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A MAXIMUM ENTROPY ANSATZ FOR NONLINEAR RESPONSE THEORY

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Abstract:

An approximate nonlinear response theory is derived by maximizing the information theoretic entropy in the space of fluctuations $J(t)$ of the degree of freedom of interest, subject to constant energy dissipation in the external field. The resulting formalism expresses all cumulant averages of $J(t)$ in an external field in terms of equilibrium averages.

Linear response theory [1-7] is fundamental to much present day work in solid state physics, chemical physics, plasma physics, etc. Its usefulness rests in the fact that the response to an external field to first order is determined solely by an equilibrium autocorrelation function. The extension of linear response theory to include higher order terms was discussed already in the 1950's [6,8]. In contrast to the linear case, the higher order terms cannot be expressed in terms of equilibrium correlation functions of the degree of freedom under consideration [9]. The exact nonlinear response theory is complex and has found little application. There is also the question of the convergence of the formal expansion in the external field. Strong fields may induce e. g. phase transitions or self organization, information about which one does not expect to be hidden in the equilibrium fluctuations. Despite these reservations, one may ask whether a theory which predicts the nonlinear response from equilibrium fluctuations does exist, at least as a reasonable approximation applicable not too far from equilibrium. In an attempt to answer this question, this paper discusses and generalizes a little known ansatz for "non-linear response theory", due to Stratonovich [10]. The ansatz is here derived from the maximum entropy principle. Some simple examples are given and the validity of the ansatz is discussed. It is argued that the ansatz represents, in a sense, the simplest way a system may become nonlinear.

The degree of freedom of interest is denoted by Q , it may be microscopic or macroscopic. The underlying dynamics is assumed to be classical and Q is assumed to be invariant under time-reversal. Q couples to an external field E via a term $-EQ$ in the Hamiltonian. Only the case of a time-independent external

field will be considered. It is convenient to discuss response and fluctuations in terms of $J(t) \equiv \dot{Q}(t)$. In general, one is interested in not only the average $\langle J \rangle_E$ but also in the external field autocorrelation function and perhaps higher order averages. To calculate these quantities one needs to know the probability of any $J(t)$ -fluctuation in the external field, $P_E[J(t)]$. As mentioned above, even a complete knowledge of the equilibrium fluctuations, $P_0[J(t)]$, in general is not enough to determine $P_E[J(t)]$. The task of finding $P_E[J(t)]$ may be viewed as a missing information problem. In an external field there is a certain rate of energy dissipation, and no more than this and $P_0[J(t)]$ is known a priori. The maximum entropy formalism is a general method that, based on the insufficient information available, gives a least biased estimate of probabilities [11, 12]. In applications to a continuous problem a reference measure is needed. In the present case, in order to have a smooth transition to the zero field case this measure must be $P_0[J(t)]$. The maximum entropy requirement leads to [11, 12]

$$P_E[J(t)] = N^{-1} P_0[J(t)] \exp \left\{ -\lambda E \int_{-\infty}^{\infty} J(t) dt \right\} \quad (1)$$

where N is a normalization constant and $\lambda = \lambda(E)$ is a Lagrangian multiplier. Normally, one would now proceed to determine λ from the constraint, i. e. the energy dissipation rate. In this case, however, λ is determined by fundamental requirements of statistical mechanics. As shown by Bochkov and Kuzovlev [13, 14], the time-reversal invariance of the microscopic equations of motion implies

$$\frac{P_E[J(t)]}{P_E[-J(-t)]} = \exp \left\{ \beta E \int_{-\infty}^{\infty} J(t) dt \right\} \quad (2)$$

where β is the inverse temperature. Equation (2) is derived by assuming thermal equilibrium at $t=-\infty$ followed by a decoupling of the heat bath and evolution according to the canonical equations of motion for the system interacting with the external field. Alternatively, the heat bath may be thought of as part of the system. These assumptions are identical to those used by Kubo in deriving linear response theory [6]. Equation (2) is also valid for any system which is described by a master equation; in this case Eq. (2) follows from the principle of detailed balance. Since $P_0[J(t)]$ is time-reversal invariant, Eqs. (1) and (2) imply $\lambda = -\beta/2$ for all E , i. e.

