

**A PHENOMENOLOGICAL APPROACH TO THE STUDY OF THE BEHAVIOR
OF THE KINETIC COEFFICIENTS NEAR THE CRITICAL POINT IN
PURE LIQUIDS**

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We consider the contribution to the kinetic coefficients which is caused by the increase of long-wave parts of the fluctuations near the transition point of a system between liquid and gas. To describe the fluctuations before averaging over them we use the equations of hydrodynamics, regarded as microscopic equations of motion of the apposite degrees of freedom. The terms for viscosity, thermal conductivity, and so on, are treated as empirical terms approximately describing the other degrees of freedom which are taken to have already been eliminated by averaging. It is assumed that before the averaging over the fluctuations there exists a region of applicability of a theory of the Landau self-consistent-field type. We make estimates of the corrections to the kinetic coefficients, and ascertain how close to the critical point these estimates can still be taken as reasonable.

EXPERIMENTS on critical phenomena in solutions have given interesting results^[1] which indicate that the viscosity increases near the critical point of a solution. Attempts to explain such anomalies have been made in a number of papers.^[2–4] The purpose of the present paper is to treat, within the framework of a phenomenological approach, the problem of the contribution to the viscosity caused by the increase of the long-wave parts of the fluctuations near the liquid-gas transition point. The existence of a large characteristic scale of length allows us to hope that the equations of hydrodynamics can be applied, regarded as microscopic equations for the degree of freedom they contain, to describe the fluctuations before one averages over them. The terms for viscosity, thermal conductivity, and so on, are then regarded as empirical terms approximately describing the effects of other degrees of freedom over which the average has already been taken. In other words, the “bare” kinetic coefficients occurring in the microscopic equations do not give the system’s response averaged over all degrees of freedom, since the averaging has been taken only over the nonhydrodynamic degrees of freedom not associated with the critical fluctuations. We assume that the exact equations of motion of the hydrodynamic degrees of freedom are of the same form as the hydrodynamic equations in which all nonlinear effects describing the interaction of the fluctuations are taken into account. In what follows we shall keep the term “hydrodynamic equations” for these microscopic equations.

In Gibbs statistical mechanics there are relations (fluctuation-dissipation theorem) relating the mobilities of the system, averaged over all fluctuations, with the corresponding correlation functions. To find the correlation functions one could try to use the exact microscopic theory, dealing with the motion of all the particles of the system. However, we do not see any mathematical way to realize such a program. On the other hand, since the system of equations that we use contains

empirical terms describing transfer of energy into nonhydrodynamic degrees of freedom, we cannot use the usual Gibbs formalism without explicit introduction of the equations of motion for the nonhydrodynamic degrees of freedom. Nevertheless we can secure a level of critical fluctuations by introducing extraneous random forces into the system. The ensemble of these forces must be such that the “artificial” fluctuations are the same as the equilibrium thermal fluctuations that result from the usual Gibbsian averaging with the full microscopic description. Moreover they must satisfy the fluctuation-dissipation theorem, and this enables us to find the characteristics of the ensemble of random forces. This will be discussed in more detail in Sec. 2.

On averaging our system over the critical fluctuations, we get a system of macroscopic equations describing the relaxation of the macroscopic average quantities of the system (the mean particle-number density, momentum density, etc.) for the case of small deviations from equilibrium. For times and distances much larger than all the characteristic fluctuational lengths and times, we can expect, on the basis of the usual arguments employed in the derivation of the hydrodynamic equations, that these equations will be of the form of the ordinary linearized hydrodynamic equations. These macroscopic equations involve the macroscopically observable kinetic coefficients (the actual viscosity, thermal conductivity, etc.). If, on the other hand, there are macroscopic changes on time and distance scale smaller than (or of the order of) the characteristic scales associated with the correlation ranges—which are increased in the critical region—the averaged equations may be of a form very different from the ordinary equations. In particular, we can expect that there will be a strong dispersion of the kinetic coefficients. In this case the interpretation of these quantities as viscosity, thermal conductivity, and so on, is not unambiguous.

