+0.025; 0.130+0.038 and 0.058+0.027.



Fig.4. The correlation between the coefficient $K(Z_2)$ and the atom ionic radius Z_2 .

The theoretical evaluation of the absolute values of coefficient "K" still seems impossible since it depends on many unknown parameters (and first of all on the unknown values of S_e for anions) \overline{V}_{capt} and the probable smooth dependence of \overline{V}_{eaut} on Z. Beside this, the adopted by us method of calculation the parameter α as well as the probable difference between Se for positive and negative particles, due, in particular, to different influence of the Debye shielding factor even for element targets is a source of uncertainties.

But the relative values of coefficient"K". for example in the series fluoride-chlorides-iodides, are carrying, undoubtedly, some information on the relative values of S_e for anions. Basing on the nature of the atomic structure factor which is responsible for stopping, one should expect a strong dependence of S_e on the radius of the anion. And really (see Fig.4) by using the obtained by us values of"K" and the values of ionic (or covalence) atom radius one can demonstrate "K"~ Z^{2.±0.5}.

Among the scanty yet examples of studying S_e in chemical compounds, overlapping with the objects of respective measurements of atomic capture the experiments defining the $S_e(H)$ of the hydrogen atom in hydrocarbons with single, double and triple bonds^{/26/} are of special interest. It appears that moving from ethane to acetylene the $S_e(H)$ reduces substantially (for low energy particles) to approximately one third and the $S_e(C)$ of the carbon nucleus becomes 1.6 times bigger, i.e., the ratio $S_e(C)/S_e(H)$ grows to approximately 5 times as much; the same conclusion follows from ref.^{/27/}, there the measurements are carried out with low energy protons.

From the measurements of the probability $W \circ f \pi$ -mesons capture by hydrogen in hydrocarbons depending on the degree

of saturation of C-C bond it is known^{28-30/} that W becomes 5 times smaller with increasing multiplicity of the bond in the series $C_2H_6-C_2H_4-C_6H_6$ (benzene is a trimer of acetylene C_2H_2). It is evident, that the interpretation of these data should be revised since relation (2) is also valid for π^- mesons and the found strong dependence of $S_e(H)$ on the type of chemical bond C-C-covers all the effect of changing the probability of π^- -meson charge exchange on the nuclei of bound hydrogene observed in the experiments on π^- -capture.

Different variants of particle stopping theory based on the use of Thomas-Fermi model of the atom (i.e., paying no attention to the shell structure of the atom) for example the one by Firsov^{31'} or by Lindhard-Schraff^{/32'}bring to smooth dependences of S_e on Z (at a given v), which describe well enough the main tendency in the change of S_e with growing $Z^{/12'}$. Thus, for example, one of the most recent works^{/33'} brings in the case of v< v₀ region to the dependence S_e~Z^{1/3}. Note, that analogous form of W(Z₁)/W(Z₂) dependence on Z₁, has been obtained in the theory of atomic capture (see^{/9/}).

Only after the modification of the charged particle stopping theory by using atomic wave functions of the Hartree-Fock-Slater model, it has commenced to describe more or less satisfactorily the oscillations $S_{e}(Z)$ (see, e.g., ref.^{34/}). Theoretical investigations in this direction are presently intensively developing. One can think that the use of the Hartree-Fock-Slater model in the theory of meson capture by atoms will enable to describe the functions $W(Z_1)/W(Z_2)$ within the framework of the concepts from the paper by Fermi-Teller^{/8/}.

The role of the theory binding the atomic capture with stopping power of the matter seems to be quite significant. It can be used as a basis for quantitative description of such values as probability and mean energy of negative meson capture by atoms and to obtain also the values of atopping power for the atoms contained in chemical compounds (for which the probability of meson capture by atoms is measured) since it is impossible to measure them directly for each individual compound.

The indication to the common nature of the oscillations in atomic capture probability and in effective stopping cross section obtained in this work necessitates new and more accurate measurements of the values of these quantities with the aim both of establishing the exact quantitative correlation between the extreme positions in the dependence of these quantities on Z and of studying the Debye shielding factor at stopping of oppositely charged elementary particles in the measurements of S_e on the element targets and in molecular compounds.

The main results of this research can be formulated as follows:

1. It is noted for the first time that within theoretical concepts of Fermi and Teller it is possible to explain the periodic oscillations of the probability of meson capture by atoms, since they are caused by the same factors as the periodic oscillations of the matter stopping power.

2. It is demonstrated that the character of oscillations in the probability of muon capture by atoms in oxides does not contradict to and in some regions even agrees with the character of periodic oscillations in the matter stopping power.

3. A linear dependence is obtained for six classes of compounds between the relative probability of muon capture by atoms on cations and anions and the atom stopping power, corrected by the redistribution of electrons at chemical coupling of atoms; this dependence agrees with the initial theoretical ideas of Fermi-Teller.

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2.4