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Dips in Spectral Lines of He-like Ions Caused by Charge Exchange in Laser-Produced Plasmas

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ABSTRACT: In our previous works we predicted theoretically and then discovered experimentally the formation of dips (called x-dips) in spectral lines of hydrogenlike ions from laser-produced plasmas. These dips are caused by charge exchange. At that time the x-dip phenomenon was considered to be possible only in spectral lines of hydrogenic systems: due to the existence of *exact* algebraic (higher than geometrical) symmetries relevant only to hydrogenic systems and to the corresponding two-Coulomb-center systems (dicenters) having one electron. In the present paper, by engaging the concept of an approximate algebraic symmetry of two-electron dicenters (and of helium-like ions), we opened up the way to significantly broaden the scope of experimental studies of the x-dip phenomenon: namely, to studies of possible x-dips in spectral line profiles emitted by He-like ions in laser-produced plasmas. We identified three prospective two-electron dicenters and calculated analytically theoretical positions of the x-dips in the corresponding He-like spectral lines (though future experimental and theoretical studies should not be limited to these two two-electron dicenters). From the shape of experimental x-dips it is possible to determine the rate coefficient of charge exchange in the corresponding dicenter, as we demonstrated previously. Therefore the results of the present paper should significantly extend the range of fundamental data on charge exchange between multicharged ions that can be obtained via the x-dip phenomenon, but not by any other method.

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1. INTRODUCTION

In our previous works we discovered a way for producing not-yet-available fundamental data on charge exchange between multicharged ions, virtually inaccessible by other experimental methods [1]. It was based on the formation of dips (called x-dips) in spectral lines of hydrogenlike ions from laser-produced plasmas [2-7]. At that time the x-dip phenomenon was considered to be possible only in spectral lines of hydrogen atoms (as first discovered in [8]) and hydrogen-like ions: due to the existence of *exact* algebraic (a.k.a. "hidden") symmetries relevant only to hydrogenic systems and to the corresponding two-Coulomb-center systems having one electron. Exact algebraic (i.e., higher than geometrical) symmetries of these quantum systems lead to exact additional conserved quantities, known as Runge-Lenz vector and its generalizations (see. e.g., the latest paper [9] on this subject).

The concept of symmetry is very general: in fact, any approximate analytical theory can be considered as a simplified version of a more complicated problem, the simplification being achieved by using some *approximate* symmetry [10]. Actually, any regularity in the energy spectrum of a quantum system reflects certain symmetry properties [10].

The thrust of the present project is to study whether the x-dip phenomenon is also possible in spectral lines of helium-like ions from laser-produced plasmas. While helium-like ions and the corresponding two-Coulomb-center systems having two electrons do not possess *exact* additional conserved quantities, they possess *approximate* additional conserved quantities [11]. This may be sufficient for the x-dips to occur also in these systems. This

would very significantly extend the range of fundamental data on charge exchange between multicharged ions that can be obtained via the x-dip phenomenon, but not by any other method.

2. GENERAL OUTLINE OF THE THEORY

We consider electron terms in the field of two stationary Coulomb centers (TCC) of charges Z_{eff} and Z' separated by a distance R. Here $Z_{eff} = Z–1$ is the effective charge of the core for the outer electron, the core being represented by the nucleus of charge Z and the inner electron in the ground state. We use atomic units throughout the paper, unless specified to the contrary.

We seek crossings (actually, avoided crossings also known as quasicrossings) of the following two types of terms. The first type of terms is where electron is primarily bound by the charge Z_{eff} and is characterized by the principal quantum number n. The second type of terms is where electron is primarily bound by the charge Z' and is characterized by the principal quantum number n'.

