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THE BORN-OPPENHEIMER METHOD IN THE THEORY OF ATOMIC COLLISIONS

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A simple formula is obtained which permits one to calculate the exchange scattering of electrons by atoms with approximately the same degree of accuracy as that yielded by the Born formula in problems in which exchange does not have to be taken into account. The excitation functions are calculated for the 2^3S and 2^3P levels in helium. The results are compared with experiments and with calculations of exchange scattering based on the classical mechanics approximation.

INTRODUCTION

IT is well known that the probability of exchange scattering of an electron by an atom falls off rapidly as the collision energy increases. Therefore calculations using the Born-Oppenheimer formula at high energies which are the only ones for which it is valid are of no particular interest. On the other hand attempts to extrapolate it to the domain of low energies lead to poor, and often even quite meaningless results[1].

The few attempts undertaken to date[2-4] to obtain satisfactory results have been associated with a further significant complication of the calculations and have not always led to the desired goal. However, from our subsequent discussion it will be seen that the deficiencies of the calculations using the Born-Oppenheimer formula are determined not by the fact that this formula is essentially bad, but only by an incorrect extrapolation into the domain of low energies. By using it as a basis we can obtain a simple formula free from these defects.

The principal idea which is carried through below consists of the following. In the derivation of the Born-Oppenheimer formula, just as in the case of the derivation of the Born formula, the electron incident on the atom and the scattered electron are both described by plane waves. This is correct if $|\mathbf{k}| \gg 1$ and $|\mathbf{k}'| \gg 1$, where $\mathbf{k}$ and $\mathbf{k}'$ are the corresponding propagation vectors (we use atomic units throughout). It is obvious that if we consider these conditions to be satisfied and expand the amplitude of the exchange scattering written in the Born-Oppenheimer approximation in a series in inverse powers of k or k', then only the first term of this series will have a real meaning. The remaining terms should be discarded since they are of a higher order of smallness, and the corresponding effects can not be taken into account within the framework of the first order of approximation of perturbation theory. But if such terms are retained, then in extrapolating into the domain of low energies they cease to be small and lead to meaningless results.

Before proceeding to the selection of the principal term of the amplitude of exchange scattering we merely note that in the case without exchange such a selection need not be carried out since the Born formula itself is the principal term of the corresponding asymptotic series.

SELECTION OF THE PRINCIPAL TERM OF THE SCATTERING AMPLITUDE AND THE GENERAL BEHAVIOR OF THE CROSS SECTION

In order to avoid unnecessary complications we shall carry out the calculations for the special case of the collision of an electron with a helium atom. A discussion of the collision of an electron with an arbitrary atom can be carried out in a completely analogous manner.

In our example the exchange scattering amplitude for the transition from the state α to the state β is given by

$$g_{\alpha\beta}(k, \vartheta, \varphi) = \frac{1}{2\pi}\int\left(\frac{1}{r_{31}} + \frac{1}{r_{32}} - \frac{2}{r_3}\right) \times e^{i(\mathbf{k}\mathbf{r}_3 - \mathbf{k}'\mathbf{r}_1)}\bar{\psi}_\beta(\mathbf{r}_2, \mathbf{r}_3)\,\psi_\alpha(\mathbf{r}_1, \mathbf{r}_2)\,d\tau_1\,d\tau_2\,d\tau_3. \quad (1)$$

Here $\psi_\alpha(\mathbf{r}_1, \mathbf{r}_2)$ and $\psi_\beta(\mathbf{r}_2, \mathbf{r}_3)$ are the atomic functions for the initial and the final states which can have the same or different symmetries depending on the term being excited. We break up $g_{\alpha\beta}$ into three terms and consider the first one of these:

$$g^{(1)}_{\alpha\beta} = \frac{1}{2\pi}\int \frac{1}{r_{31}} e^{i(\mathbf{k}\mathbf{r}_3-\mathbf{k}'\mathbf{r}_1)}\bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_3)\,\psi_\alpha(\mathbf{r}_1,\mathbf{r}_2)\,d\tau_1\,d\tau_2\,d\tau_3$$
$$= \frac{1}{2\pi}\int \psi_\alpha(\mathbf{r}_1,\mathbf{r}_2)\,I_\beta(\mathbf{r}_1,\mathbf{r}_2)\,e^{i\mathbf{q}\mathbf{r}_1}\,d\tau_1\,d\tau_2, \qquad (2)$$

$$I_\beta(\mathbf{r}_1,\mathbf{r}_2) = \int \bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_3)\frac{e^{ikr_{31}}}{r_{31}}\,d\tau_3, \qquad (3)$$

where $\mathbf{q} = \mathbf{k} - \mathbf{k}'$.

