Ionization of atoms in weak fields and the asymptotic behavior of higher-order perturbation theory

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Using the imaginary time method, we study the structure of the perturbation series for the hydrogen atom in electric *E* and magnetic *H* fields. It is shown that there is a ''critical'' value of the ratio $\gamma = \mathcal{H}/\mathcal{E}$ at which the perturbation series for the ground state changes from having a fixed sign (for $\gamma < \gamma_c$) to having a variable sign (for $\gamma > \gamma_c$). This conclusion is confirmed by direct higher-order perturbation calculations. The change in the asymptotic regime is explained by competition among the contributions of the various complex trajectories that describe the subbarrier motion of the electrons. Here the parameter γ_c depends on the angle θ between the electric and magnetic fields. © *1998 American Institute of Physics.* $[S1063-7761(98)01006-3]$

1. The problem of the hydrogen atom in external fields is of fundamental importance in quantum mechanics and atomic physics and is often encountered in applications. $1-5$

Recently, $6-8$ a semiclassical theory has been developed for the ionization of atoms and ions in constant and uniform electric $\mathscr E$ and magnetic $\mathscr H$ fields. The imaginary time method $9-11$ was used to calculate the ionization probability $w(\mathscr{E},\mathscr{H})$, as it yields a convincing description of the subbarrier motion of the particles using the classical equations of motion, but with an imaginary "time."¹⁾

The ionization probability for the atomic *s* level with binding energy $|E_0| = \kappa^2/2$ is given by ($\hbar = m = e = 1$, natural units)

$$
w(\mathcal{E},\mathcal{H}) = \kappa^2 R(\gamma,\theta) \, \exp\bigg\{-\frac{2}{3\,\epsilon} \, g(\gamma,\theta)\bigg\},\tag{1}
$$

which is asymptotically exact in the limit of weak fields $(\epsilon, h \le 1)$. Here $\epsilon = \mathcal{E}/\kappa^3 \mathcal{E}_a$ and $h = \mathcal{H}/\kappa^2 \mathcal{H}_a$ are the reduced electric and magnetic fields, θ is the angle between the fields, $\mathcal{E}_a = m^2 e^5/\hbar^4$ and $\mathcal{H}_a = m^2 c e^3/\hbar^3$ are the atomic units for the field strengths, *m* is the electron mass, $\gamma = h/\epsilon$,

$$
g(\gamma,\theta) = \frac{3}{2} \beta \left[1 - \frac{\sqrt{\beta^2 - 1}}{\gamma} \sin \theta - \frac{1}{3} \beta^2 \cos^2 \theta \right], \quad (2)
$$

 $\beta = \tau_0 / \gamma$, $\tau_0 = \tau_0(\gamma, \theta)$ is the positive root of the equation

$$
\tau^2 - \sin^2 \theta (\tau \coth \tau - 1)^2 = \gamma^2,
$$
 (3)

and, finally, R is a (rather complicated) factor introduced in Ref. 7: $R = 2^{2\eta} \epsilon^{1-2n} PQ^{\eta}$ in the notation given there. Equation (3) can be easily obtained using the imaginary time method, where τ_0 has a simple physical significance: τ_0 = $-i\omega_L t_0$, where $\omega_L = |e| \mathcal{H}/mc$ is the Larmor frequency and t_0 is the "time" (purely imaginary) for subbarrier motion of the electron. Note that for $\gamma \rightarrow 0$,

$$
\beta = 1 + \frac{\sin^2 \theta}{18} \gamma^2 + \dots,
$$

while for $\gamma \rightarrow \infty$,

$$
\beta(\gamma,\theta) = \begin{cases}\n\frac{1}{\cos\theta} - \tan^2\theta\gamma^{-1} + O(\gamma^{-2}), & 0 \le \theta < \pi/2, \\
\frac{\gamma}{2}(1+\gamma^{-2}+...), & \theta = \pi/2.\n\end{cases}
$$
\n(3a)

Thus, for the function g , which determines the principal $(ex$ ponential) factor in the ionization probability, we obtain

$$
g(\gamma, \theta) = 1 + O(\gamma^2), \quad \gamma \to 0,
$$

$$
g(\gamma, \theta) = \frac{1}{\cos \theta} - \frac{3}{2} \tan^2 \theta \cdot \gamma^{-1} + ..., \quad \gamma \to \infty
$$
 (3b)