Gershtein and Krivchenkov [12] demonstrated that the well-known Neumann-Wigner general theorem on the impossibility of crossing of terms of the same symmetry [13] is not valid for the one-electron TCC problem of $Z' \neq Z$. ("Terms of the same symmetry" is the conventional name for terms having the same projection M of the angular momentum on the internuclear axis.) Physically, crossings and quasicrossings in the TCC problem are possible only because the problem possesses a symmetry higher than the geometrical symmetry. Namely, it possesses an additional conserved quantity (integral of motion) – in addition to the energy E and the angular momentum projection M. This additional conserved quantity is the projection (on the internuclear axis) of the super-generalized Runge-Lenz vector [9].

$$\mathbf{A} = \mathbf{p} \times \mathbf{L} - L^2/R\, \mathbf{e}_z - Z\mathbf{r}/r - Z'\,(\mathbf{R} - \mathbf{r})/|\mathbf{R} - \mathbf{r}| + Z'\,\mathbf{e}_z, \quad \mathbf{e}_z = \mathbf{R}/R, \tag{1}$$

where **p** and **L** are the linear and angular momenta vectors, respectively; **r** is the radius vector of the electron. In our two-electron case, after substituting in Eq. (1) Z by Z_{eff} and treating **r** as the radius vector of the outer electron, the projection of the vector **A** on the internuclear axis is an approximately conserved quantity.

The present analytical theory (as well the corresponding analytical theory for the one-electron case [4, 5]) is valid for a typical situation where the internuclear distance R_c, at which a quasicrossing occurs, satisfies the following conditions. The first condition is

$$R_c >> \max\,(n^2/Z_{eff}\,,\, n'^2/Z'), \tag{2}$$

i.e., for R_c to significantly exceed the characteristic sizes of the two corresponding hydrogenic subsystems. Under this condition, one can use the multipole expansion for the energy terms. The second condition is

$$R_c > \max[(3n^5\, Z'/Z_{eff}^{\,3})^{1/2},\, (3n'^5\, Z_{eff}/\, Z'^3)^{1/2}]. \tag{3}$$

This condition is necessary to ensure the existence of the energy level n_{eff} of the radiating Z_{eff} –ion and of the energy level n′ of the Z' –ion. Namely, the relation (3) is necessary to ensure that under the electric field of Z'-ion at the distance R_c, the level n of the radiating Z_{eff} -ion does not merge with the level n +1, as well as to ensure that under the electric field of Z_{eff} -ion at the distance R_c, the level n' of the Z' -ion does not merge with the level n' +1. For practically all dicenters, the condition (3) is more restrictive than the condition (2).

The third condition puts an upper limit on R_c :

$$R_c < [3n^2\, Z'/(Z_{eff}\, \Delta E)]^{1/2}, \tag{4}$$

where ΔE is the size of the unperturbed multiplet of the principal quantum number n. This inequality ensures that for the Z-ion state of the principal quantum number n, the Stark splitting caused by the Z'-ion at the distance R_c significantly exceeds the unperturbed separation of the sublevels of the n-shell. This condition allows using parabolic coordinates.

It can be shown that for the restrictions on R_c to be fulfilled, the ratio Z_{eff}/Z' should slightly exceed (but not to be equal to) a small integer. This explains our choice of three prospective candidates for observing x-dips in spectral line profiles of He-like ions from laser-produced plasmas, as presented in Sect. 3.

Under the condition (4), the two terms involved in a quasicrossing can be asymptotically labeled by parabolic quantum numbers: (n_1, n_2, m) for one term and (n_1', n_2', m') for the other. The selection rule for a quasicrossing to occur is the following. For each Z'-term (n_1', n_2', m'), a quasicrossing, and consequently CE, is possible with no more than one Z_{eff} -term, namely: either with the Z_{eff} -term of the following parabolic quantum numbers [14, 15]

$$n_1 = n_1', \quad m = m', \quad n_2 = n - n_1' - |m'| - 1 \tag{5}$$

or not at all, if the set (n_1, n_2, m) given by Eq. (5) does not correspond to any Z-term. Physically, this selection rule follows from the picture of CE as the corresponding interaction of states in two adjacent potential wells (one - centered at the charge Z_{eff}, another – at the charge Z') and from the fact that for such interaction to be possible, the radial wave functions of these states should have the same number of nodes [15, 16].