For the evaluation of I_β it is convenient to go over from the variable $\mathbf{r}_3$ to the variable $\mathbf{r}_{31}$:

$$I_\beta(\mathbf{r}_1,\mathbf{r}_2) = \int \bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_1+\mathbf{r}_{31})\,e^{ikr_{31}\cos\theta}\,r_{31}\sin\theta\,dr_{31}\,d\theta\,d\chi$$
$$= \int \Psi_\beta(\mathbf{r}_2,\mathbf{r}_1,r_{31},x)\,e^{ikr_{31}x}\,r_{31}\,dr_{31}\,dx, \qquad (4)$$
$$\Psi_\beta(\mathbf{r}_2,\mathbf{r}_1,r_{31},x) = \int \bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_1+\mathbf{r}_{31})\,d\chi, \qquad x = \cos\theta.$$

We denote by r_{31}, θ and χ the spherical polar coordinates of the vector $\mathbf{r}_{31}$ with the origin chosen at the end of the vector $\mathbf{r}_1$ and the z axis directed along $\mathbf{k}$.

Integrating by parts with respect to θ and with respect to r_{31} and neglecting each time terms of higher order of smallness we obtain

$$I_\beta(\mathbf{r}_1,\mathbf{r}_2) = \frac{1}{ik}\int_0^\infty [\Psi_\beta(\mathbf{r}_2,\mathbf{r}_1,r_{31},1)\,e^{ikr_{31}}$$
$$- \Psi_\beta(\mathbf{r}_2,\mathbf{r}_1,r_{31},-1)\,e^{-ikr_{31}}]\,dr_{31} + \ldots$$
$$= k^{-2}[\Psi_\beta(\mathbf{r}_2,\mathbf{r}_1,0,1) + \Psi_\beta(\mathbf{r}_2,\mathbf{r}_1,0,-1)]$$
$$+ \ldots = 4\pi k^{-2}\bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_1). \qquad (5)$$

Here we have taken into account the fact that $\Psi_\beta(\mathbf{r}_2, \mathbf{r}_1, r_{31}, x) \to 0$ for $r_{31} \to \infty$ and $\Psi_\beta(\mathbf{r}_2, \mathbf{r}_1, 0, \pm 1) = 2\pi\bar{\psi}_\beta(\mathbf{r}_2, \mathbf{r}_1)$.

Thus, up to terms of higher order of smallness we have

$$g^{(1)}_{\alpha\beta} = 2k^{-2}\int \bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_1)\,\psi_\alpha(\mathbf{r}_1,\mathbf{r}_2)\,e^{i\mathbf{q}\mathbf{r}_1}\,d\tau_1\,d\tau_2. \qquad (6)$$

By a similar calculation we can verify that for $k \gg 1$

$$g^{(2)}_{\alpha\beta} = \frac{1}{2\pi}\int \frac{1}{r_{32}} e^{i(\mathbf{k}\mathbf{r}_3-\mathbf{k}'\mathbf{r}_1)}\bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_3)\,\psi_\alpha(\mathbf{r}_1,\mathbf{r}_2)\,d\tau_1\,d\tau_2\,d\tau_3$$
$$= O(k^{-6}), \qquad (7)$$
$$g^{(3)}_{\alpha\beta} = \frac{1}{2\pi}\int \frac{1}{r_3} e^{i(\mathbf{k}\mathbf{r}_3-\mathbf{k}'\mathbf{r}_1)}\bar{\psi}_\beta(\mathbf{r}_2,\mathbf{r}_3)\,\psi_\alpha(\mathbf{r}_1,\mathbf{r}_2)\,d\tau_1\,d\tau_2\,d\tau_3$$
$$= O(k^{-6})$$

and, consequently, there is all the more reason for neglecting them (when ψ_α and ψ_β have different symmetry we have $g^{(2)}_{\alpha\beta} = O(k^{-7})$)