(for $\theta \le \pi/2$; for $\theta = \pi/2$ the asymptote has a different form; see Eq. (16) below). The function $g(\gamma,\theta)$ increases monotonically²⁾ along with γ (Fig. 1), so raising the magnetic field (at fixed \mathscr{E}) sharply reduces the ionization probability, stabilizing the atomic level. $6,7$

Using Eqs. (1) – (3) and invoking the same considerations as before, $15,16$ one can obtain the asymptotic behavior of the higher orders of perturbation theory, which is the subject of this paper. We note that higher-order perturbation theory has been studied for use in many quantum mechanical problems: the anharmonic oscillator, 20^{-22} the Yukawa and funnel potentials, 2^{3-26} the Stark 2^{7-33} and Zeeman^{34–36} effects in the hydrogen atom, the molecular hydrogen ion, etc., as well as for $1/n$ -expansions.^{14–17} The problem examined below is of interest in that the asymptotic regime undergoes a change at a certain value of $\gamma = \gamma_c$: the perturbation series switches from a constant sign series to an alternating series, which is

FIG. 1. The function $g(\gamma,\theta)$ (smooth curves next to which the angle θ is indicated) as a function of the parameter γ . The dashed curves are the values of $|g_c(\gamma,\theta)|$ for $N=1$ and 2, corresponding to the solution of Eqs. (10)– $(12).$

explained by examining a new class of complex subbarrier trajectories besides the usual subbarrier trajectory.

The asymptotic behavior of the higher orders of perturbation theory is of interest from a general standpoint, but is also of practical importance for calculating the shifts of atomic levels and their widths $\Gamma = \hbar \omega(\mathscr{E}, \mathscr{H})$, using special procedures for summing diverging series, such as the Borel or Padé–Borel summation techniques.^{28–33,37,38}

2. In calculating the energy levels of atoms in an electric field *E*, the standard approach is to expand the energy in a perturbation series,

$$
E(\mathscr{E}) = \sum_{k=0}^{\infty} E_k \mathscr{E}^k.
$$
 (4)

According to Dyson's argument, 39 the instability of the state (complex energy $E = E_r - i\Gamma/2$, where Γ is the level width) is related to the divergence of the perturbation theory series. We shall study the behavior of the higher orders of perturbation theory in the presence of a magnetic field. To evaluate the behavior of the perturbation coefficients E_k as $k \rightarrow \infty$, we use the dispersion relation^{20,28,33}

$$
E_k = \frac{1}{2\pi i} \oint \frac{E(\mathscr{E})}{\mathscr{E}^{k+1}} d\mathscr{E} = -\frac{1}{2\pi} \int_0^\infty \frac{\Gamma(\mathscr{E})}{\mathscr{E}^{k+1}} d\mathscr{E} \tag{5}
$$

(here we have taken advantage of the familiar analytic properties of the function $E(\mathscr{E})$, in particular its behavior on a large circle³²: $|E(\mathscr{E})| \propto (\mathscr{E} \ln \mathscr{E})^{2/3}$ as $\mathscr{E} \rightarrow \infty$ for the ground state of hydrogen).

The asymptotic behavior of the higher orders of perturbation theory is determined by the level width $\Gamma(\mathscr{E})$ in an arbitrarily small field, so that it is possible to use the semiclassical Eq. (1). Equation (3) is even with respect to τ and has a pair of roots $\pm \tau_0$, for which the values of $g(\gamma,\theta)$ differ in sign. Given this, Eqs. (1) and (5) imply that

$$
E_k \approx \frac{1 + (-1)^k}{2} k! a^k k^{\beta} \bigg(c_0 + \frac{c_1}{k} + \dots \bigg), \quad k \to \infty,
$$
 (6)

$$
a=3[2\kappa^3 g(\gamma,\theta)]^{-1}
$$
 (7)

(in the case of the ground state, the odd orders of the perturbation expansion for the energy $E(\mathscr{E})$ vanish identically). In the following we examine only the even orders of the per-

turbation expansion, omitting the factor $[1+(-1)^k]/2$. In particular, for the Stark effect in the hydrogen atom, we have²⁸

$$
E_k \approx -\frac{6}{\pi} k! \left(\frac{3}{2}\right)^k \left[1 - \frac{107}{18k} + \frac{7363}{648k^2} + \dots\right], \quad a = \frac{3}{2}, \quad \beta = 0
$$
\n(6a)

(in the ground state $E_0 = -1/2$, $\kappa = \sqrt{-2E_0} = 1$).