Thus, a quasicrossing occurs when two potential wells, corresponding to separated Z_{eff} - and Z' -centers, have states, characterized by the same energies $E = E'$, by the same projections $M = M'$ of the angular momentum on the internuclear axis, but differ by the projections A_i and A'_i of the Runge-Lenz vector on the internuclear axis. Indeed, A_i and A'_i are controlled by electric quantum numbers $q = n_1 - n_2$ and $q' = n_1' - n_2'$. From the selection rule (5), by using the definition of the principal quantum number $n' = n_1' + n_2' + |m'| + 1$, it can be easily found that the corresponding q and q' differ from each other:

$$q = q' + n' - n. \tag{6}$$

There are two independent mechanisms for quasicrossings to cause the formation of the x-dips in spectral line profiles. One of them is a sharp increase of the total frequency of inelastic collisions at $R = R_c$ – because in the vicinity of R_c charge exchange provides an additional channel for the decay of the excited state of the radiating ion [8]. Another mechanism is a sharp change of the slope of the transition energy at $R = R_c$ [4]. These two mechanisms can work independently and enhance each other [1].

It should be emphasized that an x-dip has a structure consisting of the dip itself surrounded by two adjacent bumps [1]. From the experimental bump-to-dip ratio of intensities it is possible to *determine the rate coefficient of charge exchange* between the corresponding two ions using the method theoretically developed and practically implemented in paper [1].

3. DETAILED CALCULATIONS OF THE POSITIONS OF THE X-DIPS

Condition (2) combined with the upper limit on R_c allows to treat separately a set of Z_{eff} -terms (perturbed by Z') and a set of Z'-terms (perturbed by Z_{eff}), as well as to use the 1/R expansion of the energies $E(R)$ and $E'(R)$ of the Z_{eff} – and Z'-terms, respectively:

$$E(R) = -Z_{\text{eff}}^2/(2n_{\text{eff}}^2) - Z'/R + 3Z'\, n_{\text{eff}}\, q/(2Z_{\text{eff}}R^2) - (6q^2 - n_{\text{eff}}^2 + 1)\, Z'\, n_{\text{eff}}^2/(2Z_{\text{eff}}^2R^3) + \ldots \tag{7}$$

$$E'(R) = -Z'^2/(2n'^2) - Z_{\text{eff}}/R + 3Z_{\text{eff}}\, n'q'/(2Z'R^2) - (6q'^2 - n'^2 + 1)Z_{\text{eff}}\, n'^2/(2\, Z'^2R^3) + \ldots \tag{8}$$

Here $n_{\text{eff}} = n - F(Z)$ is an effective principal quantum number, where $F(Z)$ is the averaged quantum defect for the sublevels of the principal quantum number n (details on its calculations are given few paragraphs later). We remind that $q = n_1 - n_2$ and $q' = n_1' - n_2'$ are the electric quantum numbers of the terms E and E', respectively. In Eqs. (7) and (8) we presented the multipole expansions up to (including) the quadrupole term. Higher order terms of the multipole expansion can be found in [16], but they are usually unnecessary for calculating x-dip positions within the accuracy of the experimental determination of these x-dip positions.

By equating $E(R)$ to $E'(R)$ for a pair of Z_{eff} -term and Z'-term satisfying the selection rule (3), we find the distance R_c, at which the quasicrossing of these two terms occurs. Then we calculate the Stark shift of the spectral line component, corresponding to the radiative transition in the He-like ion of the nuclear charge Z from the upper sublevel (characterized by the quantum numbers n and q) to the ground state: namely, the Stark shift caused by the Z'-ion at the distance R_c from the He-like radiating ion. This Stark shift is the theoretical position of the x-dip in the profile of this spectral line component. We note that finding the theoretical positions of x-dips does not require calculating the broadening of the spectral line.

Then we proceed to the next pair of Z_{eff} -term and Z'-term satisfying the selection rule (3), go through the same steps, and find the theoretical position of the next x-dip, and so on. In almost all practical examples, including those presented below, we have $n > n'$. In this situation, the maximum possible number of x-dips is equal to $2n' - 1$, i.e., to the number of Stark sublevels of the hydrogenlike ion of the nuclear charge Z' in the state of the principal quantum number n'.

We identified three prospective candidates for observing x-dips in spectral line profiles of He-like ions from laser-produced plasmas, as presented below. Analytical calculations of the positions of the x-dips in spectral lines of He-like ions require, in particular, analytical calculations of the effective principal quantum number n_{eff}, which in its turn require analytical calculations of the quantum defect of the energy levels of He-like ions. For this purpose in present paper we use the analytical method developed by Nadezhdin and Oks [16] reproduced in Appendix. This method (just like other methods, to the best of our knowledge) calculates the quantum defect in the spherical quantization. However, under the condition (4) the Stark effect is linear, which means also that the relatively strong electric field of Z'-ion at the distance R_c from the radiating Z_{eff} -ion intermixes all the spherical eigenfunctions of the energy level n. Therefore, the effective principal quantum number n_{eff} , used then in the parabolic quantization's formula (7), should be calculated by averaging the quantum defect over all sublevels of the spherical quantization belonging to the level n.