$$P_E[J(t)] = N^{-1} P_0[J(t)] \exp\left\{\beta/2 E \int_{-\infty}^{\infty} J(t) dt\right\}. \quad (3)$$

Equation (3) allows one to calculate all higher order cumulant averages of J in the external field, in terms of equilibrium cumulants. The cumulants are defined by the generating functional

$$\begin{aligned} D[u(\tau); E] &= \ln \left\langle \exp\left\{\int_{-\infty}^{\infty} u(\tau) J(\tau) d\tau\right\} \right\rangle_E \\ &= \sum_{n=1}^{\infty} \frac{1}{n!} \int_{-\infty}^{\infty} dt_1 \dots dt_n \langle J(t_1), \dots, J(t_n) \rangle_E u(t_1) \dots u(t_n) \end{aligned} \quad (4)$$

and Eq. (3) implies

$$D[u(\tau); E] = D[u(\tau) + \beta/2 E; 0] - D[\beta/2 E; 0]. \quad (5)$$

From Eq. (5) the nonequilibrium cumulants may be found as derivatives with respect to $u(\tau)$ at $u=0$.

For the average of J one finds from Eq. (5)

$$\langle J \rangle_E = \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{\beta E}{2} \right)^n \int_{-\infty}^{\infty} dt_1 \dots dt_n \langle J(0), J(t_1), \dots, J(t_n) \rangle_0. \quad (6)$$

The first order term is that predicted by linear response theory. Note that, for a macroscopic Q , Eq. (6) is consistent in the thermodynamic limit since cumulants are additive for independently fluctuating quantities. An identity for the

$(n+1)$ 'th cumulant average of $\Delta Q(t) = Q(t) - Q(0)$, $\lim_{t \rightarrow \infty} t^{-1} \langle \Delta Q^{(n+1)}(t) \rangle_0 = \int_{-\infty}^{\infty} dt_1 \dots dt_n \langle J(0), J(t_1), \dots, J(t_n) \rangle_0$, allows one to rewrite Eq. (6) as

$$\langle J \rangle_E = \lim_{t \rightarrow \infty} t^{-1} \frac{d}{du} \ln \langle e^{u \Delta Q(t)} \rangle_0 \Big|_{u = \beta/2 E}. \quad (7)$$

As a simple example of Eq. (7), consider a particle performing a random walk in one dimension on a lattice with lattice constant a . Q is the coordinate and J the velocity of the particle.

If there are N jumps in time t it is easy to see that

$\langle e^{u \Delta Q(t)} \rangle_0 = \cosh^N(ua)$. Since N is Poisson distributed around a mean $\bar{N} = \Gamma t$, one has

$$\langle e^{u \Delta Q(t)} \rangle_0 = e^{-\bar{N}} \sum_{N=0}^{\infty} \frac{\bar{N}^N}{N!} \cosh^N(ua) = e^{\bar{N}(\cosh(ua) - 1)} \quad (8)$$

which implies

$$\langle v \rangle_E = \Gamma a \sinh(\beta/2 E a). \quad (9)$$

This is the correct result if the random walk is realized as the low temperature limit of a particle in a symmetric periodic potential obeying a Langevin equation; the well-known Eq. (9) follows from the fact that the field lowers the barrier for jumps

in the direction of E and increases the barrier for jumps opposite to E . As another example of Eq. (7), consider the case when Q is the x -coordinate of an atom in a liquid. Then Eq. (7) expresses the velocity of the atom in a field in the x -direction acting only on the atom, in terms of the equilibrium incoherent intermediate scattering function $F_s(k, t)$:

$$\langle v \rangle_E = \lim_{t \rightarrow \infty} (it)^{-1} \frac{\partial}{\partial k} \ln F_s(k, t) \Big|_{k = \frac{\beta E}{2i}} \quad (10)$$

For simple gaussian diffusion $F_s(k, t) = \exp(-Dk^2 t)$ [15]. In this case the response is exactly linear and Eq. (10) reduces to the Nernst-Einstein relation between mobility and diffusion constant. A more realistic fit to experiment is the prediction of the jump-diffusion model [15, 16] $F_s(k, t) = \exp[-Dk^2 t / (1 + Dk^2 \tau_0)]$. From this, Eq. (10) predicts the onset of nonlinearity at fields for which $E/\beta \approx 1$ where the length $l = \sqrt{D\tau_0}$. In fits to data l is of order one interatomic spacing.

