Transition to Turbulence 
in the Rayleigh-Bénard Convection

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Time evolution of convection appearing in a fluid contained in a rectangular box heated from below is investigated using a 48-mode system of equations. For the case where the Prandtl number $\sigma=2.5$ and the box aspect ratio $r_y=3.5$, with the increase of the vertical thermal gradient across the system, the fluid motion undergoes successive transitions as follows: periodic motion → quasi-periodic motion with two fundamental frequencies → quasi-periodic motion with three frequencies → turbulence. In particular, the transition to turbulence occurs when the fundamental frequencies of the above quasi-periodicity satisfy the resonance condition.

§ 1. Introduction

There have long been both theoretical and experimental interests in the Rayleigh-Bénard convection appearing in a thin fluid layer heated from below because it develops various types of spatial structures and temporal oscillations depending on the external conditions imposed on the system. In recent years quite a lot of research groups have studied the problem using high-precision measurement techniques particularly to understand how the time-dependent ordered motion of convection suffers a transition to chaos or turbulence when the vertical thermal gradient across the fluid layer, i.e., the Rayleigh number, is increased. Their results have revealed that the periodic motion turns into turbulence in quite different ways depending on the Prandtl number, the vessel aspect ratios and furthermore how the relevant convection flow is initially prepared. Along with this, there have been several theoretical attempts to understand the transition of convection to turbulence on the basis of numerical time integration of the governing evolution equations.

The purpose of the present paper is to study time evolution of a convection in a rectangular box for the case where the Prandtl number is low and the box aspect ratios are small. The experimental results have shown that under these external conditions the well-defined spatial pattern of convection persists over quite a wide range of the Rayleigh number. Hence it will be reasonable to construct a model system of evolution equations on the basis of the Galerkin procedure. In § 2 we obtain a system of ordinary differential equations for convection velocity and temperature mode variables, starting with the basic fluid
and temperature equations which obey the Boussinesq conditions. In § 3 we use a truncated system of equations including 48 mode variables to investigate how the temporal behavior of the convection modes changes as the Rayleigh number is increased. For this purpose we have integrated it on a computer. A summary and discussion are given in § 4.

§ 2. Model equations

We consider thermal convection of a fluid confined in a rectangular box heated from below. Let \(x, y,\) and \(z\) denote the rectangular coordinates with \(z\) being the vertical coordinate. We express all the physical quantities in the non-dimensional form using the length-scale \(d,\) the time-scale \(d^2/\nu\) and the temperature-scale \(xv/gad^3\) where \(d\) is the distance between the two parallel horizontal boundary plates, \(\nu\) is the thermal diffusivity, \(v\) is the kinematic viscosity, \(\alpha\) is the coefficient of thermal expansion and \(g\) is the gravitational constant. The non-dimensional parameters characterizing the system are the Rayleigh number \(R,\) the Prandtl number \(\sigma\) and the vessel aspect ratios \(\Gamma_x\) and \(\Gamma_y,:\)

\[
R = g \rho_0 d^3 \Delta T / \nu \alpha, \quad \sigma = \nu / \alpha, \quad \Gamma_x = L_x / d, \quad \Gamma_y = L_y / d,
\]

where \(\Delta T = T_0\) is the difference between the temperatures of the two horizontal plates, \(L_x\) and \(L_y\) are the horizontal dimensions of the vessel. Within the framework of the Boussinesq approximation, the dimensionless equations for the velocity \(u(x, y, z)\) and temperature \(\theta\) take the form

\[
\begin{align*}
\partial_t u_i - \sigma \Delta u_i - \alpha \lambda \partial_i \theta + \partial_j (\delta \rho / \rho_0) &= - u_j \partial_j u_i & \text{for } i = x, y, \text{and } z, \\
\partial_t \theta - \Delta \theta - R \lambda \partial_j u_j &= - u_j \partial_j \theta, \\
\partial_j u_j &= 0, \tag{2}
\end{align*}
\]