In the main we shall consider only the region of large times and distances, where the definition of the kinetic coefficients is unambiguous. In this case, by applying the hydrodynamic theory of fluctuations to the completely averaged equations, we find the expressions for the correlation functions in terms of the true viscosity, thermal conductivity, and so on. On the other hand, by calculating the correlation functions by means of an averaging over the hydrodynamic degrees of freedom, we find the expressions for the true kinetic coefficients in terms of the empirical constants. We note that it is reasonable to separate out the hydrodynamic degrees of freedom only for scales of size much larger than interatomic distances, though still smaller than the correlation range. As will be shown below, for some of the quantities there is no large contribution owing to the critical fluctuations—the main contribution is from distances of the interatomic order. From the point of view of the theory to be given here, this means that the calculation of these quantities is outside the framework of their applicability. However, though they do not show anomalous behavior in the critical region, as functions of the temperature they may have singularities in their derivatives. The appearance of such singularities is due to the mechanism described in [5].

1. THE METHOD FOR STUDYING THE CRITICAL FLUCTUATIONS

As the macroscopic characteristics of the system we introduce the following densities of conserved quantities, which have meaning also outside hydrodynamics:

$$\frac{1}{V(r)} = \rho(r) = \sum_i \delta(r - r_i), \quad j(r) = \sum_i p_i \delta(r - r_i),$$

$$H(r) = \frac{j^2(r)}{2\rho} + e(r)\rho(r).$$

Here e is the internal energy, and the summation is taken over all particles in a small macroscopic volume. Hereafter all densities will be used in the form of their Fourier components:

$$x(k, \omega) = \int \int x(r, t) e^{i(kr - \omega t)} dr dt.$$

We now define a linear combination of the variations of these quantities:

$$\delta T(k, \omega) = -\frac{1}{C_V(k, \omega)[T(\partial P/\partial T)_V - P]} \delta V(k, \omega) + \frac{1}{C_V(k, \omega)} \delta e(k, \omega),$$

which is such that its equal-time correlation with ρ is equal to zero, and the autocorrelator is

$$\langle TT \rangle_k = T^2 / C_V(k, 0).$$

We shall use T as a local “temperature.”¹⁾ We assume that the equations for these densities are

$$\frac{dj(r)}{dt} = -\nabla \sigma(r) - \nabla \sigma^{ex}(r), \quad (1.1)$$

$$\dot{\rho}(r) = -\nabla j(r), \quad (1.2)$$

$$H(r) = -\nabla J_H(r) + \nabla q^{ex}(r). \quad (1.3)$$

Here σ^{ex} and q^{ex} are external forces which reestablish the actual temperature equilibrium in the system. For a system with $|j| = 0$ the diagonal element $\sigma_{\gamma\gamma}(r)$ of the stress tensor is the “pressure” P . Moreover, $J_H(r) = \sigma \cdot j/\rho - q$, where q is the energy flux caused by the gradient of the “temperature” T .

For the complete description of the system we must adjoin to (1.1)–(1.3) an equation of state. It is assumed that before the averaging over the fluctuations there exists a region of applicability of a theory of the type of the Landau self-consistent field.^[6] Then, assuming that the spatial dispersion of the compressibility is of the form corresponding to the theory of Ornstein and Zernike, we have

$$\begin{aligned} \delta P &= \left(\frac{\partial P}{\partial \rho} \right)_T (1 + k^2 r_C^2) \delta \rho + \left(\frac{\partial P}{\partial T} \right)_\rho \delta T + \frac{1}{2!} \left(\frac{\partial^2 P}{\partial \rho^2} \right)_T (\delta \rho)^2 \\ &+ \frac{\partial}{\partial \rho} \left[\left(\frac{\partial P}{\partial T} \right)_\rho \right]_T \delta \rho \delta T + \frac{1}{2!} \left(\frac{\partial^2 P}{\partial T^2} \right)_\rho (\delta T)^2 + \frac{1}{3!} \left(\frac{\partial^3 P}{\partial \rho^3} \right)_T (\delta \rho)^3 \\ &+ \frac{1}{2} \frac{\partial}{\partial T} \left[\left(\frac{\partial^2 P}{\partial \rho^2} \right)_T \right]_\rho (\delta \rho)^2 (\delta T) + \frac{1}{2} \frac{\partial}{\partial \rho} \left[\left(\frac{\partial^2 P}{\partial T^2} \right)_\rho \right]_\rho \delta \rho (\delta T)^2 \\ &+ \frac{1}{3!} \left(\frac{\partial^3 P}{\partial T^3} \right)_\rho (\delta T)^3, \end{aligned}$$

where r_C is a quantity of the order of the correlation range. As generalized forces and coordinates we take

$$F_n = -\frac{k_\alpha}{\omega_0} \sigma_{n\alpha}^{ex}(k, \omega), \quad x_i = j_i(k, \omega),$$

$$f = \frac{k_\alpha}{\omega T_0} q_\alpha^{ex}(k, \omega), \quad y = T(k, \omega).$$

The expressions for the corresponding generalized susceptibilities are given in the Appendix.