Thus, we see that the amplitude of exchange scattering $g_{\alpha\beta}$ reduces to expression (6) which differs from the corresponding amplitude of direct scattering only by having the factor k^{-2} replacing the factor q^{-2}. However, this difference is very important, since it leads to an essentially different dependence of the cross section on the energy. Indeed, for the excitation of the triplet state from the singlet state we have

$$\sigma_{\alpha\beta} = 3\frac{k'}{k}\int |g_{\alpha\beta}(\vartheta,\varphi)|^2\sin\vartheta\,d\vartheta\,d\varphi$$
$$= \frac{24\pi}{k^6}\int_{q_{min}}^{q_{max}} |\langle\beta|\,e^{i\mathbf{q}\mathbf{r}_1}\,|\alpha\rangle|^2 q\,dq, \qquad (8)$$

and since for $k \gg 1$ the integral over q practically does not depend on k the cross section turns out to be inversely proportional to the cube of the energy.

The calculations just carried out can be repeated performing the expansion of $g_{\alpha\beta}$ not in terms of inverse powers of k, but in terms of inverse powers of k′. If $k \gg 1$, then the two results coincide. But near the threshold only that formula is applicable which is obtained by expanding in inverse powers of the larger of the two wave numbers, i.e., in powers of $1/k$ in the case of excitation and in powers of $1/k'$ in the case of deexcitation. Such a choice of the small parameter is natural from the formal point of view. It is also easy to verify that it is just in the case of such a choice that the expression for the cross section satisfies the principle of detail balancing.

From (8) it may be seen that the transition probability is determined by the value of the matrix element $\langle\beta|\exp(i\mathbf{q}\cdot\mathbf{r}_1)|\alpha\rangle$ for $q \sim 1$. Therefore among the excited levels which differ by the values of their orbital quantum number there is no single one for which the transition probability would be considerably greater than that for the others, as happens in the case of optically allowed transitions in scattering without exchange.

A more detailed investigation shows that, generally speaking, nevertheless there exists a preference for transitions without a change of l. The transitions with $\Delta l \geq 2$ are appreciably less probable. As regards the dependence of the cross section on the principal quantum number, it can be easily obtained in the case $n \gg 1$ when the atomic functions are very close to the hydrogen functions. It is well known [5] that in this case ψ_{nl} depend on n for fixed l only through the normalization factor which is proportional to $n^{-3/2}$ and, consequently,

$$\sigma_{10,\,nl} \sim n^{-3}. \qquad (9)$$

Naturally, this conclusion can also be reached without transforming (1) to (8). Such a dependence of the cross section on n is also confirmed by experimental data. It has already been noted in the literature[6]. But usually, in discussing it, reference is made to the analogy between the cross sections for the excitation of atoms by electron collisions and the oscillator strengths for which this dependence is valid, for the same reasons. But in doing this sight is lost of the fact that the analogy between the excitation cross sections and the oscillator strengths is valid only for optically allowed transitions, while (9) is valid for arbitrary changes in l which are of practical interest. Specific calculations as well as an investigation of the available experimental data show that this dependence is approximately satisfied also for low values of n.

Finally, it is of interest to compare the exchange and the direct scattering. The simplest and the most obvious example is afforded by the problem of the excitation of the hydrogen atom for which

$$\sigma^{\pm} = \frac{8\pi}{k^2} \int\limits_{q_{min}}^{q_{max}} \left(\frac{1}{q^2} \pm \frac{1}{k^2}\right)^2 |\langle\beta| e^{i\mathbf{qr}} |\alpha\rangle|^2 q\, dq. \tag{10}$$

From this formula it is seen directly that in the excitation of levels corresponding to optically allowed transitions the exchange effect does not play any appreciable role, since in this case the magnitude of the cross section is determined by the behaviour of the integrand for $q \sim q_{min}$. On the other hand, for optically forbidden transitions the region of greatest importance is $q \sim 1$, and the role played by the exchange term turns out to be considerably greater. Apparently there exist indirect experimental data[1] which confirm the validity of this deduction.