where \(\lambda = (0, 0, 1),\) \(\delta \rho\) is the pressure and \(\rho_0\) is the density of fluid. We assume that the velocity fields \(u\) satisfy the rigid boundary conditions while the disturbance temperature field \(\theta\) satisfies the conducting and insulating conditions at the horizontal and lateral wall boundaries respectively:

\[
\begin{align*}
u_x = \partial_y u_x = u_y = u_z = \partial_z \theta &= 0 \quad \text{at } x = \pm \Gamma_x / 2, \\
u_x = u_y = \partial_y u_y = u_z = \partial_z \theta &= 0 \quad \text{at } y = \pm \Gamma_y / 2, \\
u_x = u_y = u_z = \partial_z u_z = \theta &= 0 \quad \text{at } z = \pm 1 / 2. \tag{3}
\end{align*}
\]

We further assume that the basic convection pattern is the rolls whose axes are along the \(x\) direction. This will be reasonable when the Prandtl number is low, the aspect ratios \(\Gamma_x\) and \(\Gamma_y\) are small and furthermore they satisfy \(\Gamma_x < \Gamma_y.\)
On the other hand, it was pointed out by Busse and Clever that the convection develops time oscillation closely connected with the appearance of the vertical vorticity. Hence we expand the velocity field in superposition of the two sets of rolls perpendicular to each other:

\begin{align}
    u_x &= \partial_x \phi_2, \quad u_y = -\partial_x \phi_1 - \partial_x \phi_2, \quad u_z = \partial_y \phi_1, \\
    \phi_1(x, y, z, t) &= \sum_{i, k, m} \xi_{i, k, m}(t) \chi_i(x/\Gamma_x) \phi_k(y/\Gamma_y) \phi_m(z), \\
    \phi_2(x, y, z, t) &= \sum_{i, k, m} \xi_{i, k, m}(t) \phi_i(x/\Gamma_x) \phi_k(y/\Gamma_y) \chi_m(z),
\end{align}

and the temperature field in the form:

\[ \theta(x, y, z, t) = \sum_{i, k, m} \theta_{i, k, m}(t) \phi_i(x/\Gamma_x) \phi_k(y/\Gamma_y) \chi_m(z). \]

We employ the systems of the expansion functions \( \{ \phi_m \} \), \( \{ \psi_m \} \) and \( \{ \chi_m \} \) which are determined respectively by the following eigenvalue problems:

\begin{align}
    D^4 \phi_m &= -\alpha_m^2 D^2 \phi_m, \quad \text{b.c.} \quad \phi_m(\pm 1/2) = D \phi_m(\pm 1/2) = 0, \\
    D^2 \psi_m &= -\beta_m^2 \psi_m, \quad \text{b.c.} \quad D \psi_m(\pm 1/2) = 0, \\
    D^2 \chi_m &= -\gamma_m^2 \chi_m, \quad \text{b.c.} \quad \chi_m(\pm 1/2) = 0, 
\end{align}

for \( m = 1, 2, \ldots \), where \( D = d/dx \). The orthogonal relations are defined by

\begin{align}
    (D \phi_m, D \phi_n) &= \int_{-1/2}^{1/2} D \phi_m(x) D \phi_n(x) dx = \alpha_m^2 \delta_{m,n}, \\
    (\phi_m, \psi_n) &= \delta_{m,n} \quad \text{and} \quad (\chi_m, \chi_n) = \delta_{m,n}.
\end{align}