Besides the Stark expansion (4) , let us consider the expansion of the ground state energy in powers of the magnetic field:

$$
E = \sum_{k=0}^{\infty} \widetilde{E}_k \mathcal{H}^k, \quad \widetilde{E}_k = \gamma^{-k} E_k.
$$
 (8)

In the case of the Zeeman effect ($\gamma \rightarrow \infty$), the higher orders of this expansion also increase factorially:^{35,36}

$$
\widetilde{E}_k \approx (-1)^{(k+2)/2} \left(\frac{4}{\pi}\right)^{5/2} \Gamma\left(k + \frac{3}{2}\right) \left(\frac{1}{\pi}\right)^k \left[1 + O(k^{-1})\right] \tag{6b}
$$

 $(k$ even), which corresponds formally to the asymptote (6) with a purely imaginary parameter $\tilde{a} = a/\gamma = \pm (\pi i)^{-1}$. At the same time, for $\gamma \geq 1$, by virtue of Eqs. (3b) and (7), we have

$$
\tilde{a} = 3/2 \gamma g(\gamma, \theta) \approx \frac{3}{2 \gamma} \cos \theta \rightarrow 0,
$$

which is inconsistent with the previous result. This suggests the existence of other solutions (i.e., complex subbarrier trajectories for which the parameter \tilde{a} does not vanish in the limit of a strong magnetic field). We shall show that this is indeed so, by solving Eq. (3) for $\gamma \rightarrow \infty$ in the complex plane.

Taking $\tau = i\tilde{\tau}$ and $\gamma = i\tilde{\gamma}$, we rewrite Eq. (3) in the form

$$
\tilde{\tau}^2 + \sin^2 \theta (1 - \tilde{\tau} \cot \tilde{\tau})^2 = \tilde{\gamma}^2. \tag{9}
$$

There are two possibilities as $\gamma \rightarrow \infty$: either $\tilde{\tau}_0 \rightarrow \pm i \gamma / \cos \theta$ (here cot $\tilde{\tau}_0 \rightarrow \pm i$) or $\tilde{\tau}_0 \rightarrow \pm N\pi$ for integer $N \neq 0$ (cot $\tilde{\tau}_0$) $\rightarrow \infty$). The first possibility corresponds to the real solution considered above. In the second case we obtain

$$
\tilde{\tau}_0 = N\pi + N\pi \sin \theta \cdot \tilde{\gamma}^{-1} + \frac{1}{2} (N\pi)^3 \sin \theta
$$

$$
\times \left[\left(1 - \frac{2}{3} \sin^2 \theta \right) \tilde{\gamma}^{-3} + \sin \theta \left(1 - \frac{1}{3} \sin^2 \theta \right) \tilde{\gamma}^{-4} \right] + ...
$$
 (10)

 $(N=1,2,...)$, with $\tilde{\tau}_0^*$, $-\tilde{\tau}$, and $-\tilde{\tau}_0^*$ also solutions of Eq. (9) . Introducing the function

$$
G(\gamma, \theta) = \frac{2\tilde{\gamma}}{3\pi} g_c(\gamma, \theta)
$$

=
$$
\frac{2\tilde{\tau}_0^3}{3\pi \tilde{\gamma}^2} \left\{ 1 + \frac{1}{2} \sin^2 \theta \left[1 + 3 \cot \tilde{\tau}_0 \left(\cot \tilde{\tau}_0 - \frac{1}{\tilde{\tau}_0} \right) \right] \right\}
$$
(11)

and substituting Eq. (10) into it, some simple but cumbersome calculations yield

FIG. 2. Solutions of Eq. (9) in the complex plane for $\theta=30^{\circ}$, 60°, and 90° $(N=1)$. The values of the parameter $\gamma=0,2,4,...$ are indicated on the curves.