As an illustration of the method of solution of (1.1)–(1.3) let us consider the simple model of a nonlinear oscillator with friction. The equation of motion in the ω representation is of the form

$$\begin{aligned} (\omega_0^2 - \omega^2 - i\omega\gamma)x &= A \int x(\omega - \omega')x(\omega') d\omega' \\ &+ B \int \int x(\omega - \omega' - \omega'')x(\omega')x(\omega'') d\omega' d\omega'' + f^{ex}(\omega). \end{aligned}$$

Regarding the nonlinearities as small corrections, we get in first approximation

$$x_1(\omega) = \alpha_1(\omega)f^{ex}(\omega),$$

where $\alpha_1(\omega) = [\omega_0^2 - \omega^2 - i\omega\gamma]^{-1}$ is a generalized susceptibility. In higher orders of perturbation theory $x(\omega)$ will be a complicated function of f^{ex} . We define the total mobility $\alpha(\omega)$ as the variational derivative $\langle \delta x(\omega) / \delta f^{ex}(\omega) \rangle$. Here the averaging is over the ensemble of random forces. The ensemble is assumed to be such that averages calculated with it are equal to the thermodynamic averages. Selecting the strongly connected diagrams from the resulting series, we can write the analog of the Dyson equation $\alpha(\omega) = \alpha_1(\omega) \times [1 - \Sigma_1(\omega)]$ where $\Sigma_1(\omega)$ is the mass operator. The fluctuation-dissipation theorem enables us to set up a connection between the Fourier components of the total correlation functions of the fluctuating quantities and the linear responses of the system to the external actions that could cause particular fluctuations:

$$\langle xx \rangle_\omega = -\frac{kT}{2\pi\omega} \text{Im } \alpha(\omega).$$

Analogous relations must also be imposed on the higher moments of the field variables. These conditions, which

¹⁾ Here $C_V(0, 0)$, $(\partial P/\partial T)_V$, etc., are empirical constants. Only far from the critical point, where the nonlinearities in (1.1)–(1.3) can be neglected, do they have the meaning of the ordinary thermo-dynamic derivatives.

are equivalent to the requirement that the system be in thermal equilibrium, completely fix the ensemble of the forces $f^{\text{ex}}(\omega)$, which in general is not a Gaussian one. Sufficiently far from the critical point, however, where the expansion parameters of the total vertex functions are still small, the spectrum of the random forces can be regarded as Gaussian, and $\Sigma_1(\omega)$ will have the following form:

$$\Sigma_1(\omega) = \frac{1}{\omega} \left[4 \text{---} + j \text{---} + \text{---} \right] \quad (1.4)$$

Here the point and shaded circle denote vertex functions of types A and B,^[7] and a wavy line is a total correlation function $\langle xx \rangle_\omega$:

$$\text{wavy line} = \text{---} \Sigma_2(\omega) \text{---}, \quad (1.5)$$

where $-(kT/2\pi\omega) \text{Im } \Sigma_1(\omega) = \Sigma_2(\omega)$. Thus we find that $\alpha^{-1}(\omega) = \Omega_0^2 - \omega^2 + i\omega\Gamma$, where $\Omega_0^2 = \omega_0^2 - \text{Re } \Sigma_1(\omega)$ and $\Gamma = \gamma + (2\pi) \times (kT)^{-1} \Sigma_2(\omega)$.

We shall further consider a concrete liquid-gas system described by Eqs. (1.1)–(1.3).