For each of the three prospective dicenters presented below, we compared the energy, obtained from the averaging Nadezhdin-Oks' quantum defect, with the corresponding result that can be deduced from the empirical data from [17]. It turned out that for these and other dicenters, already for $n = 4$ used in the examples, the relative difference was only ~ 0.05%. Such an excellent agreement justifies our usage of the quantum defect concept for $n = 4$. Of course, for higher values of n, the accuracy would be even better.

For the examples given below, all three conditions (2)-(4) are satisfied.

1. He_γ line of Si XIII 5.405 A ($Z = 14$) perturbed by fully-stripped C ($Z' = 6$). For the corresponding experiment the solid target can be made out of silicon carbide (SiC), also known as carborundum.
2. He_γ line of Mg XI 7.4731 A ($Z = 12$) perturbed by fully-stripped B ($Z' = 5$). For the corresponding experiment the solid target can be made out of crystals of magnesium borate $(MgO)_3B_2O_3$.
3. He_γ line of S XV 4.0885 A ($Z = 16$) perturbed by fully-stripped N ($Z' = 7$). For the corresponding experiment the solid target can be made out of crystals/granuls of ammonium sulfate $(NH_4)_2SO_4$.

In each of the above three examples, there are quasicrossings of some of the Z_{eff} -terms of $n = 4$ with some of the Z'-terms of $n' = 2$ (for the 1st example, n_{eff} =3.992; for the 2nd example, n_{eff} = 3.993). More specifically, there are the following three avoided crossings:

1) (Z, n)-term of $q = -3$ with the (Z', n')-term of $q' = -1$;

2) (Z, n)-term of $q = -2$ with the (Z', n')-term of $q' = 0$;

3) (Z, n)-term of $q = -1$ with the (Z', n')-term of $q' = 1$.

Thus, there could be up to three x-dips observed in the profile of the He_γ line of the above two radiating ions. Below we will label the three possible x-dips according to the above order of quasicrossings: the 1st dip results from the 1st quasicrossing, the 2nd dip – from the 2nd quasicrossing, the 3rd dip – from the 3rd quasicrossing.

However, it should be noted that it would be unlikely to observe all three x-dips in one experiment because the range of plasma parameters, favorable for observing an x-dip, differs for the above three x-dips. It depends on the relation between the most probable distance between the radiating and perturbing ions (which depends on plasma parameters) and the distance where the corresponding quasicrossing occurs. Therefore, most probably in some experiments only the x-dips #1 and #2 could be observed, while at experiments at another range of plasma parameters only the x-dips #2 and #3 could be observed. This situation is similar to the x-dips actually observed in the L_γ line of Al XIII (Z=13) perturbed by fully-stripped C ($Z' = 6$), as reported in two different ranges of the experimental conditions [2].

For the two chosen examples of the dicenters, the detailed results can be presented as follows. All the positions of the x-dips are in the red part of the corresponding spectral line profile.

For the He_γ line of Si XIII 5.405 *A* ($Z = 14$) perturbed by fully-stripped *C* ($Z' = 6$):

- The 1st dip is at 9.0 mA, corresponding to the crossing at $R = 8.3$ a.u.
- The 2nd dip is at 6.3 mA, corresponding to the crossing at $R = 7.7$ a.u.
- The 3rd dip is at 3.0 mA, corresponding to the crossing at $R = 7.2$ a.u.

For the He_γ line of Mg XI 7.4731 *A* ($Z = 14$) perturbed by fully-stripped *B* ($Z' = 5$):

- The 1st dip is at 16.5 mA, corresponding to the crossing at $R = 8.5$ a.u.
- The 2nd dip is at 11.8 mA, corresponding to the crossing at $R = 7.8$ a.u.
- The 3rd dip is at 5.5 mA, corresponding to the crossing at $R = 7.2$ a.u.