$$
G(\gamma, \theta) = N \left\{ 1 - 2 \sin \theta \cdot \tilde{\gamma}^{-1} + \left(\sin^2 \theta - \frac{(N\pi)^2}{3} \cos^2 \theta \right) \tilde{\gamma}^{-2} + (N\pi)^2 \sin \theta \left(\frac{2}{3} \sin^2 \theta - 1 \right) \cdot \tilde{\gamma}^{-3} + (N\pi)^2 \sin^2 \theta \left(\frac{1}{3} \sin^2 \theta - 1 \right) \tilde{\gamma}^{-4} - (N\pi)^2 \sin \theta \left(\frac{1}{3} \sin^2 \theta + (N\pi)^2 \left(\frac{2}{15} \sin^4 \theta \right) - \frac{1}{3} \sin^2 \theta + \frac{1}{4} \right) \tilde{\gamma}^{-5} + \dots \right\}.
$$
 (12)

These expansions are valid for $\gamma \rightarrow \infty$. On the other hand, the value of $\tilde{\tau}_0$ at $\gamma=0$ can be determined from the equation cot $\tilde{\tau}_0$ – $1/\tilde{\tau}_0$ = $\pm i/\sin \theta$. A numerical analysis of Eq. (9) shows that as γ increases, the point $\tau_0 = i\tilde{\tau}_0(\gamma, \theta)$ describes the curves shown in Fig. 2 in the complex plane.

We shall mainly be interested in the case $N=1$, where the function $G(\gamma,\theta)$ has its minimum absolute value. The values of $|g_c| = |(3\pi/2\tilde{\gamma})G(\gamma,\theta)|$ for $N=1$ and 2 are shown in Fig. 1 as the dotted curves. For sufficiently large γ , when $|g_c| < g$, the asymptotic parameter *a* can be found from Eq. (7) by replacing *g* with g_c . Because of the existence of a pair of complex conjugate solutions $\tilde{\tau}_0$ and $\tilde{\tau}_0^*$, the asymptotes of the higher-order perturbation theory now have the form

$$
\widetilde{E}_k \sim (-1)^{k/2} \operatorname{Re}(CA^k)k!k^{\beta}, \quad A = i\widetilde{a} = 3/2|g_c(\gamma, \theta)|, \tag{13}
$$

so that the perturbation series is alternating for sufficiently large *k*.

In the limit $\theta \rightarrow 0$ (parallel *E* and *H* fields), the expansion (12) terminates at the third term, so a solution can be obtained in analytic form:

$$
G(\gamma,0) = N \left[1 - \frac{(N\pi)^2}{3\tilde{\gamma}^2} \right],
$$

\n
$$
g_c(\gamma,0) = i \frac{3N\pi}{2\gamma} \left[1 + \frac{(N\pi)^2}{3\gamma^2} \right].
$$
\n(14)

The condition $|g_c(\gamma,0)| = g(\gamma,0) = 1$ determines the "critical'' value of γ (Ref. 6):

$$
\gamma_c = \pi [(1 + \sqrt{2})^{1/3} - (1 + \sqrt{2})^{-1/3}]^{-1} = 5.270495...,
$$

N=1 (15)

(see Appendix). For $\gamma < \gamma_c$, i.e., in sufficiently strong electric fields, the dominant contribution to the asymptotic E_k is from the subbarrier trajectory with real τ_0 , corresponding to the function $g(\gamma,0)$, and the perturbation series has a fixed sign. If, however, $\gamma > \gamma_c$, then $a_c = 1.5|g_c|^{-1} > a$ $=1.5|g|^{-1}$, so the signs of the higher orders of perturbation theory should alternate according to Eq. (13). Thus, at γ $= \gamma_c$ the structure of the perturbation series changes.

We have verified this by direct calculation of the perturbation series coefficients E_k up to $k=80$ (see Table I). (For $k \le 10$ our calculations agree with an earlier paper⁴⁰ and for $\gamma=0$, with Refs. 28–33). Some of these results are shown in Fig. 3. It has been shown that between $\gamma=5$ and 5.5, the order of the signs³⁾ of the coefficients E_k does indeed change. In addition, for $\gamma < \gamma_c$ the coefficients $E_k(\gamma)$ are all of the same order of magnitude (since $g(\gamma,\theta) \equiv 1$ and the asymptotic parameter $a=3/2$ is independent of γ), while for $\gamma > \gamma_c$ they begin an additional (and very rapid!) growth in accordance with the reduction in $|g_c(\gamma)|$, which is clearly evident in Fig. 3 (see also Eq. $(A4)$).