For the external field autocorrelation function Eq. (5) implies

$$\begin{aligned} \langle J(0), J(t) \rangle_E &= \langle J(0), J(t) \rangle_0 + \frac{\beta E}{2} \int_{-\infty}^{\infty} dt_1 \langle J(0), J(t), J(t_1) \rangle_0 \\ &+ \frac{1}{2} \left(\frac{\beta E}{2} \right)^2 \iint_{-\infty}^{\infty} dt_1 dt_2 \langle J(0), J(t), J(t_1), J(t_2) \rangle_0 + \dots \end{aligned} \quad (11)$$

An example is the case when J is the electric current. For a system with frequency-independent conductivity the zeroth order term is a delta function, giving rise to white noise. In most cases the next term is zero by symmetry and the so-called excess current noise is given by the second order term. Excess current noise is ubiquitous and usually has a $1/f$ spectrum [17-19]. A

number of random walk models have been studied to gain a better understanding of the phenomenon. In these models one finds [20-23]

$$\langle J(0), J(t) \rangle_E = (\beta E)^2 \int_0^t dt_2 \int_0^{t_2} dt_1 \langle J(0), J(t_1), J(t_2), J(t) \rangle_0 + \dots \quad (12)$$

which is equivalent to Eq. (11) because in random walk models $\langle J(t_1), J(t_2), J(t_3), J(t_4) \rangle_0$ is non-zero only if $t_1 \approx t_2$ and $t_3 \approx t_4$ (or permutations thereof) where $t_1 \approx t_2$ means $t_1 - t_2$ is a microscopic time [20, 21].

The similarity between the right hand sides of Eqs. (6) and (11) suggests that a relation exists between $\langle J \rangle_E$ and $\langle J(0), J(t) \rangle_E$. It is easy to show that

$$\frac{d}{dE} \langle J \rangle_E = \frac{\beta}{2} \int_{-\infty}^{\infty} \langle J(0), J(t) \rangle_E dt. \quad (13)$$

Actually, this result is not approximate but a special case of an exact theorem due to Peterson [24].

Since higher than second order cumulants are zero for a gaussian system, the maximum entropy ansatz implies the response is exactly linear whenever the equilibrium J-fluctuations are gaussian. This is analogous to what happens in equilibrium statistical mechanics. A magnetic system, for instance, has a field independent susceptibility only if the free energy is quadratic in the magnetization. Note that nongaussian fluctuations survive in the thermodynamic limit, the central limit theorem notwithstanding. A simple example of this in statistical mechanics is energy fluctuations in the canonical ensemble. If the energy fluctuations are strictly gaussian, the average energy is linear in β so the specific heat varies as -2 . Whenever this is not the case, the energy fluctuations are

nongaussian.

The maximum entropy formalism has a long history of applications to the problem of estimating the density matrix for a nonequilibrium system. This leads to a generalization of equilibrium statistical mechanics which takes into account the limited amount of information available about the system [25-29]. Some applications of the formalism characterize the nonequilibrium system by its energy dissipation rate [30,31]. As shown above, when applied to the external field induced "nonequilibrium" in the $J(t)$ -space, this idea leads to definite predictions of the nonlinear response in terms of equilibrium fluctuations. The resulting ansatz Eq. (5) has been discussed briefly by Bochkov and Kuzovlev [13], referring to earlier work by Stratonovich [10]. Only the case $u(\tau)=\text{const.}$ was considered and the discussion was limited to Markovian systems. No justification of the ansatz was given except that it satisfies the requirement of statistical mechanics Eq. (2). The maximum entropy property of the ansatz, on the other hand, shows that it is the most reasonable, given only a knowledge of the equilibrium fluctuations. This does not guaranty the ansatz is correct, however. To look into this question, we first discuss the consequences of a phenomenological Langevin equation for Q and then an exact result.