Substitution of Eqs. (4) into Eqs. (2) and elimination of the pressure variable lead to

\begin{align}
    (\partial_t - \sigma \Delta) \phi_1 - \sigma \partial_y (\partial_x^2 + \partial_y^2) \theta \\
    &= \partial_x \partial_y F_x + \partial_y \partial_x F_y - \partial_y (\partial_x^2 + \partial_y^2) F_x, \\
    (\partial_t - \sigma \Delta) \phi_2 + \sigma \partial_x \partial_y \theta &= -\partial_y (\partial_y^2 + \partial_z^2) F_x + \partial_x \partial_y^2 F_y + \partial_x \partial_y \partial_z F_z, \\
    (\partial_t - \Delta) \theta - R \partial_y \phi_1 &= -F_\theta,
\end{align}

where \( F_i = u_i \partial_i \psi_i \) \( (i = x, y, z) \) and \( F_\theta = u_i \partial_i \theta \). Substituting Eqs. (5)~(7) into Eqs. (10)~(12), we multiply Eq. (10) by \( \chi_i \phi_k \psi_m \), Eq. (11) by \( \psi_i \phi_k \chi_m \) and Eq. (12) by \( \phi_i \phi_k \chi_m \), and integrate each over the three-dimensional domain \( (-\Gamma_x/2, \Gamma_x/2) \times (-\Gamma_y/2, \Gamma_y/2) \times (-1/2, 1/2) \). We thus obtain a system of equations which describes time evolution of the convection mode amplitudes:
\[
\begin{align*}
\sum_{i,j,n} \delta_{i,j} \delta_{k,m} \frac{\alpha_s^2}{\Gamma_x^2} \left[ \left( \frac{\gamma_i^2}{\Gamma_x^2} + \frac{\alpha_s^2}{\Gamma_y^2} \right) (\phi_m, \phi_n) + \alpha_m^2 \delta_{m,n} \right] \partial_i \xi_{i,j,l,n}^{(1)} \\
+ \sigma \sum_{i,j,n} \delta_{i,j} \delta_{k,m} \frac{\alpha_s^2}{\Gamma_x^2} \left[ \left( \frac{\gamma_i^2}{\Gamma_x^2} + \frac{\alpha_s^2}{\Gamma_y^2} \right) (\phi_m, \phi_n) \right] \\
+ \alpha_m \left[ \alpha_m^2 + 2 \left( \frac{\gamma_i^2}{\Gamma_x^2} + \frac{\alpha_s^2}{\Gamma_y^2} \right) \right] \delta_{m,n} \xi_{i,j,l,n}^{(1)} \\
+ \sigma \sum_{i,j,n} \left( \frac{\beta_i^2}{\Gamma_x^2} + \frac{\beta_j^2}{\Gamma_y^2} \right) (\chi_i, \chi_j) (\phi_k, \partial_x \phi_l) (\phi_m, \chi_n) \theta_{i,j,l,n} \\
= \sum_{p} \sum_{i', k', m'} \sum_{i'', k'', m''} \sum_{l', k', m'} U_{i', k', m'; i'', k'', m''} \xi_{i', k', m'; i'', k'', m''}^{(p)} \xi_{i', k', m'; i'', k'', m''}^{(s)}, \\
\end{align*}
\]

\[
\begin{align*}
\sum_{i,j,n} \delta_{i,j} \delta_{k,m} \frac{\alpha_s^2}{\Gamma_x^2} \left[ \frac{\alpha_i^2}{\Gamma_x^2} \delta_{i,j} + (\phi_i, \phi_j) \left( \frac{\alpha_s^2}{\Gamma_y^2} + \gamma_m^2 \right) \right] \partial_i \xi_{i,j,l,n}^{(2)} \\
+ \sigma \sum_{i,j,n} \delta_{i,j} \delta_{k,m} \frac{\alpha_s^2}{\Gamma_x^2} \left[ \frac{\alpha_i^2}{\Gamma_x^2} \delta_{i,j} + 2 \left( \frac{\alpha_s^2}{\Gamma_y^2} + \gamma_m^2 \right) \right] \delta_{i,j} \\
+ \sigma \sum_{i,j,n} \left( \phi_i, \partial_x \phi_j \right) (\phi_k, \partial_x \phi_l) (\chi_m, \chi_n) \theta_{i,j,l,n} \\
= \sum_{p} \sum_{i', k', m'} \sum_{i'', k'', m''} \sum_{l', k', m'} U_{i', k', m'; i'', k'', m''} \xi_{i', k', m'; i'', k'', m''}^{(p)} \xi_{i', k', m'; i'', k'', m''}^{(s)}, \\
\end{align*}
\]

The explicit expressions for the nonlinear coupling coefficients appearing on the right-hand sides of Eqs. (13)~(15) are given in Appendix.

