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ALTERNATIVE FORM OF GENERALIZED TRANSFER LAWS

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A reduced form of phenomenological laws involving a single (resulting) driving force is proposed that makes it possible to describe superposition effects by a smaller number of kinetic parameters.

In his attempts to formulate laws of transfer of heat, matter, charge, etc. in the most general form including the description of effects of superposition (interrelationship) of irreversible processes of different types, Onsager postulated that the generalized rate of a process (flow J_i) is linear with all thermodynamic forces X_j acting in the system [1]:

$$J_i = \sum_{j=1}^{n} L_{ij} X_j . \tag{1}$$

Here L_{ij} are constant phenomenological coefficients forming, provided that the forces X_i and X_j belong to one and the same type of time functions (even or odd), a symmetric matrix

$$L_{ij} = L_{ji} . \tag{2}$$

For more than half a century this postulate was undisputable and was reproduced in all handbooks on thermodynamics of irreverible processes making an only proviso concerning the Curie symmetry principle stating that superposition is possible only for processes of one and the same (or even) tensor rank and form [2, 3]. In doing this, the question on the interrelationship of process rates found, according to [1], as time derivatives of independent parameters of the system state* has not even been raised. Nobody has ever been embarassed by the fact that this postulate contradicted centures-old foundations of mechanics according to which a single (resulting) force corresponds to every independent process (translation, acceleration, establishment of equilibrium, etc.) that terminates when the force vanishes. This was also supported by equations of anisotropic heat transfer or electric conductivity which served, according to Onsager's own confession, as prototypes of his phenomenological laws.**

In order to demonstrate that Onsager's theory is not free from internal contradictions, we present laws (1) in the form

$$J_i = \sum_{j=1}^{n} L_{ij} X_j = \sum_{j=1}^{n} J_{ij} , \tag{3}$$

where $J_{ij} = L_{ij}X_j$ are components of the flow J_i reflecting superposition effects.

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* In this case, interrelationship of flows can emerge only as a result of additional bounds imposed that cannot be postulated *a priori*.

** In this case, X_j ($j = 1, 2, 3$) are components of a single force – the vector of the negative temperature gradient – ∇T or electric field strength **E**.

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Since, according to (1), $L_{ij} = (\partial J_i/\partial X_j)_{X_i}$ in linear systems, and $X_j = (J_j/L_{jj})_{X_i = 0}$, simultaneous consideration of these relationships shows that a certain relationship exists at $X_i = 0$ between diagonal L_{jj} and off-diagonal coefficients L_{ij}:

$$L_{ij} = L_{jj} (\partial J_i/\partial J_j)_{X_i=0} \,. \tag{4}$$

It follows from this that, if the flows J_i and J_j are not interrelated, e.g., their variations are independent, coefficients L_{ij} become zero along with the derivatives $(\partial J_i/\partial J_j)_{X_i=0}$ when $X_i = 0$. This means that the reciprocity relationships are trivially satisfied: $L_{ij} = L_{ji} = 0$, as expected. In other words, in the absence of the driving force X_i corresponding to the flow J_i, no other force can induce this flow if the latter is indeed independent.

There are also other grounds to doubt the adequateness of Onsager's postulate. According to Eq. (1), numerous thermomechanical, thermoelectrical, thermodiffusional, and other effects are explained by an interrelationship between the rates of irreversible processes taking place within the same space regions, i.e., by superposition of the flows J_i and J_j [2, 3] and their mutual entrainment [4]. At the same time, it is well known that the effects mentioned reach their maxima in so-called stationary states in which nonfixed flows vanish and therefore cannot superimpose with other flows. For example, in electrolyte solutions, in which electrical conductivity and diffusion phenomena take place, the difference of electric potentials (the Kwincke effect) is maximum when the current is turned off [3]. The same takes place in the case of the Soret effect consisting of the appearance of a concentration gradient of the k-th substance in an originally homogeneous medium when a temperature gradient is maintained in this medium, and the specified concentration gradient reaches its maximum when diffusion flows vanish [3]. Therefore, the reason for the emergence of these superposition effects should be sought not in the interaction of flows but rather in addition of different forces, as this takes place in mechanics.