In conclusion we note that formula (10) is a peculiar analogue of the well-known Mott formula for the exchange scattering of a free electron by an electron.

CLASSICAL CALCULATION

In calculations of inelastic scattering unaccompanied by exchange the general behavior of the cross section as a function of the energy and of the number of the excited state and also the order of magnitude of its absolute value can be obtained by very simple methods of classical mechanics[7,8]. In this approximation we are dealing with the calculation of the collision of two electrons regarded as classical particles, one of which revolves about the nucleus in a corresponding Bohr orbit. By definition it is assumed that the atom has been excited into its n-th state if in the course of the collision the external electron has transferred to the atomic electron an energy ϵ which satisfies the inequality $E_n \le \epsilon \le E_{n+1}$, where E_n is the excitation potential for the n-th level.

Analytically the calculations can be carried out, naturally, only if we neglect the field of the nucleus. In such a case we lose the possibility of classifying the excited states in terms of their angular momentum and the definition of the cross section given above becomes the definition of the total cross section for the excitation of all the levels lying within the given energy interval.

The success of estimates of this kind makes them useful also in the case of exchange scattering which from the point of view of a classical calculation does not exhibit any essential special features.1) A certain difference consists only of the fact that in order for exchange to occur the external electron must transfer to the atomic electron not only all the kinetic energy which it had prior to the collision, but also a part of the energy acquired in the field of the atom. We shall assume that an exchange excitation of the n-th state of the atom has occurred if in the course of the collision the atomic electron has acquired an energy ϵ which satisfies the inequality

$$E + U_{n+1} \leqslant \varepsilon \leqslant E + U_n, \tag{11}$$

where U_n is the ionization potential of the n-th level.

Since the effect of the acceleration of the external electron by the field of the atom is important for the discussion of exchange scattering, then really we should go beyond the framework of the two body approximation. However, in formulas giving only rough estimates this difficulty can be simply avoided by ascribing to the electron at the "moment of collision" and energy $E + \Delta$ instead of E, where Δ is a certain constant which is at least equal to U_n.

We have noted above that the calculations can be carried through in analytic form only in the two-body approximation. A further simplification of the problem is not important in principle, but desiring to obtain only the general characteristic features of the cross section, we shall consider here the simplest variant of the calculation and we shall assume that the atomic electron prior to the collision is not only free, but also stationary. Then the differential cross section for a collision with an energy transfer ϵ will be given by [9]

$$d\sigma = (\pi/E)\, \varepsilon^{-2}\, d\varepsilon, \tag{12}$$

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1) The possibility of such a calculation was apparently first indicated by M. Grizhinskiĭ (paper presented at the Physicotechnical Institute, Leningrad, 1962).

where E is the energy of the external electron.

Thus, for the cross section of exchange excitation of the n-th state from the m-th state we obtain from (11) and (12)

$$\sigma^{\mathrm{cl}}_{mn} = \frac{\pi}{E+\Delta}\begin{cases}(E+U_{n+1})^{-1}-(E+U_n)^{-1}, & E \geqslant U_m - U_{n+1} \\ U_m^{-1}-(E+U_n)^{-1}, & U_m - U_{n+1} \geqslant E \geqslant U_m - U_n\end{cases}. \tag{13}$$

If $U_n - U_{n+1} \ll U_n$, which occurs in the excitation of sufficiently high levels from an arbitrary initial state, or in the excitation of arbitrary states from a deep lying (as in the case of helium) initial state, then (13) takes on a particularly simple form:

$$\sigma^{\mathrm{cl}}_{mn} = \pi\,(U_n - U_{n+1})\,(E+U_n)^{-3}, \quad E \geqslant U_m - U_{n+1}. \tag{14}$$

Here Δ has been set equal to U_n. If we recall that $U_n - U_{n+1} \sim n^{-3}$, then the complete analogy with (8) becomes evident.