Suppose Q obeys a Langevin equation $\dot{Q} = C F(Q) + \zeta(t)$ where $F(Q)=F_0(Q)+E$ is the generalized force and $\zeta(t)$ is a gaussian white noise term. Then the path-probability is given by [32,33]

$$P[Q(t)] = N^{-1} \exp\left\{-\beta/(4C) \int_{-\infty}^{\infty} [\dot{Q}(t) - CF(Q(t))]^2 dt\right\}, \text{ or}$$

$$P_E[Q(t)] = N^{-1} P_0[Q(t)] \exp\left\{\beta/2 E \int_{-\infty}^{\infty} J(t) dt\right\} \exp\left\{-\beta/2 EC \int_{-\infty}^{\infty} F_0(Q(t)) dt\right\}. \quad (14)$$

The transformation from $Q(t)$ to $\dot{Q}(t)$ is linear. Therefore, Eq. (14) implies that the maximum entropy ansatz leads to correct results whenever the last term is unimportant. As an example, consider again a particle in one dimension in a periodic potential at low temperature. Because of the $P_0[Q(t)]$ factor, the important paths fluctuate for some time around a potential energy minimum before suddenly "jumping" to a new minimum. For these paths the last term of Eq. (14) is almost independent of the number of minima traversed and may be ignored for the calculation of $\langle \Delta Q(t) \rangle_E$ for large t . Thus, the maximum entropy ansatz leads to correct results in this case, as already shown in Eq. (9).

A result due to Furukawa sheds more light on the maximum entropy ansatz. He showed that, for slowly fluctuating $J(t)$, the exact microscopic equation of motion implies [34]

$$P_E[J(t)] = N^{-1} P_0[J(t)] \exp \left\{ \beta \frac{1}{2} E \int_{-\infty}^{\infty} J(t) dt + O(E^2) \right\}. \quad (15)$$

Equation (15) shows that there are two different ways a system may become nonlinear: Either 1) as an interplay between nongaussian equilibrium J -fluctuations and the first order term, or 2) as a result of the higher order terms becoming important. The first case is when the maximum entropy ansatz gives correct results; this may be referred to as a "simple" nonlinearity since it results from the very same term in Eq. (15) that gives rise to the linear response. Upon increasing the external field, a system may pass from linear through simple nonlinear response to finally the "complex" non-linear response due to the higher order terms, or it may go directly from the linear to complex nonlinear response. The maximum entropy ansatz is never correct for the

latter systems, and in the former case it should only be trusted at not too large external fields.

To summarize, it has been shown that the maximum entropy formalism in conjunction with statistical mechanics leads to unique predictions for the external field cumulant averages of $J(t)$ in terms of equilibrium averages. The ansatz Eq. (3) may be justified in other ways. It corresponds to keeping only the first order term in Eq. (15) which is exact for slowly fluctuating $J(t)$. This term is responsible for the linear response, so the ansatz is clearly the simplest way to extrapolate linear response theory into the nonlinear regime. The extrapolation has the property that the response is exactly linear if the equilibrium J -fluctuations are gaussian. If Q obeys a Langevin equation, the ansatz is correct whenever the last term of Eq. (14) is unimportant; for slowly fluctuating $J(t)$ the requirement of consistency between Eqs. (14) and (15) implies that the extra terms of both equations must be insignificant and the maximum entropy ansatz must be correct.

The maximum entropy ansatz solves a conceptual problem, but open questions remain regarding the validity and the practical value of the ansatz. One obvious problem is that the ansatz predicts a universal form of the nonlinear response while one expects the response to depend on the way heat generated by the external field is removed. On the other hand, the ansatz only makes sense in situations where the temperature is almost constant throughout the sample (because the temperature enters into Eq. (5)). In bulk cases this requirement can be satisfied for a given external field by working with a sufficiently small sample; in this limit the problem of heating becomes insignificant. -- Equation (15) implies the ansatz can only be

valid at not too large external fields. Also, Eq. (15) implies that a system may become nonlinear either by first exhibiting the "simple" nonlinearity predicted by the ansatz before becoming "complex" nonlinear, or by going directly from the linear to the complex nonlinear regime. In this connection it should be noted that all formulas given above may be generalized to several degrees of freedom. One may speculate that, by including the "mode-coupling" terms to other relevant degrees of freedom, a complex nonlinearity may be converted to a simple, thus extending the validity of the ansatz. -- Even when the maximum entropy ansatz is valid, the practical value of it is not clear at present. It is possible that the calculation of the equilibrium higher order cumulant averages requires at least as much effort as a direct calculation of the nonlinear response. Certainly, this is the case for the above discussed simple random walk in one dimension. On the other hand, Eq. (12) has provided insight into the problem of $1/f$ noise in conductors. And, for instance, Eq. (10) gives explicit predictions for the response of a single atom to an external force, based on a knowledge of the equilibrium intermediate incoherent scattering function, a quantity which has been studied extensively [15].