This means that, in the absence of additional bonds imposed, Onsager's postulate should be replaced by a proposition according to which *the generalized rate of an irreversible process (the flow J_i) depends only on the components X_{ij} of a single (resulting) force $X_i = \Sigma_j X_{ij}$*. This proposition corresponds to kinetic equations of the following form:

$$J_i = L_{ii}(X_i)\, X_i = L_{ii}(X_i) \sum_{j=1}^{n} X_{ij} \,, \tag{5}$$

where $L_{ii}(X_i)$ are kinetic coefficients that, in contrast to those from (1), can be arbitrary functions of thermostatic parameters (temperature T, pressure p, concentrations of k-th substances, etc.) and of forces X_i. The basic distinction of Eqs. (5), except for their nonlinearity, consists in the fact that each of the equations includes the only kinetic coefficient $L_{ii}(X_i)$, whereas components X_{ij} of the force X_i are determined solely by thermodynamic properties of the system.

The possibility of transforming phenomenological laws (1) into the diagonal form (with a single thermodynamic force) does not contradict the thermodynamics of irreversible processes, according to which the necessary and sufficient condition of doing this would be linearity of the laws and symmetry of the matrix of phenomenological coefficients in their notation [2, 3]. However, this procedure carried out in the thermodynamics of irreversible processes by a linear transformation of flows and forces has no advantages, since it does not reduce the number of indepedent phenomenological coefficients in original equations (1) and does not remove constraints inherent in the linear thermodynamics of irreversible processes. It would be quite another matter if we would find a possibility to immediately find an alternative form (5) of the Fourier, Ohm, Darey, Fick, Newton, and other laws that would contain a generalized driving force and would allow description of the same superposition effects employing a smaller number of kinetic coefficients without resorting to the assumption that they are constant. In Onsager's theory and in the existing thermodynamics of irreversible processes [2, 3], finding this force is hardly possible by construction of these theories. For example, it is well known that the magnetic component of the Lorentz force $\mathrm{F_L}$ responsible for thermogalvanomagnetic effects is normal to the direction of the electric current and therefore does not contribute to the entropy production σ_s [2, 3]. Therefore, the formalism of Onsager's theory, defining forces as derivatives of σ_s over the corresponding variables of state, is inapplicable in this case. This also concerns graviational, centrifugal, and Coriolis forces responsible for reversible effects. Further, the formalism of these theories does not fully take into account the *conditions of uniqueness* of the particular processes under investigation. Thus, when writing entropy balance equations for finding flows and driving forces of different processes, the thermodynamics of irreversible processes invariably uses one and the same Gibbs relationship in its classical form

$$du = Tds - pdv + \sum_{k=1}^{K} \mu_k \, dc_k \, , \tag{6}$$

which assumes the specific entropy s and the specific volume v of the system to be independent variables (coordinates of heat transfer and bulk deformation processes). At the same time, the use of the chemical potential $\mu_k \equiv (\partial u/\partial c_k)_{s,v}$ under conditions different from s, v = const (e.g., in the presence of diffusion when s and v necessarily change as a result of changes in the system's composition) leads to the fact that the forces $\nabla\mu_k$, $\nabla\mu_k/T$, $\Delta\mu_k$, and $\Delta\mu_k/T$ found based on (6) do not include the transfer heat and therefore are insufficient for describing thermal diffusion and thermal osmosis without involving other forces, despite temperature and concentration gradients for all independent components necessary for this entry in $\nabla\mu_k$. By virtue of this, the assumption on the existence of a single driving force of an independent process could not emerge in the framework of thermodynamics of irreversible processes.

At the same time, one can easily assure oneself of the existence of resultant thermodynamic forces: it suffices to put the Gibbs relationship (6) in correspondence with the uniqueness conditions for each particular process. The first step on this way is removing the presently existing arbitrary approach to definitions of the concept of heat in open systems [2, 3], which would allow characterization of the heat transfer, work, energy, and mass transfer as independent processes inducing *special phenomenologically distinct changes in the system state irreducible to other types of changes.* This requirement is satisfied by the presently accepted definition of the heat Q and work W as a portion of the heat transfer not related to the matter transfer cross the system boundary [5]. In accordance with this, to find the elementary heat δQ and work δW in open systems, one should subtract from the total change of their entropy dS and volume dV the portions of the former $\Sigma_k \bar{s}_k dn_k$ and $\Sigma_k \bar{v}_k dn_k$ due to the transport of k-th components across systems boundaries, where under $\bar{s}_k$ one means the entropy *de facto introduced into the system by a mole of the k-th substance*, i.e., the quantity called in the literature (following Agard [3]) "transported entropy," and under $\bar{v}_k$ one means the volume *de facto* introduced into the system by a mole of the k-th substance:

$$\delta Q = TdS - \sum_{k=1}^{K} \bar{s}_k \, dn_k \, ; \;\; \delta W = pdV - \sum_{k=1}^{K} \bar{v}_k \, dn_k \, , \tag{7}$$

$$\delta q = Tds - \sum_{k=1}^{K} \bar{s}_k \, dc_k \, ; \;\; \delta w = pdv - \sum_{k=1}^{K} \bar{v}_k \, dc_k \, , \tag{8}$$

where c_k is the molar fraction of the k-th component, and n_k is the number of moles of the component.

By taking into account that $s = \Sigma_k s_k c_k$, upon adding and subtracting the term $\Sigma_k T s_k dc_k$ from the right-hand side of Eq. (7), these equations can be written as follows:

$$\delta q = Tds - \sum_{k=1}^{K} \bar{s}_k \, dc_k = \sum_{k=1}^{K} Tc_k \, ds_k - \sum_{k=1}^{K} T \, (\bar{s}_k - s_k) \, dc_k \, , \tag{9}$$

$$\delta w = pdv - \sum_{k=1}^{K} p\bar{v}_k \, dc_k = \sum_{k=1}^{K} pc_k \, dv_k - \sum_{k=1}^{K} p \, (\bar{v}_k - v_k) \, dc_k \, . \tag{10}$$

The difference between the molar entropy $\bar{s}_k$ of the transferred substance and the actual increase in the entropy of the mixture s_k upon introducing a mole of this substance at constant temperature and pressure,* entering the second term of expression (9), characterizes thermal effects accompanying introducing the k-th substance. These effects were first considered already within the framework of the pseudothermostatic theory by Eastman (1926) and Wagner (1929) who referred to the difference $\bar{s}_k - s_k$ as the entropy of transfer s_k^*, and called the corresponding quantity

$$q_k^* = T(\bar{s}_k - s_k) = Ts_k^* \tag{11}$$

the heat of transfer of a mole of the k-th substance.** According to Eastman and Wagner, q_k^* is the heat that must be supplied to a system from the environment (or removed from the system) to maintain its temperature at a constant value upon introducing a mole of the k-th substance into the system. This treatment of the heat of transfer is close to its classical understanding. The thermodynamics of irreversible processes introduces the quantity q_k^* as one of the empirical coefficients L_{ij} that, owing to the reciprocity relationships (2), acquires the meaning of the energy transported by the flow of the k-th substance in the absence of a temperature gradient [2, 3]. In both cases, the heat of transfer appears to be inextricably entwined with the mass transfer, so that the former should be excluded from the definition of the process heat in its classical definition. This is realized in Eq. (9), which thus provides a unique definition of the process heat in open systems. In a similar manner, in Eq. (10) the expansion work in open systems is determined by the change in volumes of components minus volume effects $w^* = p(\bar{v}_k - v_k) = pv_k^*$ due to introducing k-th substances. The above definition of the heat and work changes the expression for the energy and mass transfer defined as a residual term in the equation for the energy balance in open systems. Naturally, this residual term appears to be different depending on conditions under which the process takes place. Thus, for diffusion under conditions of a constant volume (V, $v = \text{const}$) in the absence of heat transfer in its classical meaning (δQ, $\delta q = 0$), substitution of expressions (9) and (10) into the Gibbs relationship (6) transforms it into the form

$$du = \delta q - \delta w + \sum_{k=1}^{K} \psi_k^{\mathrm{d}}\, dc_k, \tag{12}$$

where $\psi_k^{\mathrm{d}} = \mu_k + T\bar{s}_k - p\bar{v}_k$ is the potential of the k-th component which we earlier called the diffusional potential [6] due to the fact that the above uniqueness conditions are characteristic of the diffusion process. The fact that the heat of transfer, excluded from the heat transfer expression, entered into the other part of the equation of the energy balance (the expression for the energy and mass transfer) is quite reasonable, since the energy balance should be satisfied in this case. Thus, under conditions of diffusion aggravated by thermal and bulk effects, the chemical potential transforms into the diffusional potential.