In the case considered here $(\theta=0)$, the critical value of the parameter $\gamma=h/\epsilon$ can be found analytically. It is interesting to study the structure of the perturbation series in the more general case as well, especially for mutually perpendicular fields. The value of γ_c that determines the restructuring of the perturbation series can be found from the condition $g=|g_c|$, where⁴⁾

$$
g\left(\gamma, \frac{\pi}{2}\right) = \frac{3}{8} \gamma (1 + 2\gamma^{-2} + \gamma^{-4} + \dots), \quad \gamma \to \infty,
$$
 (16)

$$
G\left(\gamma, \frac{\pi}{2}\right) = 1 - \gamma^{-2} - \frac{2\pi^2}{3} \gamma^{-4} - \frac{2i}{\gamma} \left[1 - \frac{\pi^2}{6} \gamma^{-2} + \frac{\pi^2}{6} \left(1 + \frac{3\pi^2}{20}\right) \gamma^{-4}\right] + \dots,
$$
 (17)

whence

$$
\left| g_c \left(\gamma, \frac{\pi}{2} \right) \right| = \frac{3\pi}{2\gamma}
$$

$$
\times \left[1 + 2\gamma^{-2} - \left(\frac{8\pi^2}{3} - 1 \right) \gamma^{-4} + O(\gamma^{-6}) \right]
$$
(17a)

and $\gamma_c \approx 3.54$. This simple estimate is in good agreement with the numerical calculations (see the point of intersection of the smooth and dashed $(N=1)$ curves for $\theta=90^\circ$ in Fig. 1).

Similarly, we can calculate γ_c for arbitrary angles θ . It would be interesting to confirm the existence of a switch in the asymptotic regime at $\gamma = \gamma_c(\theta)$ by direct calculation of the higher perturbation orders, as has been done above for the case of parallel fields.

Note. The table lists the coefficients in the perturbation theory series (4) for the ground state of the hydrogen atom taken with the opposite sign; *k* is the perturbation theory order; $a(b) \equiv a \cdot 10^b$.

3. Therefore, at $\gamma = \gamma_c$ there is a change in the character of the asymptotic behavior of the higher orders of perturbation theory.⁵⁾ Upon going from one asymptotic regime to the other, the perturbation series switches (when $k \ge k_0$) from an alternating series to one with a constant sign, which shows up in the position of the singularities in the Borel transformants that are closest to zero, and therefore in the choice of an efficient method for taking the sum.^{17,33,37}

The complex solutions of Eq. (3) found above correspond to complex subbarrier trajectories which, therefore, can be important in determining the asymptotic behavior of the higher orders of perturbation theory. Their physical significance can be clarified using the example of parallel *E* and *H* fields. It is known that the asymptotic behavior of the higher orders of perturbation theory is directly related to the tunneling probability for a particle in a potential with the "wrong" sign on the coupling constant, e.g., $g \rightarrow -g$ in the case of an anharmonic oscillator,

$$
V(x) = \frac{1}{2}x^2 + g\frac{x^4}{4}
$$

FIG. 3. Higher orders of perturbation theory $(Eq. (4))$ for the ground state of the hydrogen atom in parallel fields.

(the Dyson phenomenon^{39,20}). In our problem, \mathcal{H}^2 plays the role of *g*. Going to purely imaginary values of the magnetic field $(\mathcal{H} = i\tilde{\mathcal{H}})$, we obtain a potential proportional to $-(1/8)\tilde{\mathcal{H}}^2\rho^2$, decreases without bound as $\rho = \sqrt{x^2+y^2}$ $\rightarrow \infty$. It is evident that in such a potential, tunneling is possible both along the electric field (the ζ axis) and perpendicular to it. The complex solutions $(10)–(12)$ probably correspond to an analytic continuation of ''perpendicular'' subbarrier trajectories of this sort from a region of purely imaginary magnetic fields into a region of real *H*.

