Stiff-Glass Approximation of Mode-Coupling Theory

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The stiff-glass approximation of the mode-coupling theory of the glass transition—as introduced in a recent paper by Götze and Mayr for a discussion of phenomena resembling the Boson Peak and High-Frequency Sound observations made in real glass-formers—is examined in detail. It amounts to a neglect of two-phonon processes and thus provides a clear physical picture for the MCT solutions deep in the glass. In addition, the effect of additional simplifying approximations and the combination of these approximations is studied.

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I. INTRODUCTION

In a recent paper, Götze and Mayr demonstrated that the mode-coupling theory for the glass transition (MCT) applied to a model of the hard-sphere system (HSS) at relatively high packing fractions yields spectra for the density correlation functions, that display a broad asymmetric peak as well as a sound-like mode at rather high wave vectors. Similar observations have been made in real glass formers and are known as the Boson Peak (BP) and high-frequency sound (HFS), respectively. To investigate these phenomena in detail, a large number of experiments has been performed, using various techniques such as Raman spectroscopy, x-ray spectroscopy, neutron scattering, or molecular-dynamics simulations. For an account of the literature, the reader is referred to the papers cited in [1]. This work also contains a summary of the basic equations and approximations, one arrives at an equation that, while it offers some conclusions. The paper is organized as follows: Section II will present a summary of the basic equations and approximations outlined above. In Sec. III, the solutions of the SGA equations with the full q dependence taken into account will be presented. Section IV examines the effect of the GHD and schematic approximations, and Sec. V offers some conclusions.

II. EQUATIONS OF MOTION

A. Mode-Coupling Theory

The mode-coupling theory of the glass transition (MCT) describes the dynamics of a glass-forming liquid in terms of the density correlators \( \Phi_q(t) = \langle \varphi_q^*(t) \varphi_q \rangle \). Here, \( \varphi_q = \frac{1}{N} \sum_j \exp(iq \cdot r_j) / \sqrt{N} \) are the density fluctuations of wave vector \( q \), for a system of \( N \) particles at density \( \rho \). \( \langle \ldots \rangle \) denotes canonical averaging. For amorphous systems, \( \Phi_q(t) \) depends on the wave-vector only through its modulus \( q = |q| \). At short times, one has \( \Phi_q(t) = S_q - \frac{1}{2} S_q Q_2 t^2 + \ldots \), where \( S_q = \langle |\varphi_q|^2 \rangle \) is the static structure factor specifying the equilibrium structure of the system. \( Q_2 = v^2 q^2 / S_q \) are the phonon frequencies. A system of hard spheres shall be considered. The units of time and length are chosen in accordance with [4] such that the thermal velocity \( v = 2.5 \) and the sphere diameter \( d = 1 \).

The normalized density correlators \( \phi_q(t) \), given by \( \Phi_q(t) = S_q \phi_q(t) \), obey the equations of motion obtained via a Mori-Zwanzig projection operator formalism,

\[
\ddot{\phi}_q(t) + \Omega_q^2 \phi_q(t) + \Omega_q^2 \int_0^t m_q(t-t') \dot{\phi}_q(t') dt' = 0.
\]  

(1a)

Here \( m_q(t) \) is the memory kernel, a correlation function of fluctuating forces. If one introduces Fourier-Laplace transformed quantities with the convention \( \phi_q(z) =
where the fluctuation dissipation theorem has been used to connect $\phi_q(z)$ to the dynamical susceptibility $\chi_q(z)$: $\chi_q(z)/\chi^0_q = z\phi_q(z) + 1$, with $\chi^0_q$ denoting the isothermal compressibility.

Within MCT, the memory kernel $m_q(t)$ is written as the sum of a regular contribution and a so-called mode-coupling contribution, the latter describing the slow dynamics important close to the glass transition. For details, the reader is referred to the original publications \[2, 3\] and standard literature \[4\]. One gets for $m_q(t)$ a quadratic functional in $\phi(t)$, $m_q^{\text{MCT}}(t) = F_q[\phi]$,

$$m_q^{\text{MCT}}(t) = \int_{\vec{q} = \vec{k} - \vec{p}} \frac{d^3k}{(2\pi)^3} V(\vec{q}\vec{k}\vec{p}) \phi_k(t) \phi_p(t),$$  

with coupling coefficients given by the equilibrium structure of the system,

$$V(\vec{q}\vec{k}\vec{p}) = \rho S_q S_k S_p[(\vec{q}\vec{k})c_k + (\vec{q}\vec{p})c_p]^2/(2q^4).$$