EXCITATION OF THE 2^3S AND 2^3P LEVELS IN HELIUM

Figures 1 and 2 show results of calculations for the excitation of 2^3S and 2^3P levels in helium carried out in accordance with formula (8). The atomic functions were taken in the form

$$\begin{gathered}\psi_{10}(\mathbf{r}_1,\mathbf{r}_2) = \tilde{\psi}_{10}(\mathbf{r}_1)\,\tilde{\psi}_{10}(\mathbf{r}_2), \\ \psi_{nl}(\mathbf{r}_1,\mathbf{r}_2) = 2^{-1/2}\{\psi_{10}(\mathbf{r}_1)\,\psi_{nl}(\mathbf{r}_2) - \psi_{10}(\mathbf{r}_2)\,\psi_{nl}(\mathbf{r}_1)\},\end{gathered} \tag{15}$$

and for the one-electron functions we have utilized simple analytic functions with parameters calculated by Veselov et al.[10]. The same two figures also give the results of calculations in accordance with formula (1) and by utilizing the method of distorted waves taking exchange into account carried out by Massey and Moiseiwitsch[2,3], and also the experimental results. With respect to the latter the following should be noted.

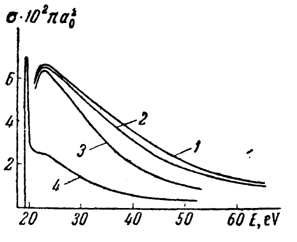

FIG. 1. Excitation of the 2^3S-level in helium: Curve 1 – calculation in accordance with formula (8), 2 – experiment [6], 3 – 1/20 of the cross section calculated in accordance with formula (1) in [2], 4 – calculation by the method of distorted waves with exchange.[2]

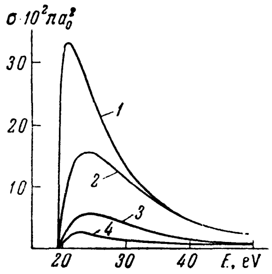

FIG. 2. Excitation of the 2^3P-level in helium: Curve 1 – calculation in accordance with formula (1) of [3], 2 – calculation by the method of distorted waves with exchange,[3] 3 – calculation in accordance with formula (8), 4 – experiment.[3]

In the immediate neighborhood of the threshold the excitation function of the 2^3S level was studied in the paper by Schulz and Fox[11] who have found a maximum equal to $4.5 \times 10^{-2}\ \pi a_0^2$ at a distance of 0.3 eV from the threshold which agrees well with the value of $5.7 \times 10^{-2}\ \pi a_0^2$ which was obtained earlier by Maier-Leibnitz[12]. At a distance of approximately 1 eV from the first maximum a second maximum was found which the authors ascribe to the excitation functions for the 2^1S and 2^3P levels. But measurements of excitation functions of the n^3S levels carried out by optical methods show that the excitation function of an individual level near the threshold can have two maxima[13,14]. As an example Fig. 3 gives a comparison of the results of Schulz and Fox with measurements for the 4^3S-level carried out by Yakhontova[13]. Here there is also observed a very sharp maximum at a distance of several tenths of a volt from the threshold and a second broader maximum at a distance of 2 ev further. If we take into account the fact that the excitation functions for levels belonging to a given series always behave in a very similar manner, then there is no basis for making an exception for the 2^3S level and for assuming that the exceedingly narrow maximum found by Schulz and Fox[11] is the principal one.

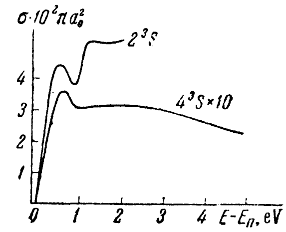

FIG. 3. Excitation functions for the 2^3S and the 4^3S levels in helium near the threshold.

It is quite clear that calculations based on the first order approximation of perturbation theory cannot pretend to give a description of the "fine structure" of the excitation function in the immediate neighbourhood of the threshold. Therefore, as an experimental curve we reproduce the curve drawn in accordance with the data of the paper of Gabriel and Heddle[6] in which the shape of the excitation functions for the n^3S levels was studied for n = 3, 4, 5, 7, while at an energy of 108 eV absolute measurements were also carried out. Assuming that $\sigma \sim n^{-3}$ one can extrapolate these measurements to n = 2.