For the He_γ line of S XV 4.0885 A ($Z = 16$) perturbed by fully-stripped *N* ($Z' = 7$):

- The 1st dip is at 5.3 mA, corresponding to the crossing at $R = 8.1$ a.u.
- The 2nd dip is at 3.7 mA, corresponding to the crossing at $R = 7.7$ a.u.
- The 3rd dip is at 1.7 mA, corresponding to the crossing at $R = 7.3$ a.u.

We should mention that there is a "reciprocity rule" concerning the dicenters and x-dips. Let us illustrate it using the above example, where we considered a quasicrossing of some of the Z_{eff} -terms of $n = 4$ with some of the Z'-terms of $n' = 2$. Above we focused at the manifestation of this quasicrossing as an x-dip in the spectral line originated from the state of $n = 4$ of the subsystem (Z_{eff}+ electron). The same quasicrossing should manifest also as an x-dip in the spectral line originated from the state of $n' = 2$ of the subsystem (Z' + electron). However, the latter type of the spectral lines is in a significantly different range of wavelengths compared to the former. This would require different spectrometers compared to spectrometers used in the previous x-dip experiments (see, e.g., [2]).

4. CONCLUSIONS

In numerous studies of the x-dip phenomenon, which started in 1995 from a high-precision experiment in the gas-liner pinch by Kunze's group [8], the paradigm was that this phenomenon is limited to one-electron dicenters, i.e., to systems consisting of two nuclei of charges Z and Z' and one electron. Such systems naturally occur in high-density plasmas – especially in laser-produced plasmas – where in the vicinity of a radiating hydrogenlike Z-ion (nucleus Z plus one electron) there is a nearest-neighbor fully-stripped ion of charge Z'. Charge exchange in one-electron dicenters manifested as x-dips in spectral line profiles emitted by the hydrogenlike Z-ion.

In the present paper, by engaging the concept of an approximate algebraic symmetry of two-electron dicenters, we opened up the way to significantly broaden the scope of experimental studies of the x-dip phenomenon: namely, to studies of possible x-dips in spectral line profiles emitted by He-like ions in laser-produced plasmas. We identified three prospective two-electron dicenters and calculated analytically theoretical positions of the x-dips in the corresponding He-like spectral lines. Naturally, future experimental and theoretical studies should not be limited to these three two-electron dicenters. Besides, future theoretical studies could be simulations incorporating the effect of the screening by plasma electrons.

From the shape of experimental x-dips (from the bump-to-dip ratio) it is possible to determine the rate coefficient of charge exchange in the corresponding dicenter. We believe that the results of the present paper would significantly extend the range of fundamental data on charge exchange between multicharged ions that can be obtained via the x-dip phenomenon, but not by any other method.

Appendix: Nadezhdin-Oks' method for calculating the quantum defect

We consider a system consisting of a nucleus of charge Z and two electrons: an inner electron in the state 1s and an outer electron in the state nl, where $n > 1$. We use atomic units $\hbar = e = m_e = 1$.

The characteristic size of the outer electron shell is noticeably greater than the characteristic size of the inner electron shell. The starting point is to treat the outer electron quasiclassically during its motion in the potential φ of a quasinucleus consisting of the nucleus Z and a spherically-symmetric charge distribution corresponding to the inner electron in the 1s state. The inner electron contribution φ_s to the total potential φ is given by

$$\varphi_s(r) = -1/r + (Z\mu + 1/r)\exp(-2Z\mu r), \tag{A.1}$$

so that the total potential for the outer electron motion is

$$\varphi(r) = (Z-1)/r + (Z\mu + 1/r)\exp(-2Z\mu r). \tag{A.2}$$

Here $\mu = mM/(M+m)$ is the reduced mass of the pair "electron-nucleus", M is the nuclear mass.

We denote the binding energy of the outer electron by E, so that its energy is $-E$ ($E > 0$). The 1-D equation for the radial wave function of the electron $\chi_{nl}(r) = rR_{nl}(r)$ has the form

$$-(1/2)d^2\chi_{nl}(r)/dr^2 + l(l+1)\chi_{nl}(r)/(2r^2) - \varphi(r)\,\chi_{nl}(r) = -E\chi_{nl}(r). \tag{A.3}$$

The quasiclassical quantization conditions are as follows.