Here $c_q$ denotes the Ornstein-Zernike direct correlation function, connected to the static structure factor by $S_q = 1/(1 - q c_q)$. If one introduces a wave-vector grid and approximates the integral as a Riemann sum, the mode-coupling functional can be written as $F_q[\phi] = \sum_{kp} V_{qkp} f_k f_p$, where $k$ and $p$ run over the set of all grid points. The coefficients $V_{qkp}$ are trivially related to the $V(\vec{q}\vec{k}\vec{p})$.

For the discussion of the MCT dynamics in the boson-peak region, Götze and Mayr \[1\] chose a model of the hard-sphere system, given by evaluating $S_q$ in the Percus-Yevick approximation, introducing a grid of $M = 300$ wave vectors with a cutoff of $q^* = 40$, and by neglecting the regular part of the memory kernel. The dependence of the solutions on these approximations will have to be investigated separately. For the structure factor used, a cutoff dependence of the solutions cannot be neglected, as was already mentioned in \[1\], and one is lead to the conclusion that the model described here is one of a soft-core fluid rather than true hard spheres. Even more so, the influence of the regular term is unclear at present. Recent calculations indicate that, at least in the white-noise approximation based on a generalized Enskog theory, the regular damping might be so large as to render the BP part of the spectrum nonexistent for a true hard-sphere system \[3\]. The question, however, remains unanswered still for regular potentials. Nevertheless, since the concern of this paper is a comparison with the results presented in \[2\], we shall choose all numerical parameters the same. Equations \[1\] and \[2\] are closed then, and since $S_q$ for the hard-sphere system does not depend on temperature, the only control parameter is the density $\rho$, which shall be written as the packing fraction $\varphi = \pi \rho/6$.

### B. Stiff-Glass Approximation

The starting point for a theoretical understanding of the MCT high-frequency dynamics is a reformulation of the equations of motion, Eqs. \[1\], within the glass state. One introduces new correlators $\phi_q(t)$ that only deal with the decay relative to the frozen structure given by the nonergodicity parameter $f_q$. $\phi_q(t) = f_q + (1 - f_q)\phi_q(t)$. Equations \[1\] are covariant in the sense that the same equations hold for the new variables,

$$\chi_q(z)/\chi^0_q = -\hat{\Omega}^2_q/\left[z^2 - \hat{\Omega}^2_q(1 - z \hat{m}(z))\right],$$

provided one replaces the static susceptibility by $\hat{\chi}^0_q = \chi^0_q(1 - f_q)$, the frequencies $\hat{\Omega}^2_q$ by

$$\hat{\Omega}^2_q = \Omega^2_q/(1 - f_q),$$

and the memory kernel by

$$\hat{m}_q(t) = F^{(1)}_q[\hat{\phi}] = \hat{F}^{(1)}_q[\hat{\phi}] + \hat{F}^{(2)}_q[\hat{\phi}],$$

$$\hat{F}^{(1)}_q[\hat{f}] = \sum_k \hat{V}_{qkp} \hat{f}_k,$$

$$\hat{F}^{(2)}_q[\hat{f}] = \sum_{kp} \hat{V}_{qkp} \hat{f}_k \hat{f}_p,$$

with new coupling coefficients given by

$$\hat{V}_{qk} = 2(1 - f_q) \sum_p V_{qkp} f_p(1 - f_k),$$

$$\hat{V}_{qkp} = (1 - f_q) V_{qkp}(1 - f_k)(1 - f_p).$$

The new variables $\hat{\phi}_q(t)$ and $\hat{m}_q(t)$ have the properties that their long-time limits vanish, thus the low-frequency limits of their Laplace transforms are regular.