Now we consider the process of osmosis of the k-th component to a closed space ($V = \text{const}$) in the absence of heat transfer($\delta Q = 0$). In this case, the Gibbs relationship is as follows:

$$du = \delta q - \delta w + \sum_{k=1}^{K} \psi_k^{\mathrm{os}}\, dc_k, \tag{13}$$

where $\psi_k^{\mathrm{os}} = \mu_k + T\bar{s}_k$ is the potential of the k-th component, which we earlier called the osmotic potential [6]. By using the well-known relationship $\mu_k = h_k - Ts_k$, one can easily show that the osmotic potential of the component equals the sum of its partial enthalpy h_k and the heat of transfer q_k^* of this component

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* This quantity, as is well known, can also be negative (as opposed to s_k).

** Other definitions of this quantity can be found in the literature. Thus, Haase [3] uses the term "entropy of transfer" for $\bar{s}_k$. We adhere to the most widely used definition of the entropy of transfer, implying that this quantity is defined as $s_k^* = q_k^*/T$.

$$\psi_k^{\text{os}} = h_k + T\,(s_k - s_k) = h_k + q_k^* . \tag{14}$$

Since the potential of the component is determined exclusively by the thermodynamic state of the system, q_k^* and w_k^* should be treated as new thermodynamic variables. Their magnitudes and signs depend on the differences $\bar{s}_k - s_k$ and $\bar{v}_k - v_k$, i.e., on thermodynamic properties of the region from which (into which) the k-th component is introduced. This corresponds to the existing experimental data [2, 3].*

One can easily notice that for noninteracting components ($\bar{s}_k = s_k$ and $\bar{h}_k = h_k$), the osmotic potential acquires the sense of the partial molar enthalpy of the transferred component h_k, and in the more special case of a single-component system it has the sense of its molar enthalpy h. Thus, the potential of the component changes not only as a result of a more precise definition of the concept of the heat transfer and the work of expansion in open systems, but also depending on thermodynamic properties of a particular system.

Owing to including the heat and work of transfer into the expression for the potential of the components, the generalized diffusion equation has the following form [7]:

$$\mathbf{J}_k = -L_{kk}\nabla\psi_k^{\text{d}} = -L_{kk}\left(\sum_{l=2}^{K}\mu_{kl}\nabla c_l + s_k^*\nabla T - v_k^*\nabla p\right), \tag{15}$$

where μ_{kl} is the shorthand notation of the derivative $\partial\mu_k/\partial c_l$. The resulting vector force $\mathbf{X}_k = -\nabla\psi_k^{\text{d}}$ includes $K + 1$ components, each of which has one and the same tensor rank, i.e., *a fortiori* satisfies the Curie principle. One of these, $\mathbf{X}_{k\text{d}} = -\Sigma_k\,\mu_{kl}\Delta c_l$ is responsible for the conventional (concentrational) diffusion, anothher one, $\mathbf{X}_{kt} = -s_k^*\nabla T$, is responsible for thermal diffusion, and another one, $\mathbf{X}_{k\text{b}} = v_k^*\nabla p$, is responsible for barodiffusion and sedimentation. In the particular case of isobaric-isothermic diffusion (∇p, ∇T=0), Eq. (15) assumes the form of the generalized Fick diffusion law [8]:

$$\mathbf{J}_k = -L_{kk}\nabla\mu_k = -\sum_{l=2}^{K} D_{kl}\nabla c_l , \tag{16}$$

where $D_{kl} = L_{kk}\mu_{kl}$ are generalized diffusion coefficients. This expression differs from the form of generalized diffusion laws adopted in the thermodynamics of irreversible processes (which includes gradients of chemical potentials of all independent components of the system [3]) by the substantially simpler form of presentation of diffusion coefficients whose consequences have experimental substantiations [8]. One of these consequences is the simple diagonal D_{kk} and off-diagonal D_{kl} diffusion coefficients

