Anisotropic van der Waals coefficients for $He(1 \ ^{1}S)-He(2 \ ^{3}P)$

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We report on the anisotropic mass-dependent dispersion coefficients $C_6(M=0,\pm 1)$ for He(1¹S)–He(2³P) interaction using two different techniques. The first is a highly accurate Hylleraas basis-set diagonalization, while the second method is a semiempirical assessment based on tabulated oscillator strengths and available photoionization cross sections. The results from the two techniques agree to within 1.5%.

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I. INTRODUCTION

High precision photoassociation and ultracold collision of atoms or molecules probe details of the interaction potentials [1]. A good deal of attention has recently been attracted to the investigation of long-range multipole terms in the electrostatic interaction of atoms and molecules and subsequent physical properties that directly result from them, such as the elastic scattering length [2]. The bulk attention has focused on calculating the interaction terms between alkali-metal atoms, and as experiments on cooling and trapping diversify to other elements, better interaction potentials are also needed for these species [3]. Lately, the Bose-Einstein condensate of metastable triplet states of helium became a reality [4], and high-precision measurements of the fine-structure constant in collision of the same metastable helium atoms with groundstate He atoms have been made [5]. Spectral broadening and shift of atomic or molecular lines in the wings are due to purely long-range interaction between the perturbing and radiating species [6]. Both classes of experiments probe the asymptotic interaction between two helium atoms [7].

In this work, we calculate the anisotropic terms in the perturbative expansion of the electrostatic interaction between $\text{He}(2^{3}P)$ and $\text{He}(1^{1}S)$ atoms, by employing first a numerical technique that in the past has proven fruitful in obtaining accurate dispersion coefficients and polarizabilities for helium and other atoms and has been used for calculating highly precise terms in the electron-nucleus interaction in atoms. Next, we analyze and assess the quality and accuracy of available discrete oscillator strengths and photoionization cross sections for the relevant He states and calculate from this analysis the best set of dynamic polarizability and aniso-tropic dispersion coefficients. The two sets of calculations are compared and we find that the agreement is better than 1.5%.

II. PERTURBATION EXPANSION OF THE INTERACTION POTENTIAL

The interaction potential between atomic (or molecular) systems A and B can be written as a multipole expansion in inverse powers of separation R as [8]

$$V = \sum_{l=0}^{\infty} \sum_{L=0}^{\infty} \frac{V_{lL}}{R^{l+L+1}} , \qquad (1)$$

$$V_{lL} = (-1)^{L} 4 \pi (l,L)^{-1/2} \sum_{m} K_{lL}^{m} T_{m}^{(l)}(A) T_{-m}^{(L)}(B).$$
(2)

The atomic multipole tensor operators are defined by

$$T_m^{(l)}(A) = \sum_i Q_i r_i^l Y_{lm}(\hat{\mathbf{r}}_i) , \qquad (3)$$

and by

$$T_m^{(L)}(B) = \sum_j q_j \rho_j^L Y_{Lm}(\hat{\boldsymbol{\rho}}_j) , \qquad (4)$$

where Q_i and \mathbf{r}_i are the electric charges and coordinates from the local center for particles in system A, while q_j and $\boldsymbol{\rho}_j$ are the electric charges and coordinates from the local center for particles in system B. The coefficient K_{lL}^m is

$$K_{lL}^{m} = \left[\binom{l+L}{l+m} \binom{l+L}{L+m} \right]^{1/2}$$
(5)

and (l, L, ...) = (2l+1)(2L+1)...

For large separation *R* the interaction potential Eq. (1) can be treated as a perturbation. If both interacting systems are neutral then the multipolar series (1) is dominated by the L = l = 1 dipole-dipole term:

$$V_{11} = -\frac{4\pi}{3} \sum_{m=-1}^{1} K_{11}^m d_m^{(A)} d_{-m}^{(B)} , \qquad (6)$$

where $d_m = T_m^{(1)}$ are the spherical components of the atomic dipole operator.