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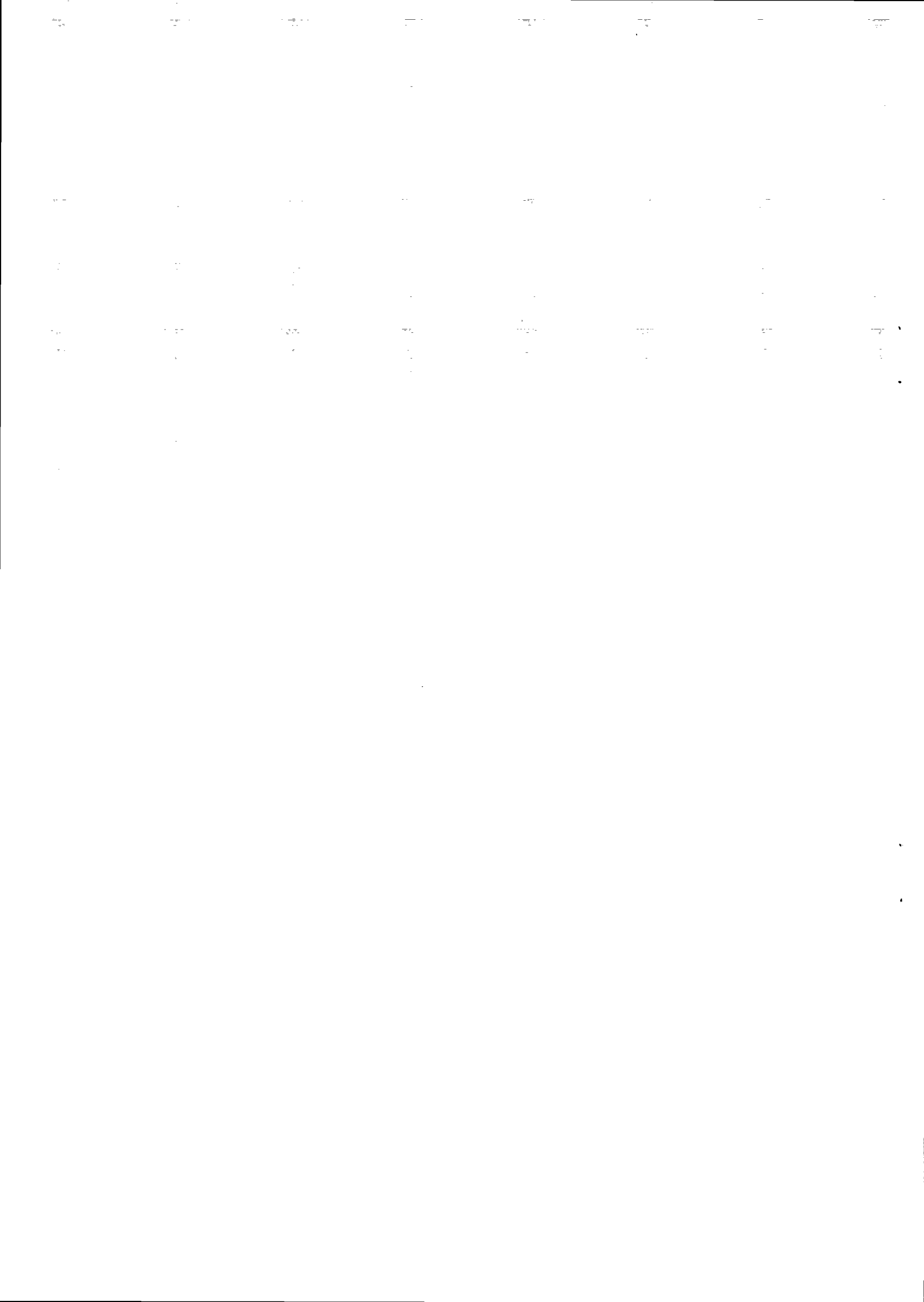
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