§ 3. Computational results

We now deal with time evolution of the convection mode amplitudes governed by Eqs. (13)~(15). We choose the system parameters as $\sigma = 2.5$, $\Gamma_x = 2.0$ and $\Gamma_y = 3.5$. Under these external conditions Gollub and Benson have observed experimentally that convection takes the form of the two rolls with their axes along the $x$ direction; that it develops with the increase of $R$ a few different types of ordered time oscillations one after another, and eventually undergoes a transition to turbulence. Hence we assume the basic flow of our system is specified by the mode variable $\xi_{i,j,l,n}^{(1)}$ where the indices $e$ and $o$ indicate the evenness and
(continued)
Fig. 1. Power spectral densities for the variable $\sum_{i=1}^{m}[\xi_{1,io10}(t)+\xi_{1,io10}(t)+\xi_{1,io10}(t)+\xi_{1,io10}(t)]$. 

(a) $r = 39.77$  
(b) $r = 39.96$  
(c) $r = 40.83$  
(d) $r = 41.04$

Fig. 2. Orbital profiles in the subspace $(\xi_{1,io10}(t), \theta_{1,io10}(t))$. 

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oddness of the corresponding mode basis functions respectively. By the use of the truncated equations consisting only of the modes \( \xi_{16,10,1e}, \theta_{16,2e,le} \) and \( \theta_{16,1e,10} \), we can describe the steady roll convection so long as their amplitudes remain sufficiently small, and furthermore determine its onset value \( R_c \). The result is given by \( R_c = 5780.7 \). In contrast, the onset value of \( R \) for steady roll convection in an infinite layer is given by \( R_{c,\infty} = 1707.8 \).

In order to describe time evolution of the convection mode amplitudes over a sufficiently wide range of \( r \), we use a truncated system of equations Eqs. (13) \~(15) which consists of the mode variables: \( \xi_{16,10,1e}, \xi_{16,10,1e}, \xi_{16,10,1e}, \xi_{16,10,1e}, \theta_{16,2e,le}, \theta_{16,2e,10}, \theta_{16,1e,1e}, \theta_{16,1e,10} \) where \( 1 \leq i \leq 6 \). The truncation contains 48 variables in all. We have numerically integrated it with respect to time for several values of \( r \) with the aid of the Adams-Bashforth-Moulton predictor-corrector method of the fourth order. Using the time-series data thus obtained, we have computed by FFT the power spectral densities (PSD) for the variable \( \Sigma_{i=1}^{6} [\xi_{16,10,1e}(t) + \xi_{16,10,1e}(t) + \xi_{16,10,1e}(t) + \xi_{16,10,1e}(t)] \) which approximately represents the local flow velocity in the \( y \) direction. We give the results in Fig. 1 where the spacing of discrete Fourier transform is \( \Delta f = 0.01526 \times 13.1x/d^2 \) (Hz). In Fig. 2 we give the orbital profiles which are obtained by mapping the phase orbits of the convection modes into the two-dimensional subspace specified by \( (\xi_{16,10,1e}(t), \theta_{16,2e,1e}(t)) \). Figure 2(a) shows that the phase orbit at \( r = 39.77 \) is a limit cycle around the two destabilized fixed points which represent the two steady convection flows whose directions are opposite to each other. Furthermore, the PSD [Fig. 1(a)] indicates that the motion is singly-periodic and characterized by a single frequency which we denote by \( p_1 \). When the Rayleigh number is increased to \( r = 39.98 \), the second frequency denoted by \( p_2 \) appears in the PSD [Fig. 1(b)] and the motion becomes quasi-periodic. The phase orbit Fig. 2(b) is now embedded in a band-like torus attractor. Incidentally, the values of the fundamental frequencies here are given by \( p_1 = 0.029 \) Hz and \( p_2 = 0.0099 \) Hz for \( x = 0.0016 \) cm \(^2\) s\(^{-1}\) and \( d = 0.79 \) cm. With a slight increase of \( r \) to \( r = 40.20 \), however, the motion develops a very slow modulation of the constituent frequencies, and in the PSD [Fig. 1(c)] each frequency component appears accompanied by another frequency which is only slightly larger or smaller than the original one. We denote this difference frequency by \( p_3 \). With further increase of \( r \), the PSD in Figs. 1(d) \~(f) show that the value of \( p_3 \) gradually increases and finally the overlapping between certain pairs of the neighboring frequencies occurs. When \( r \) is increased still further, the spectral lines in Fig. 1(g) suffer broadening and finally, as shown in Fig. 1(f), all the spectral peaks disappear from the PSD and only the featureless spectra remain. In addition, Fig. 2(d) shows the randomized irregular phase orbit. Hence we can consider that the motion is now non-periodic or turbulent.
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![Graphs showing spectral analysis for different values of r]