When the atoms A and B are not resonant, as is the case for the He(1¹S)-He(2³P) system studied here, the firstorder perturbation theory gives no correction to the energy, and the second-order corrections to the energy provide the $1/R^6$ dispersion interaction

$$\Delta E^{(2)} = -\frac{1}{R^6} \sum_{k} \frac{\langle \Psi^{(0)} | V_{11} | \Phi_k \rangle \langle \Phi_k | V_{11} | \Psi^{(0)} \rangle}{E_k - E_0} = -\frac{C_6}{R^6} , \quad (7)$$

where the index k enumerates states Φ_k in the discrete and continuous spectrum of the A-B system, with the exception, symbolized by prime in summation, of the unperturbed state $\Psi^{(0)}$. This is the state of the system at infinite separation,

where

$$\Psi^{(0)} = \frac{1}{\sqrt{2}} [\Psi_A(\mathbf{r}) \Psi_B^M(\boldsymbol{\rho}) + \boldsymbol{\beta} \Psi_A(\boldsymbol{\rho}) \Psi_B^M(\mathbf{r})], \qquad (8)$$

where Ψ_A is the ground-state He(1 ¹S) wave function and Ψ_B^M is the wave function for He(2 ³P) with the magnetic quantum number *M* corresponding to quantization along the internuclear axis. All electronic coordinates \mathbf{r}_i and $\boldsymbol{\rho}_j$ are represented by \mathbf{r} and $\boldsymbol{\rho}$, respectively. The factor $\beta = \pm 1$ describes the electronic gerade-ungerade symmetry and is irrelevant in this treatment because the two atoms are not resonant. This means that there is no allowed dipole transition between He(1 ¹S) and He(2 ³P) because of the spin conservation law, which generally forbids singlet-triplet transitions. Therefore we can take for simplicity

$$\Psi^{(0)} = \Psi_A(\mathbf{r}) \Psi_B^M(\boldsymbol{\rho}). \tag{9}$$

Similarly, the intermediate states Φ_k can be written as products of two atomic wave functions. There are four symmetry classes of states of helium atom which have nonzero dipole matrix elements with He(1¹S) or He(2³P) and contribute in summation of Eq. (7). They are denoted by ζ , σ , η and δ and are defined as

(i) $\zeta = \{ \text{He}(\zeta^{1}P) \}_{\zeta=2,3,...}$, the only states connected to the ground state $\text{He}(1^{1}S)$;

(ii) $\sigma = \{\text{He}(\sigma^3 S)\}_{\sigma=2,3,...}$, states connected to $\text{He}(2^3 P)$. State $2^3 S$ is special in that it has lower energy than $\text{He}(2^3 P)$;

(iii) $\eta \equiv \{\text{He}(\eta(pp)^{3}P)\}_{\eta=2,3,...}, \text{ double excited states connected to He}(2^{3}P);$

(iv)
$$\delta \equiv \{\text{He}(\delta^3 D)\}_{\delta=3,4,...}$$
, states connected to $\text{He}(2^3 P)$.

The contribution of each class of intermediate states can be separated in the C_6 coefficient defined by Eq. (7), such that

$$C_6(M) = C_6^{(S)}(M) + C_6^{(P)}(M) + C_6^{(D)}(M) , \qquad (10)$$

where

$$\begin{split} C_{6}^{(S)}(M) &= \frac{16\pi^{2}}{9} \sum_{\zeta,\sigma} \frac{1}{\Delta E_{\zeta} + \Delta E_{\sigma}} \sum_{m_{1},m_{2},\mu} K_{11}^{m_{1}} K_{11}^{m_{2}} \langle A | d_{m_{1}} | \zeta; \mu \rangle \\ & \times \langle \zeta; \mu | d_{m_{2}} | A \rangle \langle B; M | d_{-m_{1}} | \sigma \rangle \langle \sigma | d_{-m_{2}} | B; M \rangle, \end{split}$$

$$\begin{split} C_6^{(P)}(M) = & \frac{16\pi^2}{9} \sum_{\zeta,\eta} \frac{1}{\Delta E_{\zeta} + \Delta E_{\eta}} \sum_{m_1,m_2,\mu,\mu'} K_{11}^{m_1} K_{11}^{m_2} \\ & \times \langle A | d_{m_1} | \zeta; \mu \rangle \langle \zeta; \mu | d_{m_2} | A \rangle \langle B; M | d_{-m_1} | \eta; \mu' \rangle \\ & \times \langle \eta; \mu' | d_{-m_2} | B; M \rangle, \end{split}$$