The choice of an energy of 108 eV for carrying out absolute measurements of the excitation functions for the triplet levels cannot be regarded as fortunate, since at such an energy these excitation functions are already very small, while, for example, the filling of triplet levels as a result of a transfer of excitation from the n^1P levels as a result of atomic collisions is, on the other hand, maximal[15]. However, for the 2^3S level the error associated with this is not too great, since the curve constructed in the manner indicated above gives for the cross section at the maximum $5.6 \times 10^{-2} \pi a_0^2$. The extrapolation of data obtained by Yakhontova for the 3^3S level gives $5.2 \times 10^{-2} \pi a_0^2$. Finally, we note that if the curve of Gabriel and Heddle is normalized in such a way that it coincides with our calculation at E = 108 eV, then at the maximum we obtain $5.4 \times 10^{-2} \pi a_0^2$. Thus, the experimental curve given in Fig. 1 apparently is a bit too high, but the available data do not allow us to say by how much. In any case the true values are not exceeded by a factor of more than 1.5.

The excitation function for the 2^3P level has been measured in the unpublished work of Frost and Phelps (quoted in [3]). Extrapolation of the measurements by Yakhontova for the 3^3P level carried out in the neighbourhood of the maximum yields the same result. But the extrapolation of data of Gabriel and Heddle leads to cross sections which are larger by a factor of almost two. Although a discrepancy by a factor two does not appear to be too great when we are dealing with absolute measurements of cross sections, nevertheless this discrepancy is probably associated primarily with the already noted lack of reliability of measurements at relatively high energies. This is also confirmed by the nature of the dependence of the cross section on the energy. Although in the case of the 2^3S level the calculated and the measured cross sections fall off equally rapidly with increasing energy, nevertheless for the 2^3P level calculations show that starting with an energy E $\sim$ 50 eV the cross section falls off like E^{-3}, i.e., noticeably faster than the experimental cross section, and at 108 ev becomes by a factor two lower than the value measured experimentally by Gabriel and Heddle.

Finally, it is of interest to compare the experimental data with calculations utilizing formula (14). We have noted that such a calculation yields only the total cross section for all levels lying in the corresponding energy interval. But it can also be compared to the excitation function for the 2^3S-level, taking into account the fact that the excitation of the 2^3S-level is more probable than the excitation of the 2^3P-level, and that the calculation has the nature of only a rough estimate. Such a comparison is made in Fig. 4 which, moreover, also gives the results of a similar calculation for the 3^3S- and 4^3S-levels.

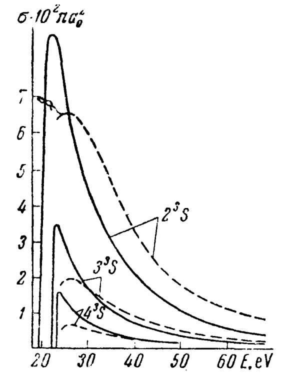

FIG. 4. Excitation functions for the n^3S levels in helium: solid curve – calculations in accordance with formula (14), dotted curve – experiment[6].

CONCLUSION

Comparison of the above calculations with experimental data shows that the calculation of exchange scattering of electrons by atoms is not as complicated as it is usually considered to be. Even formula (14) based on concepts of classical mechanics enables us to estimate correctly the probabilities of the most intense transitions. Formula (8) enables us to carry out more detailed and more reliable calculations. It is analogous to Born's formula both with respect to its structure, and with respect to the accuracy of the results obtained.

The analogy between the quantum mechanical and the classical formulas suggests a method for some further improvement in the quality of the calculations. Formula (14) took into account the acceleration of the external electron by the atomic field. Although we had assumed that the increment in the energy determined by this has the lowest possible value (4.7 eV for the 2^3S-level), taking it into account has led to a decrease in the cross section at the maximum by a factor of 1.7. It is of interest to introduce an analogous correction also into the quantum mechanical formula. Naturally, here the procedure does not reduce to a simple replacement of E by $E + \Delta$, since the effect of acceleration must play a different role for the different partial cross sections. Possibly taking this effect into account will turn out to be useful also in the case of scattering without exchange, particularly for atoms with low binding energy or for calculations of stepwise excitation.

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