2. THE CORRECTIONS TO THE KINETIC COEFFICIENTS. DISCUSSION OF RESULTS

As is well known, the poles of the Green's functions determine the spectrum of the elementary excitations, and thus in our case give a complete spectral description of the thermal fluctuations. In the critical region there are three characteristic lengths in a liquid: the isothermal correlation range $r_{CT} \sim \text{const} \cdot C_T^{-1}$, the adiabatic correlation range $r_{CS} \sim \text{const} \cdot C_S^{-1}$, and the interatomic distance $a \sim 10^{-7} \text{ cm}$. Accordingly near the critical point the characteristic times associated with the relaxation of individual formations or with undamped oscillations can be classified in the following way: a) thermal-flux times $\omega_1^{-1} \sim -\rho_0 C_P r_{CT}^2 / \kappa$; b) sound waves $\omega_2^{-1} \sim -\rho_0 r_{CS}^2 / \lambda$, $\omega_3^{-1} \sim r_{CS} / C_S$; c) times associated with the establishing of equilibrium in momentum space in a small macroscopic volume of size $a \sim 10^{-7} \text{ cm}$: $\omega_4^{-1} \sim 10^{-12} \text{ sec}$. Divergences in the kinetic coefficients, if there are any, are necessarily caused by slowly relaxing formations, including interferences of all possible kinetic processes with large wavelengths. The high-frequency fluctuation field, on the other hand, leads simply to a shift of the position of the critical point.

Using (1.1)–(1.3) and the estimates for the susceptibilities: $\alpha_{ij}^{(1)}(\omega_1, \mathbf{k}) \sim k^4$, $\alpha_i^{(1)}(\omega_1, \mathbf{k}) \sim k^3$, $\alpha^{(1)}(\omega_1, \mathbf{k}) \sim \alpha^{(1)}(\omega_2, \mathbf{k}) \sim k^2$, $\alpha_{ij}^{(1)}(\omega_2, \mathbf{k}) \sim \text{const}$, and $\alpha_i^{(1)}(\omega_2, \mathbf{k}) \sim k$, we can easily see that all the nonlinearities except those in the equation of state lead to a shift of the transition point. Consequently we can assume that the "bare" thermodynamic quantities are regular functions of the temperature in the critical region. The main contributions to the corrections to the kinetic coefficients from these nonlinearities comes from a region of \mathbf{k} and ω associated with interatomic distances. We can write, for example, some characteristic terms of this sort, taking the complete vertex functions as the bare quantities:

$$\Delta\eta_a \sim \Delta\xi_a \sim \frac{1}{\rho_0} \int \int \alpha_y(-\mathbf{k}) \frac{k_y}{\omega} \langle \rho S \rangle_{\mathbf{k}, \omega} d\mathbf{k} d\omega, \quad (2.1)$$

$$\Delta\kappa_a \sim \rho_0(kT)^{-1} \int \int [\langle ST \rangle_{-\mathbf{k}, -\omega} \langle Sp \rangle_{\mathbf{k}, \omega} + \langle \rho T \rangle_{-\mathbf{k}, -\omega} \langle SS \rangle_{\mathbf{k}, \omega}] \frac{\omega^2}{k^2} dk d\omega. \quad (2.2)$$

According to the fluctuation-dissipation theorem, the complete correlation functions are of the form

$$\begin{aligned} \langle j_B j_Y \rangle_{\mathbf{k}, \omega} &= -\frac{kT}{2\pi\omega} (2\pi)^3 \text{Im } a_{BY}(\mathbf{k}, \omega), \\ \langle j_B T \rangle_{\mathbf{k}, \omega} &= -\frac{kT}{2\pi\omega} (2\pi)^3 \text{Im } a_B(\mathbf{k}, \omega), \\ \langle TT \rangle_{\mathbf{k}, \omega} &= -\frac{kT}{2\pi\omega} (2\pi)^3 \text{Im } \alpha(\mathbf{k}, \omega). \end{aligned} \quad (2.3)$$

Using (2.3), we find from (2.1) and (2.2) that only the temperature derivatives of η , ξ , and κ can have singularities in the critical region. Inclusion of the corrections to the vertex functions does not change the behavior of $\Delta\eta$, $\Delta\xi$, and $\Delta\kappa$. The result derived by Fixman^[2] can be reconstructed on the assumption of "frozen" isothermal fluctuations. In fact, in this case we have

$$\begin{aligned} \Delta\eta &\sim kT \left(\frac{\partial P}{\partial T} \right)_P \frac{T\lambda}{\omega\rho_0^2} \int \int \frac{\omega^2}{k^4} \\ &\times \frac{C_T^2(k) - \omega^2/k^2}{[(C_T^2(k) - \omega^2/k^2)^2 + \omega^2\lambda^2/\rho_0^2]^2} dk d\omega \sim \text{const} \cdot r_{ex} \end{aligned}$$

This assumption, however, is justified only for $\lambda_0 C_P / \kappa \ll 1$. In our case, meanwhile, we must expect that the opposite inequality will hold.