$$\begin{split} C_{6}^{(D)}(M) &= \frac{16\pi^{2}}{9} \sum_{\zeta,\delta} \frac{1}{\Delta E_{\zeta} + \Delta E_{\delta}} \sum_{m_{1},m_{2},\mu,\mu'} K_{11}^{m_{1}} K_{11}^{m_{2}} \\ &\times \langle A | d_{m_{1}} | \zeta; \mu \rangle \langle \zeta; \mu | d_{m_{2}} | A \rangle \langle B; M | d_{-m_{1}} | \delta; \mu' \rangle \\ &\times \langle \delta; \mu' | d_{-m_{2}} | B; M \rangle. \end{split}$$

The inner summations over the magnetic quantum numbers can be contracted using the Wigner-Eckart theorem to give

$$C_{6}^{(S)}(-1,0,1) = (1,4,1) \frac{16\pi^{2}}{81} \sum_{\zeta,\sigma} \frac{\langle A ||d||\zeta\rangle^{2} \langle B ||d||\sigma\rangle^{2}}{\Delta E_{\zeta} + \Delta E_{\sigma}},$$
(11)

$$C_{6}^{(P)}(-1,0,1) = (5,2,5) \frac{8\pi^{2}}{81} \sum_{\zeta,\eta} \frac{\langle A ||d||\zeta\rangle^{2} \langle B ||d||\eta\rangle^{2}}{\Delta E_{\zeta} + \Delta E_{\eta}},$$
(12)

$$C_{6}^{(D)}(-1,0,1) = (19,22,19) \frac{8\pi^{2}}{405} \sum_{\zeta,\delta} \frac{\langle A ||d||\zeta\rangle^{2} \langle B ||d||\delta\rangle^{2}}{\Delta E_{\zeta} + \Delta E_{\delta}}$$
(13)

in terms of reduced dipole matrix elements $\langle A || d || \zeta \rangle$, $\langle B || d || \sigma \rangle$, $\langle B || d || \eta \rangle$, and $\langle B || d || \delta \rangle$.

We employ two different methods to evaluate the infinite summations in Eqs. (11)–(13). The first method evaluates the matrix elements and summations using a finite variational basis. Good convergence is obtained as the number of functions in our basis is increased. The second method calculates separately the discrete and the continuum contributions, using tabulated oscillator strengths and available photoionization cross sections.

A. Hylleraas basis

To calculate the dispersion coefficients $C_6(M)$, we expand the wave function in a parameteric nonorthgonal basis set in Hylleraas coordinates and transform the multipole operators Eq. (3) into the center-of-mass frame [9]. The basis set is constructed in Hylleraas coordinates

$$r_{1}^{i} r_{2}^{j} r_{12}^{k} e^{-\alpha r_{1} - \beta r_{2}} \mathcal{Y}_{l_{1} l_{2}}^{LM}(\hat{\mathbf{r}}_{1}, \hat{\mathbf{r}}_{2}) , \qquad (14)$$

where $\mathcal{Y}_{l_1 l_2}^{LM}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2)$ are the coupled spherical harmonics for the two electrons. Except for some truncations, all terms are included in the basis such that

$$i+j+k \le \Omega , \qquad (15)$$

with Ω being an integer. Furthermore, we divide our basis set into several blocks each having its own set of nonlinear parameters α and β . A complete optimization is then performed with respect to the parameters α and β . Tables I and II present the convergence study for the states He(1¹S) and He(2³P), respectively, as the size of basis set increases progressively. Compared to Drake's values [10,11], our results are accurate to about 17 and 15 digits for He(1¹S) and He(2³P) state, respectively. The spectrum of the intermediate states can be obtained by diagonalizing the Hamiltonian in a chosen basis set. Although the variational principle does not guarantee that these intermediate states correspond to the actual excited states of the system, the second-order correction Eq. (7) is exactly calculated only if this set of intermediate states is complete. The completeness of all intermediate

TABLE I. Convergence of the nonrelativistic ground-state energy of He. N denotes the number of terms in the basis set.