- (a) $r = 39.77$
- (b) $r = 39.98$
- (c) $r = 40.20$
- (d) $r = 40.41$
- (e) $r = 40.51$
- (f) $r = 40.62$

(continued)
In order to clarify in more detail how the transition to turbulence occurs, we give in Fig. 3 the PSD for the mode variable \( \xi_{1,1,0,10}(t) \). The fundamental frequencies \( \rho_1 \) and \( \rho_2 \) characterize the quasi-periodic motion of the modes \( \xi_{1,1,0,1i}(t) \) and \( \xi_{0,1,0,10}(t) \) with \( 1 \leq i \leq 6 \). Through the nonlinear interactions, these give rise to the frequencies appearing in the PSD Fig. 3(b) for the mode \( \xi_{1,1,0,10}(t) \). Figure 3(c) shows that the very low frequency \( \rho_3 \) appears at \( r = 40.20 \). Since \( \rho_3 \) increases gradually with the increase of \( r \), certain pairs of the neighboring peaks for the combination modes, e.g., \( \rho_1 - \rho_2 + \rho_3 \) and \( \rho_1 + \rho_2 - \rho_3 \), approach each other in the PSD as shown in Figs. 3(c)~(f). Figures 3(f) and (g) suggest that a slight increase of \( r \) from \( r = 40.62 \) to 40.83 causes the overlapping of several pairs of the neighboring peaks and thereby the broadening of spectral lines. Hence in our system the resonance \( \rho_2 = \rho_3 \) seems to cause the transition to turbulence.

§ 4. Summary and discussion

In this paper we have investigated the time evolution of a model system for the Rayleigh-Bénard convection to clarify how the mode motion develops turbulence when the Rayleigh number is increased but nonetheless the convection does not thereby undergo a fundamental spatial structural transition. The computational results show that the quasi-periodic motion with three frequencies \( \rho_1, \rho_2 \) and \( \rho_3 \) undergoes a transition to non-periodicity when the overlapping of certain pairs of the spectral peaks occurs, i.e., the rationally dependent relation between the frequencies is satisfied: \( \rho_2 = \rho_3 \). We have carried out similar computations for a few different model systems of the Taylor vortex flow. Also in these cases, the quasi-periodic motion with three frequencies appears before the
motion becomes non-periodic. However, we are now not able to determine to what extent these truncated finite-dimensional systems obtained using the Galerkin method describe adequately the fluid motion governed by the original equations of motion at such higher values of the Rayleigh or Reynolds number as just before the onset of turbulence. On the other hand, in the Rayleigh-Bénard experiments the quasi-periodic motion with three frequencies appears under certain experimental conditions before the flow becomes turbulent. To our knowledge, however, it has not been clarified what is the mechanism at work when the quasi-periodic motion turns into turbulence.