$$D_{kk}/\mu_{kk} = D_{kl}/\mu_{kl} = L_{kk} , \tag{17}$$

which makes it possible to study effects of kinetic L_{kk} and thermodynamic μ_{kl} diffusion factors separately [8]. By using the condition according to which the flow $\mathbf{J}_k$ and thermodynamic force $\mathbf{X}_k$ vanish simultaneously, one can obtain the relationship

$$\sum_{l=2}^{K} D_{kl}\nabla c_l = 0 , \tag{18}$$

explaining the phenomenon of the "upward" diffusion ($\nabla c_k > 0$ when $\nabla c_{l\neq k} < 0$) and alleviating the finding of hardly measurable off-diagonal diffusion coefficients [8]. In the case of nonisothermal diffusion in a system in the state of mechanical equilibrium ($\nabla p = 0$), expression (15) makes it possible to describe the stationary Soret and Dufure effects by presenting them as a consequence of the mutual cancellation of the $\mathbf{X}_{k\text{d}}$ and $\mathbf{X}_{kt}$ components [7].

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* The fact that the entropy or the density of the substance in free space fundamentally differs from those in a membrane is evidenced, in particular, by the disappearance of the Knudsen effect with increasing capillary diameter [2, 3].

By passing in (15) to bulk flow $\mathbf{J}_v = v_k \mathbf{J}_k$ and by dividing $\mathbf{X}_k$ by v_k to retain the dimensionality of the product of $\mathbf{J}_v$ and $\mathbf{X}_v = \mathbf{X}_k / v_k$, one can obtain the generalized Darcy filtration law [9]

$$\mathbf{J}_v = - D_v \, [\nabla p + (s_k^* / v_k^*) \, \nabla T + (\mu_{kk} / v_k^*) \, \nabla c_k] , \tag{19}$$

where $D_v = L_{kk} v_k^2$ is the filtration coefficient.

For systems containing charged components (conductors, electrolyte solutions, etc.), the chemical potentials μ_k, as is well known, transform into the electrochemical potential $\mu_k^* = \mu_k + e_k \varphi$, where e_k is the charge of a mole of the k-th component and φ is the electromotive potential of the region. Accordingly, the additional component $\mathbf{X}_{ke} = e_k \nabla \varphi$ appears in the expression for the driving force of transfer of the k-th component, which makes it possible to obtain, based on (15), a generalized Ohm's law for electrolytes upon passing to the electric current $\mathbf{J}_e = \Sigma_k e_k \mathbf{J}_k$ and by dividing the force $\mathbf{X}_k$ by e_k. In particular, when free electrons are the single charge-carrying component, the generalized Ohm's law assumes the form [9]:

$$\mathbf{J}_e = - \sigma \, [\nabla \varphi + (s_k^* / e_k) \, \nabla T + (v_k^* / e_k) \, \nabla p + (\mu_{kk} / e_k) \, \nabla c_k] , \tag{20}$$

where $\sigma = L_{kk} e_k^2$ is the electric conductivity of a membrane; e_k, s_k^*, v_k^*, μ_{kk}, and c_k are parameters of the electron gas. This equation makes it possible to describe *thermoelectric and electrokinetic* phenomena, in particular, to find expressions for stationary Peltier, Zeebeck, Reus, and Kwincke effects [9]. In a more general case when a conductor is placed into the magnetic field $\mathbf{B}$, the magnetic component of the Lorentz force $\mathbf{F}_\mathrm{L}$ is added to the resulting force, which results in an anisotropy of electric conductivity and induces the whole spectrum of thermomagnetic effects (the Tomson, Rigi–Leduke, Ettinshausen–Nernst, and other effects). These effects can also be explained in terms of superposition and mutual compensation of components of the resulting force [9].