In our treatment power-law estimates and divergences can be associated only with the scale r_{CT} . In the case when the contribution to the quantities in question is mainly due to small distances (of the order of interatomic distances), the hydrodynamic treatment is not correct, and in our approximation we must regard such a quantity as a constant. We note that in general corrections of the order of the value of the correlators at interatomic distances can have singularities in their temperature derivatives. The point is that the mean energy, density, and so on, are expressed in terms of such correlators. Therefore those of the kinetic coefficients which, while remaining finite, contain corrections of the type described above can show a behavior analogous, for example, to that of the resistance of a ferromagnetic substance at the Curie point, which was studied in^[5].

Let us now consider the correlations, associated with r_{CT} , to the volume viscosity λ and the thermal conductivity κ . It is obvious from (1.1)–(1.3) that the main contribution comes from the interactions described by the nonlinearities in the equation of state. We shall assume that the expansion parameters in the vertex functions are still small, so that in the diagrams for Σ we can confine ourselves to the bare vertices. Furthermore it is obvious that the behavior of $\Delta\lambda$ and $\Delta\kappa$ is mainly determined by those diagrams of Σ which contain only j -lines. For simplicity we can go over to the variables T and ρ . Then

$$\Delta\lambda \approx \rho_0^2 (kT)^{-1} (2\pi)^{-2} \left[4 \text{---} + j \text{---} + \text{---} \right], \quad (2.4)$$

where a point is $(\partial^2 P / \partial \rho^2)_T \rho_0^{-1}$, a circle with a point is $(\partial^3 P / \partial \rho^3)_T \rho_0^{-1}$, and a wavy line is $\langle \rho \rho \rangle_{\mathbf{k}, \omega}$.

As is well known, in phase transitions of the second kind the density and the thermodynamic functions are continuous. There are jumps in the derivatives of these

functions with respect to the physical parameters. Therefore near the transition point, in the expression for the expansion of the free energy F in powers of the small parameter $\delta V = \delta(1/\rho)$,

$$\delta F = -\frac{1}{2!} \left(\frac{\partial P}{\partial V} \right)_T (\delta V)^2 - \frac{1}{3!} \left(\frac{\partial^2 P}{\partial V^2} \right)_T (\delta V)^3 - \frac{1}{4!} \left(\frac{\partial^3 P}{\partial V^3} \right)_T (\delta V)^4,$$

the third-order terms are always smaller than the fourth-order terms. Otherwise there can be a jump of the particle density. Accordingly, near the transition point we can confine ourselves to the contributions from cubic nonlinearities in the equation of state. We then have

$$\lambda \approx \lambda_0 + \Delta\lambda_a + \frac{C_p C_T^{10} r_{CT}^8}{\kappa (\partial^3 P / \partial \rho^3) r^2 (kT)^2} \gamma^4 \quad (1 + \gamma^2)^{-1}.$$

Here C_p , C_T^2 , κ , and λ are macroscopically observable quantities, and γ is the expansion parameter,

$$\gamma \approx \text{wavy line} \circlearrowleft.$$

Far from the critical point, where the random-force ensemble can be taken as Gaussian, C_T^2 and r_{CT}^{-2} behave like $\tau = (T - T_C)/T_C$. Right at the transition point, where the irreducible parts of the correlations are important, C_T^{-2} and r_{CT}^2 must have a singularity of the type $\tau^{-4/3}$. Thus the value of τ at which γ comes to be of the order of unity is the boundary of the region of applicability of the calculation. How close it is to the critical point depends on the properties of the system. In fact,