Ω	Ν	$E(\Omega)$			
16	1050	-2.903 724 377 034 118 93			
17	1230	-2.903 724 377 034 119 37			
18	1430	-2.903 724 377 034 119 53			
19	1650	-2.903 724 377 034 119 57			
20	1892	-2.903 724 377 034 119 58			
Drake [10]		-2.903 724 377 034 119 598 311(1)			

states is checked with sum rules. For example, the Thomas-Reiche-Kuhn (TRK) sum rule for the He(2 ³P) state [12] reads, after letting the *z*-axis component of angular momentum M_0 be zero,

$$3S_S^1 + \frac{6}{5}S_D^1 = N_e \ . \tag{16}$$

In the above equation,

$$S_L^{\ell} = \sum_n \bar{f}_{n0}^{\ell}(L) , \qquad (17)$$

with $\overline{f}_{n0}^{\ell}(L)$ being the oscillator strength connecting the initial state $\Psi_0(L_0)$ of angular momentum L_0 and the final state $\Psi_n(L)$ of angular momentum L,

$$\overline{f}_{n0}^{\ell}(L) = \frac{8\pi}{(2\ \ell\ +1)^2(2L_0+1)} \times (E_n - E_0) |\langle \Psi_0(L_0)|| \sum_i r_i^{\ell} Y_{\ell}(\hat{\mathbf{r}}_i) ||\Psi_n(L)\rangle|^2 ,$$
(18)

and N_e is the number of electrons. For helium, $N_e=2$. Our numerical result shows that

$$3 \times (0.028\ 541\ 77) + \frac{6}{5} \times (1.595\ 354) = 2.000\ 05$$
 (19)

TABLE II. Convergence of the nonrelativistic energy eigenvalue of the state He(2 ${}^{3}P$). *N* denotes the number of terms in the basis set.

Ω	Ν	$E(\Omega)$
12	910	-2.133 164 190 779 194 2
13	1120	-2.133 164 190 779 246 8
14	1360	-2.133 164 190 779 278 8
15	1632	-2.133 164 190 779 281 2
16	1938	-2.133 164 190 779 282 7
Drake [11]		-2.133 164 190 779 283 202(5)

B. Discrete and continuum oscillator strengths assessment

An array of oscillator strengths for transitions involving $He(1 \ S)$ and $He(2 \ P)$ are tabulated by NIST [13]. The reduced dipole matrix elements required in the calculation of C_6 coefficients Eqs. (11)–(13), described in the L-S coupling, are calculated from the available fine-structure oscillator strengths as the following:

$$\frac{4\pi}{3} \langle A ||d||\zeta\rangle^2 = \frac{3}{2\Delta E_{\zeta}} f_{1}{}^{1}{}_{S \to \zeta}{}^{1}{}_{P},$$

$$\frac{4\pi}{3} \langle B ||d||\sigma\rangle^2 = \frac{9}{2\Delta E_{\sigma}} f_{2}{}^{3}{}_{P_{j} \to \sigma}{}^{3}{}_{S} \quad \text{for } j = 0, 1, 2,$$

$$\frac{4\pi}{3} \langle B ||d||\delta\rangle^2$$

$$= \frac{9}{2} \begin{bmatrix} 1 & 0 & 0 \\ 4 & \frac{4}{2} & 0 \end{bmatrix} f_{2} = \frac{9}{2} \int_{0}^{1} f_{2} \int_{0}$$

$$= \frac{4}{2\Delta E_{\delta}} \begin{bmatrix} 4 & \frac{1}{3} & 0\\ 100 & \frac{20}{3} & \frac{25}{21} \end{bmatrix} f_{2}^{3} p_{j} \rightarrow \delta^{3} D_{j'} \text{ and } j' = 1, 2, 3.$$
(20)

In the last equation, the elements in the square brackets reter in rows to j' and columns to j. The missing matrix elements $\langle B || d || \eta \rangle$ required to calculate $C_6^{(P)}$ coefficients in Eq. (12) involve double excited states and the corresponding oscillator strengths are not available. Fortunately, because of the large energy differences in the denominators in Eq. (12) and because of the nature of the double excited states the contribution of the coefficient $C_6^{(P)}$ towards the total C_6 coefficient is likely to be small. The validity of this approximation is confirmed by the variational calculation, which shows that the contribution of these states is less than 1% (Table IV).