In a similar manner, one can find numerous superposition effects in continuous (valve) systems separated by an immovable semipenetrable membrane. Under conditions of $V = \mathrm{const}$ typical for these systems, the driving force of transfer of the k-th substance is determined by a drop in the osmotic potential $\mathbf{X}_k = -\Delta \psi_k^{\mathrm{os}}$, so that the equation of transfer of this substance assumes the form [9]

$$\mathbf{J}_k = - L_{kk} \Delta \psi_k^{\mathrm{os}} = - L_{kk} \left(\sum_{l=2}^{K} \mu_{kl} \Delta c_l + s_k^* \Delta T + v_k \Delta p \right) . \tag{21}$$

One of the components of this force, $\mathbf{X}_{k\mathrm{con}} = -\Sigma_k \mu_{kl} \, \Delta c_l$, is responsible for the conventional osmosis, the other one, $\mathbf{X}_{kt} = -s_k^* \Delta T$, is responsible for the thermal osmosis, and the third one, $\mathbf{X}_{kr} = -v_k \Delta p$, is responsible for the reverse osmosis effect and development of the osmotic pressure. Mutual compensation of these forces makes it possible to immediately find analytical expressions for a group of so-called *thermomechanical* effects (the Feddersen, Knudsen, Allen–Johns, and Daunt–Mendelson effects [9]).

Finding the resulting force from the uniqueness conditions made it possible to propose an alternative method for investigation of irreversible processes [6], based on which we managed to obtain in [9] the overwhelming majority of superposition effects known in the thermodynamics of irreversible processes. The method does not require writing cumbersome entropy balance equations and does not need applying the Onsager–Kasimir reciprocity relationships. The practical significance of this method consists, first of all, in the possibility of finding stationary superposition effects in nonlinear systems where Onsager's reciprocity relationships break down. Indeed, in the stationary state ($\mathbf{X}_i = 0$) resulting from the mutual compensation of the components $\mathbf{X}_{ij}$ of the force $\mathbf{X}_i$, the character of dependences of the coefficients $L_{ii}(\mathbf{X}_i)$ on these forces in Eq. (5), e.g., the degree of their nonlinearity, is already of no significance.

On the other hand, application of the alternative form of the phenomenological laws leads to further reduction in the number of kinetic coefficients necessary for their formulation. This reduction is achieved as a result of additional relationships between diagonal and off-diagonal phenomenological coefficients which follow from (5) upon moving $L_{ii}(X_i)$ into the sum and comparing corresponding terms of the expression obtained with those from (1)

$$L_{ij} = L_{ii}\,(X_i)\,X_{ij}/X_j\,. \tag{22}$$

Expressions (17) are an example of these relationships. Since for n independent flows the number of relationships (22) equals $n(n-1)$, which is twice as large compared to the number of reciprocity relationships, Eqs. (5) make it possible to further reduce the number of kinetic coefficients from $n(n+1)/2$, as in thermodynamics of irreversible processes, to n [10]. In addition, this approach can constitute the basis for the novel method of finding nondescript thermodynamic quantities such as μ_{kl}, s_k^*, etc. by measuring the corresponding superposition effects [10]. At the same time, it becomes possible to express the effects specified exclusively in terms of thermodynamic variables and find $n(n-1)/2$ additional relationships between them. The Videman–Franz law is an example of these relationships [10]. All this emphasizes the heuristic value of finding the above-proposed alternative form of transfer laws.

NOTATION

u, h, specific internal energy and enthalpy of the system, J/kg; X_i, $\mathbf{X}_i$, scalar and vector thermodynamic force of the i-th process; J_i, $\mathbf{J}_i$,scalar and vector flows; δQ, δW, elementary amounts of heat and work, J; q^*, w^*, heat and work of transfer of a mole of the k-th substance, J/mole; T, p, absolute temperature (K) and pressure, Pa; V, v, total (m^3) and specific (m^3/kg) volumes of the system; S, s, complete (J/K) and specific (J/(kg·K)) entropies; v_k, s_k, partial molar volume (m^3/mole) and molar entropy (J/(mole·K)) of the k-th component; $\vec{v}_k^*$, $\vec{s}_k^*$, volume and entropy virtually introduced to the system by a mole of the k-th component; μ_k, c_k, chemical potential of a mole of the k-th component (J/mole) and its molar fraction (mole/mole); ψ_k^{d}, ψ_k^{os}, diffusional and osmotic potentials of a mole of the k-th substance, J/mole; φ, electric potential, V; σ_s, local entropy production rate, (W/(m^3·K)); $\mathbf{E}$, $\mathbf{B}$, electric (V/m) and magnetic (A/m) fields.

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