To study BP phenomena, the limit of high packing fraction is of interest. There the original coupling coefficients become large, formally written $c_q \to \infty$, the frequencies $\Omega^2_q$ and $\hat{\Omega}^2_q$ run over the set of all grid

\begin{equation} \label{eq:stiffglass}
\hat{\phi}^{(1)}_q(z) = -1/\left[z - \hat{\Omega}^2_q/\left[z + \hat{\Omega}^2_q \hat{m}_q^{(1)}(z)\right]\right],
\end{equation}

\begin{equation} \label{eq:stiffglass2}
\hat{m}_q^{(1)}(z) = \sum_k \hat{V}_{qk} \hat{\phi}_k^{(1)}(z).
\end{equation}

Equations similar to Eq. \[8\] were derived in a different context by other authors. Let us, for example, rewrite these equations in a notation closer to the conventions of
field theory. The dynamical susceptibility can be rewritten for positive frequencies $z = \omega^2$, $\omega > 0$, into a propagator $G(q, \omega) = -i\chi_0^G(\sqrt{z}/(\chi_0^G_2)^{1/2})$, depending on the bare dispersion relation $c(q) = \Omega^2_0(1 + \sum_k \tilde{V}_{qk})$, and on the complex self energy $\Sigma(p, \omega)\), to give

$$G(q, \omega) = \frac{1}{\omega - \epsilon(q) - \Sigma(q, \omega)},$$

$$\Sigma(q, \omega) = \int dk \tilde{V}_{qk}G(k, \omega),$$

where $\tilde{V}_{qk} = \tilde{\Omega}^2_2\tilde{\Omega}_2^G$ are positive coupling constants. This is equivalent to the equations discussed by Martín-Mayor et al. and Grieger et al. in connection with Euclidean random matrix theory, cf. Eqs. (13) and (17) of the latter paper, whereas, however, the vertices $\tilde{V}_{qk}$ are different. Equations (9) can be viewed as a special case of the Dyson equation.

Equations (9) still have to be solved numerically. Thus, Götte and Mayr proposed two additional simplifications, the first of which is the Generalized Hydrodynamic (GHD) description. One replaces the memory kernel $\tilde{m}_q^{(1)}(z)$ by its $(q \to 0)$ limit, $K(z) = \tilde{m}_q^{(1)}$. If one is only interested in a description of sound modes at small frequencies below the BP frequency resembling this finding, the smearing out of the gap due to two-phonon processes can be understood on the basis of the schematic model by including these processes in a perturbative manner, which amounts to replacing $K(\omega)$ in Eq. (11a) with $K^{(2)}(\omega) = K(\omega) + K^{(2)}(\omega)$, where the latter term reads

$$K^{(2)}(\omega) = \frac{1}{\pi} \int \frac{d\omega'}{\omega'} \tilde{K}(\omega - \omega') \tilde{K}'(\omega') d\omega',$$

with the integrated second-order coupling coefficient $w_2 = \int dk \tilde{V}_{0kk}$. $K^{(2)}(\omega)$ is then still given by Eq. (12), but one has to set $\tilde{\omega}^2 = \omega^2/\tilde{\Omega}^2 + K^{(2)}(\omega)$. Analytic solvability can be regained by evaluating this in the $\omega \to 0$ limit, $K^{(2)}(\omega) \to 0$. The full solutions show a pseudo-gap for $0 < \omega < \omega_\ast$. The full solutions have striking similarities to the spectra of the full MCT solutions in the BP regime at $\varphi = 0.6$.

Eq. (12) implies a “gap” in the kernel spectrum for $0 < \omega < \omega_\ast$. The full solutions show a pseudo-gap for frequencies below the BP frequency resembling this finding. Since arriving at Eq. (12) involved three subsequent approximations which, in principle, could be applied independently from one another, it seems worthwhile to discuss the effects of each approximation by itself. Especially going over to the schematic model description of the memory kernel as a last step is, in contrast to the GHD-SGA, uncontrolled in the sense that its derivation is not based on a well-defined mathematical procedure.

The GHD approximation was shown in [9] to be of very good quality, provided on does apply it to the transformed memory kernel $\tilde{m}_q(z)$. In other words, keeping the full $q$-dependence of the nonergodicity parameters $f_q$ is of vital importance for a description of the BP and HFS spectra. It can thus be expected to be of similar quality when applied together with the SGA.

While not leading to analytically solvable expressions, the SGA alone, Eqs. (8), has a clear-cut physical meaning: The first-order contribution to the memory kernel can be interpreted as an elastic scattering of phonons due to the frozen disorder in the glass, a mechanism thought to be dominant over two-phonon decay processes, which are represented by the second-order term and therefore ignored in the SGA. It should be noted, however, that this does not correspond to a harmonic approximation.