We have obtained our basic equations by algebraically eliminating the pressure variable. We can also eliminate it by solving an inhomogenous Neumann problem of the Poisson equation for the pressure. This approach to the problem will be reported elsewhere.

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**Appendix**

The symmetrized nonlinear coupling coefficients appearing on the right-hand sides of Eqs. (13)–(15) take the form as follows:

\[
U_{\alpha \beta}(h;i,m,q),(j,n,r) = (1/\Gamma_y^3) M_h^{\alpha} [M_{ln}^{(1)} M_{nr}^{(3)} + M_{ln}^{(3)} M_{nr}^{(3)} + M_{ln}^{(14)} M_{nr}^{(2)}]
\]

\[
- (1/\Gamma_y^2) (M_{ln}^{(15)} M_{nr}^{(1)} + M_{ln}^{(16)} M_{nr}^{(1)} + M_{ln}^{(16)} M_{nr}^{(1)})
\]

\[+ (\gamma_h/\Gamma_x^2) (M_{ln}^{(2)} M_{nr}^{(1)} + M_{ln}^{(3)} M_{nr}^{(1)} + M_{ln}^{(3)} M_{nr}^{(1)})\frac{1}{2}, \quad (A \cdot 1)
\]

\[
U_{\alpha \beta}(h;i,m,q),(j,n,r) = (2/\Gamma_x^2 \Gamma_y^3) [M_h^{(8)} M_{ln}^{(1)} - M_{ln}^{(14)} M_{lr}^{(3)} - M_{ln}^{(7)} M_{ln}^{(3)} - \gamma_h^2 M_{ln}^{(2)} M_{ln}^{(3)}] M_{lr}^{(5)}, \quad (A \cdot 2)
\]

\[
U_{\alpha \beta}(h;i,m,q),(j,n,r) = (1/\Gamma_x \Gamma_y^3) M_h^{\alpha} [(M_{ln}^{(1)} + M_{ln}^{(2)}) M_{nr}^{(3)} + (M_{ln}^{(14)} + M_{ln}^{(3)}) M_{nr}^{(5)}]
\]

\[+ (1/\Gamma_y^2) M_{ln}^{(16)} - (\gamma_h/\Gamma_x^2) M_{ln}^{(3)} M_{nr}^{(1)}
\]

\[+ (1/\Gamma_x \Gamma_y^3) M_h^{(10)} [M_{ln}^{(2)} M_{nr}^{(7)} + (M_{ln}^{(3)} - M_{ln}^{(3)}) M_{nr}^{(6)}]
\]

\[+ [(1/\Gamma_y^2) M_{ln}^{(15)} - (\gamma_h/\Gamma_x^2) M_{ln}^{(2)} M_{nr}^{(5)}], \quad (A \cdot 3)
\]
\[ U^{21}_{i,m,q}; (i,m,q); (j,n,r) \]
\[ = \frac{2}{\Gamma \gamma_3} \left[ M^{(1)}_{mn} M_{\alpha \beta \gamma} - M^{(2)}_{mn} M^{(1)}_{\alpha \beta \gamma} - M^{(2)}_{mn} M^{(2)}_{\alpha \beta \gamma} \right], \quad (A \cdot 4) \]

\[ U^{22}_{i,m,q}; (i,m,q); (j,n,r) \]
\[ = \left( 1 + \frac{1}{\Gamma \gamma_3} \right) \left[ \frac{2M^{(1)}_{ijl}}{\gamma_p M^{(2)}_{mn}} - \frac{1}{\Gamma \gamma_3} M^{(1)}_{mn} \right] 
- \left[ \frac{a^2}{\Gamma \gamma_3^2} + \gamma_p^2 \right] M^{(1)}_{ijl} M^{(2)}_{mn} M^{(3)}_{ijl} M^{(3)}_{mn} 
+ \left( 1 + \frac{1}{\Gamma \gamma_3^2} \right) \left[ M^{(2)}_{ijl} M^{(3)}_{mn} - M^{(3)}_{ijl} M^{(2)}_{mn} \right] M^{(4)}_{opq}, \quad (A \cdot 5) \]