Ignoring the contribution from double excited states the dispersion coefficients Eq. (10) are then

$$C_{6}(-1,0,1) = \frac{27}{4} \sum_{\zeta} \left(\frac{(1,4,1)}{9} \sum_{\sigma} \frac{f_{\zeta} f_{\sigma}}{\Delta E_{\zeta} \Delta E_{\sigma} (\Delta E_{\zeta} + \Delta E_{\sigma})} + \frac{(19,22,19)}{90} \sum_{\delta} \frac{f_{\zeta} f_{\delta}}{\Delta E_{\zeta} \Delta E_{\delta} (\Delta E_{\zeta} + \Delta E_{\delta})} \right)$$
$$= \frac{3}{4} (1,4,1) W^{(S)} + \frac{3}{40} (19,22,19) W^{(D)}.$$
(21)

The anisotropic van der Waals coefficients for the corresponding molecular symmetries are now

$$C_6^{\Sigma} = C_6(0) = 3W^{(S)} + \frac{66}{40}W^{(D)},$$
$$C_6^{\Pi} = C_6(\pm 1) = \frac{3}{4}W^{(S)} + \frac{57}{40}W^{(D)}.$$
(22)

The averaged dispersion coefficient is

TABLE III. Convergence characteristics of $C_6(M)$ for the system He(1 ¹S)–He(2 ³P). N₁_S, N₃_P, N_{σ}, N_{ζ}, N_{η} and N_{δ} denote, respectively, the sizes of bases for the two initial states and the four clasess of intermediate states of symmetries ³S, ¹P, (pp) ³P, and ³D. The nuclear mass is set to be infinite.

$N I_S$	$N {}^{3}P$	N_{σ}	Nζ	N_{η}	N_{δ}	$C_6(M=0)$	$C_6(M=\pm 1)$
1430	1360	560	1360	1230	853	47.725 886 615	26.708 670 783
1650	1632	680	1632	1430	1071	47.725 886 732	26.708 670 884
1892	1938	816	1938	1650	1323	47.725 886 745	26.708 670 895

$$\overline{C}_6 = \frac{1}{3}C_6^{\Sigma} + \frac{2}{3}C_6^{\Pi}.$$
(23)

To write $W^{(S)}$ and $W^{(D)}$ in terms of atomic polarizabilities we use single-center integrals [8] to obtain

$$W^{(S)} = 2 \frac{f_{\sigma^*}}{\Delta E_{\sigma^*}} \sum_{\zeta} \frac{f_{\zeta}}{\Delta E_{\zeta}^2 - \Delta E_{\sigma^*}^2} + \frac{2}{\pi} \int_0^\infty \sum_{\zeta} \frac{f_{\zeta}}{\Delta E_{\zeta}^2 + u^2} \sum_{\sigma} \frac{f_{\sigma}}{\Delta E_{\sigma}^2 + u^2} du = 2 \frac{f_{\sigma^*}}{\Delta E_{\sigma^*}} \alpha_A (\Delta E_{\sigma^*}) + \frac{2}{\pi} \int_0^\infty \alpha_A (iu) \alpha_B^{(S)}(iu) du, \quad (24)$$

where the state σ^* refers to the He(2 ³*S*) state whose transition energy to the He(2 ³*P*) state is negative, i.e., $\Delta E_{\sigma^*} < 0$, and $\alpha(\omega)$ are atomic dynamic dipole polarizabilities at frequency ω . For the ground-state helium atom, we use the dynamic polarizability at imaginary frequencies as obtained by Chan and Dalgarno [14]. For comparison, the value for the static polarizability using this representation is $\alpha_A(0)$ = 1.378, whereas the exact value is 1.383 [15].

The expression for $W^{(D)}$ follows as

$$W^{(D)} = \frac{2}{\pi} \int_0^\infty \alpha_A(iu) \,\alpha_B^{(D)}(iu) \,\,du.$$
 (25)