### III. Solution of the SGA Equations

To obtain numerical solutions of Eqs. (8), an algorithm adapted from the one used in [1] was employed. It solves the initial-value problem for the coupled integro-differential equations corresponding to Eqs. (8) in the time domain. Regarding the stability of this method, proofs are available [4], which apply to the SGA as a
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tions 200
m
ory kernel spectra \( \hat{m}^{''}(\omega) \).

Let us start the discussion by a comparison of the mem-
ory kernel spectra \( \hat{m}^{''}(\omega) \), obtained from the solution of the
full MCT equations, Eq. (3), and of the SGA equa-
tions, Eq. (8). Figure 1 shows such a comparison for
packing fraction \( \varphi = 0.6 \). One notes a dominant broad
and asymmetric peak responsible for the BP features of
the memory kernel spectra. The SGA solutions still re-
cover the shape of that peak reasonably well; an obser-
ration together with its form being reminiscent of the
semi-ellipse known from the schematic model further just-
ifying the analogy put forward by the schematic model
description. At frequencies \( \omega \lesssim 40 \), a pseudo-gap can be
seen. As discussed in \( \[ \), the SGA spectrum is not ex-
actly zero within this gap; rather one obtains Rayleigh’s
law, \( \hat{m}^{''}(\omega) = R_\omega \omega^2 + O(\omega^3) \). The prefactor of the
Rayleigh contribution is, however, so small that it is in-
visible in the plot shown. Comparing with the full MCT
solution, one clearly notes the filling of the gap due to
the two-phonon modes.

On the high-frequency wing, the spectra obtained from
the full solutions show a nontrivial decay to zero, includ-
ing, for larger \( q \) vectors, a second small peak at frequen-
cies 200 \( \lesssim \omega \lesssim 250 \). Interestingly enough, this peak is
also reproduced by the SGA memory kernels.

Since the SGA reproduces the memory kernel of the
full MCT solutions resonnably well in the frequency range
of importance for the discussion of BP-like phenomena, it
comes as no surprise that it also gives a basically correct
description of the density correlator spectra \( \Phi^{''}(\omega) \). Figure 2
comparisons these spectra to those of the full MCT
equations. The BP spectra are, with some quantitative
differences, largely correct within the SGA, and the same
holds for the HFS modes for those \( q \) vectors, where the
sound damping is dominated by the BP background. At

\( q \sim q_D \), where \( q_D \approx 4 \) is the Debye frequency \( \[ \), the SGA
sound-mode damping clearly is too small. This failure is
to be expected from the above discussion of the memory
kernel, since in this case the sound resonance maximum
occurs at frequencies above the BP spectrum, where the
SGA memory kernel spectrum is too small. Similarly,
the pseudo-gap at low frequencies and the rapid decay at
large \( \omega \) within the SGA are a result of the same features
of the memory kernel spectra.

The SGA is based on the two-mode contributions of
order \( O(\eta^2) \) being negligible in comparison to one-mode
contributions of order \( O(\eta) \). The quality of this approxi-
mation is expected to decrease with lowering the packing
fraction, since \( \eta \) increases with decreasing \( \varphi \). Figures 3
and 2 repeat the comparison made in Fig. 1, now for
packing fractions \( \varphi = 0.5676 \) and \( \varphi = 0.54 \), respectively.
One can identify the increasing intensity for low frequen-
cies, eventually becoming so strong as to dominate over
the BP background for \( \varphi = 0.54 \). Nevertheless, the SGA
gives the qualitatively correct picture even for \( \varphi = 0.54 \)
at frequencies \( \omega \gtrsim 25 \), the number depending somewhat
on the wave vector. One also notices the shift of both
the sound resonance and the BP maximum to lower fre-
quencies as the packing fraction decreases. This further
demonstrates two features typical for the BP and the
HFS \( \[ \).
One notes that for $\varphi = 0.54$ and $\varphi = 0.5676$ the true HFS maximum positions are higher than those read off the SGA solutions, especially for $q$ vectors where the SGA significantly underestimates the damping. This effect is also known as level repulsion. For a discussion of the effects of the SGA on the HFS modes, Figs. 3 and 5 show the maximum positions of the spectra $\phi_q''(\omega)$ and the resonance widths versus wave vector $q$ for different packing fractions. Two maximum positions are noted whenever the BP background and the HFS mode could be identified as distinct, separated by a spectral minimum. The full widths at half maximum (FWHM) were obtained by marking the lowest and the highest frequency where the correlator spectrum reaches half the value of its global maximum.