\[ U^{12}_{i,m,q}; (i,m,q); (j,n,r) \]
\[ = \left( 1 + \frac{1}{\Gamma \gamma_3} \right) \left[ M^{(3)}_{ijl} M^{(4)}_{mn} + M^{(1)}_{mn} M^{(1)}_{\alpha \beta \gamma} \right] 
+ \left( 1 + \frac{1}{\Gamma \gamma_3^2} \right) \left[ M^{(2)}_{ijl} M^{(2)}_{mn} M^{(2)}_{\alpha \beta \gamma} \right] 
+ \left( 1 + \frac{1}{\Gamma \gamma_3} \right) \left[ \gamma_p^2 M^{(2)}_{ijl} M^{(3)}_{mn} - (1 + \frac{1}{\Gamma \gamma_3^2}) M^{(3)}_{ijl} M^{(2)}_{mn} \right] M^{(4)}_{opq}, \quad (A \cdot 6) \]

where the expressions (A \cdot 1), (A \cdot 2), (A \cdot 4) and (A \cdot 5) are symmetrized with respect to the pairs of the indices (m, q) and (n, r) so that for the case (m, q) = (n, r) each of these should be further multiplied by the factor 0.5. The quantities \( M^{(n)}_{ijl} \) (n = 1, \ldots, 16) are expressed using \( \langle F \rangle = \int_{-1/2}^{1/2} F(x) dx \) in the form

\[ M^{(1)}_{ijl} = \langle (D \phi_k) \phi_s \phi_i \rangle, \quad M^{(2)}_{ijl} = \langle (D \phi_k) (D \phi_s) (D \phi_i) \rangle, \]
\[ M^{(3)}_{ijl} = \langle (D^2 \phi_k) \phi_s \phi_i \rangle, \quad M^{(4)}_{ijl} = \langle (D^2 \phi_k) (D \phi_s) \phi_i \rangle, \]
\[ M^{(5)}_{ijl} = \langle \phi_k \phi_s \phi_i \phi_t \rangle, \quad M^{(6)}_{ijl} = \langle (D \phi_k) (D \phi_s) \phi_i \phi_t \rangle, \]
\[ M^{(7)}_{ijl} = \langle (D \phi_k) \phi_s (D \phi_i) \phi_t \rangle, \quad M^{(8)}_{ijl} = \langle (D \phi_k) \phi_s \phi_i \phi_t \rangle, \]
\[ M^{(9)}_{ijl} = \langle (D \phi_k) \phi_s \phi_i \phi_t \rangle, \quad M^{(10)}_{ijl} = \langle (D^2 \phi_k) \phi_s \phi_i \phi_t \rangle, \]
\[ M^{(11)}_{ijl} = \langle (D \phi_k) \phi_s \phi_i \phi_t \rangle, \quad M^{(12)}_{ijl} = \langle (D \phi_k) (D^2 \phi_s) \phi_i \phi_t \rangle, \]
\[ M^{(13)}_{ijl} = \langle (D \phi_k) (D^2 \phi_s) \phi_i \phi_t \rangle, \quad M^{(14)}_{ijl} = \langle (D \phi_k) (D^3 \phi_s) \phi_i \phi_t \rangle, \]
\[ M^{(15)}_{ijl} = \langle (D^2 \phi_k) (D \phi_s) (D \phi_i) \phi_t \rangle, \quad M^{(16)}_{ijl} = \langle (D^2 \phi_k) (D^3 \phi_s) \phi_i \phi_t \rangle. \quad (A \cdot 9) \]

References