$$\gamma \sim \frac{v^2}{c^2} \frac{a^3}{r^3} \tau^{-4},$$

where $v^2 a^3 \sim kT/\rho_0$, c is the velocity of sound far from the transition point, and r is the range of the interaction far from the transition point. Accordingly, the larger the range of the interaction in the system, the broader the region of applicability of our estimates. If we assume that $c \sim 10^5$ cm/sec, $T_C \sim 3 \times 10^{20}$ K, $\lambda \sim 10^{-1}$ g cm⁻¹ sec⁻¹, $\rho_0 \sim 1$ g·cm⁻³, and $r \sim \kappa/C_p C_T \rho_0 \sim 10^{-6}$ cm, then for $\tau \sim 10^5$ we have $\gamma \sim 10^{-3.5}$ and $\Delta\lambda \sim \lambda_0 \times 10^{-2}$. But because the estimates are sensitive to the numerical values of the coefficients they must be specially worked out for each concrete system. For κ we have, in analogy with (2.4)

$$\begin{aligned} \kappa - \kappa_0 &\approx \Delta\kappa_a + \frac{\partial}{\partial T} \left[\left(\frac{\partial^2 P}{\partial \rho^2} \right)_T \right] \frac{(\partial^2 S / \partial \rho^2)_T}{\rho_0 k} \\ &\int \int \int \int \frac{\omega^2}{k^2} \langle \rho^2 \rangle_{-k-q, -\omega-\Omega} \langle \rho^2 \rangle_{k, \omega} \langle \rho^2 \rangle_{q, \Omega} dk d\Omega d\omega d\Omega. \end{aligned} \quad (2.5)$$

It is not hard to see from (2.3) and (2.5) that κ is finite for $T \rightarrow T_C$ as long as $\gamma < 1$. The corrections to the

shear viscosity are the same as before, since the nonlinearities in the equation of state merely renormalize the vertex function. For the critical point of a solution the consideration of such a mechanism of increase of the viscosity basically leads to the replacement of $\kappa/\rho_0 C_p$ by the diffusion coefficient $D = (\alpha/\rho_0) \times (\partial \mu / \partial C)_T p$. A more detailed treatment of this case will be given elsewhere. The work of Kadanoff differs from ours through the use of quadratic nonlinearities in the equation of state to describe the critical fluctuations, and evidently also in a different definition of the kinetic coefficients.

APPENDIX

$$\begin{aligned} a_{ij}^\perp &= -i\omega\rho_0^2(\eta k^2 - i\omega\rho_0)^{-1} \left(\delta_{ij} - \frac{k_i k_j}{k^2} \right), \\ a_{ij}^\parallel &= -\omega^2\rho_0 \left[C^2(k, \omega) - \frac{i\omega\lambda}{\rho_0} - \frac{\omega^2}{k^2} \right]^{-1} \frac{k_i k_j}{k^4}, \\ a_i &= i \left(\frac{\partial P}{\partial T} \right)_\rho \frac{\omega^2 T_0}{\kappa k^4} \left(C_T^2 - \frac{i\omega\lambda}{\rho_0} - \frac{\omega^2}{k^2} \right)^{-1} \left(1 - \frac{k_i^2}{k^2} \right)^{-1} k_i, \\ a &= -\frac{i\omega T_0}{\kappa k^2} \left(1 - \frac{k_i^2}{k^2} \right)^{-1} \end{aligned}$$

Here $C^{-2}(k, \omega)$ is the "compressibility" at frequency ω :

$$\begin{aligned} C^2(k, \omega) &= \left(C_T^2 - \frac{i\omega\lambda}{\rho_0} - \frac{\omega^2}{k^2} \right) \left(1 - \frac{k_i^2}{k^2} \right) \left(1 - \frac{k_x^2}{k^2} \right)^{-1} + \frac{i\omega\lambda}{\rho_0} + \frac{\omega^2}{k^2}, \\ \frac{k_i^2}{k^2} &= \frac{i\omega\rho_0 C_V}{\kappa k^2} \left(C_S^2 - \frac{i\omega\lambda}{\rho_0} - \frac{\omega^2}{k^2} \right) \left(C_T^2 - \frac{i\omega\lambda}{\rho_0} - \frac{\omega^2}{k^2} \right)^{-1}, \\ \frac{k_x^2}{k^2} &= \frac{i\omega\rho_0 C_V}{\kappa k^2}, \quad \lambda = \frac{4}{3}\eta + \xi, \end{aligned}$$

α_{ij}^\parallel denotes the longitudinal part of the generalized susceptibility, and α_{ij}^\perp the transverse part.

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