For the HFS maxima as well as the BP maxima at $\varphi = 0.6$, Fig. 5, good agreement between the full solutions, reproduced from [1], and the SGA solutions is found. This even holds in the low-$q$ regime, where only a damped sound mode can be seen in the spectra. Here the shift in the maximum position due to damping is still small, such that an underestimation of the damping by the SGA does not necessarily lead to a wrong description of the position. The agreement of the FWHM values also is good, with the reservation that the SGA values are too small in general. The lower frequency for which the spectrum reaches half the maximum intensity is described better than the higher one within the SGA, since here the pseudo-gap marks the dominant rise in the spectrum.

Comparing the maximum positions and FWHM at $\varphi = 0.5676$, one notes that for the BP maximum, the agreement between full MCT and SGA still is very good. Expectedly, the description of the FWHM is worse, mostly due to the appearance of the pseudo-gap and due to the underestimation of the sound-mode damping around $q_D$.

The sound damping in the regime of the HFS is illuminated in more detail in Fig. 7. Here, full width at half maximum values $\Gamma_q$ obtained from the full solutions of Eq. (3) and from within the SGA, Eq. (8), are compared for the two packing fractions $\varphi = 0.6$ and $\varphi = 0.5676$. For $q \leq 0.6$ (0.467) at $\varphi = 0.6$ (0.5676), the SGA line widths could not be determined reliably. This reflects a limitation of the numerical procedure used, which is not adequate for very weakly damped oscillations, as occurring within the SGA. Nevertheless, the errors introduced by the numerical algorithm for low $q$ do not significantly affect the accuracy of the solutions for larger $q$, since the small-$q$ contribution to the memory kernel is negligible.

The dashed lines in Fig. 7 show the low-$q$ asymp-
In the $q$ range from $q \approx 0.6$ to $q \approx 1.4$, the data for $\varphi = 0.6$ can be approximately described by some power law, with its exponent being close to 4. To demonstrate this, the dotted line was included in Fig. 3 representing a $q^4$ law. While in some investigations, no indication of such a $q^4$ law was found [14], it has been argued that this is behaviour is connected to Rayleigh’s law [15]. In the present case, however, the approximate $q^4$ power law is not an indication for Rayleigh scattering. As already noted in Fig. 2, the Rayleigh contribution $R_0$ is much lower. It is included in Fig. 3 as the dot-dashed line. Note that a magnification by a factor of 1000 was necessary to make this contribution visible in the plot. Thus one concludes, that the $q^4$ law for the high-frequency sound is a mere numerical coincidence. The possible appearance of a pseudo-power law for the HFS line width can be seen more clearly for $\varphi = 0.5676$, where an exponent between 2 and 4 would be obtained.

Fig. 3 again demonstrates the validity of the SGA. At and below $q \approx 0.6$, deviations are to be expected, since the SGA line widths do not obey the asymptotic $q^2$ law. Rather one expects the SGA damping for $q \to 0$ to be given by Rayleigh’s law noted above. Despite this fact, the overall description of $\Gamma_q$ by the SGA is quite good.

IV. EFFECT OF ADDITIONAL APPROXIMATIONS

Having discussed the quality of the SGA in comparison to a full MCT solution, we shall now turn to a discussion of the additional simplifications outlined in the introduction. Since the $q$ dependence of the memory kernel spectra is weak, cf. Fig. 1, one is lead to the combination of both the GHD approximation and the SGA. Figure 8 shows a comparison of thus obtained density-correlator spectra with selected full-MCT and SGA spectra reproduced from Fig. 2. Indeed, good agreement is found for the experimentally relevant $q$-vector range at $q \lesssim q_0$.