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APPENDIX

The perturbation theory coefficients (4) for the energy of the ground state of the hydrogen atom are polynomials in γ^2 :

$$
E(\mathcal{E}, \mathcal{H}) = \sum_{i,j} c_{ij} \epsilon^{2i} h^{2j} = \sum_{k=0}^{\infty} E_{2k}(\gamma) \epsilon^{2k},
$$
 (A1)

$$
E_{2k}(\gamma) = \sum_{j=0}^{k} c_{k-j,j} \gamma^{2j},
$$
 (A2)

where 2*k* is the order of perturbation theory, $\gamma = h/\epsilon$ $= \alpha \mathcal{H}/\mathcal{E}$, and $\alpha = e^2/\hbar c$ is the fine structure constant. Several of the lowest orders of perturbation theory are known exactly, i.e., in the form of rational fractions, as

$$
E_0 = -\frac{1}{2}
$$
, $E_2 = -\frac{1}{4}(9 - \gamma^2)$,

$$
E_4 = -\frac{1}{64} \left(3555 - 318 \gamma^2 + \frac{53}{3} \gamma^4 \right),
$$

\n
$$
E_6 = -\frac{1}{512} \left(2512 779 - 254 955 \gamma^2 + \frac{49195}{3} \gamma^4 - \frac{5581}{9} \gamma^6 \right),
$$

\n
$$
E_8 = -2^{-12} \cdot \left(\frac{13012 777 803}{4} - \dots - \frac{12368 405}{9} \gamma^6 + \frac{21577 397}{540} \gamma^8 \right),
$$
 (A3)

and were used to monitor the numerical calculations. The outer coefficients c_{k0} and c_{0k} in Eq. (A2) correspond to the Stark²⁷⁻³¹ and Zeeman³⁴ effects, while the cross terms $(1 \le j \le k-1)$ were taken from Johnson *et al.*⁴⁰ and Lambin *et al.*⁴¹ Here $c_{k-j,j} = 2^{-3j} \epsilon^{(k-j,j)}$, where the $\epsilon^{(ij)}$ are coefficients tabulated (for the case of parallel fields) by Johnson *et al.*⁴⁰

The asymptotes of the higher-order perturbation theory can be written in the form

$$
E_k(\gamma) \approx -k! \left\{ \frac{6}{\pi} \, c_0 a^k + (-1)^{k/2} \left(\frac{4}{\pi} \right)^{5/2} c_1 a_c^k k^{1/2} \right\},\quad \text{(A4)}
$$

where

$$
a = \frac{3}{2}, \quad a_c = \frac{\gamma}{\pi} \left(1 + \frac{\pi^2}{3\gamma^2} \right)^{-1}, \tag{A5}
$$

 $c_0 = \gamma / \sinh \gamma$, and for c_1 we obtained (numerically) $c_1 \approx 1$ $-12.03\gamma^{-2}$ for $\gamma \ge 1$. The condition $a=a_c$ yields a cubic equation whose solution (according to the Cardano formula) leads to Eq. (15) .

- ¹⁾The imaginary time method was developed for the theory of multiphoton ionization of atoms and ions in strong optical fields,^{9,10,12} and has also been used in the problem of electron–positron pair production from the vacuum in a variable electric field.^{11,13} This method has been used^{14–16} to study the asymptotic behavior of the higher orders of the 1/*n*-expansion in multidimensional quantum mechanics problems, including the two-center Coulomb problem (another approach to this problem has appeared recently 17,18).
- ²⁾This function was first calculated by Kotova et al.¹⁹
- ³⁾Numerical calculations show that sign $E_{2k} = (-1)^{k+1}$ for $\gamma \ge 5.5$ and 2*k* ≤ 80 . On the other hand, for $\gamma \leq 5$ the coefficients E_{2k} < 0 for sufficiently large $k \ge k_0$, where k_0 depends on γ and increases rapidly as it approaches γ_c . Thus, $2k_0=0, 0, 4$, and 68, respectively, for $\gamma=0, 2, 4$, and 5 (see Table I).
- ⁴⁾The first expansion follows from Eqs. (2) and (3) , the second, from Eq. (12) with $\overline{N}=1$. The parameter in these expansions is γ^{-2} , with γ_c^{-2} $\sim 0.08 \ll 1$
- 5 An analogous phenomenon occurs in the $1/n$ -expansion in the problem of two Coulomb centers.^{14–16} In this case the role of the parameter γ is played by the internuclear distance *R*.

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