For $l = 0$:

$$(2^{1/2}/\pi)\int_0^{r_{max}} dr[(Z-1)/r + (Z\mu+1/r)\exp(-2Z\mu r) - E]^{1/2} = n_r + 1. \tag{A.4}$$

For $l > 0$:

$$(2^{1/2}/\pi)\int_{r_{min}}^{r_{max}} dr[(Z-1)/r + (Z\mu+1/r)\exp(-2Z\mu r) - l(l+1)/(2r^2) - E]^{1/2} = n_r + 1/2. \tag{A.5}$$

Here r_{min} and r_{max} are the classical turning points.

In the limit $E \to 0$, Eqs. (A.4) and (A.5) allow analytical solutions. Let us first consider the case of $l = 0$. When $E \to 0$, the integral in Eq. (A.4) scales as $1/E^{1/2}$ and would diverge at $E = 0$. The convergence at small, but not zero values of E is related to the integration over large values of r – since $r_{max} \to \infty$ when $E \to 0$. Indeed, at $r >> 1/(Z\mu)$, the term containing $\exp(-2Z\mu r)$ can be neglected and the integral can be estimated as

$$(2^{1/2}/\pi)\int_0^{(Z-1)/E} dr[(Z-1)/r - E]^{1/2} = (Z-1)/E^{1/2}. \tag{A.6}$$

The next term in the expansion of the integral from Eq. (A.4) in powers of E is independent of E and we denote it $F_0(Z, m)$. It can be obtained by subtracting the integrand of Eq. (A.6) from the integrand of Eq. (A.4) and then setting $E = 0$:

$$F_0(Z,\mu) = (2^{1/2}/\pi)\int_0^{\infty} dr\{[(Z-1)/r + (Z\mu+1/r)\exp(-2Z\mu r)]^{1/2} - [(Z-1)/r]^{1/2}\}. \tag{A.7}$$

From Eqs. (A.4), (A.6), (A.7) we find that in the limit $E \to 0$, the quantization condition has the form

$$(Z-1)/E^{1/2} + F_0(Z, \mu) = n_r + 1 = n, \tag{A.8}$$

where n is the principal quantum number, so that

$$E_{n0} = (1/2)\,(Z-1)^2/[n - F_0(Z,\mu)]^2. \quad \text{(A.9)}$$

From Eq. (A.9) it is clear that $F_0(Z, \mu)$ has the meaning of a quantum defect.

Similarly, for $l > 0$ we obtain

$$E_{nl} = (1/2)\,(Z-1)^2/[n - F_l(Z,\mu)]^2, \quad \text{(A.10)}$$

where the quantum defect $F_l(Z, \mu)]$ is given by

$$F_l[(Z,\mu)] = (2^{1/2}\pi)\int_{r_{\min}}^{\infty} dr\{[(Z-1)/r + (Z\mu+1/r)\exp(-2Z\mu r) - l(l+1)/(2r^2)]^{1/2} - [(Z-1)/r - l(l+1)/(2r^2)]^{1/2}\}. \quad \text{(A.11)}$$

Thus, this method yields explicit analytical results for the energy levels expressed through a simple one-fold integral. We note that the method has not only an excellent accuracy for $n >> 1$, but also a good accuracy for relatively low values of n, even for $n = 2$. We illustrate this by calculating energies of He atoms. Since the method ignores exchange effects, the comparison of the results of the method for the He atom should be made with the reference data averaged between singlet and triplet configurations. In the Table 1 below we used the most current reference data from NIST.

Table 1
Binding Energy of States of the He Atom (in a.u.). The Reference Data from NIST is the Average between Singlet and Triplet Configurations

nl	*Reference data*	*Our analytical results*	*Accuracy of our results*
20	0.1606	0.1571	2%
21	0.1285	0.1267	1%
30	0.06497	0.06452	0.7%
31	0.05661	0.05606	1%
32	0.05562	0.05556	0.1%
40	0.03504	0.03492	0.3%
41	0.03169	0.03146	0.7%
42	0.03128	0.03125	0.1%
43	0.031251	0.031250	0.003%

From Table 1 it is seen that the method has a good accuracy even for relatively low values of n. It shows also that the relative error decreases as n and/or l increase – as should be expected.

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