The GHD-SGA solution fails to produce reasonable sound modes for wave vectors where the position of the sound maximum is at about the upper cutoff for the BP memory kernel. This is due to the sensitivity of the shape of the memory kernel in this frequency region. As noted in connection with Fig. 1, the SGA still reproduces the nontrivial second peak in $m^0_q(\omega)$ also seen in the full MCT solutions. Since this peak only evolves for higher $q$ vectors, the GHD approximation obviously misses this feature, resulting in only weakly damped sound modes there. Still, the GHD-SGA combination is valuable for a discussion of BP-like spectra and the hybridization effects occurring between HFS modes and the BP spectra. Note that the maximum positions obtained from the GHD-SGA description show a qualitatively correct behavior. This can be seen from Figs. 3 and 3. Within the hybridization regime, the FWHM values (not shown in the figures) also are almost quantitatively correct.

A comparison between the full solutions and those ob-
obtained by applying the GHD only, still including two-phonon modes, was given in Fig. 6 of [1].

There it can be seen that the GHD approximation is, for \( q \lesssim q_D \), at least as good as the SGA, at the expense of still requiring the computation of a nonlinear memory kernel. Only the combination of both SGA and GHD seems to be too crude to deal with high-frequency sound modes for all wave vectors. For higher \( q \) vectors, the GHD-only approximation of [1] and consequently also the GHD-SGA description somewhat miss the shape of the BP background spectra, as is evident from the lower panel of Fig. 8. On the other hand, the SGA discussed here regains better quality for those \( q \)-vector regions where the HFS mode again shows hybridization with the BP-like spectrum. This shows that in this region, one-mode contributions from higher \( q \) vectors are important to fully explain the shape of the BP phenomena.

As a last step, one can also compare the memory kernel spectra resulting from the SGA in the \( q \to 0 \) limit with the schematic model description proposed by Götzte and Mayr, Eqs. (11). Fig. 13 of [1] presented a comparison at \( \varphi = 0.6 \) between the schematic model semi-ellipse and the full-MCT kernel spectrum at \( q = 0 \). Let us extend the discussion by adding the corresponding SGA spectrum. This is done in Fig. 9, while Figs. 10 and 11 carry the same comparison to lower packing fractions, \( \varphi = 0.5676 \) and \( \varphi = 0.54 \), respectively. The averaged frequency was chosen to be \( \tilde{\Omega} = 120 \) for \( \varphi = 0.6 \), in accordance with [1], and \( \tilde{\Omega} = 85 \) (60) for \( \varphi = 0.5676 \) (0.54). One could improve on these values, but this would not give further physical insight, nor would it significantly alter the results discussed below.

The success of the semi-ellipse description at \( \varphi = 0.6 \) is based largely on the fact, that in the relevant \( q \) range, the variations of \( \tilde{\Omega} \) are suppressed as \( \eta \) becomes small. For lower packing fractions, the relative variations in \( \tilde{\Omega} \) are larger. Together with the increasing two-mode contributions, this leads to stronger deviations of the full-MCT spectra from the schematic semi-ellipse form.

As explained above, the two-mode contributions can within the schematic description be accounted for approximately by including a frequency-independent damp-
The stiff-glass approximation (SGA) to the mode-coupling theory of the glass transition (MCT) was discussed and shown to provide a good description of the spectra in the boson-peak regime. It holds valid even for packing fractions lower than \( \varphi = 0.6 \), where the analytical formula of Götte and Mayr [1]. Eq. (12), cannot describe the memory kernel any longer. This failure of Eq. (12) is due to increasing variations with \( q \) in the phonon dispersion \( \hat{\Omega}_q \), which in the schematic model is neglected. The shape of the SGA memory kernel shows more structure than a simple semi-ellipse form. This leads to a substantially correct description of high-frequency sound and sound damping in this approximation.

Even the combination of the SGA with the generalized hydrodynamic description (GHD) gives reasonable results for the memory kernels as well as for the density correlators, for \( q \) vectors where the damping of the sound mode is dominated by contributions from the BP-like memory kernel. Outside this window, one gets weakly damped sound waves, an artifact introduced in the intermediate \( q \) range mainly by the GHD, not the SGA. Nevertheless, the GHD-SGA gives a basically correct description of the BP spectra. This finding further corroborates the explanation of the HFS as a result of the BP spectra.

As a side effect, it was demonstrated, that equations of the form of Eqs. (8) can be solved relatively easy in the time domain. At first glance, this seems paradox, since in Eq. (8), both \( q \) and frequency \( \omega \) only appear as parameters, while in the time domain one has to solve a convolution integral.

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