The partial polarizabilities evaluated at imaginary frequency, $\alpha_B^{(S)}(iu)$ and $\alpha_B^{(D)}(iu)$, are obtained according to

$$\alpha_{B}^{(S)}(x) = \sum_{\sigma = (2-6,8,10)} \frac{f_{\sigma}}{\Delta E_{\sigma}^{2} - x^{2}} + \sum_{\sigma = (7,9,11,\cdots)} \frac{f_{\sigma}}{\Delta E_{\sigma}^{2} - x^{2}} + \int_{0}^{\infty} \frac{df_{\sigma}}{dE} \frac{dE}{\Delta E_{\sigma}^{2} - x^{2}} = \alpha^{(ST)}(x) + \alpha^{(SE)}(x) + \alpha^{(SC)}(x),$$
(26)

where the first sum is taken over the values obtained from

the NIST tables [13], the second sum is obtained by extrapolating the oscillator strengths and the transition energies through scaling with the principal quantum numbers, and the integral term is obtained from photoionization cross sections [16] by ensuring that at the spectral heads, the scaled discrete oscillator strengths agreed with the zero-energy limits of the photoionization cross sections. A similar expression also holds for $\alpha_B^{(D)}$, where now the discrete oscillator strengths for $\delta=3-10$ levels are taken from the NIST table [13]. At the spectral heads, the scaled oscillator strengths are $f_{\infty}^{(S)}$ $=\lim_{\sigma\to\infty}\sigma^3 f_{\sigma}=0.240$ and $f_{\infty}^{(D)}=\lim_{\delta\to\infty}\delta^3 f_{\delta}=3.352$, while the equivalent values obtained from the continuum [16] are $\lim_{\epsilon\to0}(df_i^{(S)}/d\epsilon)=0.231$ and $\lim_{\epsilon\to0}(df_j^{(D)}/d\epsilon)=3.550$.

The oscillator strength sum rule for the He(2³*P*) state is satisfied to within 0.2%, i.e., $\Sigma_n f_{2^3P \to n} = 1.002$, with 0.765 units in the discrete part of the spectrum. The static dipole polarizability for this state estimated with our semiempirical method is $\alpha_{2^3P}(0) = 47.08$ a.u., which compares well with the accurate value of Yan [17] of 46.7 a.u. Finally, upon substituting for the dynamic polarizabilities and transition energies, we obtain

$$W^{(S)} = 7.869$$
 and $W^{(D)} = 14.310$ a.u. (27)

III. RESULTS AND SUMMARY

Table III contains the convergence behavior of $C_6(M)$. It is clear that the convergence is smooth against the sizes of Hylleraas basis sets and variations are in the last two significant digits. Table IV shows the contributions to $C_6(M)$ from the intermediate states of different symmetries. Note that the doubly excited (pp) states contribute at the level of 0.16% and 0.73%, respectively, to the total values for the $C_6(0)$ and $C_6(\pm 1)$ coefficients. Table V shows the effect of different nuclear masses on C_6 . There is a systematic increase in the values for the coefficients $C_6(M)$ with decreasing nuclear mass. In general, the results for the van der Waals coeffi-

TABLE IV. Contributions to $C_6(M)$ for the system He(1¹S)-He(2³P) from the He(¹P, ³S), (¹P,(pp)³P), and (¹P, ³D) symmetries. The nuclear mass is set to be infinite.

Symmetries	$C_{6}(0)$		$C_6(\pm)$		
	Hylleraas	semiempirical	Hylleraas	semiempirical	
$({}^{1}P, {}^{3}S)$	23.855 483 835 6(1)	23.607	5.963 870 958 89(1)	5.902	
$({}^{1}P,(pp){}^{3}P)$	0.079 109 536 86(3)		0.197 773 842 09(9)		
$({}^{1}P, {}^{3}D)$	23.791 293 39(2)	23.612	20.547 026 09(1)	20.392	

TABLE V. Effect of different nuclear masses on $C_6(M)$ for the atom system He(1 ¹S)-He(2 ³P)

Mass	$C_{6}(0)$))	$C_6(\pm)$		
	Hylleraas	semiempirical	Hylleraas	semiempirical	
$+\infty$	47.725 886 76(2)		26.708 670 89(1)		
⁴ He	47.752 349 75(3)	47.210	26.723 515 42(2)	26.290	
³ He	47.761 010 19(2)		26.728 373 55(2)		

cients from the two techniques agree to within 1.5%.

In this work, we investigate the anisotropic long-range interaction between two helium atoms in the ground and the first excited triplet *P* state. We employ two complimentary methods to obtain the $C_6(M)$ coefficients. The agreement is quite satisfactory, providing a logical check on the methods.

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